Thermally altered coal from Upper Silesian Coal Basin

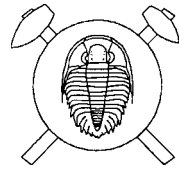## Tepelně alterované černé uhlí z Hornoslezské černouhelné pánve (Czech summary)

(*10 text-figs*)

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The paper is devoted to Professor Miloslav Dopita on the occasion of his 70th birth day

Chemical composition of organic matter of coal, minerals and chemical composition of inorganic elements of coal were studied from 33 coal samples of coal seam 512(39), where thermally altered coal are present. Volatile matter and combustion heat were determined from other 300 samples from coal seams 530(38), 512(39), and 504(40). Using this data the process of coal alteration was interpreted. The coal seams 530(38) and 504(40) were altered at low temperature conditions while coal seam 512(39) was altered at high temperature condition. Also coal altered both thermally and by oxidation was found in this coal seam. It is supposed that source of heat was coal oxidation (undersurface combustion) of a part of coal seams 512(39) deposited in space of present red beds body which burned out.

Key words: coal, red beds, oxidation, thermal alteration

Introduction

Variegated (red) beds in Upper Silesian Coal Basin (USCB) and altered coal seams deposited in the vicinity to the red beds bodies were described before (e. g. Dopita et al. 1997). Recently Klika (1999) studied coal altered by oxidation at low temperature. Except oxidative altered coal the coal altered at high temperatures and at different oxidative condition is present in USCB. According to tentative classification the altered coals have been divided into eight subtypes based on the relative grade of thermal and oxidative alteration (Klika – Kraussová 1993). They are:

I[*]   : Nonaltered coal
I   : zero–low oxidative and zero–low thermal alteration
II/1 : zero–low oxidative and high thermal alteration
II/2 : zero–low oxidative and the highest thermal alteration
III/1 : medium oxidative and zero–low thermal alteration
III/2 : medium oxidative and low–medium thermal alteration
IV/1 : high oxidative and zero–low thermal alteration
IV/2 : high oxidative and medium thermal alteration

While the coal altered at low temperature (subtypes I, III/1 and IV/1) is abundant, the medium and high thermal alteration grade subtypes III/2, IV/2, II/1 and II/2 are rarely found in USCB. In this paper thermally altered coal from Lazy Mine and its characteristics of organic and inorganic matter are presented.

Locality, samples and methods

In the Lazy Mine the red beds body follows the contours of the Sedlové Members which are of Namurian B age (Dopita – Kumpera 1993, Dopita et al. 1997). The red beds body reaches from the Carboniferous surface through a rock environment deeply into the Carboniferous massif (Fig. 1). The coal seams are denoted according

to correlation tables presented by Dopita et al. (1997). The Sedlové Members are composed mainly of coarse grained clastic sediments with a lot of tectonic faults. Therefore they have high permeability and enable the intensive oxidation of rocks and coal seams.

The same tectonic disturbances as illustrated in Fig. 2 for coal seam 512(39) are present in coal seams 530(38) and 504(40). At present the paleorelief of Carboniferous is covered by Miocene calcareous clays.

In Lazy Mine easterly from the overturned flank of the Orlová structure the coal seams pass gradually into the red beds body (Fig. 1). In contact with and in the vicinity of this red beds body, the altered coals were formed. A reduction in thickness of coal seams is observed when they pass into the red beds body. After a distance of some meters, the coal seams completely pinch out (Fig. 3.). Beyond the red beds body the coal seams onset and they have again their original thickness (Fig. 4.). The thickness of nonaltered coal seam 530(38) is usually from 4 to 5 meters and it is deposited about 300 meters or more below the Carboniferous paleorelief. Coal seam 512(39) is 10 meters below coal seam 530(38) and its thickness in the nonaltered region is from 5 to 6 meters. Coal seam 504(40) lies 50 meters below coal seam 512(39) and it is partly incised. The thickness of the not denudated part of coal seam is from 1 to 3 meters. In time of coal sampling the coal seam 538(37) was already extracted and closed.

About 100 samples of coal (in an approximately regular lattice) were taken from each coal seam in the vicinity of the red beds body. The volatile matter and combustion heat were determined from these samples. Moreover 33 coal samples were also taken from 512(39) coal seam (coal corridor 39706) in the vicinity of the red beds body (Figs 4 and 5). The coal samples were taken from 18 profiles. The distance between them is approx. 2–5 meters. On each profile 1 to 3 samples were taken. The sample of nonaltered coal was taken from the same coal seam from coal corridor 39313, approximately 1 km of red beds body. Samples from coal corridor 39706

NW   SE
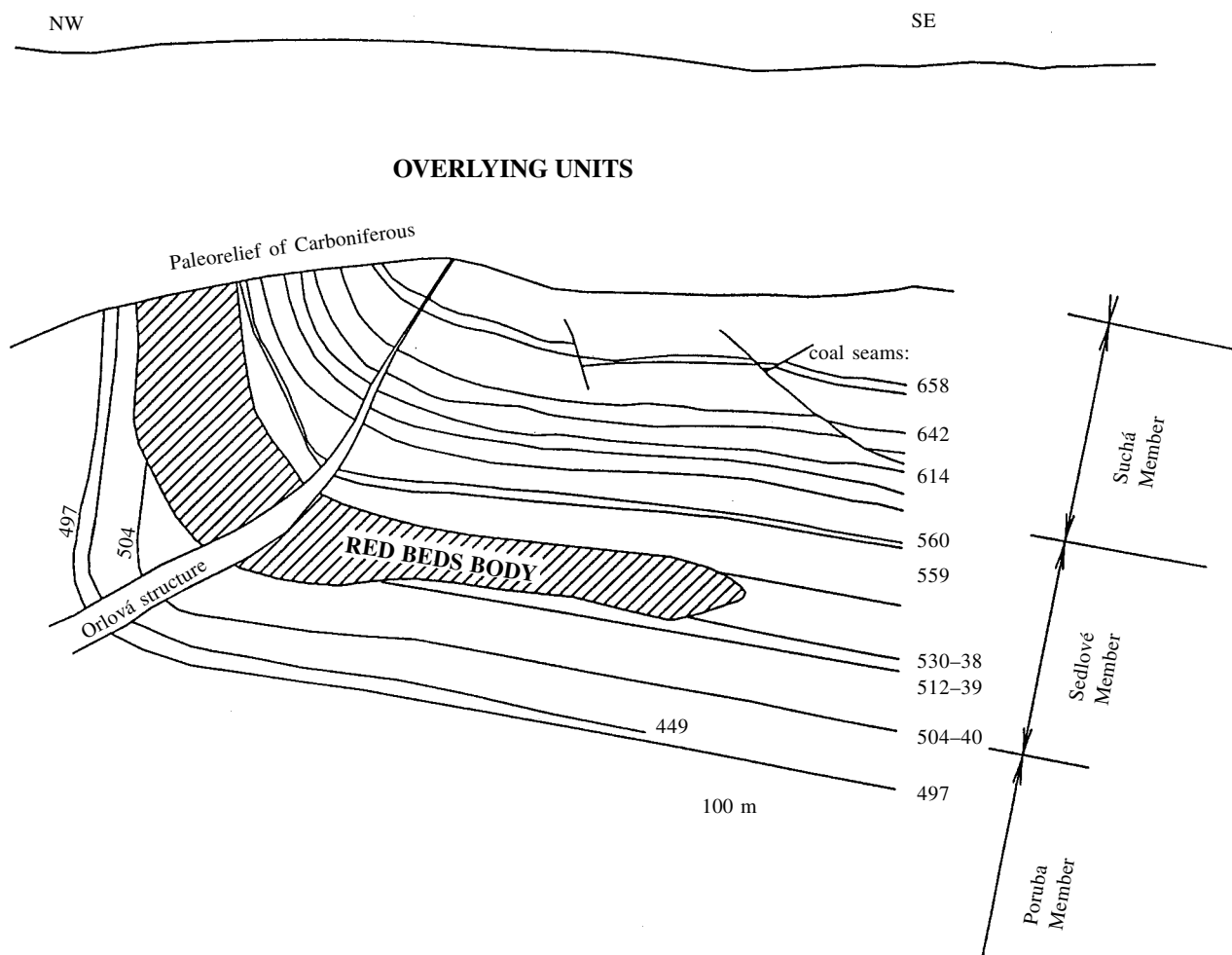
OVERLYING UNITS

Fig. 1. Schematic cross-section of red beds body in coal-bearing strata in Lazy Mine.

(Fig. 4) have been altered by intensive thermal and oxidative processes which also reduced the thickness of the coal seam. The original nonaltered coal seam 5 meters thick was reduced between profiles VIII and IX to approx. 0.8 meter (Fig. 5).

The methods used for coal and ash samples preparation as well as those for mineral, major and trace elements determination and methods used for characterization of organic matter of coal were described previously (Klika 1999). As parameters characterizing coal samples or their organic matter the following are used: the contents of ash A^d (wt %), moisture W^a (wt %), volatile matter V^{daf} (wt %) and humic acids HA^{daf} (wt %), combustion heat Q^{daf} (MJ/kg), mean reflectance of vitrinite (R_o) and concentration of elements C^{at}, H^{at}, O^{at}, N^{at}, S^{at} (all in atom. %) and Meffert's index of weathering defined as $MI=[(O+N)/H]^{at}$ (Stach et al. 1982).

Results

Alteration of coal

The thermal alteration of coal seams in proximity to the red beds was traced using the volatile matter (V^{daf}) as a characteristic parameter. Generally, coal altered by oxidation at lower thermal conditions shows increased values of V^{daf} (e. g. subtypes III/1 and IV/1 in Doubrava Mine have mean V^{daf} approx. 36 wt %) while thermally altered coal has much lower V^{daf} values (e. g. coal types II has V^{daf} even 5 wt %, Klika – Kraussová 1993). In Lazy Mine, in proximity to the red beds body according to V^{daf} values, all altered coal types and subtypes coexist (Fig. 6). The isolines of V^{daf} are plotted in Fig. 6A for the 530(38)-th, in Fig. 6B for the 512(39)-th and in Fig. 6C for the 504(40)-th coal seams respectively. In coal seam 530(38), the values of V^{daf} range from 29 to 32 wt %; in coal seam 512(39), from 5 to 32 wt %; and in coal seam 504(40), from 24 to 36 wt %. In coal seam 512(39), where the biggest difference in thermal alterationn is observed, the V^{daf} isolines are approximately parallel to the red beds body. Outside the red beds body, the values of V^{daf} increase, i. e., intensity of thermal alteration is gradually decreasing. Coal type II is located in the vicinity of the red beds body while at a distance greater than approximately 500 meters the nonaltered coal type I* is present. According to chemical characteristics, the altered coal samples in the locality of coal seam 512(39) were classified into various coal subtypes. Their distribution
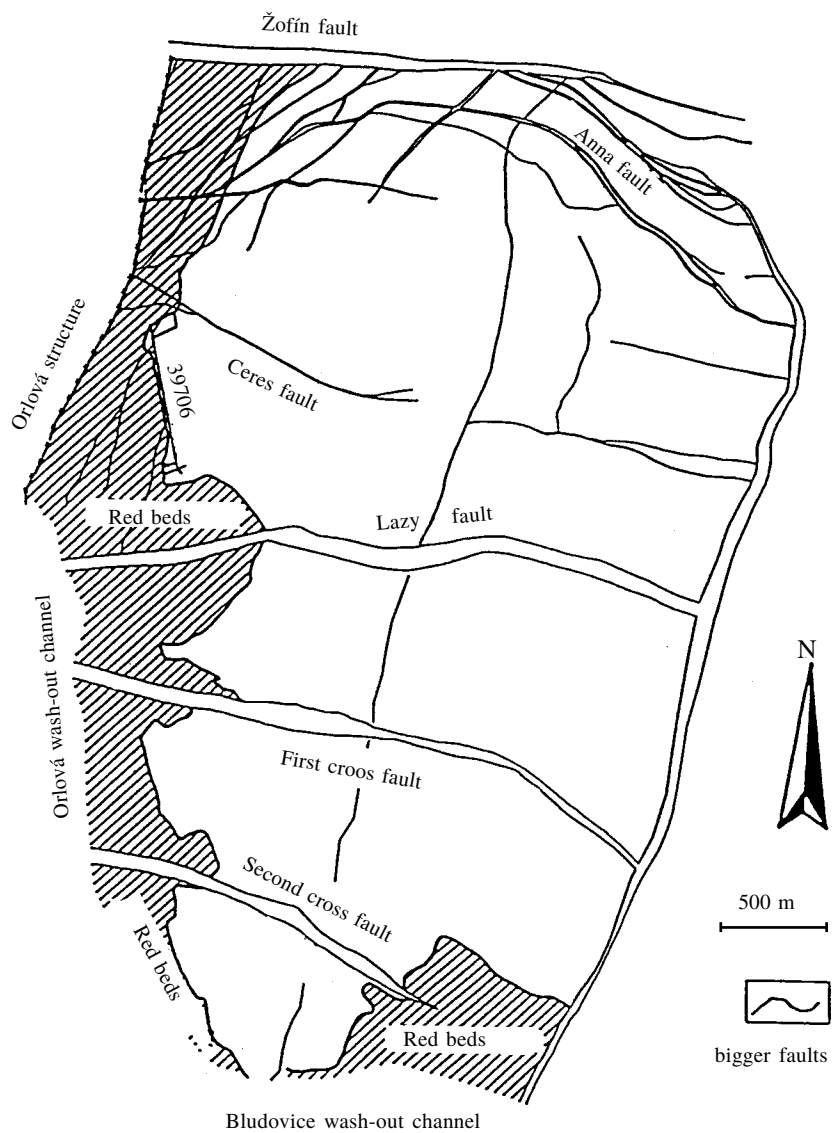
Fig. 2. Scheme of faults linked with red beds body
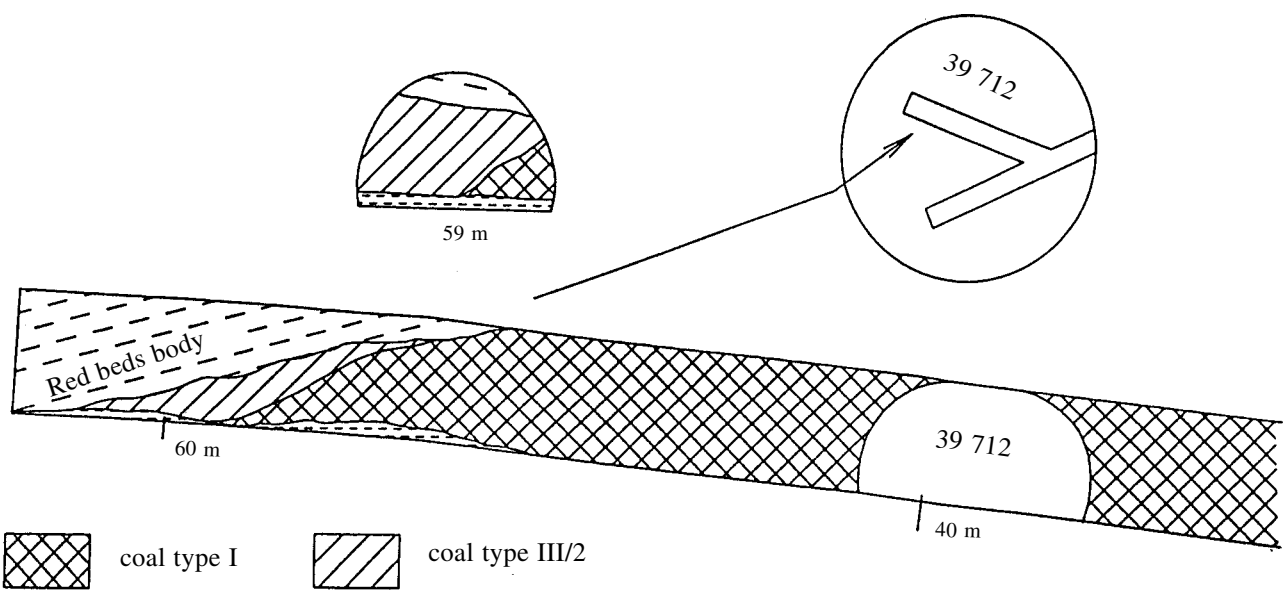in coal seam 512(39).

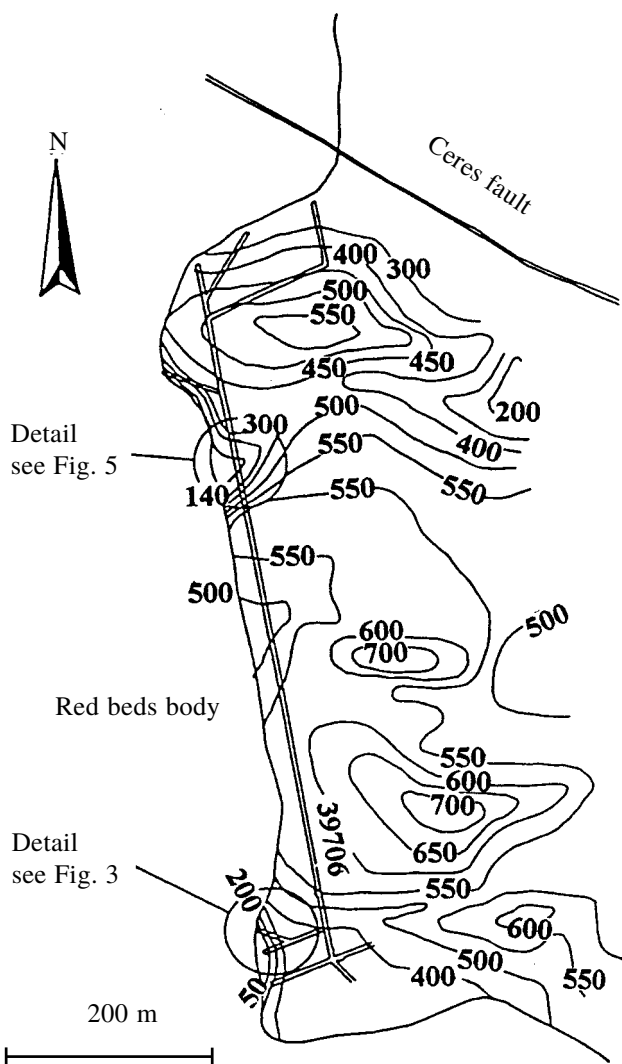Fig. 3. Tailing off the coal corridor 39712.

Fig. 4. Thickness (cm) of the coal seam No. 512(39) and denotation of coal corridors 39706 (detail in Fig. 5) and 39712 (detail in Fig. 3).

at a studied locality is denoted on Fig. 5. Coal subtypes III/2 and IV/2 were found in the upper part of the center of the coal corridor 39706 and subtypes II/1 and II/2 along its sides. The subtype III/2 is less altered by oxidation then subtype IV/2. Statistical characteristics of altered coal sub-

types ($\bar{x}$ – arithmetic mean, s – standard deviation) were evaluated and they are presented in Table 1.

Two principally different types of altered coal can be identified in the coal samples. They are:

– thermally altered coal without influence of oxidation (coal type II, both subtypes II/1 and II/2)

– thermally altered coal with intensive influence of oxidation (coal subtypes III/2 and IV/2).

(a) The chemical composition of the thermally altered coal without influence of oxidation is characterized by lower concentration of hydrogen. The reduction of hydrogen is associated with relative increase of carbon and oxygen concentrations in coal subtypes II/1 and II/2 (Table 1). Meffert's index MI = $[(O^{at}+ N^{at})/H^{at}]$ of this coal is higher than that for nonalterd coal (I^*) because of much lower concentration of hydrogen in altered type II. The similar concentrations of oxygen (O^{at}) in types (I^*) and II suggest that coal type II (subtypes II/1 and II/2) were not oxidized. The coal type II is also characterized by very high reflectance of vitrinite R_0 (4.0 for subtype II/1 and 5.5 for subtype II/2 respectively) and by very low mean values of volatile matter (8.1 for subtype II/1 and 3.9 for subtype II/2 respectively). The chemical composition and also R_0 and V^{daf} of coal type II are similar to the composition and properties of anthracites. The subtype II/2 is of a higher metamorphic grade than subtype II/1.

(b) The composition of coal types III (present here as subtype III/2) and IV (present here as subtype IV/2) quite differs from coal type II. Coal subtypes III/2 and IV/2 may be characterized by an extremely high concentration of oxygen (O^{at}), high Meffert's index (MI) and also by retrogradely formed humic acids (HA^{daf}). Also the concentration of nitrogen N^{at} is higher. The content of retrogradely formed humic acids is much lower than that in coal subtypes III/1 and IV/1 (Klika 1999). They also differ in colour. While humic acids separated from coal subtypes III/1 and IV/1 are of black colour, those separated from coal subtypes III/2 and IV/2 are of yellow colour. The difference in colour is in direct relation to molecular weight and reflects thermal conditions at which humic acids were formed. The high reflectance of vitrinite R_0 (apr. 1.8 for subtype III/2 and 2.0 for subty-
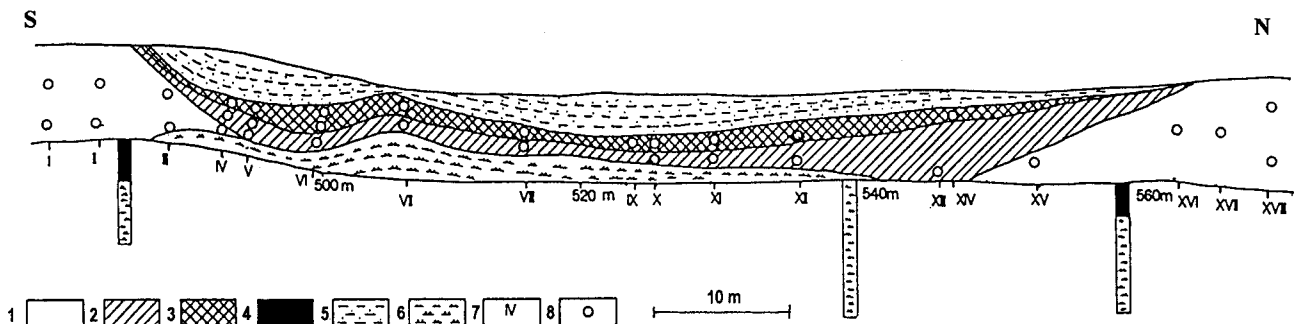

Fig. 5. Scheme of the studied part in coal seam 512(39) (coal corridor 39706).
1 – coal type II (subtypes II/1 and II/2); 2 – coal subtype III/2; 3 – coal subtype IV/2; 4 – coal seam verified by drilling; 5 – coloured sandy claystone; 6 – grey siltstones; 7 – number of profile; 8 – sample.
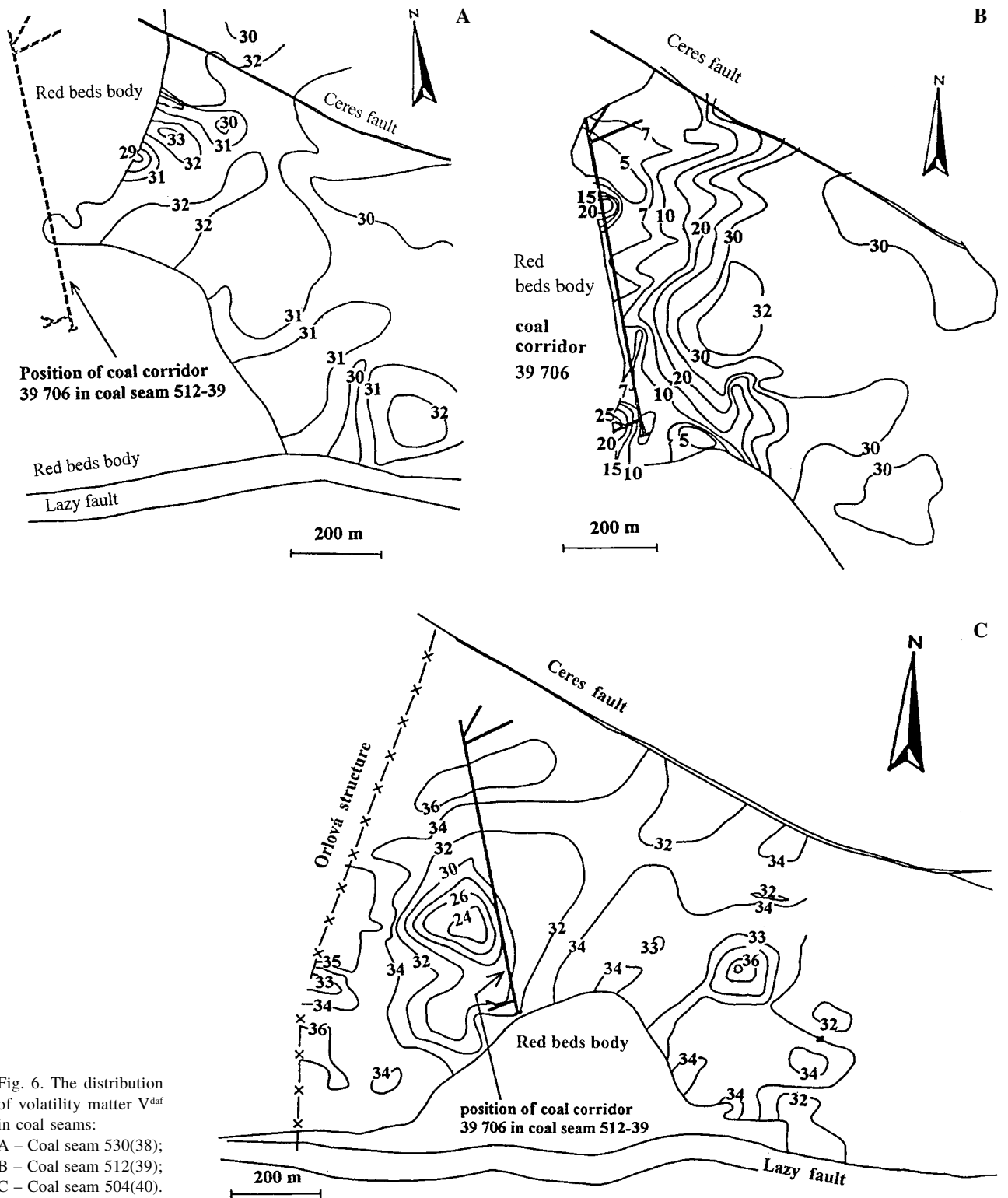
Fig. 6. The distribution of volatility matter V^{daf} in coal seams:
A – Coal seam 530(38);
B – Coal seam 512(39);
C – Coal seam 504(40).

pe IV/2 respectively) is further evidence of oxidation at higher temperatures.

The relation between the Meffert's index (MI) and ratio of $(C/H)^{at}$ is plotted for all samples in Fig. 7. The first parameter (MI) characterizes the oxidation grade whereas the aromaticity index $(C/H)^{at}$ characterizes the relation between the aromatic and nonaromatic part of

coal matter. For all altered coals both parameters (MI and $(C/H)^{at}$) are higher than those for nonaltered coal. The diagram (Fig. 7) shows two different sequences of a possible alteration. The first one is a thermal alteration under zero oxidative conditions (a) :

Coal types (subtypes):   II/2 —— I* —— II/1
A^d [%]: (4.76)—(2.68)—(8.04)

T a b l e 1 . Basic statistical characteristics of parameters of coal types from Lazy Mine.

Type		W^a		A^d		V^{daf}		HA^{daf}		R_o	
Subtype	n	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s
I*	1	2.11	–	2.68	–	29.10	–	0.00	–	1.01	–
II/1	9	7.73	3.17	8.04	2.68	8.12	2.52	0.00	0.00	4.00	0.57
II/2	3	8.51	0.52	4.76	0.64	3.86	1.57	0.00	0.00	5.50	0.22
III/2	9	19.05	3.96	9.15	9.89	20.34	3.88	0.55	0.41	1.75	0.51
IV/2	11	17.46	1.78	14.28	3.95	33.73	3.97	1.13	0.33	1.96	0.32

Type		C^{at}		H^{at}		N^{at}		O^{at}		S^{at}		MI	
Subtype	n	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s	$\bar{x}$	s
I*	1	56.94	–	39.28	–	0.80	–	2.97	–	0.01	–	0.10	-
II/1	9	74.69	2.13	20.47	1.75	0.93	0.06	3.67	1.26	0.05	0.03	0.22	0.07
II/2	3	86.68	0.98	10.16	0.65	0.83	0.03	2.34	0.65	0.07	0.04	0.31	0.06
III/2	9	67.68	2.67	23.68	1.73	1.16	0.11	7.15	1.58	0.20	0.12	0.35	0.06
IV/2	11	63.93	3.50	20.20	2.12	2.24	0.65	13.31	1.62	0.31	0.12	0.77	0.10

Locality: Lazy Mine, $\bar{x}$ – arithmetic mean, s – standard deviation, n – number of samples

and the second one is a thermal treatment followed by an oxidative alteration (b):

Coal types (subtypes):   I* —— II/1 — III/2 —— IV/2
A^d [%]: (2.68)—(8.04)—(9.15)—(14.28)

Taking into account ash content (A^d) we can suppose that in the first stage the coal was altered by thermal treatment (without the contribution of oxygen (subtypes II/1 and II/2). Then the oxidation took place and coal subtypes III/2 and IV/2 were formed.

Petrographic study of altered coal from coal corridor 39706 was performed by Kraussová (1987). The coal samples from this locality were divided among three basic macrotypes which were also described by Klika and Kraussová (1993).

Inorganic matter of altered coal

Alteration of organic matter of coal (during coal thermal and oxidative alteration) is accompanied by alteration of minerals and by differences in chemical composition of inorganic matter of coal (Simonova – Shendrik 1995, Hurley – Schobert 1993, Benson – Harb 1993). The coal seam 512(39) is not rich in inorganic matter. Ash content of nonaltered coal (I*) is about 3 wt %. The principal minerals present in this coal (Table 2) are carbonates (dolomite/ankerite and siderite) and sulphides (pyrite) or silicates (kaolinite).

During thermal alteration, which resulted in formation of coal types II, carbonates were not thermally decomposed. Therefore, we can suppose that the temperature of alteration was lower than approx. 850 °C. Unlike carbonates, sulphides are not present in coal type II (Table 2) because they were thermally decomposed. The temperature at which sulphides decompose is approximately 540 °C for pyrite and 515 °C for marcasite. The stability of carbonates and decomposition of pyrite suggest that the temperature of thermal alteration of coal type II ranged from 540 to 850 °C.

Samples of coal type III (subtype III/2) and IV (subtype IV/2) exhibited substantial oxidation. Ash content in these types is higher then for previous coal types I* and II and is approx. 9 wt % for subtype III/2 and 14 wt % for subtype IV/2, respectively. Oxidation also causes mineral alteration. The original carbonate mineralization was completely replaced by sulphates (e. g. gypsum) and halite (Table 2). Laboratory water leachates were prepared from the altered coal by extraction of coal with water warmed on approx. 90 °C for 15 minutes. After evaporation of the water, the minerals halite, gypsum, hexahydrite, hydrobasalunite and thenardite were crystallized. Gypsum and halite were usually the most abundant minerals. Content of salts dried at 105 °C recalculated on ash base of subtypes II/1 and II/2 ranged from 0.89 to 4.71 wt %, for coal subtype III/2 from 1.8 to 17.5 wt % and for subtype IV/2 from 3.30 to 28.2 wt %.

Chemical analyses of major and trace elements of coal samples was presented in Klika (1999). Nonaltered coal (I*) and coal subtypes II/1 and II/2 contain very high concentrations of CaO, Fe_2O_3 and CO_2. Concentrations of sulphur (presented here as SO_3) in ash from nonaltered coal and coal subtypes III/2 and IV/2 is about 4 wt % while in ash from coal subtypes II/1 and II/2 it is very low. These sulphur concentrations are in agreement with pyrite content. The relation between concentration of CO_2 (determined in coal and recalculated on ash base) and concentration of total sulphur is plotted in Fig. 8. The carbonate minerals (dolomite/ankerite, siderite resp. calcite) originally present in nonaltered coal have been in coal subtypes III/2 and IV/2 replaced by sulphate and/ or halite.

The mass differences of elements before and after alteration of coal were evaluated by the mass balance described by Klika (1998). Similarly as in previous paper (Klika 1999) the values of i-th mass element increase/decrease $10^2[(\Delta m_{ALT})/(m_{I*})]_{IN,i}$ was used. This mass ratios were calculated using A^d values from Table 1 and concentrations of elements given in Klika (1998). In
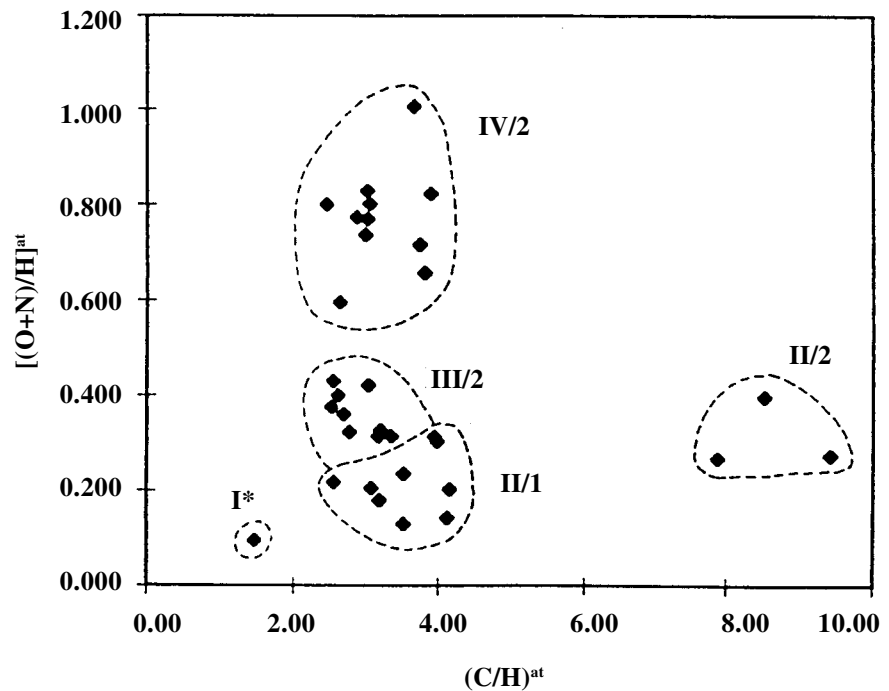
Fig. 7. Distribution of oxidative altered coal subtypes in diagram Meffert's index $[(O + N)/H]^{at}$ versus aromaticity index $(C/H)^{at}$. I^* – nonaltered coal, II/1, II/2, III/2 and IV/2 – altered coal subtypes.

Fig. 9A the values $10^2[(\Delta m_{ALT})/(m_{I^*})]_{IN,i}$ are plotted for major elements and in Fig. 9B. for selected trace elements. Calculated increase of mass of most major elements (e. g. the typical elements of phyllosilicates SiO_2 and Al_2O_3) is small in altered coal corridor 39706. The highest increases of major elements masses were observed for Na_2O, K_2O and also TiO_2 but they are much lower than those which were calculated for Doubrava Mine (where e. g. mean increase of major elements mass is approx. 350 %).

Percentages of trace element mass increase is much higher than those for major elements. For trace elements (B, As, Sr, Ba) and ammonia which show the highest mass increases are their values plotted in Fig. 9B. Their concentrations in nonaltered coal type I^* were: 196 ppm B, 22 ppm As, 853 ppm Sr, 2590 ppm Ba and 50 ppm NH_3. High concentrations of these elements and ammonia in altered coal is usually connected with increased concentration of Na which originates from underground percolating sodium chloride waters. High mobility of some trace elements (e. g. As, S, etc.) during the self-burning of coal seams was also found by Goodarzi (1990). It is worth mentioning that the trace elements which were the most enriched in Lazy Mine are almost

the same as in Doubrava Mine (Klika 1999) where coal was altered by oxidation.

Discussion

The foregoing data analysis has shown that the thermal source for the coal alteration in Lazy Mine was probably heat produced by oxidation (under-surface combustion) in parts of coal seams 530(38), 512(39) and 504(40) deposited in red beds body (Fig. 1). The impact of combustion heat from the thermal source on surrounding coal seams 530(38), 512(39) and 504(40) (outside red beds body) is different. The coal seams 530(38) and 504(40) were altered under low temperature conditions during which coal types I and III/1 were formed while coal seam 512(39) was altered at high temperature condition forming coal subtypes II/1 and II/2.

The weak thermal alteration of coal seams 530(38) and 504(40) (outside the red beds body) probably occurred at non-adiabatic conditions. Numerous faults in overlying rocks above coal seams outside the red beds body made possible supply of air, very good heat transfer, outlet of products of coal oxidation, etc. In agreement with results of Francis (1961) and taking into account our laborato-

Table 2. Mineral composition of altered coal

Type /Subtype	n	Q	K	I	Ank/Do	Si	Ka	Py	Su	Ha
I*	1	–	++	–	+++	+	–	++	–	–
II/1	9	+	–	–	+++	+++	(+)	(+)	–	–
II/2	3	(+)	–	–	+++	+++	+	(+)	(+)	–
III/2	9	(+)	(+)	(+)	+	+	–	++	++	+
IV/2	11	(+)	(+)	–	+	+	–	++	++	+

Locality: Lazy Mine,
Q – quartz, K – kaolinite, I – illite, Ank/Do – ankerite/dolomite, Si – siderite, Ka – calcite, Py – pyrite, Su – sulphate(gypsum), Ha – halite
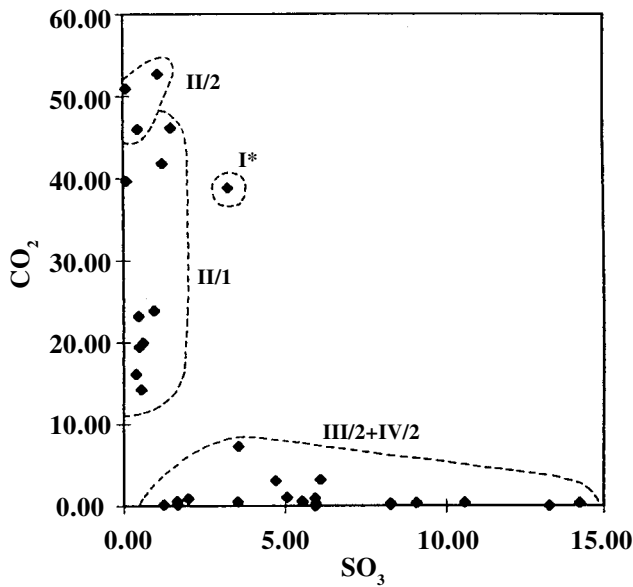Concentrations of minerals: (–) below detection limit, (+) 3–5%, + 5–10%, ++ 10–25%, +++ above 25%

Fig. 8. Relation between CO_2 and SO_3 in nonaltered coal (I^*) and altered coal subtypes II/1, II/2, III/2 and IV/2.

ry results (Klika 1999) the coal subtypes III/1 and IV/1 were formed at temperature below 150 °C.

The high thermal alteration of the 512(39)-th coal seam (outside the red beds body) probably occurred at adiabatic conditions (i.e. no faults in overlying rocks above coal seam outside the red beds body, no air to oxidation, negligible heat transfer, etc.).

In coal corridor 39 706 (coal seam 512(39)) in addition to coal subtypes II/1 and II/2, coal subtypes III/2 and IV/2 have also been found. It was demonstrated that they have been formed by intensive oxidation of previously thermally heated coals. The high temperature of alteration (above 515 °C) is confirmed by decomposition of pyrite originally present in coal. Oxidative conditions for subtypes III/2 and IV/2 are also (except of high concentration of O^{at} and origin of humic acids) confirmed by formation of sulphate mineralization and high concentration of soluble salts. The difference among subtypes III/1, IV/1 and III/2 and IV/2 is shown in Fig. 10. The parameters characterizing oxidation grade of coal subtypes (A – oxygen and B – humic acids) are plotted on y-axis and parameter characterizing thermal alteration of coal subtypes (reflectance of vitrinite R_0) is plotted on the x-axis. The data of 99 coal samples from USCB (Doubrava and Lazy mines and other localities were used for preparation of Fig. 10. Oxidation that gave origin to coal subtypes III/2 and IV/2 run at higher temperatures (higher values R_0) than that at which the coal subtypes III/1 and IV/1 were formed. This idea is also supported by lower content of humic acids in subtypes III/2 and IV/2. According to ash content the subtypes III/2 and IV/2 originated from subtypes II/1 or II/2 in coal seam 512(39). The formation of humic acids from anthracite-like coals (subtypes II/1 and II/2) by oxidation is much slower process than formation of HA from bituminous coal (Klika 1998). Mo-
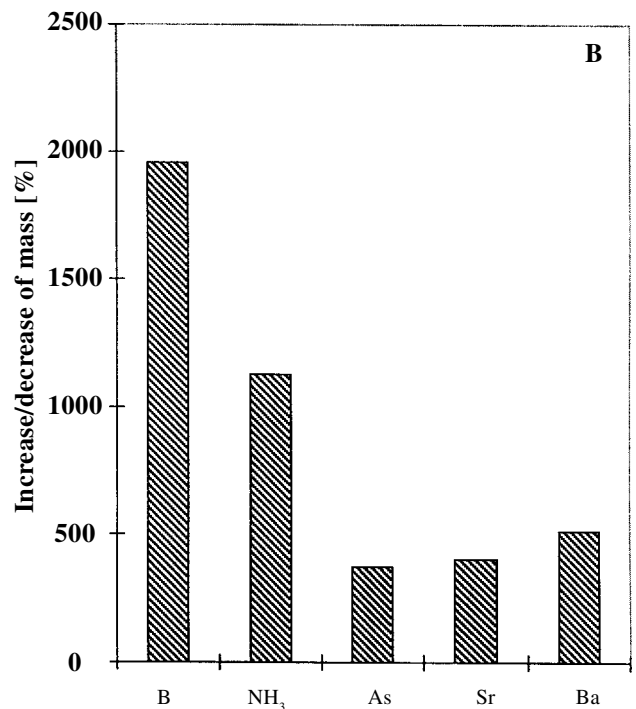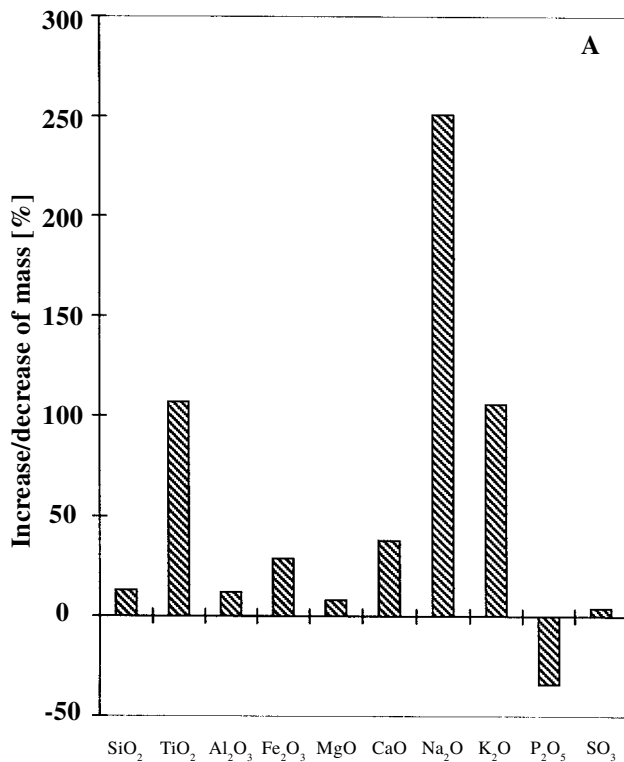

Fig. 9. Changes of elements relative mass (wt%) in coal ashes from altered coal.
A – Major elements, B – Trace elements.

reover, if the oxidation of the coal run at temperature higher than 250 °C the partial decomposition of humic acids took place (Klika 1998). Therefore the transformation of subtypes II/1 and II/2 to III/2 and IV/2 had to run at temperatures below 250 °C.

Conclusion

The high thermal alteration and formation of coal subtypes II/1 and II/2 (outside the red beds body) could occur in 512(39) coal seam (Lazy Mine) under the conditions of adiabatic system. It is supposed that heat was liberated from oxidative degradation of coal seam deposited in red beds body. Faults in overlying rocks outside the red beds body and above such coal subtypes are not present and therefore coal type II can be formed at high temperature and without presence of oxygen. If oxygen was present, the coal was altered additionally by oxidation forming the coal subtypes III/2 and IV/2.

The idea about coal combustion heat as a source for coal alteration is in an agreement with Králík (1980, 1982) and Dopita (1994) but not in an agreement with opinion of Gabzdyl and Probierz (1987) who supposed endogenic source for thermal coal alteration. The fact that oxidative altered coal of subtype III/1 is present in a coal seam 504(40) while the subtypes II/1 and II/2 are present in coal seam 512(39), which is deposited above coal seam 504(40) (Figs 5 and 6), give also rather evidence for thermal coal combustion source. We suppose that formation of the red beds body and alteration of coal seams occurred approximately at the same period (from Jurassic to Cretaceous; Krs et al. 1993, Dopita et al. 1993) when the paleorelief of Carboniferous was denuded and the Orlová and Michálkovice structures and next tectonic faults were already formed.
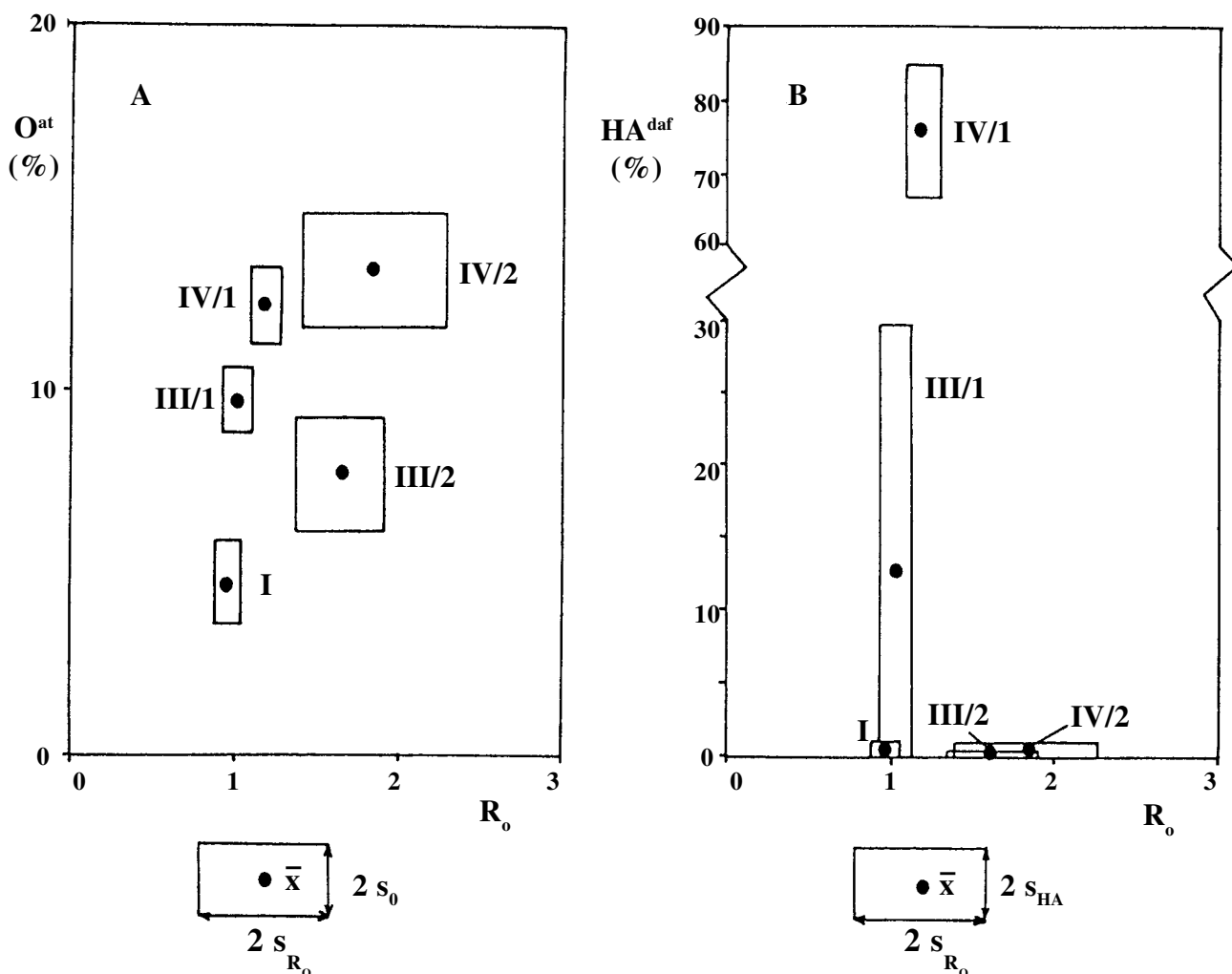
Fig. 10. Subtypes of oxidative and thermally altered coals.
A – Relation between the oxygen content (atom. %) and vitrinite reflectance.
B – Relation between the humic acids content (% related to organic combustible matter) and vitrinite reflectance. *Black points* in the middle of oblongs – arithmetic means of characteristics on figures coordinates, dimensions of the oblongs are equal to value of 2s (two standard deviations) each of the characteristics).

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Tepelně alterované černé uhlí z Hornoslezské černouhelné pánve: souhrn

Z uhelné sloje 512(39), v které se vyskytuje tepelně alterované uhlí, bylo odebráno 33 vzorků uhlí na kterých bylo studováno složení organické a minerální hmoty. V dalších 300 vzorcích uhlí odebraných ze studované 512(39) a okolních uhelných slojí (530(38) a 504(40)), byla stanovena prchavá hořlavina a spalné teplo. Za použití těchto dat byl interpretován proces, který vedl ke vzniku tepelně alterovaného uhlí. Na rozdíl od uhelné sloje 512(39), která byla alterována při vysoké teplotě, sloje 530(38) a 504(40) byly alterovány pouze při nízké teplotě. V uhelné sloji 512(39) bylo rovněž nalezeno uhlí alterované jak za vysoké teploty, tak v intenzivním oxidačním prostředí. Předpokláme, že zdrojem tepla byla intenzivní oxidace (podzemní požár) té části uhelné sloje 512(39) která by se v současné době vyskytovala v tělese pestrých vrstev. Tato část uhelné sloje byla touto oxidací zcela zlikvidována.