



In seam variation of element-oxides and trace elements in coal from the eastern Ordos Basin, China

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ABSTRACT

This paper investigated the mineralogical and geochemical compositions of the upper Pennsylvanian coal from Hedong Coalfield, in the eastern Ordos basin, China, using SEM, XRD, ICP-MS, ICP-CCT-MS, DMA-80 and ISE methods. The bituminous coal samples were collected from Nos. 3 and 4 Coal, which had low ash yield and were medium volatile, and had low sulfur content and low hazardous trace elements concentrations (Be, F, As, Hg, U and Pb). Maceral compositions were dominated by vitrinite (up to 90.2%), and to a lesser extent by inertinite and liptinite, all subgroups were detected in the coal samples. Minerals included quartz, clay minerals, sulfide and other oxide minerals. SiO₂ and Al₂O₃ were the dominant major elements-oxides in both coal seams with much higher concentrations than in Chinese coal, while other major elements-oxides (Na₂O, K₂O, Fe₂O₃, MgO) had average values compared to Chinese coal. No. 3 Coal was enriched in Li and Zr, and No. 4 Coal was enriched in Li, Be, Cu, Se, and In. Elemental occurrence was studied using statistical methods, and Li, Rb, Zr, Cs, Ba, Hf, Tl, Pb, and Th were inorganic affinities, while F, V, Cr, Ga, Ni, As, In, Ta occurred as inorganic–organic affinities and Fe and S may have occurred in pyrite. Coal facies indicators (GI, TPI, GWI, and VI) plots suggested that the two coal seams were deposited in different environments: No. 3 Coal was deposited in a wet forest swamp, while No. 4 Coal was formed in a mesotrophic to ombrotrophic environment. The geochemistry of these coal samples (Al₂O₃/TiO₂, TiO₂/Zr, Zr/Sc and Th/Sc ratios) indicated that the coal samples sediment was sourced from intermediate and felsic igneous rocks.

1. Introduction

Coal is the most abundant fossil fuel in China, and is a reliable long-term fuel source for China (Dai et al., 2017c) and other countries, including Turkey (Erarslan and Örgün, 2017; Erarslan et al., 2014) and South Africa (Wagner and Hlatshwayo, 2005). With the increasing use of coal in China, a large amount of pollutants has been produced, not only in the form of gas emissions but also as ash residues. Studies on the elemental geochemistry of coal have demonstrated the environmental impacts of coal. The geochemical and mineralogical compositions of coals are also mainly controlled by the sediment source and sedimentary mire environments. Previous investigations have studied the geochemistry and mineralogy of coal deposits around the world, such as coal producing areas in northern China (Dai et al., 2015a; Dai et al., 2015b; Sun et al., 2016), southern China (Dai et al., 2017b; Zhao et al., 2017), Iran (Rajabzadeh et al., 2016), Ukraine (Misch et al., 2016),

Russia (Dai et al., 2016) Canada (Goodarzi, 1987, 1988; Pollock et al., 2000) and the USA (Hower et al., 2017).

China's Liulin district, located at the Hedong Coalfield, eastern Ordos basin, is famous coal producing area, because the coal is characterized by low ash yield and low harmful elements concentrations. Previous work has focused on the relationships between coal facies and pore structure (Zhang et al., 2010). The geochemistry and mineralogy of coal in these areas have rarely been reported.

In this study, we collected “super-clean” coal from the Liulin, Hedong Coalfield, and described the petrological, geochemical and mineralogical characteristics. The main goal was to identify coal features, investigate the sedimentary conditions during peat accumulation, and study the sediment source.

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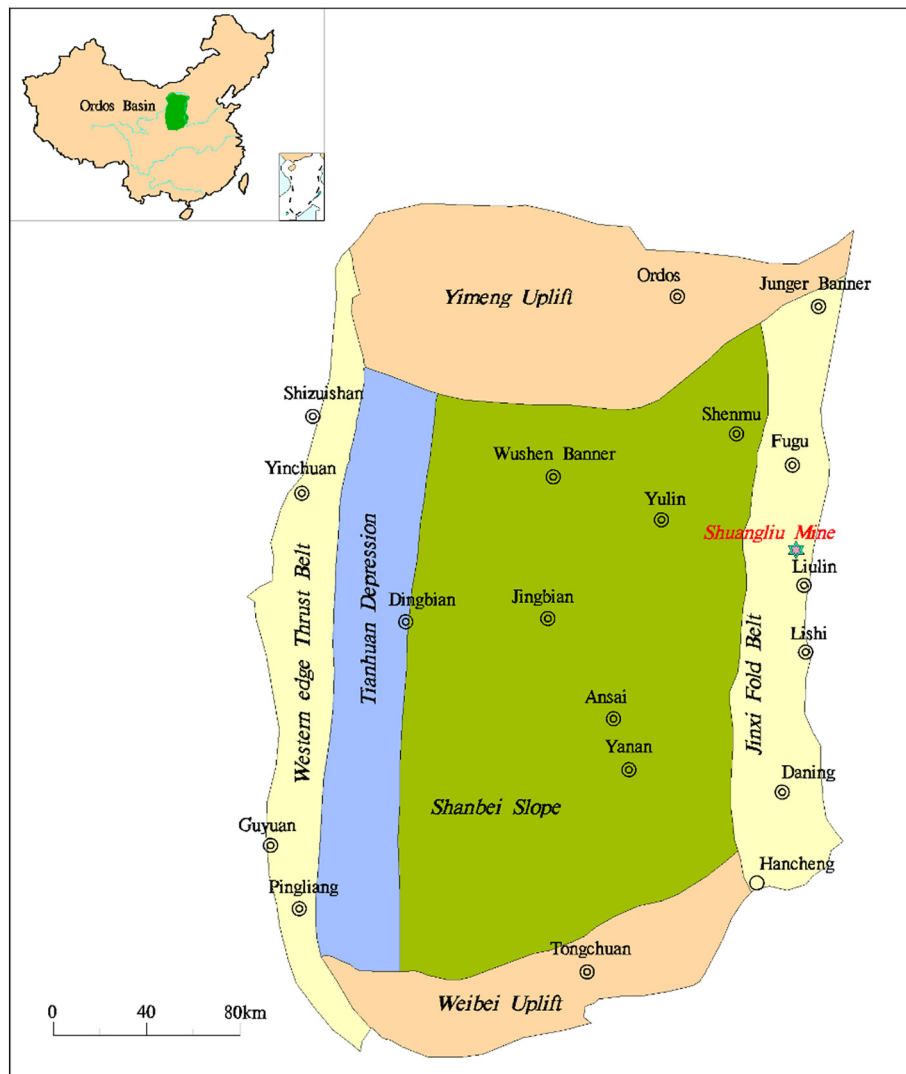


Fig. 1. The geological map of Ordos Basin and sampling location.

2. Geological setting

The Ordos basin is the oldest craton, the second largest sedimentary basin in China (Dai et al., 2006a). The Shuangliu coal mine is in the Liulin county, in the middle of Hedong coalfield, located along the eastern margin of the Ordos basin (Fig. 1). The geological setting is simple and faults are less well developed in this area (Xu et al., 2015). The stratigraphic strike is nearly NE-SW, and dips westward at 3 to 10°.

The main coal-bearing strata in the Liulin area are the upper Pennsylvanian Taiyuan Formation (C_{2t}) and the lower Permian Shanxi Formation (P_{1s}) which have a total thickness of 137–138 m and nine coal seams (Zhang et al., 2010). The main coal seams in the Taiyuan Formation are No.8 and No.9 Coal, and in the Shanxi Formation are No.3, No. 4 and No.5 Coal (Fig. 2a).

The samples in this study were collected from Nos. 3 and 4 Coal (Fig. 2b), which developed together in Shuangliu Mine and have a total thickness of 3.6 m, buried at a depth of 400 m.

3. Samples and analytical methods

The samples were collected from the faces of Shuangliu Mine along the eastern margin of the Ordos basin and included 15 coal samples (Nos.3&4 coal) and three coal bed roof and floor samples from the early Permian Shanxi formation.

Proximate analyses (Aad, Mad, and Vdaf,) of the coal samples were performed in accordance with ASTM standards (ASTM D3173-11, 2011a; ASTM D3175-11, 2011b and ASTM D3174-11, 2011c). Total sulfur was determined according to the ASTM D3177-02, 2011d standard. Mean vitrinite reflectance (%R_o) measurements and maceral analyses (500 points) were performed on the same polished section of the coal samples based on Taylor et al. (1998) and the ICCP System 1994 (ICCP, 1998, 2001). The scanning electron microscope (SEM) was used to study the distribution of minerals in the coal and the accelerating voltage was set to 20 KV, with a beam current of 10⁻¹⁰ A.

Samples were grounded to finer than 200 mesh for the major and trace elements analyses. X-ray diffraction (XRD) was performed for selected samples using a D/max-2500/PC powder diffractometer (Rigaku, Tokyo, Japan) with Ni-filtered Cu–K radiation and a scintillation detector to examine the mineral content. The major elements oxides (i.e., SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MgO, CaO, MnO, Na₂O, K₂O, and P₂O₅) were analyzed using X-ray fluorescence (XRF) spectrometry (ARL ADVANT' XP+), and the trace elements (except for As, Se, Hg and F) were determined by inductively coupled plasma mass spectrometry (X series II ICP-MS). While in pulse counting mode (three points per peak), the samples were digested in an UltraClave Microwave High Pressure Reactor (Milestone, Sorisole, Italy). More details are given in previous reports (Dai et al., 2015a; Dai et al., 2012b; Yang et al., 2016). Arsenic and Se were analyzed using a collision/reaction cell technology for

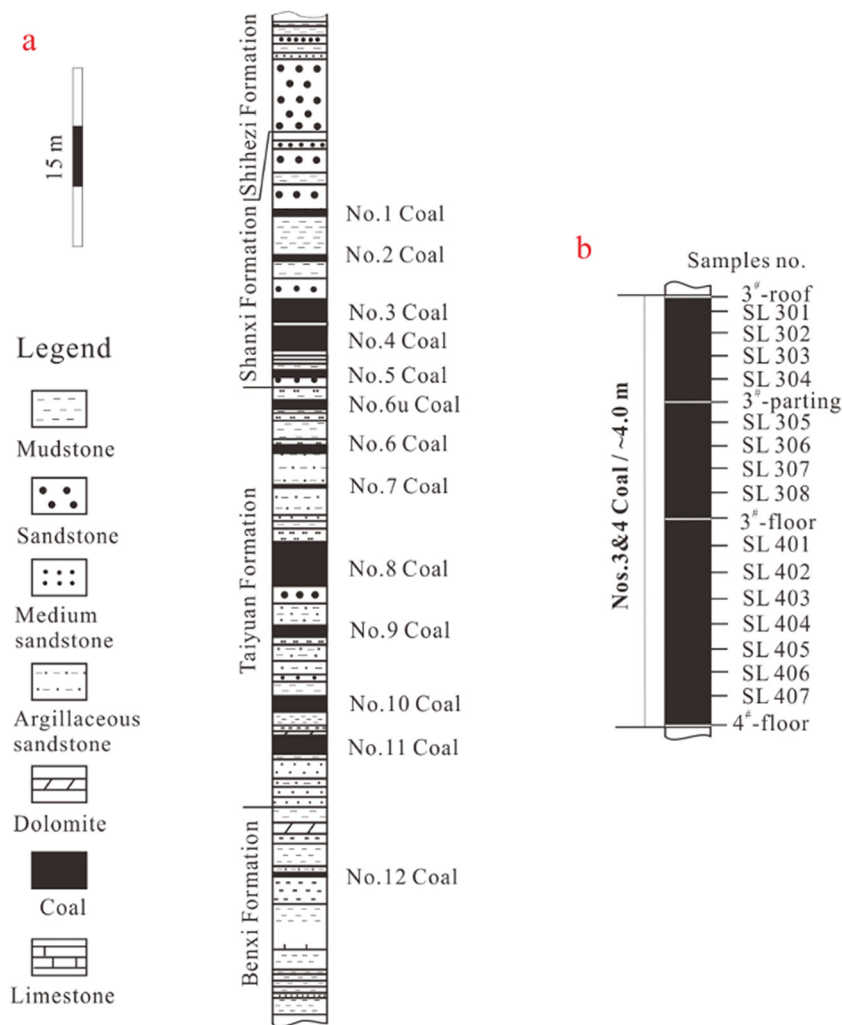


Fig. 2. Generalized stratigraphic columns for: (a) Pennsylvanian-Permian coal-bearing sequence in the eastern Ordos basin, China and (b) Nos. 3&4 Coal in the Liulin mine.

inductively coupled plasma mass spectrometry (ICP-CCT-MS), as described by (Li et al., 2014). Fluorine was determined using the ion-selective electrode (ISE) method, and Mercury was determined using a Milestone DMA-80 Hg Analyzer (Milestone). Elemental determinations was completed at the State Key Laboratory of Coal Resources and Safe Mining, at the China University of Mining and Technology (Beijing).

4. Results

4.1. Coal chemistry

The results for proximate analysis and sulfur concentrations are listed in Table 1. The ash yield of No. 3 Coal ranged from 9.89–20.70%, and from 0.26–15.1% in No.4 Coal. They are extremely low ash coal samples based on Chinese Standards GB/T 15224.1–2004 (Ash yield < 10% for very low ash coal, 10–20% for low ash coal, 20–30% for medium ash coal, 30–40% for medium-high ash coal, and 40–50% for high ash coal). Both the No.3 and No.4 coal samples had low moisture contents (0.53–0.72%) based on Chinese Standards MT/T 850–2000 (< 5% for low moisture coal, 5% to 15% for medium moisture coal, and > 15% for high moisture coal) (MT/T 850–2000). The volatile matter yields for Nos.3 and 4 Coal ranged from 10.93–37.88% across the coal-seam section, suggesting they were medium volatile bituminous coal samples based on Chinese Standards MT/T 849–2000. We observed that the ash yield of No.3 Coal was much

Table 1

Proximate analysis and sulfur concentrations in eastern Ordos basin coals (%).

Sample ID	M _{ad}	Ad	V _{daf}	S _{t,d}
3 [#] -roof	1.27	80.08	24.16	0.23
SL301	0.64	20.70	14.87	0.37
SL302	0.65	10.21	16.48	0.43
SL303	0.72	14.62	14.87	0.38
SL304	0.68	23.27	14.85	0.30
3 [#] -parting	0.70	48.96	10.50	0.15
SL306	0.67	15.63	14.09	0.40
SL307	0.63	12.00	12.84	0.40
SL308	0.63	9.89	10.93	0.36
3 [#] -floor ^a	0.78	46.28	10.49	0.18
SL401	0.61	15.01	18.66	0.36
SL402	0.57	7.55	24.89	0.46
SL403	0.52	7.65	22.90	0.40
SL404	0.54	1.18	21.64	0.46
SL405	0.57	0.63	24.49	0.52
SL406	0.57	0.26	23.82	0.54
SL407	0.53	2.47	21.94	0.59
4 [#] -floor	0.99	79.04	66.27	2.42

higher than that of No.4 Coal and it increased vertically from the floor to roof.

The total sulfur content for Nos.3 and 4 Coal ranged from 0.30–0.40% and 0.36–0.59% respectively, which corresponded to super-low sulfur coal based on Chinese standards GB/T 15224.2–2004

Table 2

Coal-petrography results of the coal samples from the eastern Ordos basin.

Sample	T	CT	DC	GC	CG	VD	T-V	SF	F	PF	ID	Ma	Mi	Scl	T-I	TPI	GI	VI	GW
SL301	8.5	19.8	8.9	1.2	5.0	8.0	51.4	17.5	4.2	1.0	3.3	5.0	2.3	0.2	33.5	1.66	2.26	4.20	0.71
SL302	2.5	46.6	26.4	0.4	2.1	1.5	79.5	1.2	0.8	1.4	1.9	0.8	5.9	0.2	12.2	1.56	20.59	1.86	0.14
SL303	2.5	21.7	19.9	4.2	3.3	4.8	56.4	8.2	5.6	2.2	3.3	11.5	7.9	1.6	40.3	0.89	3.97	1.80	0.32
SL304	3.4	31.7	22.9	1.0	3.8	2.0	64.8	7.4	3.8	2.6	2.8	5.5	1.0		23.1	1.25	5.02	1.90	0.27
SL306	2.6	17.0	28.8	3.8	2.6	5.3	60.1	7.7	1.4	1.8	4.2	9.3	4.0	0.2	28.6	0.57	5.22	0.94	0.44
SL307	2.0	18.2	36.3	0.8	2.6	4.4	64.3	5.7	2.2	2.8	2.2	6.3	3.8	0.6	23.6	0.54	6.99	0.81	0.26
SL308	5.1	3.3	13.2	1.6	14.6	1.6	39.4	9.7	1.4		3.2	27.9	4.3	0.4	46.9	0.32	4.71	1.20	1.22
SL401	3.0	12.6	26.2	1.8	4.1	1.8	49.5	14.2	8.4	0.2	4.7	10.2	3.3	0.6	41.6	0.81	2.19	1.28	0.33
SL402	0.8	62.6	19.4		0.4	2.0	85.2	1.6	1.4	1.0	1.6	2.4	1.0		9.0	2.57	19.04	3.25	0.04
SL403	0.8	39.3	29.1		1.4	0.8	71.4	4.4	3.6	0.6	2.8	5.0	3.2		19.6	1.23	7.07	1.55	0.08
SL404	5.1	58.3	6.9	0.2	2.2	1.6	74.3	1.2	0.2	0.6	2.6	5.3	11.6		21.5	3.48	19.90	7.01	0.07
SL405	1.6	79.7	11.1		0.2	0.4	93.0	0.6	0.2	0.8	0.6	2.6	0.8		5.6	5.51	68.29	7.05	0.01
SL406	0.8	73.6	10.5	0.2	2.2	1.6	88.9		0.8		1.2	5.3	1.6	0.2	9.1	3.62	47.10	6.06	0.05
SL407	0.6	75.4	12.6		1.6		90.2	0.2	0.6	1.2	1.2	3.2	1.0	0.2	7.6	4.13	46.70	5.68	0.03

T, telinite; CT, collotelinite; DC, desmocollinite; GC, gelocollinite; CG, corpogelinite; VD, vitrodetrinite; T-V, total vitrinite; SF, semifusinite; F, fusinite; PF, Pyrofusinite; ID, inertodetrinite; Ma, macrinite; Mi, micrinite; Scl, sclerotinite; T-I, total inertinite.

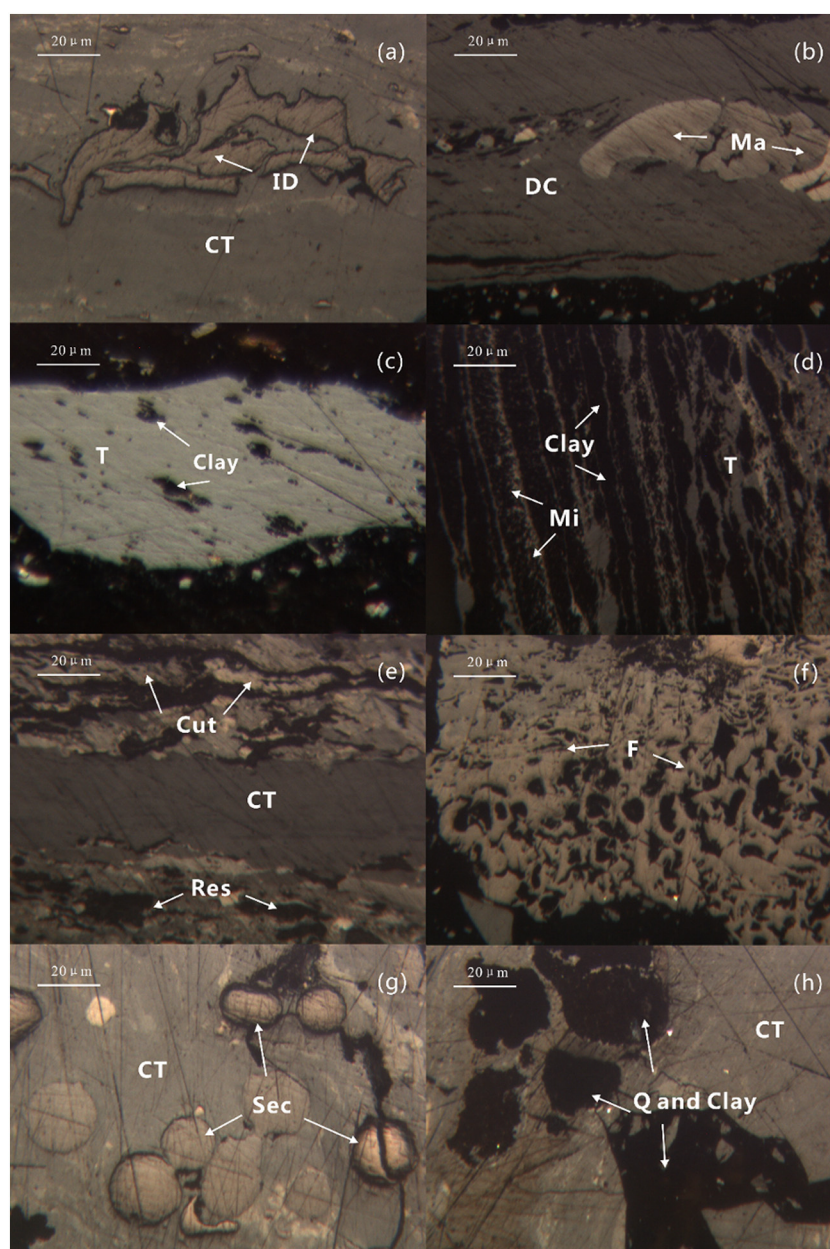


Fig. 3. Photomicrographs of eastern Ordos coal taken on polished blocks with reflected light, oil immersion, and 500× total magnification.

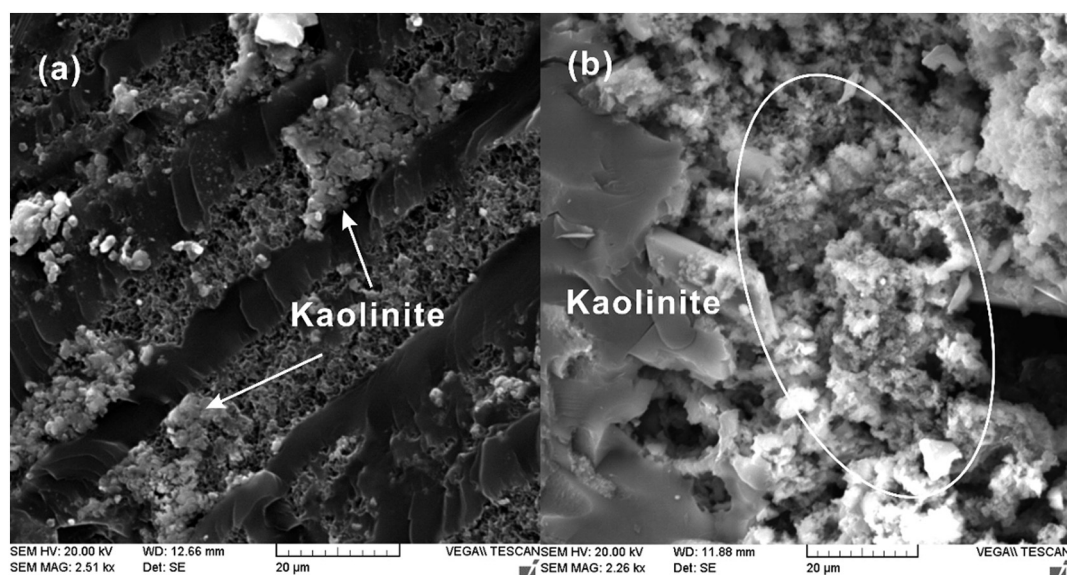


Fig. 4. SEM images of minerals in Liulin.

Table 3

Major element oxides (%) of the coals and partings in the Liulin Mine, eastern Ordos basin (on whole-coal basis).

Samples	SiO ₂	Al ₂ O ₃	TiO ₂	CaO	Fe ₂ O ₃	K ₂ O	MgO	Na ₂ O	MnO	P ₂ O ₅	LOI
3 [#] -roof	57.93	34.98	1.20	0.15	2.14	2.43	0.44	0.23	0.014	0.10	19.92
SL301	55.23	39.73	2.14	0.24	0.88	0.97	0.20	0.16	0.002	0.09	79.30
SL302	53.72	40.95	2.13	0.41	1.31	0.48	0.17	0.18	0.004	0.11	89.79
SL303	52.51	41.21	1.57	0.52	2.29	0.45	0.32	0.37	0.007	0.22	85.38
SL304	52.53	41.95	1.93	0.34	1.72	0.29	0.28	0.28	0.003	0.26	76.73
3 [#] -parting	52.27	42.21	2.35	0.22	1.63	0.18	0.26	0.16	0.003	0.28	51.04
SL306	52.68	39.84	1.14	1.51	2.05	0.62	0.25	0.27	0.007	1.10	84.37
SL307	48.23	38.74	1.20	4.35	2.56	0.35	0.28	0.30	0.007	2.82	88.00
SL308	49.47	41.43	0.55	2.02	1.98	0.15	0.21	0.53	0.012	2.05	90.11
3 [#] -floor*	51.45	41.13	2.79	0.15	3.09	0.22	0.54	0.19	0.003	0.09	53.72
SL401	49.81	40.95	4.95	0.28	2.24	0.12	0.24	0.35	0.002	0.35	84.99
SL402	49.61	41.27	1.18	0.44	5.10	0.12	0.53	0.52	0.003	0.57	92.45
SL403	51.29	42.11	2.16	0.42	2.39	0.13	0.21	0.51	0.006	0.16	92.35
SL404	43.13	31.74	12.74	1.90	5.63	0.34	0.63	1.12	0.015	0.26	98.82
SL405	45.75	33.67	5.43	2.38	6.44	0.56	0.68	1.46	0.032	0.39	99.37
SL406	43.45	35.39	6.00	2.18	6.93	0.34	0.54	1.57	0.037	0.38	99.74
SL407	50.14	36.67	3.62	0.82	4.14	1.73	0.46	0.69	0.020	0.19	97.53
4 [#] -floor	55.95	30.44	1.58	0.17	5.71	4.42	0.73	0.31	0.004	0.06	20.96
Average	49.83	38.98	3.34	1.27	3.26	0.48	0.36	0.59	0.01	0.64	89.92
Chinese ^a	8.47	5.98	0.33	1.23	4.85	0.19	0.22	0.16	0.015	0.092	–
World ^b	–	–	–	–	–	–	–	–	–	–	–

^a From Dai et al. (2012a).^b From Ketris and Yudovich (2009).

(< 0.5% for super-low sulfur coal, 0.51%–0.9% for low-sulfur coal and 0.9%–1.50% for medium-sulfur coal).

4.2. Coal petrology and mineralogy

Proportions of vitrinite, liptinite, and inertinite macerals varied across the two seams (Table 2). Vitrinite was the dominant maceral group (up to 90.2 vol% of the total), and its proportions in the No.4 seam were slightly higher than that in the No.3 Coal seam. Both coal seams were characterized by high collotelinite (Fig. 3a) content with the highest value recorded at the lower part of the No.4 seam (79.7 vol %). Desmocollinite (Fig. 3b) was the second highest enrichment subgroup for both seams. No.3 Coal samples had high concentration of desmocollinite with the highest value of 36.3 vol% (sample SL307). Other vitrinite subgroups were present in average quantities (Fig. 3c–f).

Inertinite proportions in No.3 Coal were relatively high, with the highest value of 46.9 vol%, and ranging from 5.6–21.5 vol% in No.4

Coal with a wide variability (Table 2). The inertinite macerals were represented by semifusinite, fusinite (Fig. 3f), pyrofusinite, inertodetrinite, macrinite, macrinite (Fig. 3b) and sclerotinite, while nearly all kinds of inertinite subgroups were detected in the samples. All subgroups concentrations were higher in No.3 Coal than in No.4 Coal. Liptinite in these coal samples was represented by macroporinite, microsporinite, cutinite, and resinite (Fig. 3e).

The minerals were made up of clay minerals (Fig. 3c, d, h), quartz (Fig. 3h), sulfide, and oxide minerals, but clay minerals were the main constituents (Fig. 4).

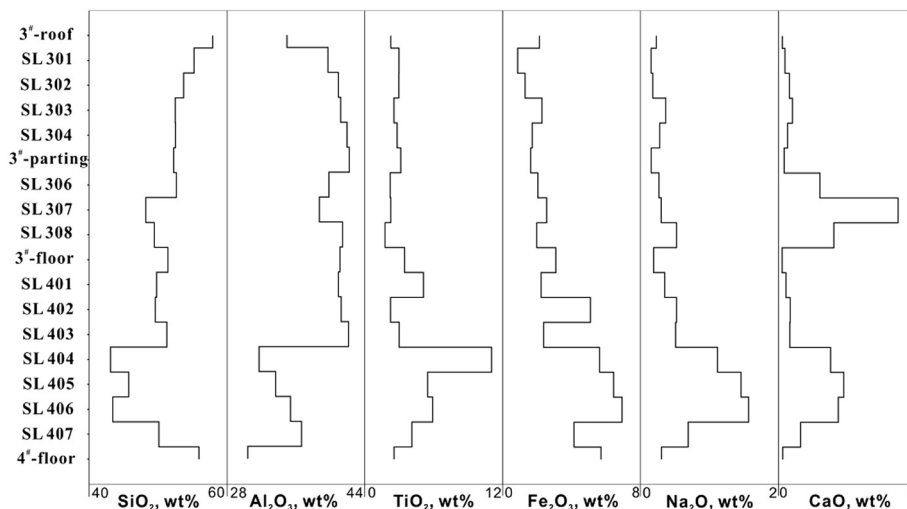
4.3. Geochemical patterns

The percentage of major-elements oxides and trace-elements concentration in Nos. 3&4 Coal from the eastern Ordos basin are listed in Tables 3 and 4, in comparison with average values for Chinese coals (Dai et al., 2012a) and global hard coals (Ketris and Yudovich, 2009).

Table 4

Trace elements (mg/kg) concentrations of the coals and partings in the Liulin Mine, eastern Ordos basin (on whole-coal basis).

Samples	Li	Be	F	Sc	V	Cr	Co	Ni	Cu	Zn	Ga	As	Se	Rb	Sr	Zr
3 [#] -roof	200.0	8.9	475.0	17.3	51.0	53.0	58.0	56.9	33.9	112.0	35.0	22.0	3.5	67.0	153.0	282.0
SL301	58.0	12.7	119.0	10.0	22.6	11.3	10.2	16.2	16.6	12.6	11.4	0.8	7.0	4.9	55.7	328.0
SL302	27.0	6.3	58.0	3.8	15.9	7.7	16.0	14.6	35.1	17.5	18.8	0.6	2.7	1.5	46.3	70.7
SL303	61.8	1.9	71.0	5.8	18.1	7.5	4.3	9.4	37.3	24.1	15.0	0.4	4.0	2.3	64.1	165.0
SL304	145.0	2.6	100.0	14.6	44.3	21.4	3.0	13.5	51.3	38.8	12.3	0.4	3.4	2.9	136.0	383.0
3 [#] -parting	269.0	3.2	164.0	21.3	31.3	28.4	0.5	7.8	21.4	21.3	26.2	0.5	3.7	3.2	322.0	395.0
SL306	68.0	2.6	94.0	4.9	15.5	12.8	4.5	9.5	39.3	24.5	16.9	0.5	3.2	2.9	201.0	111.0
SL307	52.1	2.0	95.0	3.0	9.8	13.3	1.8	7.3	39.8	19.2	18.6	0.6	3.4	1.4	252.0	325.0
SL308	48.6	2.0	182.0	2.5	7.7	8.1	1.2	6.9	37.3	18.5	11.2	0.6	2.9	0.5	488.0	41.1
3 [#] -floor*	278.0	2.2	144.0	14.7	33.2	28.2	0.4	4.3	57.2	28.1	19.1	0.3	5.3	3.9	94.6	395.0
SL401	79.6	1.4	61.0	6.8	33.8	15.7	1.1	2.7	51.7	21.8	10.7	0.3	4.2	1.0	164.0	180.0
SL402	35.9	0.7	40.0	2.5	11.1	7.7	1.6	3.5	40.5	19.5	11.6	0.2	4.0	0.6	121.0	58.4
SL403	39.4	1.1	28.0	2.8	12.0	8.1	1.7	6.6	46.8	24.5	5.4	0.2	4.4	0.5	49.7	63.1
SL404	3.9	2.9	4.0	1.2	8.7	8.7	1.5	5.3	45.5	23.3	4.0	0.2	2.6	0.3	31.2	34.5
SL405	2.3	5.4	5.0	1.1	6.0	5.3	2.2	5.4	16.2	14.0	8.0	0.1	2.2	0.2	19.6	16.0
SL406	1.8	6.5	3.0	2.5	7.8	6.2	2.6	5.4	17.7	15.2	8.9	0.2	1.7	0.2	21.4	20.4
SL407	5.2	10.3	46.0	18.1	49.4	20.8	3.4	11.6	27.8	20.0	10.6	0.4	4.4	1.9	25.6	59.2
4 [#] -floor	121.0	4.1	580.0	18.5	130.0	139.0	25.3	67.0	31.0	69.0	26.0	19.7	4.7	138.0	159.0	203.0
Average	44.9	4.2	64.7	5.7	18.8	11.0	3.9	8.4	35.9	21.0	11.7	0.4	3.6	1.5	119.7	132.5
Chinese ^a	31.8	2.11	130	4.38	35.1	15.4	7.08	13.7	17.5	41.4	6.55	3.79	2.5	9.25	140	89.5
World ^b	12.0	1.6	88.0	3.9	25.0	16.0	5.1	13.0	16.0	23.0	5.8	8.3	1.3	14.0	110	36.0
3 [#] -roof	32.0	2.8	0.3	0.2	3.1	5.2	465.0	8.7	0.8	1.9	308.2	0.8	53.8	0.7	23.6	4.6
SL301	16.8	0.7	0.1	0.1	1.1	0.3	87.2	8.3	0.8	0.5	35.3	0.1	31.6	0.4	13.0	3.0
SL302	3.5	1.2	0.0	0.1	0.9	0.1	37.6	1.8	0.2	0.6	8.9	0.0	10.8	0.1	2.9	0.8
SL303	9.3	0.7	0.1	0.2	0.9	0.1	54.0	4.3	0.4	0.5	47.9	0.1	20.2	0.2	7.9	2.0
SL304	10.1	0.7	0.2	0.2	1.1	0.3	92.2	9.6	0.6	0.9	62.2	0.1	43.0	0.4	13.9	5.4
3 [#] -parting	28.8	1.1	0.2	0.3	0.6	0.3	133.0	12.7	1.9	2.3	24.0	0.1	77.9	1.1	32.0	4.4
SL306	7.5	0.7	0.1	0.1	0.9	0.2	54.9	3.1	0.4	0.4	22.3	0.1	26.1	0.3	6.7	3.0
SL307	26.7	0.7	0.1	0.1	0.9	0.1	42.3	8.4	0.8	0.3	45.7	0.1	20.0	0.1	11.4	3.5
SL308	3.4	0.4	0.0	0.1	3.4	0.1	62.5	1.1	0.2	0.1	8.0	0.1	13.1	0.1	1.3	1.9
3 [#] -floor*	26.4	1.5	0.2	0.2	0.4	0.3	124.0	11.4	1.6	2.4	22.8	0.1	28.2	1.1	28.0	4.0
SL401	19.2	1.1	0.1	0.1	1.5	0.1	45.6	4.8	1.4	1.4	20.9	0.1	18.1	0.5	8.6	3.7
SL402	2.9	0.7	0.0	0.1	0.7	0.1	23.2	1.6	0.2	0.7	9.7	0.0	12.8	0.2	3.1	1.5
SL403	4.3	0.6	0.1	0.1	0.3	0.0	21.1	1.6	0.3	0.5	45.9	0.0	11.8	0.2	4.2	1.4
SL404	3.6	0.6	0.0	0.1	0.4	0.0	13.5	0.9	0.3	0.7	32.0	0.0	5.4	0.2	2.3	0.6
SL405	1.6	0.5	0.0	0.1	0.2	0.0	11.6	0.5	0.1	0.7	8.4	0.0	4.7	0.1	0.9	0.4
SL406	2.1	0.6	0.0	0.0	0.4	0.0	10.8	0.7	0.2	0.7	20.3	0.0	6.0	0.1	1.4	0.6
SL407	6.5	0.5	0.0	0.1	1.2	0.2	20.0	1.5	0.2	0.5	60.5	0.0	14.1	0.3	2.8	5.5
4 [#] -floor	25.8	2.9	0.2	0.2	0.6	9.7	581.0	6.7	0.7	2.5	1648.6	3.2	40.4	0.5	20.5	5.9
Average	8.4	0.7	0.1	0.1	1.0	0.1	41.2	3.4	0.4	0.6	30.6	0.1	17.0	0.2	5.7	2.4
Chinese ^a	9.44	3.1	0.3	0.05	0.84	1.1	159	3.71	0.62	1.08	163	0.5	15.1	0.79	5.84	2.43
World ^b	3.7	2.2	0.2	0.03	0.92	1.0	150	1.2	0.28	1.1	100	0.6	7.8	0.97	3.3	2.4

^a From Dai et al. (2012a).^b From Ketris and Yudovich (2009).**Fig. 5.** Vertical distribution for several major element-oxides from Nos.3 and 4 Coal in the eastern Ordos basin.

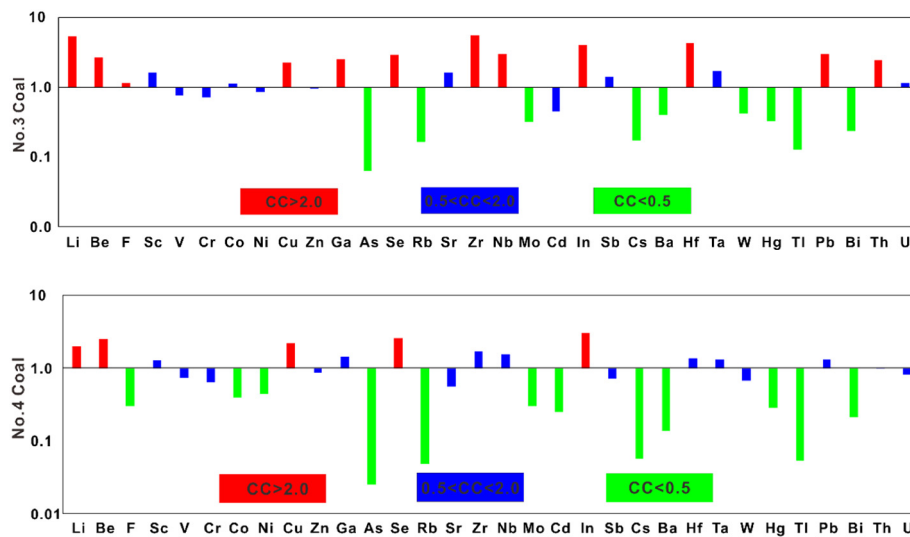


Fig. 6. Trace elements concentration coefficients (CC) in Liulin coal, eastern Ordos basin.

Table 5

Rare earth elements and yttrium (REY) concentrations of the coals in the Liulin Mine, eastern Ordos basin (on whole-coal basis) (mg/kg).

Samples	La	Ce	Pr	Nd	Sm	Eu	Gd	Tb	Dy	Y	Ho	Er	Tm	Yb	Lu	REY	LREY	MREY	HREY
SL301	17.30	35.00	3.70	13.90	2.88	0.56	2.56	0.61	4.30	31.80	0.99	3.07	0.59	3.98	0.57	121.8	72.78	39.84	9.20
SL302	21.70	41.80	4.77	16.00	2.58	0.61	2.64	0.48	2.83	18.50	0.56	1.56	0.26	1.57	0.23	116.1	86.85	25.05	4.17
SL303	19.90	39.80	4.14	16.60	2.96	0.62	2.80	0.53	2.96	18.10	0.59	1.68	0.31	1.57	0.24	112.8	83.40	25.01	4.39
SL304	24.30	51.90	5.31	20.10	4.41	0.87	4.13	0.87	5.15	30.60	1.14	3.14	0.57	3.31	0.44	156.3	106.0	41.62	8.61
SL306	27.70	52.40	5.34	21.10	4.12	0.62	3.82	0.71	3.63	21.80	0.79	2.08	0.39	2.09	0.28	146.9	110.7	30.58	5.63
SL307	30.70	50.20	5.21	19.90	4.29	0.45	3.53	0.67	4.00	22.50	0.75	2.06	0.38	2.06	0.28	146.9	110.3	31.14	5.53
SL308	56.20	95.50	11.20	36.20	5.96	0.75	5.00	0.82	4.19	20.00	0.81	2.07	0.36	2.09	0.25	241.4	205.1	30.75	5.58
SL401	24.20	44.40	4.31	16.60	3.22	0.53	2.85	0.54	2.87	18.50	0.61	1.55	0.29	1.77	0.24	122.5	92.73	25.28	4.46
SL402	15.80	27.10	2.95	9.88	1.67	0.26	1.35	0.21	1.17	6.27	0.22	0.66	0.11	0.69	0.10	68.4	57.40	9.26	1.78
SL403	9.46	22.90	2.57	10.10	2.05	0.30	1.69	0.26	1.52	7.75	0.32	0.79	0.16	0.95	0.13	60.9	47.08	11.52	2.35
SL404	7.15	14.70	1.41	5.42	0.97	0.16	0.99	0.16	1.00	9.78	0.23	0.66	0.12	0.78	0.11	43.6	29.65	12.08	1.90
SL405	4.50	9.12	1.17	4.49	0.98	0.21	1.09	0.22	1.56	17.30	0.38	1.17	0.18	1.16	0.17	43.7	20.26	20.38	3.07
SL406	4.17	9.30	1.03	4.69	1.02	0.24	1.40	0.30	2.02	22.40	0.52	1.61	0.32	1.70	0.26	50.9	20.21	26.35	4.41
SL407	2.21	4.57	0.61	2.56	0.98	0.25	1.43	0.44	3.41	34.60	0.99	3.32	0.63	3.90	0.65	60.6	10.93	40.12	9.49
Average	18.95	35.62	3.84	14.11	2.72	0.46	2.52	0.49	2.90	19.99	0.63	1.82	0.33	1.97	0.28	106.6	75.24	26.36	5.03
Chinese ^a	22.5	46.7	6.42	22.3	4.07	0.84	4.65	0.62	3.74	18.2	0.96	1.79	0.64	2.08	0.38	136	102	28	5.85
World ^b	11.0	23.0	3.5	12.0	2.0	0.47	2.7	0.32	2.1	8.4	0.54	0.93	0.31	1.0	0.2	68.4	51.6	13.7	3.07
UCC ^c	30.0	64.0	7.1	26.0	4.5	0.88	3.8	0.64	3.5	22.0	0.8	2.3	0.33	2.2	0.32	169	133	30.6	5.74

^a From Dai et al. (2012a).

^b From Ketris and Yudovich (2009).

^c From Taylor and McLennan (1985).

4.3.1. Major element-oxides

Major-elements oxides in the Nos.3 and 4 Coal samples were mainly dominated by SiO₂, Al₂O₃, with respective averages of 49.83 wt% and 38.98 wt%, which were much higher than that of Chinese coal (8.47 wt %, 5.98 wt%) (Dai et al., 2012a). The main carriers were quartz and clay minerals (Dai et al., 2015e; Yang et al., 2015). To a lesser extent, TiO₂, Fe₂O₃ were also present. Trace values for Na₂O, K₂O, CaO, and MgO were average compared to Chinese coal (Table 3). In the two coal seams, SiO₂ and Al₂O₃ had relatively higher concentrations in No.3 Coal than in No.4 Coal, but other major-elements oxides (TiO₂, Fe₂O₃, Na₂O, CaO) had higher contents in No.4 Coal (Fig. 5). The SiO₂/Al₂O₃ ratios for both coal seams (1.28 on average) were a little lower than that of Chinese coal (1.42) (Dai et al., 2012a) and similar to the theoretical ratio for kaolinite (1.18). This indicated that kaolinite was the primary mineral in these coal samples and free SiO₂ existed, which was consistent with the mineral compositions (Fig. 3).

A significant relationship was observed between ash yield and most major oxides in the No.3 and No.4 Coal samples. Values for SiO₂ and Al₂O₃ had significant positive correlations with ash yield ($r = 0.78$, 0.71), because they are found in aluminosilicate minerals and quartz,

which constitute the bulk of ash. Values for Fe₂O₃, MgO, Na₂O and MnO had positive correlations with sulfur ($r_{Fe-S} = 0.77$, $r_{Mg-S} = 0.71$, $r_{Na-S} = 0.70$ and $r_{Mn-S} = 0.76$). Fe₂O₃, MgO, Na₂O had positive correlations with TiO₂ ($r_{Fe-Ti} = 0.62$, $r_{Mg-Ti} = 0.66$, $r_{Na-Ti} = 0.66$), and MgO, Na₂O also had significantly positive correlations with Fe₂O₃ ($r_{Mg-Fe} = 0.95$, $r_{Na-Fe} = 0.93$). These findings suggest that the major-element oxides occurred in sulfides in No.3 and 4 Coal samples from the eastern Ordos basin.

4.3.2. Trace elements

The trace elements concentrations in these coal samples were compared with coal worldwide (Ketris and Yudovich, 2009), and Chinese coal (Dai et al., 2012a) (Table 4). Based on the environmental impact of trace elements found in coals (Goodarzi, 2002, 2006), elements were divided into several categories: Elements of prime environmental concerns, including As, Cd, Cr, Hg, Mo, Ni, Pb, Se, V and Zn, presented average concentrations or depleted. Elements Cr, V, and Zn were close to global average concentrations ($0.5 < CC < 2.0$), elements As, Hg, and Mo were depleted (Fig. 6). Elements of some environmental interest, including Ba, Be, Co, U, Th, F, indicated

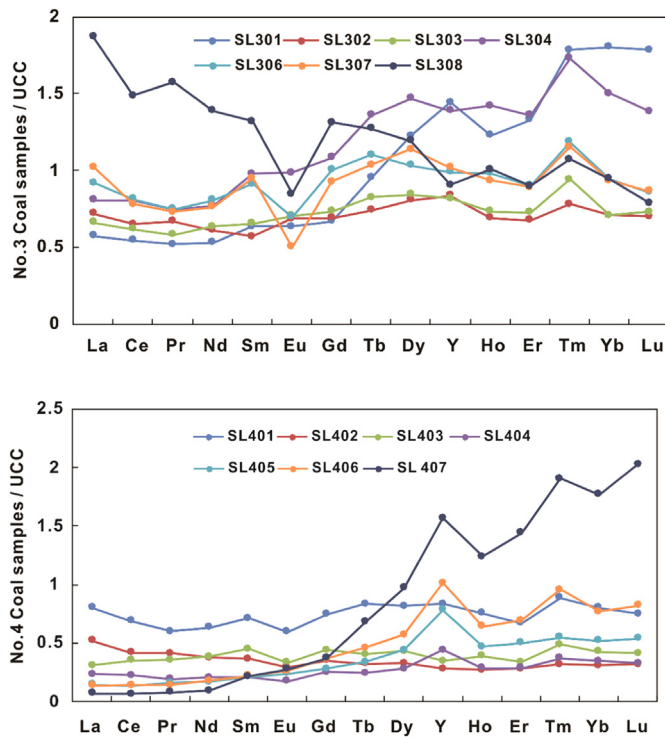


Fig. 7. Upper Continental Crust-normalized REY patterns of samples from the Liulin Nos. 3 and 4 Coal samples, eastern Ordos basin.

different degree of enrichment. Be was slightly enriched in studied coal, U was on average content and Ba was depleted in coal samples. For other elements, Li and Zr were enriched ($CC > 5.0$), Cu, Ga, Se, Nb, In, and Hf were slightly enriched ($2.0 < CC < 5.0$), Sc, Sr, Sb, and Ta were close to global average concentrations ($0.5 < CC < 2.0$). Nearly all trace elements had relatively higher concentrations in No.3 Coal than in No. 4 Coal.

Most elements occurred as inorganic affinities due to their positive correlations with ash yield. The relationship between trace elements and Al is thought to indicate the associations with aluminosilicate or clay minerals (Chen et al., 2016). In this study, many elements (Li, F, Co, Ni, As, Se, Rb, Cs, Ba, Pb and Th) had positive correlations with SiO_2 and Al_2O_3 , suggesting primarily aluminosilicate affinities. The affinities of these elements in No. 3 and No. 4 Coal samples were similar to coals from Pingshuo (Yang et al., 2016) and Junger (Dai et al., 2006a; Xiao et al., 2016). Be and Sc had strongly positive correlations with K_2O ($r = 0.80, 0.68$, respectively). Elemental assembly in these coal samples indicated similar occurrence: for example, F, Zr, Cd, Ba, Hf, Pb, and Th may have had similar occurrence with Li, V, Cr, Cs, and U, showing significant correlations with Sc.

4.3.3. Rare earth elements and yttrium (REY)

The concentrations and Upper Continental Crust (UCC)-normalized REY distribution patterns of the Liulin coal samples are shown in Table 5 and Fig. 7. The REY concentrations ranged from 43.6 mg/kg to 241.4 mg/kg, with an average of 106.6 mg/kg, which were slightly lower than that of Chinese coal (136 mg/kg) (Dai et al., 2012a).

The REY classification used in this study included LREY (La, Ce, Pr, Nd, and Sm), MREY (Eu, Gd, Tb, Dy, and Y), and HREY (Ho, Er, Tm, Yb, and Lu) (Seredin and Dai, 2012). Three enrichment types were identified (Seredin and Dai, 2012): L-type (light-REY; $La_N/Lu_N > 1$), M-type (medium-REY; $La_N/Sm_N < 1$, $Gd_N/Lu_N > 1$), and H-type (heavy REY; $La_N/Lu_N < 1$). No.3 Coal samples had high LREY concentrations, ranging from 72.78 to 205.1 mg/kg, and the No.4 Coal upper part samples (SL401, SL402, SL403 and SL404) also had high LREY concentrations. Samples from the lower part (SL406 and SL407) had high

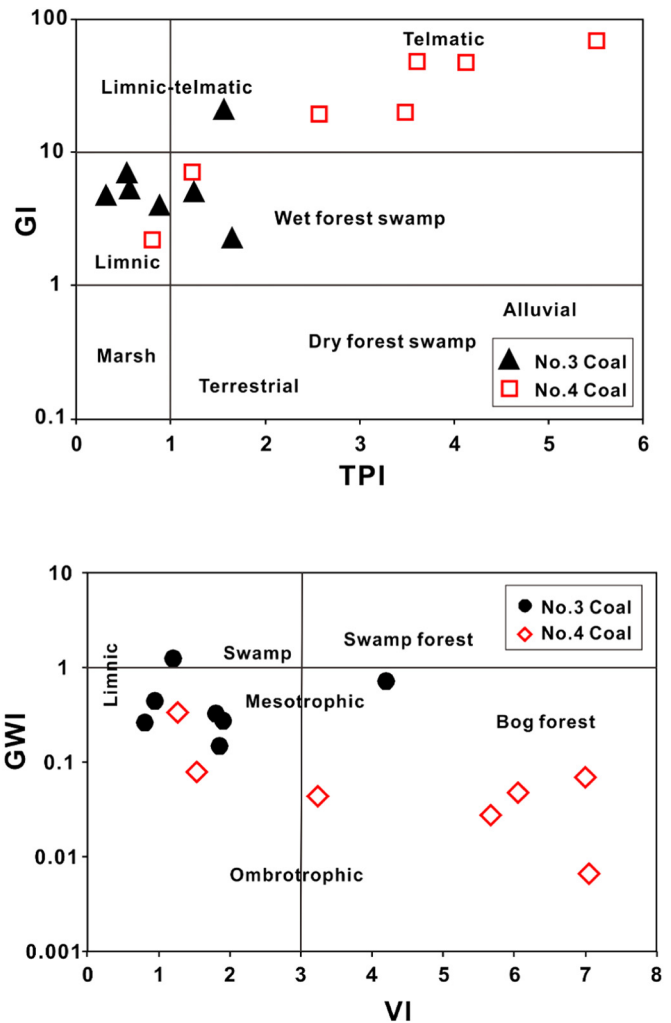


Fig. 8. GI vs. TPI plot of coal samples from the Liulin district of the eastern Ordos basin, modified from Diessel (1986); GWI vs. VI plot of coal samples modified from Calder et al. (1991).

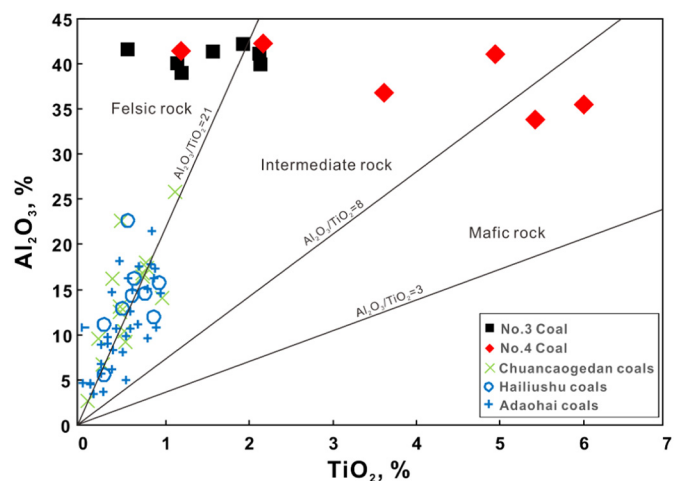


Fig. 9. The plot of Al_2O_3 versus TiO_2 in the No.3 and No.4 Coal samples from the Liulin district in the eastern Ordos basin, China, compared with coal from Adaozhai, Hailiushu and Chuancaogedan.

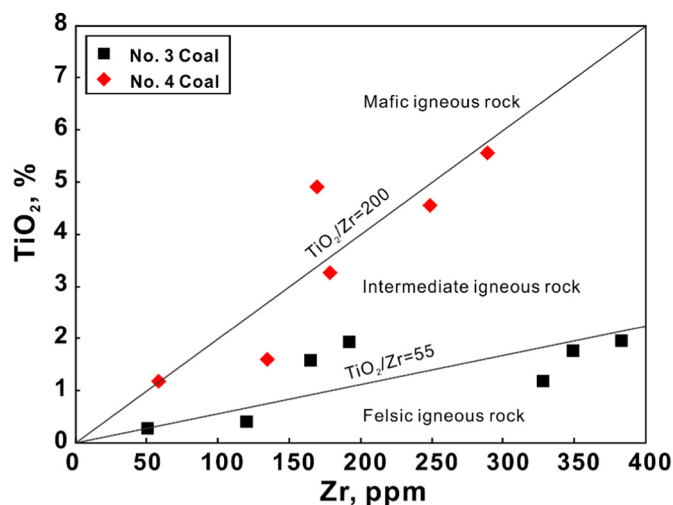


Fig. 10. A plot of TiO_2 versus Zr in the No.3 and No.4 Coal samples.

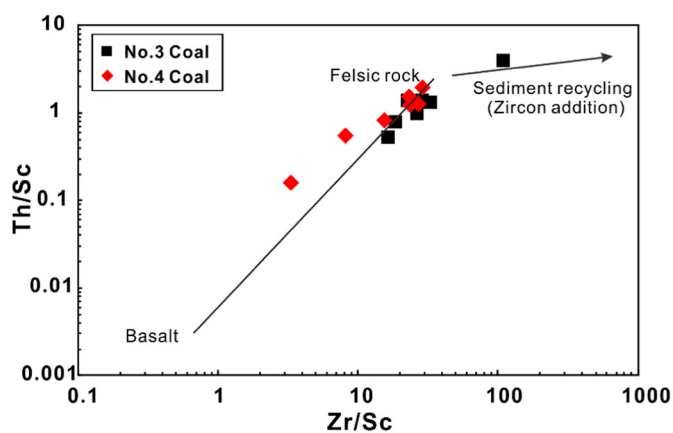


Fig. 11. Th/Sc versus Zr/Sc bivariate plot.

Table 6

Elements affinities deduced from correlation coefficients.

Correlation with ash yield
rash: from 0.7 to 1.0
SiO ₂ (0.78), Al ₂ O ₃ (0.71), Li (0.91), Rb (0.77), Zr (0.83), Cs (0.75), Ba (0.94), Hf (0.84), Tl (0.82), Pb (0.92), Th (0.88),
rash: from 0.4 to 0.69
F (0.67), V (0.40), Cr (0.46), Ni (0.52), Zn (0.47), Ga (0.50), As (0.62), Se (0.55), Nb (0.57), In (0.61), Ta (0.60), Bi (0.61), U (0.55)
rash: from -0.5 to +0.4
CaO (-0.37), K ₂ O (-0.11), P ₂ O ₅ (0.07), Be (-0.06), Sc (0.39), Co (0.27), Cu (0.36), Sr (0.30), Mo (0.37), Sb (0.31), W (0.09), Hg (0.31)
rash: from -0.51 to -1.0
TiO ₂ (-0.56), Fe ₂ O ₃ (-0.84), MgO (-0.74), Na ₂ O (-0.82), MnO ₂ (-0.77), St (-0.86),
Correlation coefficients between selected pairs of elements
Li-Al ₂ O ₃ (0.68), Li-SiO ₂ (0.58), F-Al ₂ O ₃ (0.59), Li-SiO ₂ (0.57), St-Fe ₂ O ₃ (0.77), U-Hg (0.69), U-Pb (0.73), F-As (0.80),

MREY concentrations (Table 5). The UCC-normalized REY distribution patterns for the Nos. 3 and 4 Coal samples (Fig. 7), showed that all samples (with the exception of samples SL302 and SL303) had negative Eu anomalies, No. 3 Coal (except for SL 308) had HREY enrichment.

5. Discussion

5.1. Coal facies indices

Macerals in coals, combined with mineralogical, petrological and

geochemical data, can provide information about the paleoenvironment.

Macerals compositions and various indices, such as the gelification index (GI), tissue preservation index (TPI), groundwater index (GWI) and vegetation index (VI), can reflect coal-forming plants, mire conditions and sedimentary environments. The GI is a ratio of gelation to non-gelation components that reflects the degree of wetness and duration of peat mire, where a high GI value indicates wet mire, and sediments from a dry basin margin will have low GI values. The TPI is the ratio of wood in coal forming plants, and can reflect the pH value, because microbial activity is weak and plants can be preserved very well in a low pH environment, creating in a high TPI value. GI and TPI can indicate a balanced ratio of plant growth and peat accumulation versus rise in groundwater table (Calder et al., 1991; Diessel, 1986). Ash content is an important factor when using this model to determine paleoenvironment. High GI and low TPI values suggest that peat accumulated in a dominantly herbaceous, continuously wet elevated bog for low ash content, which is believed to indicate telmatic to limnetic peat for high ash content. Low GI and high TPI values represent peat derived from a forested elevated bog for low ash content, or a forested swamp that was subjected to occasional dry periods for high ash content (Sen et al., 2016). The GWI is a value based on gelification and mineralal inputs, which reflects the level of water during peat accumulation. A GWI value of < 0.5 means it is ombrotrophic from raised mires to mesotrophic fens; values between 0.5 and 3 are described as mesotrophic; between 3 and 5 indicates rheotrophic mires in a limnetic environment; and > 5 signifies flooding of the mire surface, or prone to siliciclastic inundation (Calder et al., 1991; Sen et al., 2016). High VI values (VI > 3) represent a large quantity structured tree-derived material as well as a more terrestrial environment, and low values (VI < 3) indicate of low quantities of preserved structured material and a more limnic environment dominated by marginal aquatic or herbaceous plants (Sen et al., 2016).

The differences between two coal seams were significant. The TPI values for No. 3 Coal were higher than those for No. 4 Coal (Table 2), implying that the No. 4 Coal was deposited in a lower pH value environment. Because GI values likely reflect the degree of wetness and duration of peat mire, the high GI values from No. 3 Coal samples indicated that they were deposited in a wet mire and the low GI values of No. 4 Coal suggested that they were formed in a relative dry environment. GWI values from No. 3 Coal ranged from 0.14 to 1.22, which means that these coals were formed in ombrotrophic to mesotrophic depositional environments. The No. 4 Coal samples had low GWI values (0.01–0.33), demonstrating that they were deposited in an ombrotrophic environments, in raised mires to mesotrophic fens. High VI values (VI value > 3) imply that coal was formed in more terrestrial environment and low VI values suggest a limnic environment, so the No. 4 Coal was deposited in limnic environment. Based on GI-TPI and GWI-VI plots (Fig. 8), we also concluded that No. 3 Coal was deposited in a wet forest swamp and No. 4 Coal was formed from telmatic peat, in a mesotrophic to ombrotrophic environment.

5.2. Coal geochemistry and sedimentary source rocks

The geochemistry of the sediment-source region on the margin of a coal basin is the primary factor for determining the background values of elements in both coal and non-coal normal sedimentary materials (Dai et al., 2015c; Dai et al., 2006b). Some major elemental ratios (such as $\text{K}_2\text{O}/\text{Na}_2\text{O}$, $\text{Al}_2\text{O}_3/\text{SiO}_2$, $(\text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO})/(\text{SiO}_2 + \text{Al}_2\text{O}_3)$, and $\text{Al}_2\text{O}_3/\text{TiO}_2$), trace elements (Th, Sc, and Zr), and REE have been used to indicate the tectonic setting and provenance of sedimentary rocks (Bhatia, 1985; Hayashi et al., 1997; Jiang et al., 2015).

The $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio could indicate the provenance of sedimentary rocks (Hayashi et al., 1997) and sediment source region for coal deposits (Dai et al., 2015b; Dai et al., 2015e), due to the similar value of these elements in sedimentary rocks compared to the source rocks.

Values for $\text{Al}_2\text{O}_3/\text{TiO}_2$ in mafic, intermediate, and felsic igneous rocks range from 3 to 8, 8–21, and 21–70, respectively (Hayashi et al., 1997). The $\text{Al}_2\text{O}_3/\text{TiO}_2$ ratio in coal samples for this study were 22.6 on average, with a density of 18–30 (Fig. 9), similar to coal from Chuancaogedan (Yang et al., 2015), Hailiushu (Dai et al., 2015a) and Adaohai (Dai et al., 2012b), indicating that the source region for No. 3 and No. 4 Coal was mainly composed of felsic and intermediate rock.

The TiO_2/Zr ratio can also be used to indicate sediment provenance, values > 200 indicate mafic rocks, values from 199 to 55 indicate intermediate rocks, and values < 55 indicate felsic igneous rocks (Hayashi et al., 1997; Vosoughi Moradi et al., 2016). The TiO_2/Zr ratio values for these samples were in the intermediate and felsic igneous rock range (Fig. 10), suggesting of an intermediate and felsic source composition.

Variability in mineral composition, degree of sorting, and heavy mineral content is reflected in sediment Zr/Sc and Th/Sc ratios (McLennan et al., 1993). For the coal samples in this study, these ratios ranged from 3.27 to 109.80 and 0.16 to 1.91, respectively. A crossplot of Zr/Sc versus Th/Sc indicated that the studied coal samples were mainly derived from intermediate and felsic igneous rocks and had not experienced sediment recycling (Fig. 11), which was consistent with the above conclusions.

5.3. Modes of occurrence of trace elements

The presence of trace elements and their modes occurrences in coal have been studied extensively (Dai et al., 2015d; Dai et al., 2017a; Dai et al., 2017b). Several methods are used for determining elemental occurrence, and one of the most common is statistical methods, which would be more effective when accompanied by direct methods such as SEM-EDX (Karayıgıt et al., 2017; Li et al., 2016).

The modes of occurrence for trace elements in Nos. 3 and 4 coal samples were preliminarily investigated using correlation coefficients. Elemental affinities derived from the correlation coefficients between the concentrations of each element in coal and ash yield indicated their modes of occurrence in the coal samples for this study. Many elements had positive correlations with ash yield (Table 6), such as SiO_2 , Al_2O_3 , Li, Rb, Zr, Cs, Ba, Hf, Tl, Pb, and Th. Their correlation coefficients ($r > 0.7$) with ash yield suggested inorganic affinities. The elements Li and F had positive correlations with SiO_2 and Al_2O_3 , indicating that they occurred in aluminosilicate in the coal samples. Other elements (F, V, Cr, Ga, Ni, As, In, and Ta) had inorganic–organic affinities, based on correlation coefficients that ranged from –0.5 to 0.4. While other selected elements had similar affinities, Fe and S had negative correlations with ash yield and the correlation coefficient between Fe and S was 0.77, suggesting that Fe and S had similar affinities and may have occurred in pyrite.

6. Conclusion

The Nos. 3 and 4 Coal samples were classified as low ash yield (0.26–20.7%), low moisture (0.53–0.72%), medium volatile (10.93–37.88%) bituminous coal with low sulfur contents (0.30–0.59%). Vitrinite was the dominant maceral group with concentration up to 90.2% and lower proportion in No. 3 Coal than in No. 4 Coal. Inertinite content in No. 3 Coal was higher than that of No. 4 Coal. Nearly all subgroups were detected in coal samples. Major minerals in both coal seams included quartz, clay minerals, sulfide and other oxide minerals.

Major elements in coal samples were dominated by SiO_2 (average on 49.83%) and Al_2O_3 (38.98%), whose average concentrations were much higher than other coals in China (8.47%, 5.98%); these occurred in quartz and clay minerals. Other major elements (Na_2O , K_2O , Fe_2O_3 , and MgO) had average values compared to Chinese coal and may have occurred in sulfides based on their correlations with sulfur content. Compared to Chinese coal, No.3 Coal was enriched in Li and Zr, and No.

4 Coal was enriched in Li, Be, Cu, Se, and In. Correlation coefficients between elements and ash yield suggested that Li, Rb, Zr, Cs, Ba, Hf, Tl, Pb, and Th occurred in inorganic affinities, while F, V, Cr, Ga, Ni, As, In, and Ta had inorganic–organic affinities, and Fe and S may have occurred in pyrite. The REY concentrations ranged from 43.6 to 241.4 mg/kg in the coal samples with negative Eu anomalies.

Based on the macerals compositions, coal facies indicators (GI, TPI, GWI, and VI) suggested that No. 3 Coal was deposited in a wet forest swamp and No. 4 Coal was formed from telmatic peat, in a mesotrophic to ombrotrophic environment. Elemental indicators, such as the ratios of $\text{Al}_2\text{O}_3/\text{TiO}_2$, TiO_2/Zr , Zr/Sc and Th/Sc, indicated that the studied coal sediments were mainly derived from intermediate and felsic igneous rocks.

Acknowledgements

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