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Sulphur and carbon isotopic composition of power supply coals in the Pannonian Basin, Hungary

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Abstract

The present work is an attempt to establish the stable isotope database for Mesozoic to Tertiary coals from the Pannonian Basin, Hungary. Maceral composition, proximate analysis, sulphur form, sulphur isotopes (organic and pyritic), and carbon isotopes were determined. This database supports the assessment of the environmental risks associated with energy generation, the characterization of the formation and the distribution of sulphur in the coals used.

The maceral composition, the sulphur composition, the C, S isotopic signatures, and some of the geological evidences published earlier show that the majority of these coals were deposited in freshwater and brackish water environments, despite the relatively high average sulphur content. However, the Upper Cretaceous, Eocene, and Lower Miocene formations also contain coal seams of marine origin, as indicated by their maceral composition and sulphur and carbon chemistry.

The majority of the sulphur in these coals occurs in the organic form. All studied sulphur phases are relatively rich in ^{34}S isotopes ($\delta^{34}S_{organic}=+12.74‰$, $\delta^{34}S_{pyrite}=+10.06‰$, on average). This indicates that marine bacterial sulphate reduction played a minor role in their formation, in the sense that isotopic fractionation was limited. It seems that the interstitial spaces of the peat closed rapidly during early diagenesis due to a regime of high depositional rate, leading to a relative enrichment of the heavy sulphur isotopes.

Keywords: Coal; Coal petrology; Stable sulphur and carbon isotopes; Pannonian Basin; Hungary

1. Introduction

The sulphur content of coal is a main contributor of air pollution, which has adverse environmental impacts, in relation to the acidification of soil and water. SO_2 emission is regulated by the Geneva Convention (1979) which prescribes a 653 kt/year emission limit for Hungary by the year 2010. The European Union member states' financial aids help the coal industry to adjust to environmental protection standards (Commission of the European Communities, 2001) to reach this goal. Another Communication from the Commission of the European Communities (2002) acknowledges that the security of energy supply in the Community requires the use of multiple energy sources.

Hungary covers the largest central part of the Pannonian Basin and has long-standing traditions in solid fossil fuel exploitation, without paying any attention to the genesis and distribution of sulphur in

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Table 1
Summarised organic petrographic data of studied samples

Mine	S/S no.	Age	T. vitrinite MMB (vol.%)	D. vitrinite MMB (vol.%)	Gelinite MMB (vol.%)	T. L. MMB (vol.%)	A.+A. MMB (vol.%)	L. d. MMB (vol.%)	Inertinite MMB (vol.%)	Pyrite MMB (vol.%)	Clay MMB (vol.%)	Qvarz MMB (vol.%)	Vitrinite MM free (vol.%)	Liptinite MM free (vol.%)	Inertinite MM free (vol.%)	R_o(%)
Szászvár	2	J_1	34.3	35.7	2.4	2.4	0	1.8	19.6	0.8	3.0	0	69.9	5.7	24.5	2.89
Szászvár	6	J_1	65.6	23.7	2.3	1.5	0	0.0	3.8	0.8	2.3	0	93.1	6.6	0.3	1.16
Pécs-Vasas	11/1	J_1	48.7	41.9	2.6	0.9	0	0.0	0.0	0.0	5.9	0	99.1	0.9	0.0	
Pécs-Vasas	11/6	J_1	59.7	22.8	7.0	0.8	0	5.3	0.0	3.5	0.9	0	93.6	6.4	0.0	
						0.0	0		0.0							

Mine	S/S no.	Age	T. huminite MMB (vol.%)	D. huminite MMB (vol.%)	Gelinite MMB (vol.%)	T. L. MMB (vol.%)	A.+A. MMB (vol.%)	L. d. MMB (vol.%)	Inertinite MMB (vol.%)	Pyrite MMB (vol.%)	Clay MMB (vol.%)	Qvarz MMB (vol.%)	Huminite MM free (vol.%)	Liptinite MM free (vol.%)	Inertinite MM free (vol.%)	R_o(%)
Ajka	6/1	Cr_3	47.2	32.6	0.0	6.1	0	1.1	10.9	0.0	2.1	0.0	81.6	7.1	11.2	0.47
Ajka	6/2	Cr_3	19.2	41.3	5.8	1.0	0	0.0	30.8	1.0	0.9	0.0	67.6	1.0	31.4	
Ajka	6/3	Cr_3	18.1	35.7	2.7	13.1	0	4.8	25.6	0.0	0.0	0.0	56.5	17.9	25.6	
Ajka	5/4	Cr_3	33.1	33.0	0.9	8.7	0	0.0	24.3	0.0	0.0	0.0	67.0	8.7	24.3	
Ajka	5/5	Cr_3	30.4	27.5	2.0	22.5	0	0.0	15.7	1.0	0.9	0.0	61.1	22.9	16.0	
Ajka	5/6	Cr_3	20.6	29.4	8.8	9.8	0	0.0	27.4	2.0	2.0	0.0	61.2	10.2	28.6	
Ajka	5/7	Cr_3	11.9	31.7	5.9	1.9	0	4.0	43.6	0.0	1.0	0.0	50.0	6.0	44.0	
Ajka	4/8	Cr_3	3.7	36.1	1.9	4.7	0	1.8	48.1	1.9	1.8	0.0	43.3	6.7	49.9	
Ajka	4/9	Cr_3	50.5	24.2	13.9	3.9	0	0.0	4.1	0.0	3.4	0.0	91.7	4.0	4.2	
Ajka	3/10	Cr_3	48.5	23.8	11.9	5.4	0	1.5	5.9	2.3	0.7	0.0	86.8	7.1	6.1	0.48
Ajka	3/11	Cr_3	16.3	33.7	14.4	12.1	0	2.3	20.2	0.0	1.0	0.0	65.1	14.5	20.4	
Ajka	3/12	Cr_3	18.4	29.2	5.8	18.4	0	0.0	27.2	0.0	1.0	0.0	53.9	18.6	27.5	

Ajka	1/13	Cr_3	22.0	23.9	4.5	0.7	0	2.1	10.1	1.8	34.9	0.0	79.6	4.4	16.0	
Ajka	1/14	Cr_3	10.2	36.1	7.2	7.8	0	0.7	11.8	2.8	23.4	0.0	72.5	11.5	16.0	
Ajka	1/15	Cr_3	22.8	27.5	9.0	3.9	0	1.3	16.4	6.1	13.0	0.0	73.3	6.4	20.3	
Balinka	2	E_2	28.6	45.6	8.6	6.0	0.7	3.8	3.8	2.9	0.0	0.0	85.3	10.8	3.9	0.44
Balinka	6	E_2	29.1	32.3	14.2	8.6	1.6	8.7	1.6	0.0	3.9	0.0	78.7	19.7	1.7	
Doba	2/1	E_2	10.4	33.8	5.2	7.7	13.9	7.8	0.4	5.2	15.6	0.0	62.4	37.1	0.5	
Doba	2/5	E_2	7.5	53.8	11.3	9.5	0	2.8	5.7	1.9	7.5	0.0	80.1	13.6	6.3	
Mány I/A	fo/1	E_2	33.3	47.6	8.6	4.7	0	2.9	1.0	0.0	1.9	0.0	91.2	7.7	1.0	
Mány I/A	fo/4	E_2	40.0	29.6	13.0	5.9	1.7	5.4	0.9	1.7	1.8	0.0	85.6	13.5	0.9	0.42
Vértessomló	1	Ol_2	42.8	29.9	5.9	8.0	0	4.9	0.6	0.0	7.9	0.0	85.3	14.0	0.7	0.36
Vértessomló	2	Ol_2	43.8	18.1	4.8	6.7	0	6.6	0.0	0.0	20.0	0.0	83.4	16.6	0.0	
Vértessomló	3	Ol_2	34.0	28.4	25.7	2.7	0	2.8	0.0	2.8	3.6	0.0	94.1	5.9	0.0	
Székvölgy	3/4	M_1	60.4	15.5	8.2	5.5	1.1	1.6	0.4	4.9	2.4	0.0	90.7	8.8	0.4	
Székvölgy	3/5	M_1	38.9	37.2	6.2	3.9	2.5	2.4	0.9	2.7	5.3	0.0	89.5	9.6	1.0	
Vadna	5/2	M_1	48.7	32.1	10.1	3.6	0.9	1.9	0.9	1.8	0.0	0.0	92.6	6.5	0.9	
Vadna	5/6	M_1	66.7	18.0	8.1	2.7	0	2.7	0.0	0.0	1.8	0.0	94.5	5.5	0.0	0.30
Edelény	4/4	M_1	27.5	36.2	2.0	6.9	9.8	3.9	2.0	0.0	11.7	0.0	74.4	23.3	2.3	0.33
Edelény	2/2	M_1	68.7	3.9	9.7	1.1	0	0.0	0.0	0.0	0.0	16.6	98.7	1.3	0.0	0.29
Várpalota	2	M_2	28.2	42.7	10.0	9.1	0.9	1.8	4.5	1.0	1.2	0.6	83.2	12.1	4.6	
Várpalota	6	M_2	17.9	43.4	5.7	6.6	6.6	1.9	10.4	1.9	5.6	0.0	72.4	16.3	11.2	0.32
Bükkábrány	1	M_3	38.5	31.7	14.9	4.0	1	1.0	6.9	2.0	0.0	0.0	86.8	6.1	7.0	
Bükkábrány	2	M_3	20.0	47.3	3.6	16.4	0	2.7	4.5	0.9	4.6	0.0	75.0	20.2	4.8	0.29
Bükkábrány	3	M_3	29.5	38.1	2.9	9.6	1	5.6	3.8	1.9	7.6	0.0	77.9	17.9	4.2	0.27
Bükkábrány	4	M_3	69.1	7.3	3.6	18.2	0	0.9	0.0	0.0	0.9	0.0	80.7	19.3	0.0	

Legend: T. vitrinite—telovitrinite; D. vitrinite—detrovitrinite; T. L.—sum of terrestrial liptinite; A.+A.—alginite + amorphinite; L. d.—liptodetrinite; T. huminite— telohuminite; D. huminite—detrohuminite; MMB—calculated on mineral matter basis; MM free—calculated on mineral matter free basis.

Table 2
The depositional environment and chemical composition of Pannonian Basin coals

Mine	S/S	Age	Dep. environment		$\delta^{13}C$ (daf %)	$\delta^{34}S_{org}$ (daf %)	$\delta^{34}S_{pyr}$ (daf %)	S_{org} (daf %)	S_{pyr} (daf %)	S_{sulph} (daf %)	S_{total} (daf %)	Ash (daf %)	TOC (daf %)
a) The depositional environment and the chemical composition of the Mesozoic coals													
Szászvár*	2	J_1	FW	U.D. FP	−25.09	9.10	10.47	0.81	0.04	0.13	0.98	25.63	*
Szászvár**	3	J_1	FW	U.D. FP	−27.58	*	*	*	*	*	*	*	*
Szászvár**	4	J_1	FW	U.D. FP	−24.54	*	*	*	*	*	*	*	*
Szászvár**	5	J_1	FW	U.D. FP	−25.61	*	*	*	*	*	*	*	*
Szászvár	6	J_1	FW	U.D. FP	−25.73	7.99	4.34	3.90	1.53	0.67	6.10	27.90	*
Szászvár	7	J_1	FW	U.D. FP	−26.32	*	*	*	*	*	*	*	*
Pécs-Vasas	11/1	J_1	FW	U.D. FP	−26.19	7.51	6.94	0.59	0.02	0.01	0.62	17.01	*
Pécs-Vasas*	11/4	J_1	FW	U.D. FP	−27.50	*	*	*	*	*	*	*	*
Pécs-Vasas*	11/5	J_1	FW	U.D. FP	−25.79	*	*	*	*	*	*	*	*
Pécs-Vasas	11/6	J_1	FW	U.D. FP	−26.55	16.27	16.43	4.00	1.00	0.28	5.28	14.80	*
Ajka	6/1	Cr_3	FW	U.D. FP	−25.20	*	*	*	*	*	*	*	*
Ajka	6/2	Cr_3	FW	U.D. FP	−24.12	*	*	*	*	*	*	*	*
Ajka	6/3	Cr_3	FW	U.D. FP	−23.92	12.09	9.50	4.55	0.43	0.23	5.21	14.13	58.10
Ajka	5/4	Cr_3	FW	BB SP	−24.30	*	*	*	*	*	*	*	*
Ajka	5/5	Cr_3	FW	BB SP	−24.07	*	*	*	*	*	*	*	*
Ajka	5/6	Cr_3	FW	BB SP	−24.66	*	*	*	*	*	*	*	*
Ajka	5/7	Cr_3	FW	BB SP	n.a.	*	*	*	*	*	*	*	*
Ajka	4/8	Cr_3	BR	BB SP	−24.05	*	*	*	*	*	*	*	*
Ajka	4/9	Cr_3	BR	BB SP	−24.72	*	*	*	*	*	*	*	*
Ajka	3/10	Cr_3	BR	BB SP	−24.14	12.97	8.86	3.59	0.79	1.14	5.52	11.60	60.50
Ajka	3/11	Cr_3	BR	BB SP	−24.45	*	*	*	*	*	*	*	*
Ajka	3/12	Cr_3	BR	BB SP	−24.63	*	*	*	*	*	*	*	*
Ajka	1/13	Cr_3	M	BB SP	−24.81	*	*	*	*	*	*	*	*
Ajka	1/14	Cr_3	M	BB SP	−24.11	*	*	*	*	*	*	*	*
Ajka	1/15	Cr_3	M	BB SP	−24.34	*	*	*	*	*	*	*	*
b) The depositional environment and the chemical composition of the Palaeogene coals													
Balinka	1	E_2	BR	BB SP	−24.67	*	*	*	*	*	*	*	*
Balinka	2	E_2	BR	BB SP	−25.71	19.05	15.77	4.49	0.78	0.11	5.38	11.43	*
Balinka	3	E_2	BR	BB SP	−24.93	*	*	*	*	*	*	*	*
Balinka	4	E_2	BR	BB SP	−25.16	*	*	*	*	*	*	*	*
Balinka	5	E_2	BR	BB SP	−24.80	*	*	*	*	*	*	*	*
Balinka	6	E_2	BR	BB SP	−25.70	17.50	16.20	4.34	0.85	0.10	5.29	12.05	*
Balinka	8	E_2	BR	BB SP	−25.09	*	*	*	*	*	*	*	*
Doba	1/6	E_2	FW	D. FP	−24.50	*	*	*	*	*	*	*	*
Doba	2/1	E_2	BR	BB SP	−25.84	7.31	−3.52	4.40	2.05	1.55	8.00	42.12	30.40
Doba	2/2	E_2	BR	BB SP	−25.35	*	*	*	*	*	*	*	*
Doba	2/3	E_2	BR	BB SP	−24.68	*	*	*	*	*	*	*	*
Doba	2/4	E_2	BR	BB SP	−24.72	*	*	*	*	*	*	*	*
Doba	2/5	E_2	BR	BB SP	−25.80	16.18	15.12	3.53	0.29	0.05	3.87	10.38	56.90
Mány I/A	fo/1	E_2	FW	U.D. FP	−25.38	18.28	17.15	2.60	0.20	0.04	2.84	6.09	40.00
Mány I/A	fo/2	E_2	FW	U.D. FP	−24.77	*	*	*	*	*	*	*	*

Mány I/A	fo/3	E_2	FW	U.D. FP	−25.98	*	*	*	*	*	*	*	*
Mány I/A	fo/4	E_2	FW	U.D. FP	−25.29	16.33	13.71	1.77	0.48	0.25	2.50	10.15	62.60
Mány I/A	fo/5	E_2	FW	U.D. FP	−25.41	*	*	*	*	*	*	*	*
Mány I/A	fo/6	E_2	FW	U.D. FP	−26.16	*	*	*	*	*	*	*	*
Mány I/A	fo/7	E_2	FW	U.D. FP	−24.9	*	*	*	*	*	*	*	*
Vértessomló	1	Ol_2	FW	U.D. FP	−26.78	*	12.14	0.47	0.22	0.06	0.69	20.05	59.30
Vértessomló	2	Ol_2	FW	U.D. FP	−26.94	*	*	*	*	*	*	*	*
Vértessomló	3	Ol_2	FW	U.D. FP	−26.47	7.44	7.04	0.66	0.24	0.10	1.00	18.84	58.20
Véretssomló	4	Ol_2	FW	U.D. FP	−28.53	*	*	*	*	*	*	*	*
Vértessomló	5	Ol_2	FW	U.D. FP	−26.59	*	*	*	*	*	*	*	*
c) The depositional environment and the chemical composition of the Miocene coals													
Székvölgy	3/1	M_1	FW	BB LCP	−27.79	*	*	*	*	*	*	*	*
Székvölgy	3/2	M_1	FW	BB LCP	−25.61	*	*	*	*	*	*	*	*
Székvölgy	3/3	M_1	FW	BB LCP	−25.71	*	*	*	*	*	*	*	*
Székvölgy	3/4	M_1	FW	BB LCP	−26.75	−2.41	−4.88	3.26	3.30	2.75	9.31	22.12	46.90
Székvölgy	3/5	M_1	FW	BB LCP	−26.98	10.50	4.66	1.23	0.38	0.60	2.21	23.69	44.50
Székvölgy	3/6	M_1	FW	BB LCP	−27.35	*	*	*	*	*	*	*	*
Székvölgy	3/7	M_1	FW	BB LCP	−26.87	*	*	*	*	*	*	*	*
Vadna	5/1	M_1	FW	BB LCP	−26.58	*	*	*	*	*	*	*	*
Vadna	5/2	M_1	FW	BB LCP	−26.34	14.20	12.38	2.31	0.64	0.87	3.82	21.26	*
Vadna	5/3	M_1	FW	BB LCP	−26.54	*	*	*	*	*	*	*	*
Vadna	5/4	M_1	FW	BB LCP	−25.32	*	*	*	*	*	*	*	*
Vadna	5/5	M_1	FW	BB LCP	−26.50	*	*	*	*	*	*	*	*
Vadna	5/6	M_1	FW	BB LCP	−24.39	13.77	12.99	1.60	0.14	0.05	1.79	60.86	*
Edelény	2/1	M_1	FW	BB LCP	−25.20	*	*	*	*	*	*	*	*
Edelény	2/2	M_1	FW	BB LCP	−24.24	18.20	13.22	1.81	0.38	0.57	2.76	10.32	*
Edelény	2/3	M_1	FW	BB LCP	−26.37	*	*	*	*	*	*	*	*
Edelény	2/4	M_1	FW	BB LCP	−24.91	*	*	*	*	*	*	*	*
Edelény	4/1	M_1	FW	BB LCP	−27.23	*	*	*	*	*	*	*	*
Edelény	4/2	M_1	FW	BB LCP	−25.00	*	*	*	*	*	*	*	*
Edelény	4/3	M_1	FW	BB LCP	−26.84	*	*	*	*	*	*	*	*
Edelény	4/4	M_1	FW	BB LCP	−25.79	8.41	1.38	2.18	0.88	1.26	4.32	34.62	*
Edelény	4/5	M_1	FW	BB LCP	−26.45	*	*	*	*	*	*	*	*
Várpalota	0	M_2	FW	BB	−26.05	*	*	*	*	*	*	*	*
Várpalota	1	M_2	FW	BB	−25.55	*	*	*	*	*	*	*	*
Várpalota	2	M_2	FW	BB	−27.01	28.08	27.87	2.29	0.98	0.65	3.89	12.43	*
Várpalota	3	M_2	FW	BB	−25.41	*	*	*	*	*	*	*	*
Várpalota	4	M_2	FW	BB	−24.91	*	*	*	*	*	*	*	*
Várpalota	5	M_2	FW	BB	−25.47	*	*	*	*	*	*	*	*
Várpalota	6	M_2	FW	BB	−26.75	18.81	10.57	3.40	1.05	1.96	6.47	27.45	*
Várpalota	7	M_2	BR	BB	−25.36	*	*	*	*	*	*	*	*
Várpalota	8	M_2	BR	BB	−24.74	*	*	*	*	*	*	*	*
Bükkábrány	1	M_3	FW	L.D. I-D	−26.47	*	*	*	*	*	*	*	*
Bükkábrány	2	M_3	FW	L.D. I-D	−27.21	*	*	*	*	*	*	*	*

(continued on next page)

Table 2 (*continued*)

Mine	S/S	Age	Dep. environment		$\delta^{13}C$ (daf %)	$\delta^{34}S_{org}$ (daf %)	$\delta^{34}S_{pyr}$ (daf %)	S_{org} (daf %)	S_{pyr} (daf %)	S_{sulph} (daf %)	S_{total} (daf %)	Ash (daf %)	TOC (daf %)
Bükkábrány	3	M_3	FW	L.D. I-D	−28.54	3.63	5.94	1.13	0.98	1.34	3.45	30.77	*
Bükkábrány	4	M_3	FW	L.D. I-D	−27.62	*	*	*	*	*	*	*	*
Bükkábrány	5	M_3	FW	L.D. I-D	−27.87	*	*	*	*	*	*	*	*

Mine	Seam/ sample no.	Age	Dep. environment		Facies	Lithotype	$\delta^{13}C$ (daf %)	$\delta^{34}S_{org}$ (daf %)	$\delta^{34}S_{pyr}$ (daf %)	S_{org} (daf %)	S_{pyr} (daf %)	S_{sulph} (daf %)	S_{total} (daf %)	Ash (daf %)	TOC (daf %)
Balinka	1	E_2	BR	BB SP		Coaly shale	−24.67								
Balinka	2	E_2	BR	BB SP	Marsh	Bunded dull	−25.71	19.05	15.77	4.49	0.78	0.11	5.38	11.43	n.a
Balinka	3	E_2	BR	BB SP		Banded coal	−24.93								
Balinka	4	E_2	BR	BB SP		Banded coal	−25.16								
Balinka	5	E_2	BR	BB SP		Banded Bright	−24.80								
Balinka	6	E_2	BR	BB SP	Marsh	Banded coal	−25.70	17.50	16.20	4.34	0.85	0.10	5.29	12.05	n.a
Balinka	8	E_2	BR	BB SP		Banded Bright	−25.09								
Doba	1/6	E_2	FW	D. FP		Banded Bright	−24.50								
Doba	2/1	E_2	BR	BB SP	Aquatic	Banded coal	−25.84	7.31	−3.52	4.40	2.05	1.55	8.00	42.12	30.40
Doba	2/2	E_2	BR	BB SP		Banded dull	−25.35								
Doba	2/3	E_2	BR	BB SP		Banded coal	−24.68								
Doba	2/4	E_2	BR	BB SP		Banded Bright	−24.72								
Doba	2/5	E_2	BR	BB SP	Marsh	Banded Bright	−25.80	16.18	15.12	3.53	0.29	0.05	3.87	10.38	56.90
Mány I/A	fo/1	E_2	FW	U.D. FP	Marsh	Braned coal	−25.38	18.28	17.15	2.60	0.20	0.04	2.84	6.09	40.00
Mány I/A	fo/2	E_2	FW	U.D. FP		Banded Bright	−24.77								
Mány I/A	fo/3	E_2	FW	U.D. FP		Banded coal	−25.98								
Mány I/A	fo/4	E_2	FW	U.D. FP	Int.-swamp	Banded Bright	−25.29	16.33	13.71	1.77	0.48	0.25	2.50	10.15	62.60
Mány I/A	fo/5	E_2	FW	U.D. FP		Banded coal	−25.41								
Mány I/A	fo/6	E_2	FW	U.D. FP		Banded coal	−26.16								
Mány I/A	fo/7	E_2	FW	U.D. FP		Banded Bright	−24.9								
Vértessomló	1	$O1_2$	FW	U.D. FP	Int.-swamp	Banded Bright	−26.78		12.14	0.47	0.22	0.06	0.69	20.05	59.30
Vértessomló	2	$O1_2$	FW	U.D. FP	Aquatic	Banded coal	−26.94								
Vértessomló	3	$O1_2$	FW	U.D. FP	Aquatic	Banded dull	−26.47	7.44	7.04	0.66	0.24	0.10	1.00	18.84	58.20
Vértessomló	4	$O1_2$	FW	U.D. FP		Shally coal	−28.53								
Vértessomló	5	$O1_2$	FW	U.D. FP		Banded coal	−26.59								
Székvölgy	3/1	M_1	FW	BB LCP		Banded Bright	−27.79								
Székvölgy	3/2	M_1	FW	BB LCP		Banded Bright	−25.61								

Székvölgy	3/3	M_1	FW	BB LCP		Banded Bright	−25.71							
Székvölgy	3/4	M_1	FW	BB LCP	W. Swamp	Banded Bright	−26.75	−2.41	−4.88	3.26	3.30	2.75	9.31	22.12	46.90
Székvölgy	3/5	M_1	FW	BB LCP	Marsh	Banded coal	−26.98	10.50	4.66	1.23	0.38	0.60	2.21	23.69	44.50
Székvölgy	3/6	M_1	FW	BB LCP		Bright coal	−27.35								
Székvölgy	3/7	M_1	FW	BB LCP		Banded Bright	−26.87								
Vadna	5/1	M_1	FW	BB LCP		Banded Bright	−26.58								
Vadna	5/2	M_1	FW	BB LCP	W. Swamp	Banded Bright	−26.34	14.20	12.38	2.31	0.64	0.87	3.82	21.26	n.a
Vadna	5/3	M_1	FW	BB LCP		Banded coal	−26.54								
Vadna	5/4	M_1	FW	BB LCP		Banded dull	−25.32								
Vadna	5/5	M_1	FW	BB LCP		Banded Bright	−26.50								
Vadna	5/6	M_1	FW	BB LCP	W. Swamp	Banded Bright	−24.39	13.77	12.99	1.60	0.14	0.05	1.79	60.86	n.a
Edelény	2/1	M_1	FW	BB LCP		Shally coal	−25.20								
Edelény	2/2	M_1	FW	BB LCP	W. Swamp	Bright coal	−24.24	18.20	13.22	1.81	0.38	0.57	2.76	10.32	n.a
Edelény	2/3	M_1	FW	BB LCP		Banded Bright	−26.37								
Edelény	2/4	M_1	FW	BB LCP		Banded Bright	−24.91								
Edelény	4/1	M_1	FW	BB LCP		Banded Bright	−27.23								
Edelény	4/2	M_1	FW	BB LCP		Banded Bright	−25.00								
Edelény	4/3	M_1	FW	BB LCP		Banded coal	−26.84								
Edelény	4/4	M_1	FW	BB LCP	Marsh	Banded coal	−25.79	8.41	1.38	2.18	0.88	1.26	4.32	34.62	n.a
Edelény	4/5	M_1	FW	BB LCP		Banded Bright	−26.45								
Várpalota	0	M_2	FW	BB		Shally coal	−26.05								
Várpalota	1	M_2	FW	BB		Dull coal	−25.55								
Várpalota	2	M_2	FW	BB	Marsh	Banded dull	−27.01	28.08	27.87	2.29	0.98	0.65	3.89	12.43	n.a
Várpalota	3	M_2	FW	BB		Banded Bright	−25.41								
Várpalota	4	M_2	FW	BB		Banded dull	−24.91								
Várpalota	5	M_2	FW	BB		Banded Bright	−25.47								
Várpalota	6	M_2	FW	BB	Marsh	Banded dull	−26.75	18.81	10.57	3.40	1.05	1.96	6.47	27.45	n.a
Várpalota	7	M_2	BR	BB		Shally coal	−25.36								
Várpalota	8	M_2	BR	BB		Banded Bright	−24.74								
Bükkábrány	1	M_3	FW	L.D. I-D	Marsh	Light, fibrous	−26.47								
Bükkábrány	2	M_3	FW	L.D. I-D	Marsh	Light, fibrous	−27.21								
Bükkábrány	3	M_3	FW	L.D. I-D	Marsh	Light, fibrous	−28.54	3.63	5.94	1.13	0.98	1.34	3.45	30.77	n.a
Bükkábrány	4	M_3	FW	L.D. I-D	W. Swamp	Dark, fibrous	−27.62								
Bükkábrány	5	M_3	FW	L.D. I-D		Dark, fibrous	−27.87								

Legend: S/S—seam/sample No.; LT—lithotype; **—thermally altered coal; *—data not available; FW—fresh water; BR—brackish water; M- marine; U.D. FP—upper delta flood plain; BB SP—back barrier strand plain; BB LCP – back barrier lower coastal plain; BB—back barrier; L.D. I-D—lower delta plain interdistributary plain; DFS—dry forest swamp; WFS—wet forest swamp; IS—intermediate swamp-marsh complex; R—reworked; A—aquatic; MA—marsh; Ba—banded coal; Bb—banded bright coal; Bd—banded dull coal; Br—bright coal; D—dull coal; Sc—shally coal; Cs—coaly shale; Lf—light fibrous; Df—dark fibrous; $\delta^{34}S_{org}$ - $\delta^{34}S_{organic}$; $\delta^{34}S_{pyr}$ - $\delta^{34}S_{pyrite}$; S_{org} - $S_{organic}$; S_{pyr}—S_{pyrite}; S_{sul}—$S_{sulphate}$.

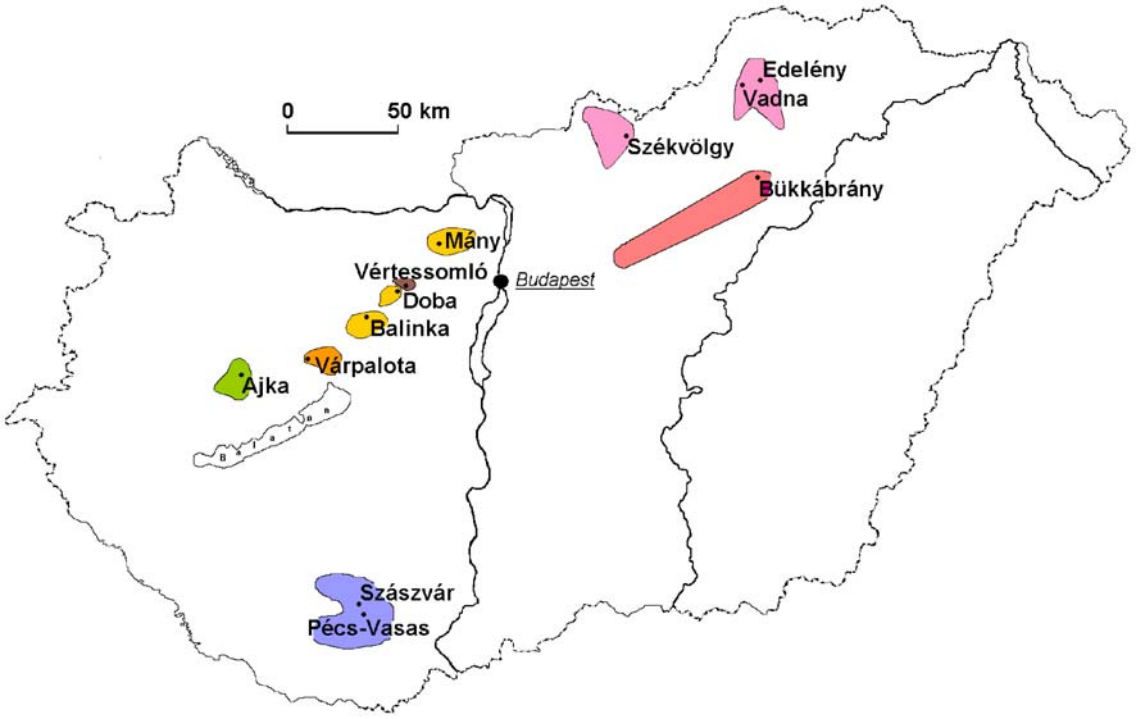

Fig. 1. Location map of the studied basins.

coals in order to mitigate its local and regional environmental impacts.

The first and only country-wide coal geology volume was published in Hungarian by Vadász (1952). A survey of sulphur in coal was published in 1989 by the Central Office of Geology, which set a general figure of the mean sulphur values in the different coal basins, without considering their genesis (Fejér et al., 1989). There are studies that deal with the sulphur diagenesis of organic-rich sediments and kerogens (Hámor, 1994; Vetõ et al., 1994; Pápay, 1998; Vetõ et al., 2000) or the organic (Brukner-Wein and Sajgó, 1990; Sajgó et al., 1993, 1995) and inorganic geochemistry of coals (Tomschey, 1990; Sachsenhofer and Tomschey, 1992; Pápay, 1993) from the Pannonian Basin, but a focused study on the present topic was lacking.

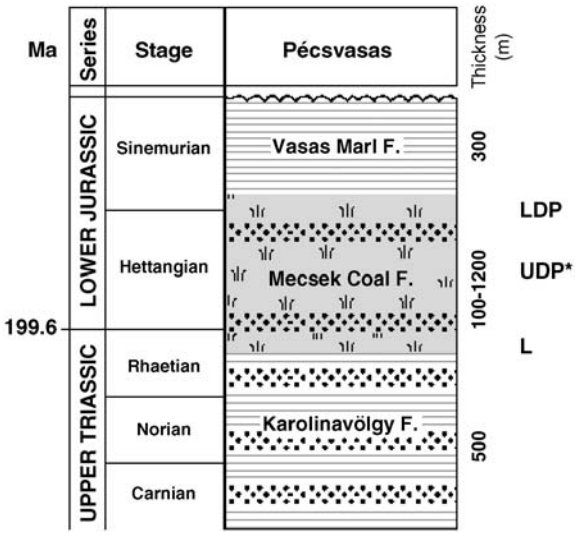

Fig. 2. Stratigraphic column of the Lower Jurassic Mecsek Coal Formation. Legend: L — limnic; UDP — upper delta plain; LDP — lower delta plain; other symbols at Fig. 8.

There are a few studies on carbon isotopes, but no sulphur isotope studies have been carried out for the coals in this study (Holzacker et al., 1981), even though lignites and bituminous coals were important sources of energy between 1950 and 1990.

Modern coal petrographical studies were carried out for the Lower Miocene lignite seams in the Borsod Basin and the Upper Miocene lignites of North Hungary (Hámor-Vidó, 1992, 1993; Hámor-Vidó et al., 2003). However, up-to-date maceral composition data is not available for most coal basins in the Pannonian Basin.

The present study is the first modern review of coal geology and a major step in establishing the detailed sulphur data base of Pannonian coals by providing a representative chemical and isotopic data set emplaced

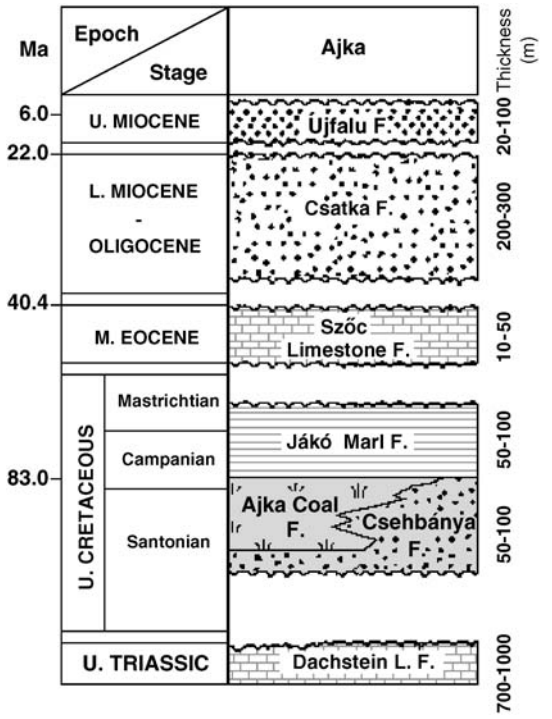

Fig. 3. Stratigraphic column of the Upper Cretaceous Ajka Coal Formation. See legend at Fig. 8.

in a context of general geological and organic petrographical information and to extend the carbon isotopic database. This data allows for the clarification regarding the paleoenvironmental setting of the sampled sequences.

2. General geological setting

The Pannonian Basin, surrounded by the Dinarides, the Alps and the Carpathians, is a back-arc basin of the continent–continent type collision and subduction of the African and Eurasian plates (Pogácsás, 1984). The onset of formation of the Pannonian Basin began in the Early Miocene. The synrift phase began in the Middle Miocene. The major, postrift phase of basin formation, resulting in a 2–4 km thick, brackish lacustrine sequence, lasted 12 Ma (Hámor and Pogácsás, 2001). Coal-bearing formations occur in both the Mesozoic basement units (Lower Jurassic Mecsek Coal Formation, Upper Cretaceous Ajka Coal Formation) and in the prerift (Eocene Dorog Coal Formation, Oligocene Mány Formation, Lower Miocene Salgótarján Brown Coal Formation), synrift (Middle Miocene Hidas Brown Coal Formation), and postrift (Upper Miocene Bükkalja Lignite Formation) basin sequences.

3. Methodology

During a systematic field campaign, 12 coal mines were visited (three-fourth of all coal mines that were in operation at that time) and ca. 100 coal samples were collected, representing the seams utilised in power plants. The samples were taken from walls exposed by the daily operations, were sealed in three-fold nylon bags and nylon sack packing and transported to the laboratory. 85 samples were selected for the $\delta^{13}C$ study. Financial limitation of the project budget defined that the analyses were carried out on 40 samples for detailed maceral analysis (Table 1) and 23 samples were selected for the proximate and the $\delta^{34}S$ studies (Table 2a, b, c). Macroscopic pyrite concretions were removed from the samples by hand (e.g. Szászvár and Vadna samples).

For the macroscopic description of the Mesozoic to Lower Miocene coals, the hard coal lithotype terminology of Diessel (1965, 1992) was used. For the younger, less-coalified Middle and Upper Miocene lignites, lithotypes were modified for the characteristic physical properties such as colour and structure (Table 2c). Organic petrography was carried out on polished

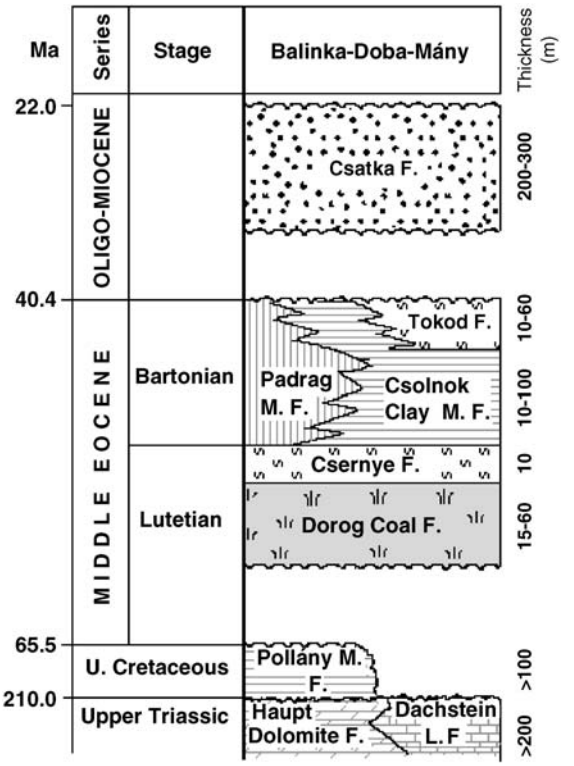

Fig. 4. Stratigraphic column of the Middle Eocene Dorog Coal Formation. See legends at Fig. 8.

sections of crushed and homogenized bulk rocks with reflected light microscopy in oil immersion. The maceral terminology applied in this study is this adopted by the International Committee for coal and Organic Petrology (ICCP, 1998, 2001; Sýkorová et al., 2005) Both white light and fluorescence (UV and blue excitation, 420–490 nm; barrier filter 515 nm) were applied using a Leica DM-RX microscope.

For chemical and isotopic analyses samples were ground in a Fritsch ball mill. Ash content was determined by burning at 700 °C. For all other analysis, carbonates were removed from the samples using standard techniques. The acid-soluble sulphate sulphur was determined using the HCl and the $BaCl_2$ separation method (Smith and Batts, 1974).

Stable isotope ratio analysis was performed on a custom built stable isotope ratio mass spectrometer (Hertelendi et al., 1986) by precipitating barium sulphate, which is combusted to produce sulphur dioxide. The carbon isotope results are given in the δ-notation relative to the PDB standard. The reproducibility of the carbon isotope ratio data is better than ±0.2‰. The sulphur isotope results are given in the δ-notation relative to the CDT standard. The reproducibility of the sulphur isotope ratio data is better than ±0.2‰.

4. Results

4.1. Geological and petrographical characterization of the coal formations

4.1.1. Mecsek Coal Formation, MCF

4.1.1.1. Geologic setting. Hungary's only basin of the highest rank, from high to medium volatile bituminous coal with a production of coking coals, lies in the Mecsek Mountains (Fig. 1). Lower Liassic coal measures evolve continuously from Upper Triassic fluvio-lacustrine sandstone and deltaic shale layers (Nagy, 1969). Coal formation shows a transition from upper delta conditions to lagoon environments (Fig. 2). The lowermost coal seam accumulated in a limnic environment. The productive sequences of the coal measures were formed on the upper delta flood plain (Nagy, 1969). The marine deposits of the overburden are the result of the stepwise change from lagoon into shallow-water, pelagic facies. The thickness of the Mecsek Formation (MF) varies between 100–400 m with 10–30 coal seams thickening westwards. Coal seams are identified on the basis of their relative position to two intercalated rhyolitic tuff beds and by the relative volume of interbedded sediments. Gross calorific values are between 18.5–30 MJ/kg, the average thickness of coal seams is 1.5 m. Lower Cretaceous sub-volcanic basalt intrusions coked the coal seams frequently. Later, intense Miocene tectonism folded, and inclined the coal seams 20–30°. The coal seams studied in Pécsvasas and Szászvár are from the same interval of the coal measures between the two marker rhyolitic tuff beds, indicating the deposition in the central part of the delta system.

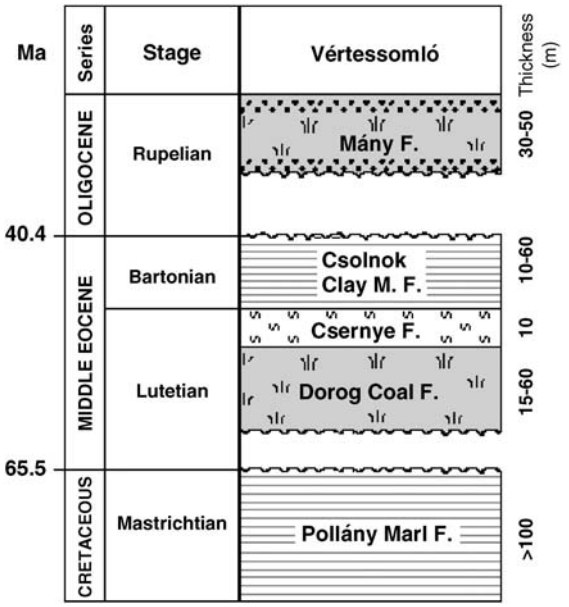

Fig. 5. Stratigraphic column of the Oligocene Mány Formation. See legend at Fig. 8.

4.1.1.2. Petrography. Macroscopically these bituminous coals (0.79–1.25% R_o) are well-banded and bright (80%) to dull or weakly banded. Coal balls of tectonic origin are frequent in Pécsvasas samples. Microscopically, vitrinite constitutes more than 70% of the samples (Table 1). At the two sampling sites telovitrinite is the dominant maceral, and the content of detrovitrinite increases significantly in the uppermost beds. Liptinite content is usually below 5%. Sporinite and liptodetrinite are common, cutinite and resinite seldom occur. Inertinite is found in Szászvár samples, as fusinite and inertodetrinite in vitrinertite microlithotype.

The Pécsvasas samples have higher rank due to post-depositional orogenic upheating (Varga and Horváth, 1986). The lower, Szászvár S-2 heat affected sample is collected in the vicinity of a basalt intrusion that increased vitrinite reflectance to 2.89%.

4.1.2. Ajka Coal Formation, ACF

4.1.2.1. Geologic setting. Ajka Coal Formation is situated in the western part of the Transdanubian Mountain Range. The peat formed on the pre-Senonian basement in a terrestrial fluvio-lacustrine depositional environment (Haas et al., 1992). The pre-Senonian formations are Upper Triassic, Lower Jurassic, and locally Middle Cretaceous bauxite or clay. Senonian freshwater limestone and clay directly underlay the coal sequence in the studied area (Fig. 3). The continuous transition from freshwater environment to marine conditions during peat formation was shown by palynological studies (Góczán et al., 1986). The thickness of beds, calorific value, and ash content decrease from the lower seams up to the top of the section.

The thickness of ACF is 40 to 110 m (100 m average). The coal measures generally consist of seven coal seams, of which five are of economic interest. Clay and marl interbeds with mollusc debris are common within coal seams. Gross calorific value and ash content are between 8–16.7 MJ/kg and 12–30%, respectively. The numbering of coal seams starts from the bottom.

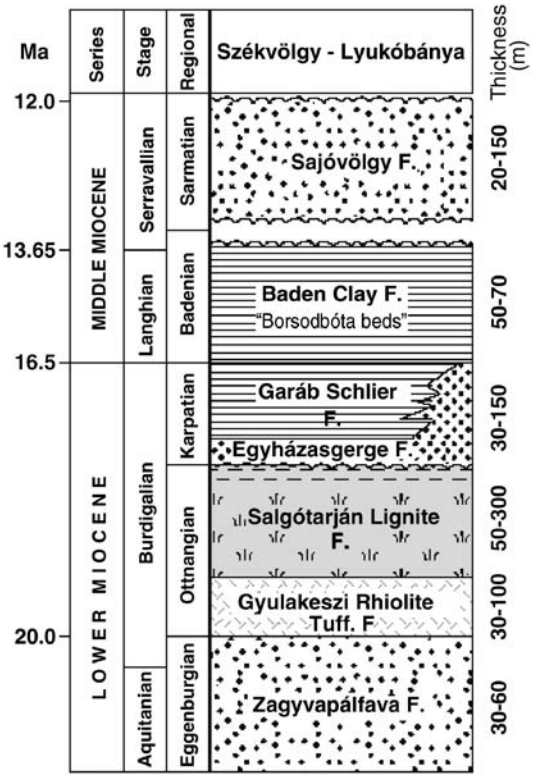

Fig. 6. Stratigraphic column of the Lower Miocene Salgótarján Brown coal Formation. See legend at Fig. 8.

The lowermost sequence consists of five coal seams (Nos. 6, 5, 4, 3, 2). The four lower 1.5–3.5 m thick seams are the most economic. In the middle coal-bearing sequence the 4–8 m thick coal seam No. 1 has high mineral content.

4.1.2.2. Petrography. These subbituminous coals (0.47 to 0.48% R_o) are hard to medium hard, weakly banded, or unbanded, dull or with a slight lustre (Table 1). In seams Nos. 6, 5 and 1 the banded, slightly banded beds are found in the lower parts of the coal seams in contrast to Nos. 4 and 3, where the banded layers appear at the top. In the lower sequence mollusc shells occur within the coal at the top of the No. 6, and in the middle of the No. 5 coal seams. In the upper part of seam No. 4, a few-mm thick layer of gypsum is characteristic in the whole area. In seam No. 5, 1–2 mm amber grains were recognized.

Detrohuminite and ulminite are the characteristic macerals. Gelinite and corpogelinite are subordinates among huminites. Distribution of liptinites are uneven, in samples Nos. 6/3, 5/5, 3/11, 3/12 resinite content is extremely high with more than 9.6%. Amber grains are of 0.1–2.0 mm size. They show rounded sections and often oxidation rims on the polished surface and are cemented by densinite.

Inertinite is a characteristic maceral group of ACF. Fusinite and semifusinite contents increase from the bottom up to seam No. 4. Their maximum concentration is in samples 5/7 and 4/8 with 43.6% and 48.1%, respectively. The inertinite content correlates with the resinite concentration in samples 6/3, 5/5, 3/11 and 3/12 (Table 1).

4.1.3. Dorog Coal Formation, DF

4.1.3.1. Geologic setting. In the north-east part of the Transdanubian Mountain Range subbituminous coals were exploited in five Middle Eocene coal fields, three of which are studied in this paper: NE-Bakony (Balinka), Vértes (Doba) and Tatabánya (Mány) (Fig. 1). They were deposited in the same paleoenvironmental realm 40–42 Ma ago. The underlying sediments are Lower Cretaceous marls or bauxite in the central part, and Upper Triassic limestone and dolomite in the north-east (Mány). Coal deposits are included in the lower section of Eocene transgressive sediments (Fig. 4). The basis of the formation is grey or variegated clay layers overlying Mesozoic sequences. Coal formation took place in different parts of the estuary system of strand plain to karstic peat-forming environments, frequently represented by marine nannoplankton assemblages in the

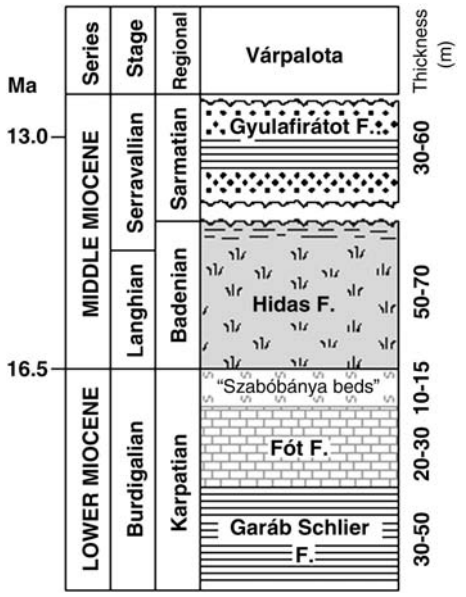

Fig. 7. Stratigraphic column of the Middle Miocene Hidas Brown Coal Formation. See legend at Fig. 8.

upper part of the coal seams, where the direction of marine transgression was SW–NE (Báldi-Beke, 2003). The thickness of the coal measures is 15–60 m, thicker in NE and thinner in SW, usually consisting of one or two seams. The thickness of the coal seams is 0.8–4.0 m, in the Tatabánya coal field the main seam reaches 30 m exceptionally. The gross calorific value is 9.5–24 MJ/kg. The sulphur content is the highest among Hungarian coals (2.4–7.0%).

Coal seams are separated by grey marls with molluscs and foraminifers, and some places by freshwater limestones. The overlying sediments of the coal sequence are marls with molluscs and foraminifers.

Although the inclination of seams is only 3–10°, the intensive fracturing due to tectonics and the unsealed direct contact to the underlying karst system mean hazardous conditions for the exploitation.

4.1.3.2. Petrography. These subbituminous coals (0.43% R_o) are bright or weakly banded (Table 1). Detrohuminite and gelohuminite, in the form of porigelinite, along elevated quantity of liptodetrinite dominate. The telohuminite content is the lowest in the Doba samples compared to the other coals studied in this work. Besides liptodetrinite, sporinite, and alginite in high quantity also occur. In the Doba 2/1 sample a high concentration of solitary algae was found. In Balinka and Doba 2/1 samples amorphinite with weak, brownish yellow fluorescence are also present. Inertinite occurs very seldom, in most cases in the form of funginite. Inertodetrinite is a minor constituent, reaching 2.8% in Balinka 2.

4.1.4. Mány Formation, MF

4.1.4.1. Geologic setting. The Vértessomló Coal Member of the Mány Formation (MF) deposited in isolated basins of limnic–paralic back barrier environment (Gyalog et al., 1996). The coal measures are found in the lower part of the Upper Oligocene Mány Formation. This coal basin developed on the denudation surface of Mesozoic sediments, which were uplifted in the Upper Cretaceous. The underlying sediments are Lower Jurassic, Upper Triassic limestone and in the central part Lower Cretaceous marl and limestone with Middle Eocene coal-bearing sequences (Fig. 5). The average thickness of the coal measures is 40 m. The direct basis of the seam is limnic clay; in the roof sandy clay and clay represent brackish conditions. MF coals have the highest humic acid content among Pannonian Basin coals. For this reason a part of the exploited coal is used as a chemical filter. The thickness of the seam is 0.2–2.4-m; interbeddings of clay, silt and travertine divide the seam into 20–40 cm thick beds. The average gross calorific value is 8.5–21 MJ/kg.

4.1.4.2. Petrography. These lignites (0.36% R_o) are well-banded with 4–5-mm thick bright bands alternating with 6–8 mm dull bands. In the telohuminite

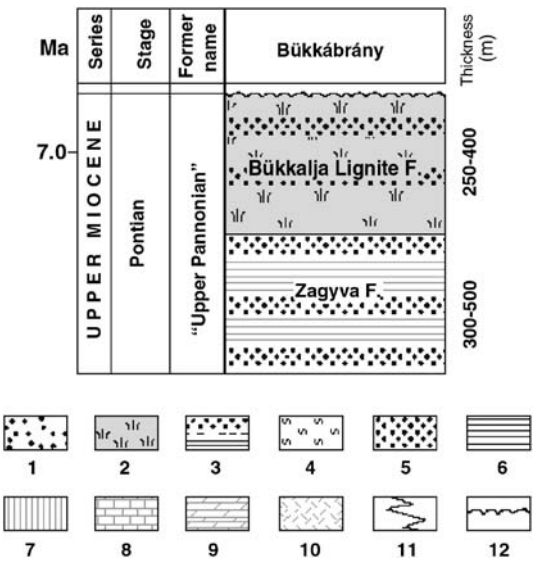

Fig. 8. Stratigraphic column of the Upper Miocene Bükkalja Lignite Formation. Legend: 1: gravel; 2: coal; 3: fluvial; 4: calcareous marl; 5: sandstone; 6: marl; 7: clay; 8: limestone; 9: dolomite; 10: rhyolitic tuff; 11: heteropic facies; 12: erosion unconformity.

Table 3
Summarised chemical and isotopic data of the studied coals

Age		$\delta^{13}C$	$\delta^{34}S_{org}$	$\delta^{34}S_{pyr}$	S_{org}	S_{pyr}	S_{sul}	S_{total}	Ash
		‰	‰	‰	%	%	%	%	%
L. Jurassic	Average	−26.09	10.22	9.55	2.33	0.65	0.27	3.25	21.34
	Minimum	−27.58	7.51	4.34	0.59	0.02	0.01	0.62	14.80
	Maximum	−24.54	16.27	16.43	4.00	1.53	0.67	6.10	27.90
	STD	0.91	3.54	4.53	1.63	0.65	0.25	2.47	5.54
	Number	10	4	4	4	4	4	4	4
U. Cretaceous	Average	−24.39	12.53	9.18	4.07	0.61	0.69	5.37	12.87
	Minimum	−25.20	12.09	8.86	3.59	0.43	0.23	5.21	11.60
	Maximum	−23.92	12.97	9.50	4.55	0.79	1.14	5.52	14.13
	STD	0.35	0.44	0.32	0.48	0.18	0.46	0.16	1.27
	Number	14	2	2	2	2	2	2	2
M. Eocene	Average	−25.24	15.78	12.41	3.52	0.78	0.35	4.65	15.37
	Minimum	−26.16	7.31	−3.52	1.77	0.20	0.04	2.50	6.09
	Maximum	−24.50	19.05	17.15	4.49	2.05	1.55	8.00	42.12
	STD	0.48	3.92	7.20	1.02	0.62	0.54	1.86	12.11
	Number	20	6	6	6	6	6	6	6
Oligocene	Average	−27.06	7.44	9.59	0.57	0.23	0.08	0.85	19.45
	Minimum	−28.53	7.44	7.04	0.47	0.22	0.06	0.69	18.84
	Maximum	−26.47	7.44	12.14	0.66	0.24	0.10	1.00	20.05
	STD	0.75	0.00	2.55	0.10	0.01	0.02	0.16	0.61
	Number	5	1	2	2	2	2	2	2
Miocene	Average	−26.27	12.43	8.89	2.20	1.07	1.25	4.53	22.83
	Minimum	−28.54	−2.41	−4.88	1.13	0.38	0.57	2.21	10.32
	Maximum	−24.24	28.08	27.87	3.40	3.30	2.75	9.31	34.62
	STD	1.00	8.94	9.13	0.78	0.88	0.72	2.16	7.84
	Number	35	8	8	8	8	8	8	8
Total	Average	−25.74	12.74	10.06	2.61	0.80	0.67	4.07	19.31
	Minimum	−28.54	−2.41	−4.88	0.47	0.02	0.01	0.62	6.09
	Maximum	−23.92	28.08	27.87	4.55	3.30	2.75	9.31	42.12
	STD	1.10	6.52	7.13	1.37	0.73	0.71	2.27	9.02
	Number	84	21	22	22	22	22	22	22

Legend: STD—standard deviation; Number—number of samples calculated in the statistic; explanation of other abbreviations is available in Table 2.

subgroup, ulminite is the common maceral. Textinite occurs only sporadically in Vértessomló 3 sample. In this latter sample the highest amount of gelinite (25.7%), in the form of porigelinite, is observed. In Vértessomló 1 sample one-third of the humodetrinite is of leaf origin. Aquatic conditions are indicated by the high quantity of liptodetrinite (2.8–6.6%) and by the concentration of sporopollenin layers in Vértessomló 2.

4.1.5. Salgótarján Brown Coal Formation, SBF

4.1.5.1. Geologic setting. The Lower Miocene Salgótarján Brown Coal Formation (SBF) located in Northern Hungary, is mainly exposed in outcrops, underground, and open pit mines in the Borsod Basin. The Borsod Basin is surrounded by the Paleozoic–Mesozoic basement blocks of the Bükk Mt., Uppony Mt., Rudabánya Mt., and Szendrõ Mt. (Fig. 1). The formation of the Borsod Basin is associated with the early "synrift" phase of the development of the Pannonian Basin (Pogácsás, 1984). The Borsod Basin was a set of tectonically preformed, narrow seaways and their heteropic sedimentary facies (Hámor, 2001) including peat-forming environments on the north-east margin of the so-called Central Paratethys ca. 18–20 Ma ago.

This formation is a 200–300 m siliciclastic sequence of cyclic repetition of sand, silt, and silty clay; including five paralic lignite seams in the east (Vadna, Edelény) and three lignite seams in the north-west part of the basin (Székvölgy). It was deposited unconformably on Paleogene sedimentary rocks or on the Lower Miocene Gyulakeszi Rhyolite Tuff Formation (Fig. 6). SBF was deposited in a coastal plain environment and was covered by a marine turbidite sequence. The coal seams are numbered from the top Nos. 1, 2, 3, 4, 5, respectively. The average thickness of lignite seams is ca. 2 m. The two lowermost seams (Nos. 4, 5) are intercalated by marine and reworked tuffitic sediments. They split the lignite

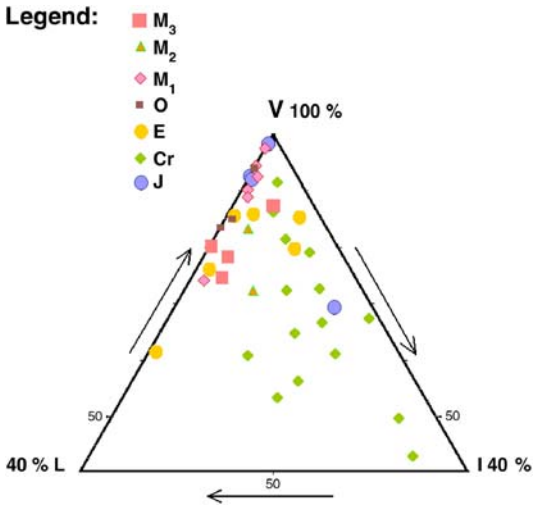

Fig. 9. Ternary diagram of maceral composition.

seam into 0.2–0.6 m thick beds. In seams Nos. 5 and 2, the uppermost layers and the layers underlying tuff intercalations are often silicified, the thickness of these petrified lignite layers varies between 0.1–0.3 m. The gross calorific value of the SBF is 10.5–23.5 MJ/kg.

4.1.5.2. Petrography. These lignites (0.26 to 0.33% R_o) are hard, dark brown or black with bright banding, and banded coal lithotypes (Hámor-Vidó, 1992).

Telohuminite with texto-ulminite and eu-ulminite dominate (Hámor-Vidó, 1992, 1993). The degree of preservation is characterized by the appearance of texto-ulminite with resinite cells of *Taxodiacea* tissues (e.g. Székvölgy 3/4 and Edelény 4/4 samples). The highest vegetation index (Table 1) of the Edelény 2/2 sample is most likely due to the diagenetic silification affecting one-fourth of the macerals. The wettest conditions existed during the formation of Edelény 4/4, as indicated by *Botryococcus*-type algae. Degradinite was found only in Székvölgy 3/4 and Edelény 4/4 samples (1.1% and 7.8%, respectively). Funginite represents the inertinite group, in agreement with the works of Juhász (1970, 1988).

4.1.6. Hidas Brown coal Formation, HB

4.1.6.1. Geologic setting. The Middle Miocene Hidas Brown Coal Formation (HB) is situated in the central part of the Transdanubian Mountain Range. Coal measures formed in back barrier, strand plain environment. The peat-forming environment developed in paralic-type lagoons of oscillating shoreline and water-depth (Kókay, 1987). The thickness of the coal measures is 70–140 m, the overlying sediments are marine limestones (Fig. 7). In the NE at Várpalota one seam is developed to a thickness of 10 m. This seam is divided into two to three layers in SW direction. The overburden of the coal seam is a 0.5 m thick, finely laminated oil shale. Gross calorific value of HB is 9–18 MJ/kg.

4.1.6.2. Petrography. These lignites (0.32% R_o) are in transition between soft and bright brown coal (Stach et al., 1982). Usually they are consolidated dark brown, dull coals, but finely laminated or light lithotypes also occur (Table 2a, b, c). Detrital material dominates the huminites, the ratio of attrinite to densinite being 1:1. Amorphinite and sporinite, mostly dissacate pollens, are characteristic for HB. These macerals with laminated alginite in Várpalota 6 sample represent aquatic conditions during the peat formation.

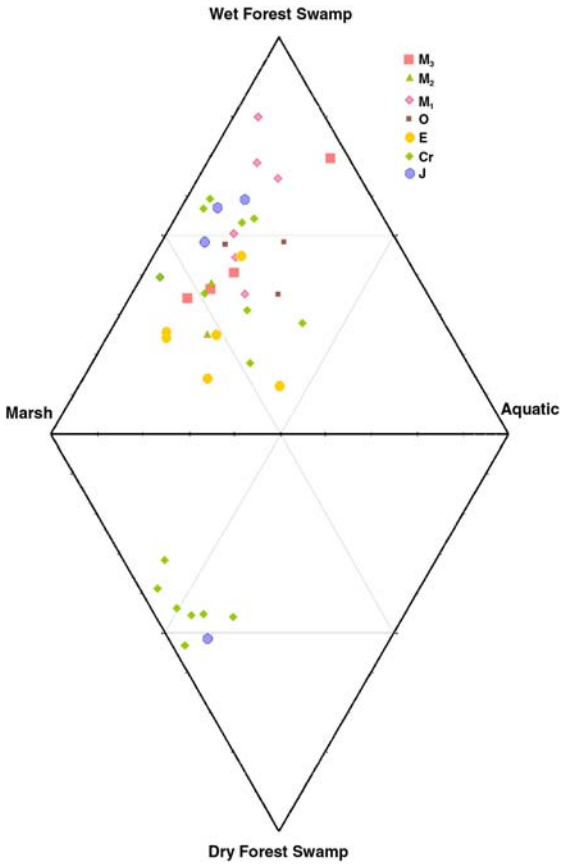

Fig. 10. Ternary facies diagram. The depositional environment is indicated by the organic and mineral matter composition. Modified after Hámor-Vidó, 1992. Legend: M_3 — Bükkalja Lignite Formation, BL; M_2 — Hidas Brown Coal Formation, HB; M_1 — Salgótarján Brown Coal Formation SBF; O — Mány Formation, MF; E_2 — Dorog Coal Formation, DF; Cr_3 — Ajka Coal Formation, ACF; J_1 — Mecsek Coal Formation, MCF.

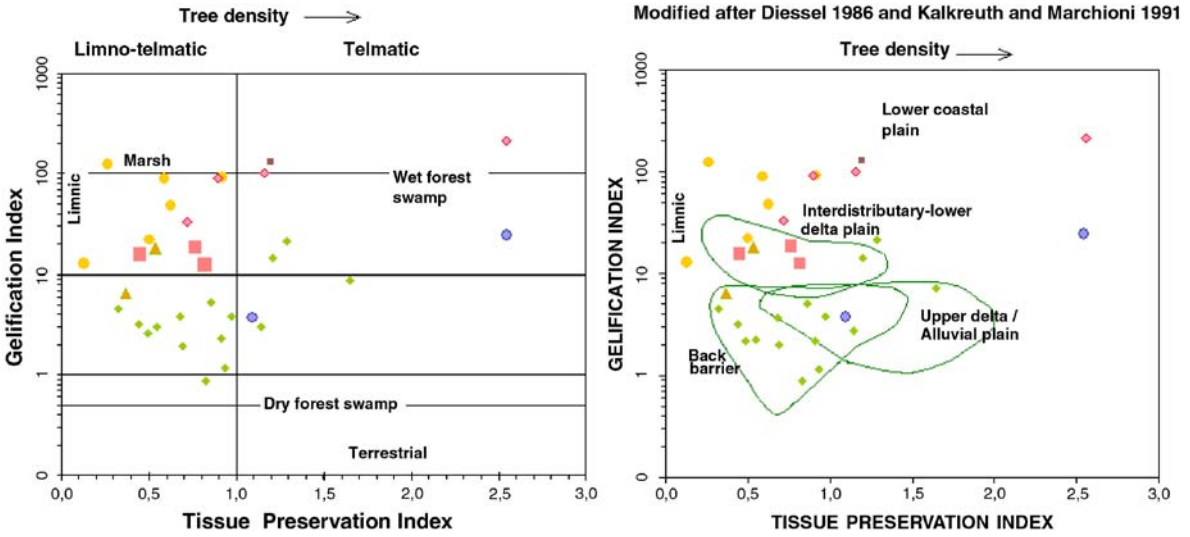

Fig. 11. TPI/GI facies diagram. Modified after Diessel (1986) and Kalkreuth and Marchioni (1991).

4.1.7. Bükkalja Lignite Formation, BL

4.1.7.1. Geologic setting. Lignite formation took place during the Late Miocene (ca. 8.8–6.5 Ma; Lantos et al., 1992) in the delta system of the Pannonian Basin. The productive area of the Bükkalja Lignite Formation (BL) is in the southern foreland of the North Hungarian Uplands in an E–W range of 120 km length and 10 km width. The denudation surface of Lower Miocene rhyolitic tuff and fluvial sediments was the basement of peat formation (Fig. 8). The thickness of coal sequence is ca. 300–400 m, the number of seams is 15 to 30. The average thickness of lignites is 1.5 m, coaly shale and clay intercalations are frequent. Two to three seams are economic; the gross calorific value is 8.5–18 MJ/kg.

4.1.7.2. Petrography. These lignites (0.27 to 0.29% R_o) are light fibrous lithotypes, where the woody texture is well recognisable or medium-dark earthy and dull. The lignites consist of huminite, in which texto-ulminite, eu-ulminite and attrinite are the dominant macerals. Gelinite usually is a minor part of the huminites except in Bükkábrány 1 sample (14.9%). Texto-ulminite frequently contains cellulose and in some samples (e.g. Bükkábrány 4) the gymnosperm woody tissue is strongly degraded. It shows a bright-yellowish fluorescence and is gelified, often structureless. Resinite, cutinite and sporinite represent liptinite. In Bükkábrány 3 sample, liptodetrinite and *Botryococcus*-type alginite occur in high amounts. Inertinite, mainly semifusinite and fusinite, is characteristic in the upper third of the two sections.

4.2. Analytical chemical data

The ash content of the studied coals and associated organic-rich sediments varies between 6.09–42.12% (mean value: 19.31%; Table 2a, b, c). Miocene coals are characterized by the highest ash volume (mean = 22.83%), Upper Cretaceous coals contain the least (mean = 12.87%). The total sulphur content is rather high (4.07% mean value), most of these coals qualify as high-sulphur coals according to the international practice. There are significant differences within the sequences (0.3–9.7%; Tables 2a, b, c and 3). Upper Cretaceous coals are extremely rich in sulphur (5.37%); Lower Jurassic, Eocene and Miocene coals have high sulphur content as well (3.25%, 4.53%, 4.22%); and only the Oligocene samples qualify as low-sulphur coals (0.85%).

The major part of the sulphur content is associated to the organic matter (2.61% mean value), which coincides with the reported global average (Casagrande and Ng, 1979). Significantly less sulphur is sulphide (0.80% mean value) and the smallest fraction is sulphates (0.67% mean value). Exceptions from this trend are coals of Székvölgy III/4, where pyrite sulphur content is higher than organic-bond sulphur; and Jókai III/10, Székvölgy III/5, Edelény IV/4, Edelény II/2, Vadna V/2, Várpalota 6, Szászvár 2, Bükkábrány 3, where sulphate sulphur exceeds sulphide sulphur content. It is very much likely that despite the careful sampling and storage, these latter coals were exposed for too long time to weathering in the mine or during oxidation in the laboratory. However, the sum of the sulphate and sulphide contents is still lower

than the organic sulphur content in most of these samples. The average S_{pyrite} + $S_{sulphate}$ content in coals of different ages is rather similar (0.92–2.32%), while Oligocene coals have a significantly lower value of 0.31%.

4.3. Carbon isotopes

The carbon isotope ratios of Pannonian Basin coals vary over a wide range of values from −23.92 to −28.54‰ (Table 2a, b, c). The mean values of coals of different ages fit well to the secular variation of terrestrial organic matter for the correlative time period but the range is somewhat higher than the relevant data in the literature (e.g. for Oligocene–Miocene TOC in; Pagani et al., 1999). The average $\delta^{13}C$ value is most depleted in the Oligocene coals (−27.06‰), while the Upper Cretaceous coals represent the other extreme (−24.39‰) (Fig. 12). As it is mentioned above, the high sulphate content relative to sulphide sulphur in eight coal samples indicates most likely an oxidation of the reduced sulphur phases. According to Lewan (1983), the slow oxidation of the coals is not accompanied by significant fractionation.

4.4. Sulphur isotopes

The $\delta^{34}S$ ratios of both organic and sulphide phases show a moderate enrichment of the light sulphur isotopes relative to the isotopic composition of seawater sulphate, with a mean value of 12.74‰ for $\delta^{34}S_{organic}$ and 10.06‰ for $\delta^{34}S_{pyrite}$, respectively (Table 3, Fig. 13). The heaviest average sulphur isotopic compositions are in Eocene coals in both organic and sulphide fractions, 15.78‰ and 12.41‰, respectively.

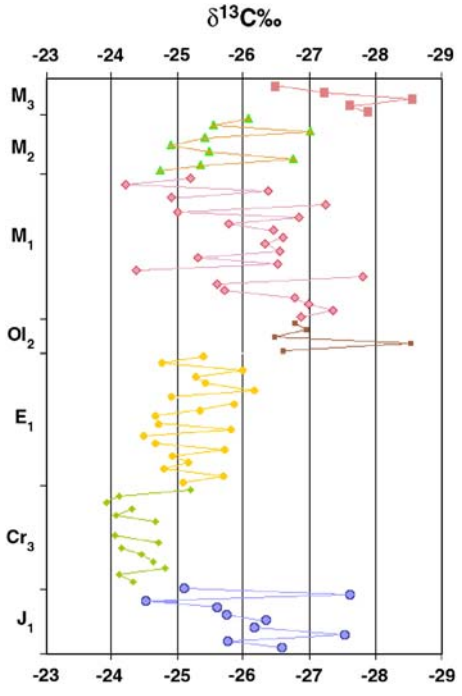

Fig. 12. Stable carbon isotope ratio changes with stratigraphic position.

5. Discussion

5.1. Organic petrology and the depositional environment

Maceral composition of the selected 40 samples represents the huminite character of the above mentioned coal formations (Fig. 9). The average huminite/vitrinite, liptinite and inertinite contents are 78.2%, 11.3% and 10.4%, respectively. The same type and character, i.e. relatively low liptinite and inertinite contents, are observed in the Jurassic, Oligocene and Miocene coals. Eocene coals reveal on average the highest liptinite content with a high portion of sporinite and liptodetrinite and a low abundance of inertinite in the form of inertodetrinite. With regard to maceral composition the most distinct coal is this of ACF. This coal is usually rich in inertinite in the form of inertodetrinite and in some beds characterized by elevated amount of allochthonous resinite. The depositional environment is indicated by the double ternary diagram of Hámor-Vidó (1992) and by the modified facies indices of Diessel (1986, 1992) and Kalkreuth et al. (1991).

We used both ternary and TPI/GI diagrams to distinguish in parallel peat-forming environments because the application of TPI/GI diagram is limited when inertinite content is too low or not present. The two facies diagrams give similar results for interpretation and complement each other. For example, some MCF, MF, SBF, BL samples deposited in wet forest swamp conditions can be displayed only in the ternary diagram. On the contrary, the depositional environment of the Upper Cretaceous ACF coals could be delineated exclusively on the TPI/GI diagram.

According to the ternary diagram, Lower Miocene SBF and Jurassic MCF coals were formed dominantly under wet forest swamp conditions together with some samples of Cretaceous ACF coals (Fig. 10). Upper Miocene BL, Middle Miocene HB and Oligocene MF coals deposited in intermediate swamp–marsh complex environment under limno-telmatic conditions.

Cretaceous ACF coals show extreme depositional environments from the wet forest–swamp to marsh and dry forest–swamp environments depending on the magnitude of organic matter transportation and/or reworking. The other extreme is the Middle Eocene

DF, where coals formed in marsh or in the vicinity of intermediate swamp–marsh complex environment and the characteristic coal types are of limnic, limnotelmatic conditions after the TPI/GI diagram (Fig. 11). The published sedimentological and paleontological interpretations of the formations generally correspond well with the TPI/GI depositional environments.

5.2. Carbon chemistry

There are some systematic data available in the literature on carbon isotopic variation in coal (Deines, 1980; Whiticar, 1996; Lücke et al., 1999; Bechtel et al., 2001, 2002, 2003a,b). Coals are of initially heterogenous material composition, the bulk isotopic composition is the result of its various constituents. For example, immature brown coals consisting of different macrolithotypes and chemical composition can show considerable isotope variations (Rigby et al., 1981; Smith et al., 1982; Bechtel et al., 2001, 2002, 2003a,b). In more mature coals isotopic differences between petrographic varieties are no longer detectable (Geissler and Belau, 1971). During the later catagenesis stage of coalification, coals do not change their carbon isotopic composition (Lewan, 1983).

Coal facies is predetermined by the peat-forming vegetation, water and nutrient supply, marine influence, and fire (Diessel, 1986). Some recent studies outline isotopic differences between gymnosperm-and angiosperm-dominated plant communities. The differences have been demonstrated on the basis of fossil wood and wood cellulose from Tertiary lignite deposits (see Lücke et al., 1999; Bechtel et al., 2003a). Stable carbon isotope ratio measurements are widely used to map the above mentioned paleoenvironmental characteristics of peat-formation and coal genesis.

Atmospheric carbon dioxide is the ultimate source of all natural carbon phases. The photosynthetic assimilation of carbon from the atmospheric carbon is accompanied by a kinetic isotope fractionation in which ^{13}C becomes depleted in plants. Because of the different pathways in photosynthetic biochemistry, C3 and C4 plants discriminate differently against the heavier carbon isotope (Emrich et al., 1970). Consequently, tissues of C3 plants, such as trees, have an average $\delta^{13}C$ value of −27‰, whereas C4 plants, which are mainly grasses, have an average of −12‰. In topogenous mire of terrestrial environment coals consists mainly of tree and bushes. Marine coal deposits may contain grass or herbaceous plant communities. Within both plant types, genetic and environmental factors such as temperature, precipitation, and soil water saturation can cause more subtle variability in the carbon isotopic composition (Smith et al., 1976; Leavitt and Long, 1982; Spiker and Hatcher, 1984; Dupouey et al., 1993). The nature of the CO_2 source itself is one of the most important environmental factors. For example, significant spatial carbon isotope gradients can exist across the same ecosystem between different species and also within a single species due to the recycling of locally available depleted CO_2 (Martinelli et al., 1991; Raymond and Bauer, 2001).

Some recent studies outline isotopic differences between gymnosperm-and angiosperm-dominated plant communities. The differences have been demonstrated on the basis of fossil wood and wood cellulose from Tertiary lignite deposits (see Lücke et al., 1999; Bechtel et al., 2003a). These differences may also contribute to the variability seen in ä^{13}C of total organic carbon of the coals.

$\delta^{13}C$ values of subbituminous coal can provide information on the distribution of plants within a regional flora, as organic matter derived from individual plant families show different isotope signals (Smith, 1972; Smedley et al., 1991). Variations of carbon isotopes in plant organic matter have been related to changes in atmospheric carbon dioxide concentrations (Beerling, 1997), while in other studies isotopic shifts were attributed to climatic changes (Holmes et al., 1991) and even orbital forcing (Jones et al., 1996).

Limited attention was paid to the application of carbon isotopes in studying the diagenesis of coal. The few relevant works showed that the preservation of organic matter strongly depends on early diagenetic physico-chemical processes such as the transformation of cellulose and lignin to fulvic and humic acids (Hedges et al., 1985; Spiker and Hatcher, 1987; Benner et al., 1987; Opsahl and Benner, 1995). Similar conclusions were drawn by Hámor-Vidó and Hertelendi (1996). Studies on recent and ancient wood samples (Hatcher and Breger, 1981) showed that there is a linear correlation between chemical composition and $\delta^{13}C$ ratio changes during early diagenesis (Spiker and Hatcher, 1987; Rimmer et al., 2006-10-31). Degradation and loss of cellulose in buried wood may result in 1–2‰ of $\delta^{13}C$ fractionation.

The $\delta^{13}C$ pattern of Pannonian Basin coals shows a significant change with stratigraphic setting (Fig. 12). However, this is not due to global variation of OM composition but to the paleoenvironmental changes. A major point of argument is that, according to the maceral composition, the original OM source differed a lot in these formations. For example Cretaceous coals display a separate cluster on the maceral composition diagram

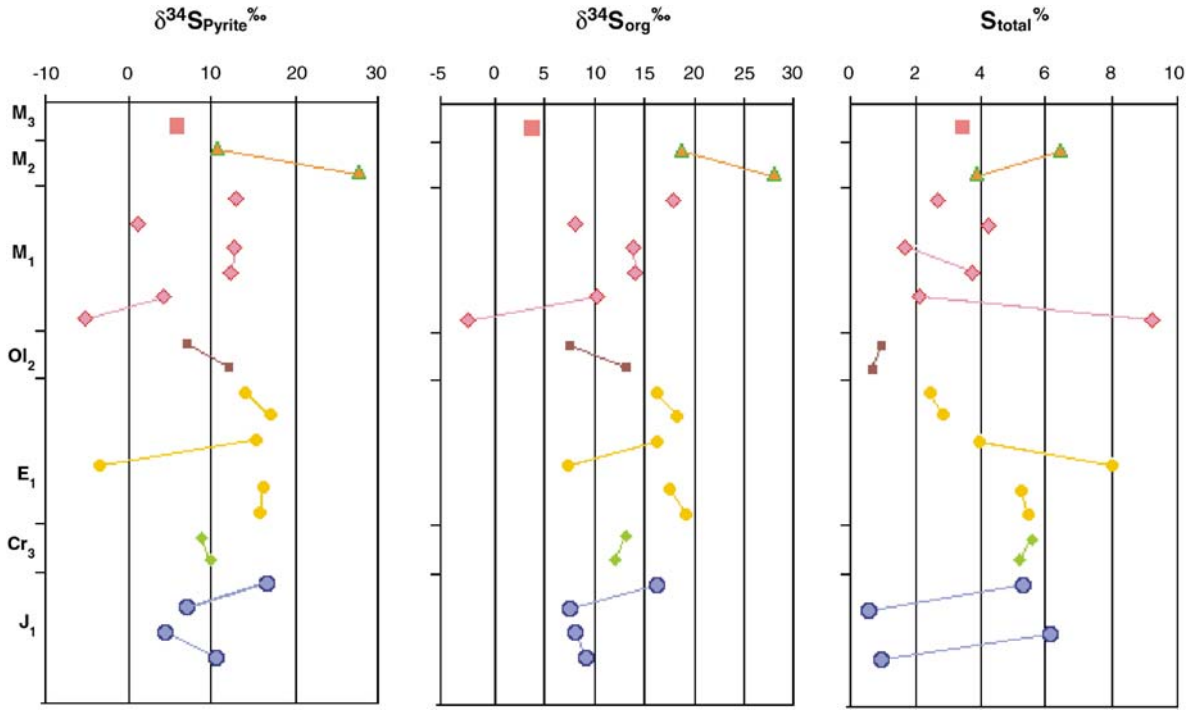

Fig. 13. Stable sulphur isotope ratio changes of different sulphur phases and the total sulphur content changes with stratigraphic position.

(Fig. 9), having the least vitrinite content indicating the dominance of a vegetation with less depleted $\delta^{13}C$ figures (Fig. 12). It is known that especially fusinite is enriched in ^{13}C with respect to the non-oxidized organic matter. The abundant inertinite present in Cretaceous coal can be a reason for the high $\delta^{13}C$ values of the coals.

The Eocene coals have similar character, 1‰ more depleted on average, and both formations reveal high detrovitrinite and liptinite content, an additional evidence of the herbaceous vegetation origin. The $\delta^{13}C$ values of Jurassic, Oligocene and Miocene coals are significantly more depleted than these corresponding to the original woody material of C3 photosynthesis (Fig. 12, Table 3).

The less depleted −23‰, −24‰ $\delta^{13}C$ ratios of some layers might show the effect of marine incursion of the SBF, DF and ACF samples (Fig. 12). An alternative explanation is the shift of the deposition and peat formation to seaward, heteropic sedimentary facies of normal marine water regime. For example Székvölgy 3/4, Doba 2/1 and Ajka 6/3 samples appear in the lower delta plain domain in the TPI/GI diagram, show depleted $\delta^{34}S_{pyrite}$ and relatively enriched $\delta^{13}C$ ratios. A good evidence of a marine incursion is provided by the high percentage (20%) of alginite and liptodetrinite

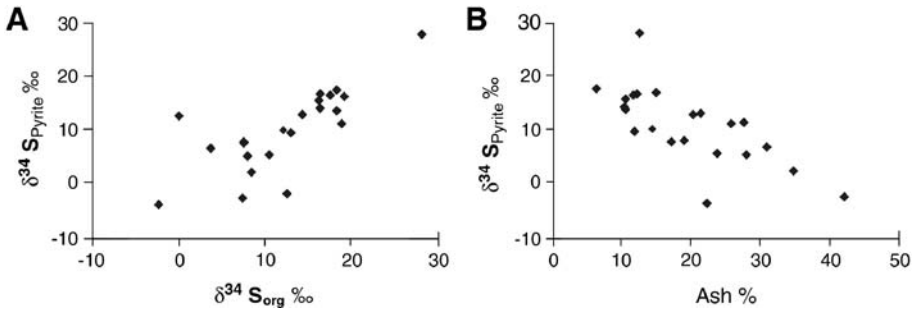

Fig. 14. Relationship between the $\delta^{34}S_{organic}$ and $\delta^{34}S_{pyrite}$ and crossplot of the ash content *versus* $\delta^{34}S_{pyrite}$.

in Doba 2/1 layer. Another evidence that Sachsenhofer and Tomschey (1992) published, is the extremely high boron composition in the seam that corresponds to the Ajka 6/3 sample.

In general, this is in good accordance to the relevant palynological and sedimentological data. Statistically there is no correlation found between maceral composition and isotopic variations. The studied coal seams represent a significant time span; peat formation and coalificiation are spatially and temporally integrating, homogenizing processes, thus isotopic values recorded in bulk coal samples represent taxonomic, sedimentary and preservation variations at large.

5.3. Sulphur chemistry

Sulphur in coal occurs in several forms, including iron mono-and disulphides, sulphates, elemental sulphur, and organically bound sulphur (Renton and Bird, 1991). Pyrite and marcasite are the most abundant minerals (Smith and Batts, 1974; Casagrande, 1987). While the formation of iron sulphides is well-established (Berner, 1984), the processes of incorporation of sulphur into plants, peat, and coal are less well known. Organically-bound sulphur can be either preserved from the original plant material or derived from reduction of sulphur contained in the organic debris through microbial processes (Chambers and Trudinger, 1979). Thiols, sulphides and disulphides, and thiophene and their derivatives are thought to be several kinds of sulphur compounds that occur in coals (Dai, 2000).

The primary depositional environment plays an eminent role in the overall quantity and distribution of sulphur among organic and sulphide phases (Altschuler et al., 1983; Phillips and Bustin, 1996) and, to a certain extent, in their sulphur isotopic distribution. The major source of sulphur in peat is the sulphate dissolved in the water and in the sedimentary pore water of the peat-forming depositional system (Price and Casagrande, 1991). Sulphate concentrations are typically a hundred times higher in seawater than in freshwater; therefore peat formed under marine influence, in general, reveals higher sulphur contents than freshwater peat. According to Chou (1997) sulphur available during plant growth is the principal source of sulphur in low-sulphur (<1% total sulphur) coals and there the $\delta^{34}S_{organic}$ values range between 2–8‰. In contrast, in medium- (1–3% total sulphur) and high-sulphur (>3% total sulphur) coals overlain by a marine roof, most of the sulphur is derived from seawater sulphate (Diessel, 1992). The assimilation of sulphur by plants does not involve significant isotopic fractionation (Chukhrov et al., 1980).

The mechanism responsible for the major fractionation of sulphur isotopes during diagenesis is the bacterial sulphate reduction (e.g. Altschuler et al., 1983; Berner, 1984; Casagrande, 1987; Bottrell and Raiswell, 2000; Dellwig et al., 2000). Sulphate-reducing bacteria discriminate against the heavy ^{34}S isotopes, which leads to the generation of hydrogen sulphide depleted by 20–40‰ relative to the original sulphate. The hydrogen sulphide produced is a major source of incorporated sulphur in peat. The available reactive iron phases react with the hydrogen sulphide and, through a precursor monosulphide phase, iron-disulphide (dominantly pyrite) forms with a negligible isotopic fractionation. However, in sedimentary environments of rapid deposition interstitial spaces are sealed rapidly, thus inhibiting the continuous sulphate supply during the early diagenesis. This process results in less depleted and even positive $\delta^{34}S$ ratios of the pyrite. In certain depositional settings, for example in coastal swamps of a carbonate platform, the poor availability of reactive iron phases can limit the formation of iron-sulphides by letting H_2S to escape, thus leading to relatively heavy sulphur isotopic values of both organic and sulphide phases.

In most of the Pannonian Basin coal formations the $\delta^{34}S_{pyrite}$ and $\delta^{34}S_{organic}$ ratios are highly positive, enriched in the heavy sulphur isotopes (Table 2a, b, c). There is a positive correlation between them (Fig. 14a), which is most likely an indication of a common sulphur source and isotope fractionation history. The correlation of the S_{pyrite} and $S_{organic}$ contents suggests similar formation pathways as well. Similar $\delta^{34}S_{organic}$ ratios are reported from the Everglades (Price and Casagrande, 1991) and from Australian coals (Smith and Batts, 1974). Similar and even more positive $\delta^{34}S_{pyrite}$ values were measured in the Herrin Coal Member of Illinois (Westgate and Anderson, 1984). The difference between the isotopic composition of organic and sulphide sulphur is typically +1–3‰, but −2.15‰ occurs in low-sulphur Oligocene coals (MF) of lacustrine facies (Fig. 13). However, these differences are within the range reported for low-sulphur Permian coals by Hunt and Smith (1985).

$\delta^{34}S_{pyrite}$ *versus* ash content (Fig. 14b) shows a good negative correlation, which is most likely a consequence of higher availability of sulphate and the resulting higher fractionation during pyrite formation in a more open system with respect to SO_4^{-2} supply, characterised by a higher ash (i.e. sediment and pyrite) content.

The highly positive $\delta^{34}S_{pyrite}$ ratios, with a maximum value of +27.87‰ in BL, are most likely the result of the dominantly closed diagenetic pore spaces due to the

high deposition rates in freshwater of deltaic setting. The general paleogeographic scheme and the maceral analysis data support this. The dominant part of the studied coal seams does not contain epigenetic sulphur phases, except of the Százsvár and Vadna samples, where the veins of pyrite were hand-picked before analytical works. The sampled sites were located on the marginal parts of the Tethys during the Mesozoic and of the Paratethys during the Neogene. The maceral data indicate a freshwater regime for the peat formation in the majority of the samples. The same explanation for the origin of organic-sulphur might stand but the isotopic composition of the original plant material and the incorporation of elemental sulphur during early diagenesis can result in the same heavy isotopic composition. For certain stratigraphic intervals (Permian, Triassic, Miocene) $\delta^{34}S$ data of primary sulphate evaporites are available (Hámor, 1996). Most of those data fit well to the global secular curve of the sulphur isotopic composition of seawater sulphate but the high deviation in $\delta^{34}S$ of the Miocene evaporites of the Pannonian Basin was interpreted as the result of the marginal paleoenvironmental location affected by freshwater inflow, and high depositional rates. Similar conclusions were drawn by Hámor and Hertelendi (1991) when studying the sulphur isotopic pattern of the different mineral forms of pyrite in Pannonian Basin formations. In harmony with the above character, the isotopic ratios of Miocene coals of our study are the most scattered ($\delta^{34}S_{organic}$ from −2.41 to +28.08‰, and $\delta^{34}S_{pyrite}$ from −4.88 to +27.87‰).

Sulphur isotopic fractionation is a record of the diagenetic history of the given sedimentary pore space which is a function of the characteristics of the sedimentary environment (marine *vs*. freshwater, organic-rich *vs*. organic-poor, or deposition rate, availability of reactive iron phases) that governs the supply, enrichment, and incorporation of sulphur isotopes into sulphide phases.

The mean chemical and isotopic values of the coal formations (Table 3) and the interpretation of sedimentary facies on the basis of maceral composition (Table 2a, b, c) give a more detailed insight into peat formation and corroborate each other in the proposed general model. Oligocene coals represent one extreme end member, deposited in a lacustrine environment. MF is characterised by the most depleted $\delta^{13}C$ ratios, the lowest S_{total} and S_{pyrite} contents. Upper Cretaceous coals indicate deposition in the most marine-influenced environment. These samples have the least depleted $\delta^{13}C$, the most depleted $\delta^{34}S_{pyrite}$ ratios, the least ash content and the highest S_{total} content. However, there are individual seams which are exceptional in the general trends. For example the Doba 2/1 seam of the Eocene DF and the Székvölgy 3/4 seam of Miocene SBF have the most depleted $\delta^{34}S_{pyrite}$ values (−3.52‰ and −4.88‰, respectively) and the highest total sulphur content with 8.00% and 9.31% respectively, which is a strong indication of marine influx, in spite of the general character of their formations.

There might be an alternate interpretation for the sulphur isotopic values obtained. In general, the coastal basin setting can produce local brackish water environments by the intermittent influx of sulphate from marine flooding or storm surges. In addition, H_2S can readily escape such a system and if the rate of escape is matched to the rate of replenishment by storms then the shift in sulphur isotopic ratios will have a similar pattern. However, at the present sampling frequency of the sections and in lack of other decisive methods, such as detailed micropaleontology and high resolution sedimentology, this latter explanation can not be excluded as a complementary to the described general paleogeographic model. Nevertheless, the sample set and the generated data make a further refinement of the facies analysis feasible.

6. Conclusions

The maceral composition, the chemical and C, S isotopic data based on the detailed sampling campaign of the Pannonian Basin coals allowed to draw general conclusions on the paleoenvironment, including the early diagenetic history, of the peat deposition. The maceral composition, the sulphur composition, and the C and S isotopic signatures show that the majority of these coals were deposited in a dominantly freshwater, to a lesser extent brackish water environment, in spite of the relatively high sulphur content. However, some of the Upper Cretaceous, Middle Eocene, and Middle Miocene formations include coals of marine origin, as indicated by their maceral composition and sulphur and carbon contents. The major part of the sulphur contained is associated with the organic matter. All sulphur phases are rich in ^{34}S isotopes; this indicates that marine bacterial sulphate reduction played a minor role in their formation and/or the interstitial spaces of the peat were rapidly closed during the early diagenesis thus inhibiting the replenishment of dissolved sulphate and leading to a relative enrichment of the heavy sulphur isotopes.

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