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Geochemistry of Middle Jurassic Coals from the Dananhu Mine, Xinjiang: Emphasis on Sediment Source and Control Factors of Critical Metals

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Abstract: In recent years, coal-type critical metal deposits have become a research hotspot in coal geology. As a major coal-accumulating basin in the Xinjiang area, the Turpan-Hami Basin contains abundant coal resources and has the potential to become a large coal-type critical metal deposit. However, previous studies on the enrichment characteristics of critical metal elements in coal are few and need further research. Based on SEM-EDS, XRF, and ICP-MS experiments, this study investigates the coal petrology, mineralogy, and geochemistry of the No. 22 coal of the Xishanyao Formation from the Dananhu Coal Mine, Xinjiang, to identify the sediment source, depositional environment, and controlling factors of the critical metal elements of the No. 22 coal. The results showed that the Dananhu coals are characterized by a low ash yield, low total sulfur content, high volatile yield, and high inertinite proportions. Quartz, kaolinite, and illite are the main minerals in the coal. Compared with the world's low-rank coals, Ni, Co, Mo, and Ta are slightly enriched, Li, Rb, Cs, Ba, Tl, Bi, and Ge are depleted, and the concentrations of other trace elements are comparable to the average values of the world's low-rank coals. The REY of the Dannanhu coals exhibited high fractionation, with its enrichment patterns characterized by the H-type and M-H-type. Although most of the critical metals are not enriched in the Dannanhu coals, the Ga, Zr (Hf), and Nb (Ta) concentrations in the coal ash of the Dannanhu coals have reached the economic cut-off grade and have the potential to be a substitute for rare metal resources. The terrigenous detrital sources of the Dannanhu coals mainly come from the Paleozoic dacite, andesite, and a small amount of granite from the Harik Mountain and Eastern Bogda Mountain in the Turpan-Hami Basin. The Dannanhu coals are generally in a dry and hot depositional environment, with high salinity and weak reduction-oxidation. The low source input and weak reduction-oxidation environment have resulted in low concentrations of critical metal of the No. 22 coal from the Dananhu Coal Mine.

Keywords: geochemistry; Jurassic coal; critical metals; sediment source; control factors

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1. Introduction

Coal is a special sedimentary rock or sedimentary deposit mainly composed of organic matter [1]. By the end of 2022, China's proven coal reserves were 2070.12 × 10^8 t, and coal consumption was 30.40 × 10^8 t, accounting for 56.2% of the total primary energy consumption [2]. This indicates that the dominant position of coal in China's energy composition will not change for a long time [1].

Coal has reduction and adsorption, and under certain geological conditions, critical metal elements can be enriched in coal and even form "coal-type critical metal deposits" [1,3,4]. With the upsurge in the research and development of critical metal ore deposits, the metal deposits associated with coals have received great attention. At present, the critical metal deposits that have been found in coal include coal-Li, coal-Ga (Al), coal-Ge, coal-Be, coal-Nb (Ta) and coal-REY (rare earth elements) [3,5–10].

Li resources in coal in China are mainly distributed in Carboniferous–Permian coal fields in North China and Late Permian coalfields in South China [5,6]. The average concentration of Be in the Dazhai Mine, Yunnan Province, is 343 μg/g, which has potential industrial value [9]. The Wulantuga coalfield's average concentration of Ge in Inner Mongolia is 274 μg/g, and the Ge metal reserves are 1700 t. The average Ge content in the Lincang coalfield in Yunnan Province is 850 μg/g, and the Ge metal reserves are 1620 t [11,12]. Ga-rich coal in China is mainly distributed in Carboniferous–Permian and Jurassic coal-bearing basins, and the Ga content in Guanbanwusu Coal Mine in the Jungar coalfield, Inner Mongolia is 3.40–59.0 μg/g [6]. Ga content in the Haerwusu coal mine is 7.40~54.0 μg/g [13].

The Turpan-Hami Basin is one of the large coal-accumulating basins in China, with abundant coal resources (over 5000 × 10^8 t). The Jurassic Xishanyao Formation and Badaowan Formation are the main coal-bearing strata in the Turpan-Hami Basin [14]. In recent years, a large number of studies have been made on Jurassic coal in the Turpan-Hami Basin, but the studies have mainly focused on mineral characteristics, geochemistry, sedimentary and tectonic evolution, etc., and there are few reports on the critical metal elements in coal [15–20]; therefore, it is necessary to conduct in-depth research on the enrichment characteristics, sediment source, and control factors of critical metal elements in coal.

Based on this, the mineralogy, geochemistry, and distribution of critical metal elements, the sediment source, and control factors of the No. 22 coal from the Dananhu coalfield were discussed by SEM-EDS, XRF, and ICP-MS.

2. Geological Setting

The Turpan-Hami Coalfield is named after the Turpan-Hami Basin, which is located in eastern Xinjiang (Figure 1a) and is an east–west-oriented narrow and elongated intermountain basin-type coalfield (Figure 1b) [21]. The coalfield consists of eight sub-tectonic units, including five depressions, two uplifts, and one slope [22]. The Dananhu Mine is located at the southern Turpan-Hami coalfield (Figure 1a) [23], which is tectonically located at the northern edge of the Dananhu depression (Figure 1b). The coal-bearing strata is the Middle Jurassic Xishanyao Formation (Figure 1c) [24]. In the late Early Jurassic, the Dananhu Depression began to subside drastically from the status of ancient uplift [14,25,26]. In the middle of the Xishanyao Formation, the delta-lake depositional system was formed. Peat swamps developed steadily, resulting in the extensive development of extra-thick coal seams [14,25,26]. In the late Xishanyao Formation, lake intrusion led to a coal accumulation stop. The depositional environment of the Xishanyao Formation in the Dananhu Mine is overall dominated by the delta-lake environment [25].

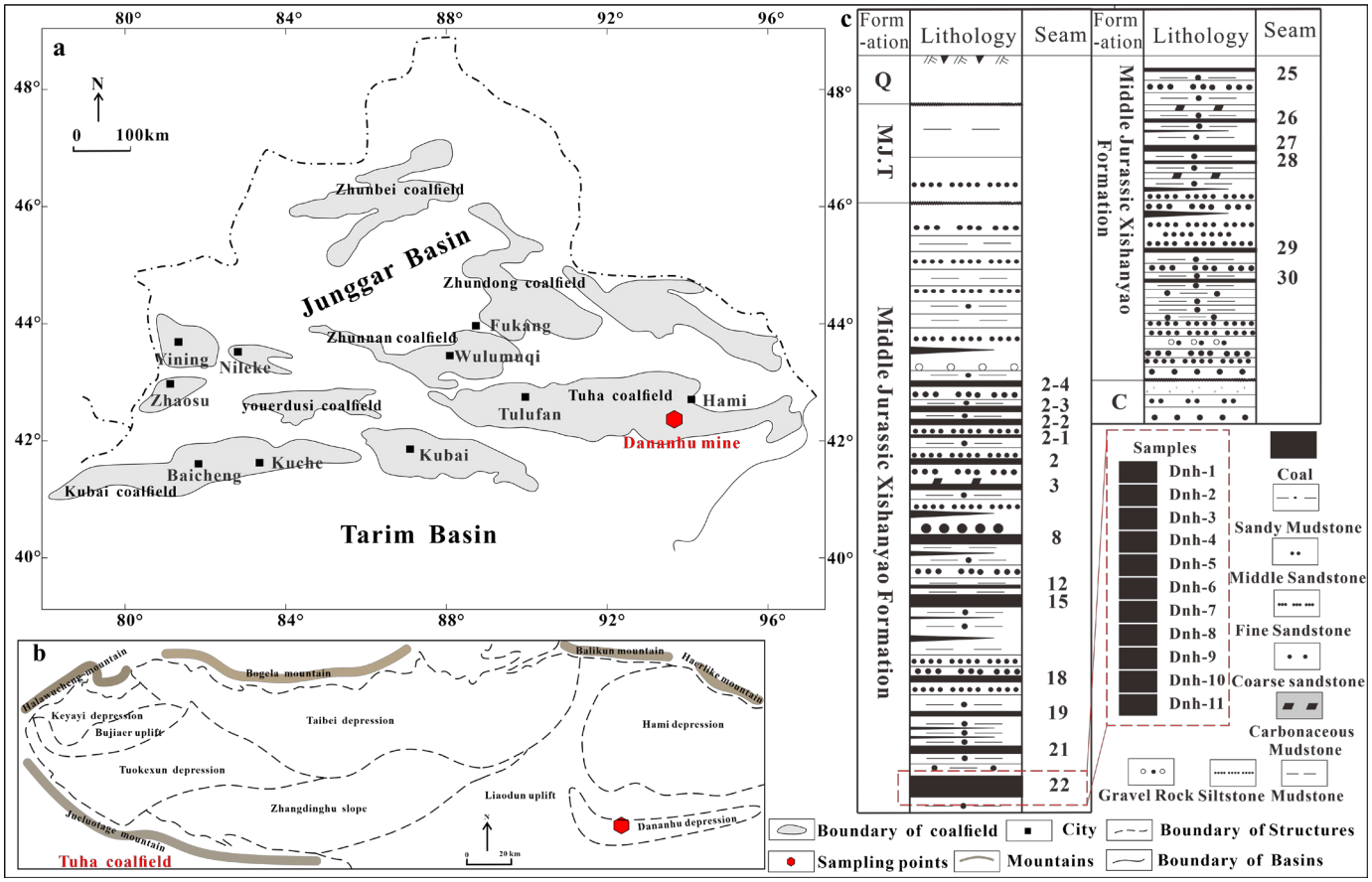


Figure 1. Location and structure of the study area, stratigraphic column and sampling profile of the No. 22 coal of the Dananhu coal mine. (**a**) The location of the Dananhu mine; (**b**) The structure map of the Dananhu mine; (**c**) Stratigraphic column and sampling profile of No.22 coal of the Dananhu mine. Q, quaternary; MJ.T, middle Jurassic Toutunhe formation; C, Carboniferous.

3. Samples and Analytic Methods

In this study, 11 coal samples (designated as Dnh-1 to Dnh-11) with a size of 10 cm × 10 cm × 10 cm were taken of the No. 22 coal from the Dananhu Mine (from the top to the bottom, spacing 0.5 m) according to the Chinese Standard method GB/T 482-2008 (2008) [27]. They were sealed immediately to prevent the samples from oxidizing.

Each sample was ground to pass through 200-mesh (elemental and chemical analyses) and 18~40-mesh (0.425 mm~1.00 mm, polished grain mounts, maceral and mineralogical analysis) sieves.

The 18~40-mesh samples were made into polished particle holders with epoxy resin and curing agents. The maceral of No. 22 coals was observed and identified by the oil-immersed microscope (instrument model: Zeiss Axio Imager M1m) according to the ICCP system and Pickel et al. [28–30]. A small number of 18~40-mesh samples were fixed on a conductive adhesive and plated with gold. The chemical element composition and morphology of the minerals were further examined by field emission scanning electron microscopy and energy spectroscopy (SEM-EDS, Hitachi SU8220 with an accelerated voltage and a beam current of 20 kV and 5 μA).

The standards ASTM D3173-11 (2011), D3174-11 (2011), and D3175-11 (2011) were used as the standard test for ash yield, moisture, and volatile yield for approximate analysis of the coal samples [31–33]. The total sulfur content was tested according to the international standard ASTM D3177-02 (2011) [34].

After the coal sample was ashed by a plasma asher (K1050X, EMITECH, Laughton, UK) at a low temperature (~120 °C), 0.3 g of the low-temperature ash product was analyzed using a Smartlab SE powder X-ray diffractometer (XRD, Rigaku, Japan, with a Cu

anode target and scanning angle ranging from 5° to 70°). The resulting spectra were subjected to qualitative and quantitative mineral analysis using the Siroquant 3.0 commercial interpretation software developed by Taylor [35].

After obtaining the high-temperature ash products of the samples using a muffle furnace at 815 °C, X-ray fluorescence (XRF, instrument model: ARL Perform X4200) was used for the determination of major element oxide content in the high-temperature ash after the coal ash was pressed into approximately 1 cm thick pellets with boric acid as a base, using an electric pellet press (PrepP-01A). For the determination of trace elements, 50 mg samples were first digested by microwave digestion. Then, through acid driving, extraction, and volume fixing, the detection solution was obtained. Finally, the trace element content of the coal was determined by inductively coupled plasma mass spectrometry (ICP-MS).

4. Results

4.1. Coal Chemistry

The results of the proximate analyses and total sulfur analysis of the Dannanhu coals are shown in Table 1. The average content of moisture and fixed carbon of the Dannanhu coals were 23.08% (17.39%~27.89%) and 51.67% (43.21%~58.37%), respectively, indicating that the coals were characterized by high moisture (>20%) and extra-low to low fixed carbon (<55%). The ash yields of the coals were lower than 10%, except for Dnh-1 and Dnh-11. The average ash yield of the coals was 8.17% (4.30%~18.19%), which belonged to the low ash coal [36]. The volatile yield of the Dannanhu coals ranged from 35.82% to 50.24% (43.76% on average), with associative values of the reflection of vitrinite (on average 0.39%, Table 1); therefore, the Dannanhu coals can be classified as high volatile matter brown coal [37]. The total sulfur content was very low (ranging from 0.10% to 0.30%), with all samples having a total sulfur content of less than 0.5% (Table 1).

Table 1. The results of the macerals and coal chemistry of the Dananhu coal (%).

Samples	M_{ad}	A_d	V_{daf}	FC_d	$S_{t,d}$	V	I	L	Mi	$R_{o,max}$
Dnh-1	18.70	18.19	47.18	43.21	0.16	36.80	55.10	7.10	1.00	0.42
Dnh-2	17.39	9.05	35.82	58.37	0.15	39.60	52.30	7.50	0.60	0.45
Dnh-3	24.64	6.63	50.24	46.46	0.23	28.40	63.30	6.40	1.90	0.36
Dnh-4	26.06	6.29	45.97	50.64	0.16	26.80	65.10	6.80	1.30	0.44
Dnh-5	24.03	6.09	38.31	57.93	0.11	30.50	64.70	4.80	0.00	0.32
Dnh-6	22.68	6.79	38.78	57.06	0.10	32.80	58.90	7.40	0.90	0.37
Dnh-7	27.89	6.27	49.19	47.62	0.18	34.70	61.20	3.20	0.90	0.41
Dnh-8	18.63	9.77	43.84	50.67	0.16	29.70	67.20	2.10	1.00	0.39
Dnh-9	24.06	6.12	42.42	54.06	0.30	22.60	72.00	3.90	1.50	0.37
Dnh-10	25.63	4.30	45.19	52.45	0.24	29.20	62.00	6.80	2.00	0.39
Dnh-11	24.19	10.37	44.37	49.86	0.22	35.40	55.20	8.30	1.10	0.42
Average	23.08	8.17	43.76	51.67	0.18	31.50	61.55	5.85	1.11	0.39

M_{ad}, moisture on air dry basis; A_d, ash yield on dry basis; V_{daf}, volatile matter on dry and ash-free basis; FC_d, fixed carbon on dry basis; $S_{t,d}$, total sulfur; V, vitrinite; I, inertinite; L, liptinite; Mi, minerals; $R_{0,ran}$, the vitrinite random reflectance.

4.2. Maceral and Minerals

In No. 22 coal, the highest content of maceral was inertinite (61.55% on average, Table 1). The inertinite was composed of fusinite (Figure 2a), semifusinite (Figure 2b,c), macrinite (Figure 2d), and inertodetrinite (Figure 2e,f). Vitrinite was the second maceral in No. 22 coal, with an average content of 31.5% (from 22.60% to 39.60%, as shown in Table 1), which was mainly composed of ulminite (Figure 2c), attrinite (Figure 2c), densinite (Figure 2d), phlobaphinite (Figure 2e), corpohuminite (Figure 2f), and Gelinite (Figure 2f). The content of liptinite was low, ranging from 2.10% to 8.30% (5.85% on average), which was the Sporinite (Figure 2c).

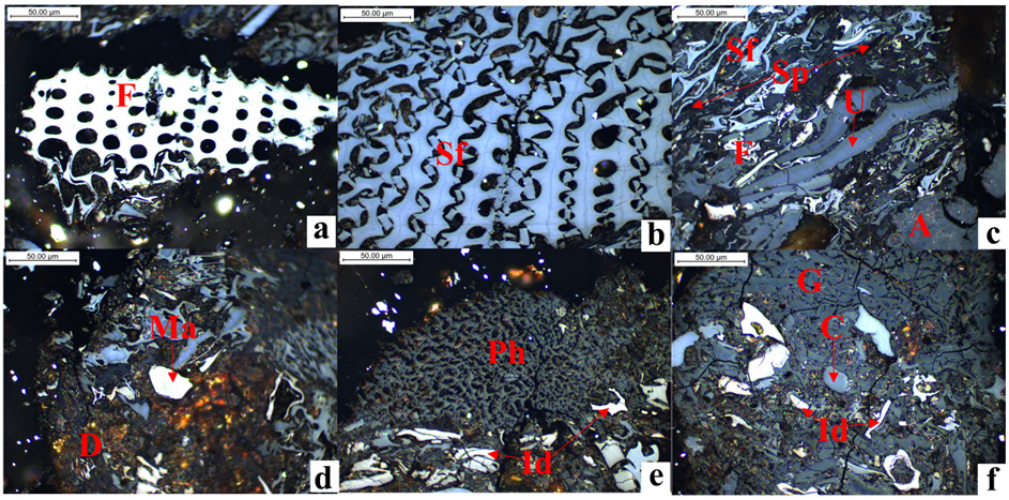


Figure 2. Macerals in No. 22 coals (under the reflected light). (**a**), fusinite in Dnh-3; (**b**), semifusinite in Dnh-3; (**c**), fusinite, semifusinite, ulminite, attrinite and sporinite, in Dnh-7; (**d**), macrinite and densinite in Dnh-4; (**e**), phlobaphinite and inertodetrinite in Dnh-11; (**f**), corpohuminite, gelinite and inertodetrinite in Dnh-9. F, fusinite; Sf, semifusinite; U, ulminite; A, attrinite; Sp, sporinite; Ma, macrinite; D, densinite; Id, inertodetrinite; Ph, phlobaphinite; C, corpohuminite; G, gelinite.

The mineral contents of the No. 22 coal were shallow (0.60%~2.00%, with an average of 1.11%, Table 1). The mineralogy of the No. 22 coal was dominated by quartz (43.3%), kaolinite (37.0%), and illite (19.7%) (Figure 3).

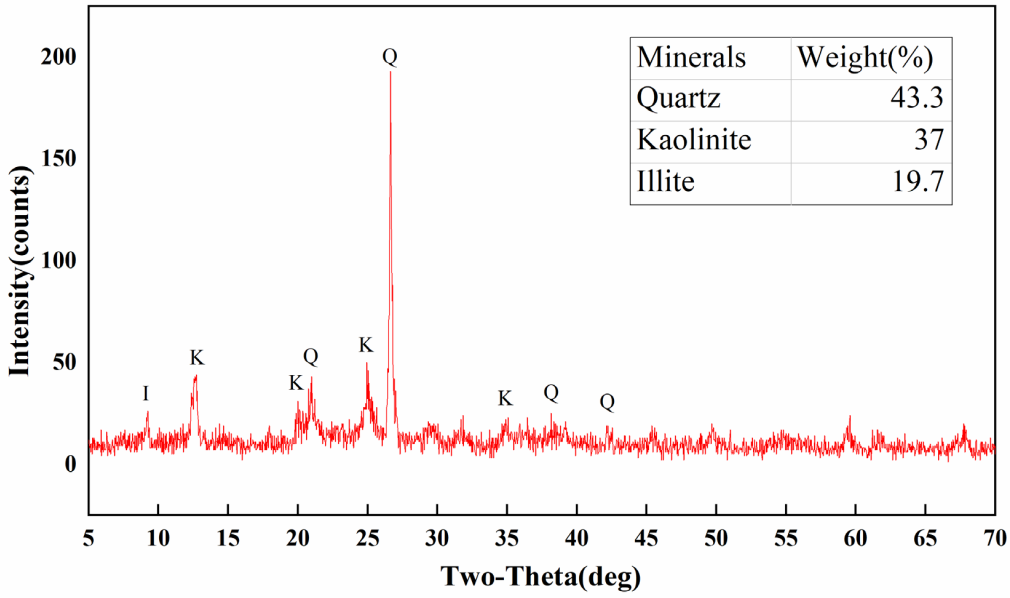


Figure 3. X-ray diffraction patterns of the Dnh-5. Kln: kaolinite, Qz: quartz, I: Illite.

Quartz is distributed in cracks in the form of small particles or a tetrahedral structure (Figure 4a,c,j), and quartz in organic matter is distributed in the form of lamellar structures (Figure 4b,d,k,l) or dispersed particles (Figure 4f), which is an autogenesis. Two types of kaolinite were found. The authigenic kaolinites were attached to organic matter by a flocculent structure (Figure 4a). Another kind of kaolinite was distributed as irregular masses on organic matter (Figure 4d), which belongs to the terrestrial clastic origin [38]. Ilmenite was mainly distributed in a flocculent structure (Figure 4e). The calcite was mainly filled the cell lumen of fusinite (Figure 4f). In addition, small amounts of chalcopyrite, anatase, and zircon were detected by SEM-EDS (Figure 4h,i).

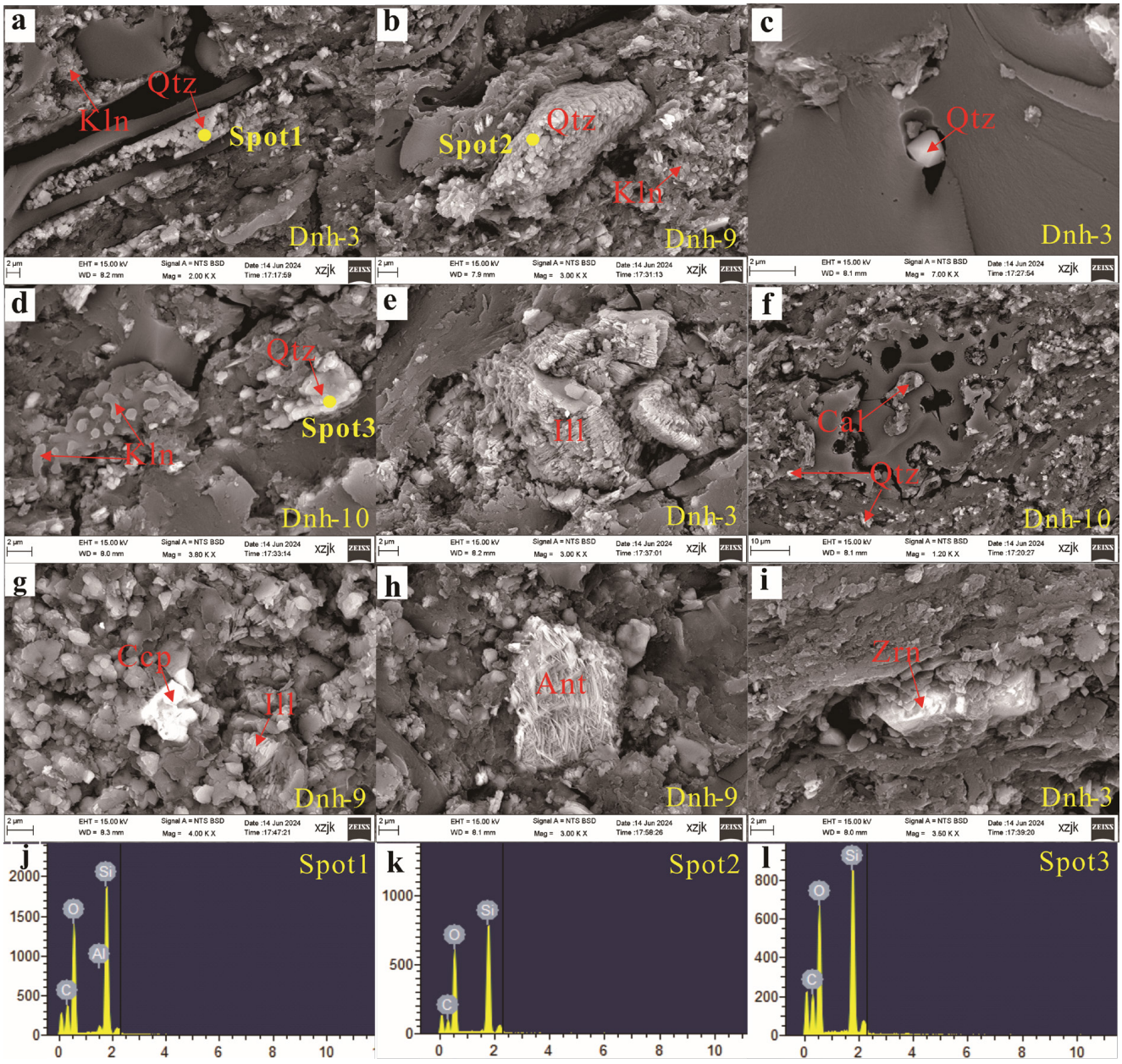


Figure 4. SEM back-scattering images of minerals in samples. (**a**) Quartz and kaolinite in Dnh-3; (**b**) Quartz and kaolinite in Dnh-9; (**c**) Quartz in Dnh-3; (**d**) Quartz and kaolinite in Dnh-10; (**e**) Illite in Dnh-3; (**f**) Quartz and calcite in Dnh-10; (**g**) Illite and chalcopyrite in Dnh-9; (**h**) Anatase in Dnh-9; (**i**) Zircon in Dnh-3; (**j**) The EDS data of Spot1; (**k**) The EDS data of Spot2; (**l**) The EDS data of Spot3. Qtz, Quartz; Kln, Kaolinite; Ill, Illite; Cal, Calcite; Ccp, Chalcopyrite; Ant, Anatase; Zrn, Zircon.

4.3. Major Element Oxides in Raw Coal

Table 2 provides the content of the major element oxides of the Dananhu coals. SiO_2, CaO, and Al_2O_3 were the dominant major element oxides in the coals, accounting for 2.324%, 1.301%, and 1.257%, respectively, with the following contents of Fe_2O_3 (0.631%), MgO (0.389%), and Na_2O (0.374%). The rest of the major element oxides (K_2O, MnO, TiO_2, and P_2O_5) were extra-low, with a very low content of less than 0.1%.

Table 2. The major element oxide contents of the Dananhu coal (%, on a whole coal basis).

Samples	SiO_2	Fe_2O_3	Al_2O_3	CaO	MgO	K_2O	Na_2O	MnO	TiO_2	P_2O_5	LOI
Dnh-1	9.06	0.22	3.96	1.25	0.34	0.03	0.40	0.01	0.31	0.01	84.41
Dnh-2	2.59	0.11	1.58	1.59	0.45	0.03	0.38	0.01	0.04	0.03	93.20
Dnh-3	1.74	0.18	1.27	1.09	0.35	0.02	0.41	0.01	0.05	0.01	94.89
Dnh-4	1.35	0.43	0.77	1.18	0.36	0.04	0.38	0.01	0.07	0.00	95.42
Dnh-5	1.38	0.20	0.88	1.40	0.42	0.02	0.31	0.01	0.03	0.00	95.35
Dnh-6	1.54	0.12	0.95	1.54	0.44	0.03	0.30	0.01	0.03	0.00	95.04
Dnh-7	0.55	1.85	0.46	1.05	0.36	0.03	0.29	0.03	0.04	0.00	95.34
Dnh-8	3.49	0.22	1.57	1.48	0.42	0.03	0.42	0.01	0.12	0.00	92.24
Dnh-9	1.32	0.36	0.80	1.38	0.40	0.03	0.42	0.01	0.05	0.00	95.23
Dnh-10	0.32	0.60	0.42	1.27	0.39	0.02	0.37	0.01	0.03	0.00	96.58
Dnh-11	2.24	2.66	1.17	1.08	0.36	0.04	0.43	0.05	0.10	0.01	91.88
Average	2.32	0.63	1.26	1.30	0.39	0.03	0.37	0.01	0.08	0.01	93.60
Avg-C	8.47	4.85	5.98	1.23	0.22	0.19	0.16	0.015	0.33	0.09	--

Avg-C, the average content of the major element oxides for the Chinese coals [6].

Compared with the average content of major element oxides for the Chinese coals [6], the average contents of CaO, MgO, and Na_2O were slightly higher, with concentration coefficients (CC) of 1.06, 1.77, and 2.34, respectively. Particularly, Na_2O was characterized by a slight enrichment. Apart from CaO, MgO, and Na_2O, the average contents of the major element oxides in the coals were significantly lower than the average content of the Chinese coals. Moreover, the content of SiO_2 in the sample Dnh-1 was slightly higher than the average value of the Chinese coal with a concentration coefficient of 1.07, and the MnO contents of Dnh-7 and Dnh-11 were 0.032% and 0.048%, with concentration coefficients of 2.13 and 3.20, which all reached a slight enrichment (Table 2, Figure 5).

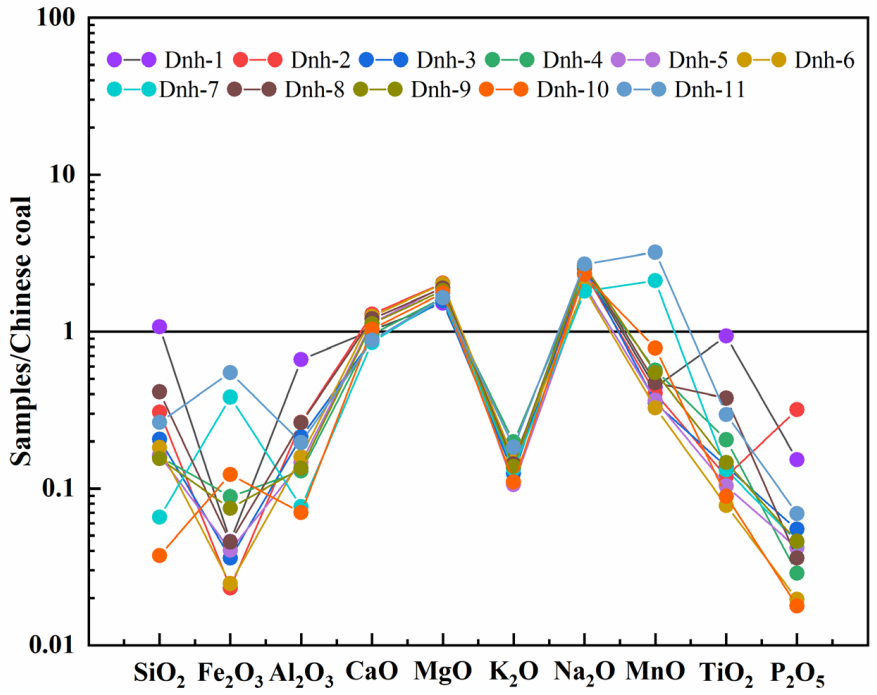


Figure 5. Spider diagram of the concentration of major element oxides in the Dananhu coals.

4.4. Trace Element in Raw Coal

The trace element contents of the Dannanhu coals are shown in Table S1. Sr is the element with the highest average content, averaging 212.18 μg/g (from 136 μg/g to 327 μg/g), followed by REY, V, B, Ni, and Zr, with an average content of 38.1, 29.36, 25.99, 25.35, and 25.00 μg/g, respectively. Apart from Li (1.61 μg/g on average) and Nb (1.66 μg/g on

average), the average contents of Be, Sc, V, Cr, Co, Ni, Cu, Zn, Ga, Rb, Sr, Mo, Cd, In, Cs, Ba, Hf, Ta, W, Tl, Pb, Bi, Th, U, and Ge were extremely low, below 1.0 μg/g (Table S1). Based on the classification standard of the trace element enrichment level in coal proposed by Dai et al. [39], compared with the average contents of the trace element of the world's low-rank coals [40], the Dannanhu coals were slightly enriched in Ni, Co, Mo, and Ta, with CCs of 2.82, 2.74, 2.12, and 2.75, respectively. The average contents of Li, Rb, Cs, Ba, W, Tl, Pb, Bi, Th, U, Ge, and B were lower than the world's low-rank coals (CC < 0.5), especially Rb, Cs, Ba, Tl, Bi, and Ge. The contents of other trace elements were comparable to the world coal average with the CC ranging from 0.5 to 2 (Figure 6).

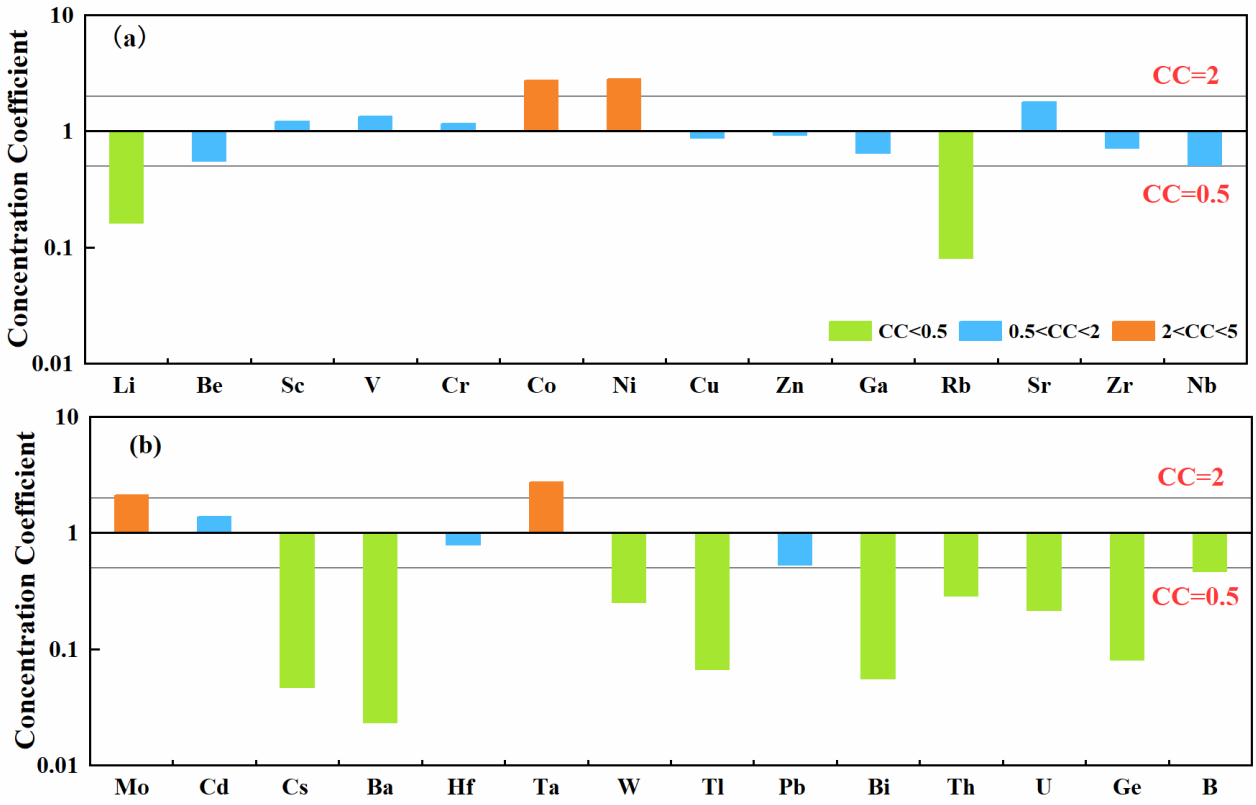


Figure 6. Concentration coefficient of trace elements of the Dannanhu No. 22 coals. (**a**) Concentration coefficient of Li~Nb; (**b**) Concentration coefficient of Mo~B.

4.5. Rare Earth Element (REY)

4.5.1. Concentration, Enrichment, and Vertical Variation of REY

The average content of the rare earth element (REY) in the Dannanhu coals was 38.12 μg/g (17.14–71.02 μg/g) (Table S1), which was apparently lower than the average content of the world's low-rank coals (65.27 μg/g) [40] and slightly higher than that of the No. 4 coal of the Sandaolin Mine (15.21 μg/g) [41]. Compared to the world's low-rank coals [40], the REY of the Dannanhu coals fall within the normal range (0.5 < CC = 0.58 < 2) (Figure 7a). Ce was the most abundant rare earth element (mean 10.67 μg/g), followed by Y, La, and Nd (on average contents of 8.01 μg/g, 5.84 μg/g, and 5.36 μg/g, respectively). Eu, Tb, Ho, Er, Tm, Yb, and Lu were extremely low, with average contents below 1.00 μg/g (Table 3). The content of Eu was generally interfered with by Ba (if Ba/Eu > 1000) [42]. A negative correlation between Eu and Ba (R^2 = 0.2376, Figure 7b) and low Ba/Eu ratios (from 4.27 to 42.20, mean 15.73) suggest that Eu of the Dannanhu coals was not interfered with by Ba.

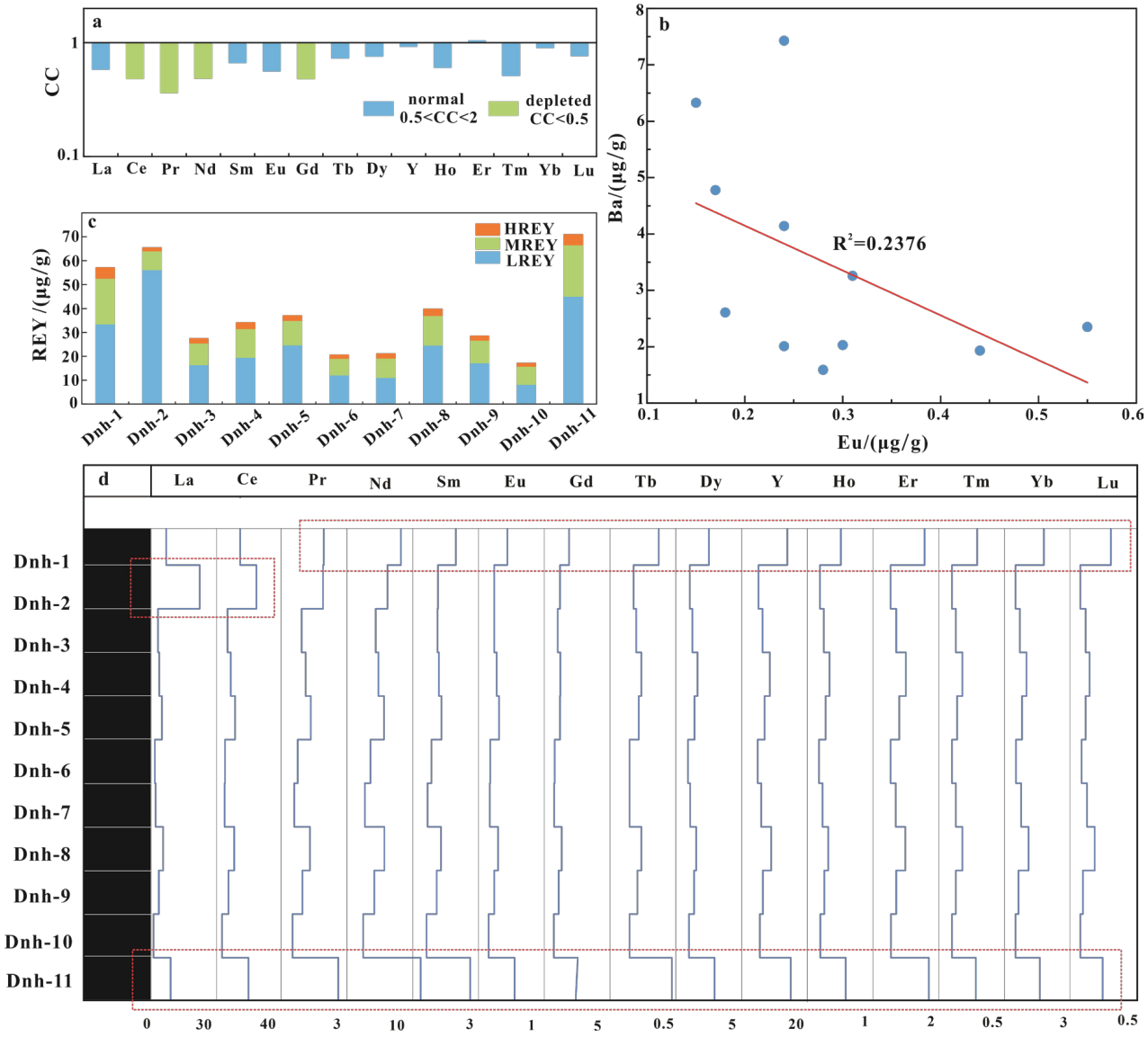


Figure 7. CC of individual REY (**a**), the relationship between Ba and Eu (**b**), concentrations of LREY, MREY, and HREY (**c**), and vertical variations of REY (**d**) in the Dananhu No. 22 coals.

Table 3. REY geochemical parameters of the Dananhu coal.

Samples	LREY (μg/g)	MREY (μg/g)	HREY (μg/g)	L/M	L/H	M/H	$(La/Lu)_N$	$(La/Sm)_N$	$(Gd/Lu)_N$	δCe	δEu
Dnh-1	33.48	19.11	4.48	1.75	7.47	4.27	0.42	0.67	0.45	1.06	0.82
Dnh-2	56.23	7.83	1.40	7.18	40.14	5.59	5.23	3.65	1.06	0.81	0.81
Dnh-3	16.40	9.12	1.90	1.80	8.63	4.80	0.50	0.53	0.62	1.07	0.79
Dnh-4	19.49	11.99	2.66	1.63	7.33	4.51	0.47	0.59	0.60	1.15	0.84
Dnh-5	24.73	10.20	2.13	2.42	11.61	4.79	0.79	0.72	0.73	1.17	0.90
Dnh-6	12.11	7.00	1.43	1.73	8.49	4.91	0.40	0.40	0.69	1.17	0.79
Dnh-7	11.04	8.13	1.94	1.36	5.69	4.19	0.33	0.62	0.43	1.12	0.86
Dnh-8	24.60	12.46	2.76	1.97	8.91	4.51	0.55	0.81	0.51	1.06	0.85
Dnh-9	17.25	9.45	1.84	1.83	9.39	5.14	0.69	0.62	0.82	1.07	0.80
Dnh-10	8.18	7.55	1.41	1.08	5.82	5.37	0.32	0.38	0.70	1.09	0.79
Dnh-11	45.08	21.51	4.43	2.10	10.18	4.86	0.67	0.64	0.75	1.11	0.78
Average	24.42	11.30	2.40	2.26	11.24	4.81	0.94	0.88	0.67	1.08	0.82

L/M = LREY/MREY; L/H = LREY/HREY; M/H = MREY/HREY; *N*, normalization; La_N = La(coal)/La(UCC); Sm_N = Sm(coal)/Sm(UCC); Lu_N = Lu(coal)/Lu(UCC); $\delta Ce = Ce_N/(0.5 \times a_N + 0.5 \times Pr_N)$; $\delta Eu = Eu_N/(0.67 \times Sm_N + 0.33 \times Tb_N)$.

Rare earth elements are generally categorized into LREY (La, Ce, Pr, Nd, and Sm), MREY (Eu, Gd, Tb, Dy, and Y), and HREY (Ho, Er, Tm, Yb, and Lu) [43]. LREY of the Dananhu coals were dominant, with an average content of 24.42 μg/g (8.18~56.23 μg/g), followed by MREY (8.18~56.23 μg/g, average 24.42 μg/g) and HREY (ranging from 1.40 to 4.48 μg/g, average 2.40 μg/g) (Figure 7c). Figure 7d demonstrates the vertical variation of individual REY contents of the Dananhu coals. Due to the similar geochemical properties of REY, REY had a similar trend in general terms, indicating that the REY has the same mode of occurrence and origin [44]. In addition, obviously raised peaks of REY were shown in Danh-11. La and Ce showed significantly elevated peaks in sample Dnh-2 and an elevated peak from Pr to Lu in sample Dnh-1 (Figure 7d).

4.5.2. Geochemical Parameters and Distribution Patterns of REY

The REY are usually derived from sediment source areas, and their geochemical parameters could reveal the source characteristics and depositional environments [44,45]. The geochemical parameters of REY of the Dannanhu coals are shown in Table 3. The average values of LREY/MREY, LREY/HREY, and MREY/HREY were 2.26, 11.24, and 4.81, respectively, all of which were greater than 1, indicating that REY in the Dannanhu coals had been clearly fractionated.

Seredin and Dai [43] classified the REY enrichment types into an LREY-enriched (L-type, (La/Lu)N > 1), MREY-enriched (M-type, (La/Sm)N < 1, (Gd/Lu)N > 1), and HREY-enriched (H-type, (La/Lu)N < 1). The average values of (La/Lu)N, (La/Sm)N, and (Gd/Lu)N were 0.94, 0.88 and 0.67, respectively (Table 3). The Dnh-2 belonged to the L-type with (La/Lu)N, (La/Sm)N, and (Gd/Lu)N contents of 5.23, 3.65, and 1.06, respectively (Table 4), while the other samples were H-type. In addition, the average values of δCe and δEu in Danannhu coals were 1.08 (from 0.81 to 1.17) and 0.82 (from 0.78 to 0.90), indicating that Ce showed a weak positive anomaly and a weak negative anomaly for Eu (Table 3).

Table 4. Average contents of critical elements of the Dananhu coal (μg/g, on an ash basis).

Samples	Li_2O	Ga	Ge	$(Nb, Ta)_2O_5$	$(Zr, Hf)_2O_5$	REO
Dnh-1	190.90	13.74	0.17	40.02	1094.91	377.18
Dnh-2	139.41	11.60	7.52	50.22	273.68	867.03
Dnh-3	163.20	34.06	1.74	49.84	505.02	496.64
Dnh-4	55.31	79.66	0.48	76.69	659.52	653.19
Dnh-5	78.25	104.85	6.19	151.86	863.70	731.00
Dnh-6	65.27	11.93	1.59	28.07	257.74	363.65
Dnh-7	31.33	57.11	1.94	70.71	409.99	405.31
Dnh-8	78.67	41.15	1.11	47.72	679.10	490.12
Dnh-9	75.13	39.85	0.82	35.26	487.96	560.41
Dnh-10	71.71	83.20	2.77	68.32	491.58	480.34
Dnh-11	109.31	69.25	0.26	119.37	1381.10	823.43
Average	96.23	49.67	2.24	67.10	645.84	568.03

Based on the REY distribution patterns, REY in the Dannanhu coals could be divided into three categories (Figure 8): (i) Dnh-2 exhibited an apparent LREY enrichment, which showed a significant Ce-negative anomaly and a slight Eu-positive anomaly. However, this enrichment type resulted from obvious HREY depletion (Figure 8a); (ii) Dnh-1, Dnh-8, and Dnh-9 showed obvious HREY enrichment accompanied by a slightly negative anomaly in Ce, a slightly positive anomaly in Eu, and a positive anomaly in Dy and Tm. This type was nevertheless caused by a depletion in LREY in contrast to (i) (Figure 8b); (iii) Dnh-3, Dnh-4, Dnh-5, Dnh-7, Dnh-7, Dnh-10, and Dnh-11 exhibited a significant MREY–HREY enrichment, with a slightly positive anomaly of Ce and Eu and salient positive anomaly of Dy and Tm (Figure 8c).

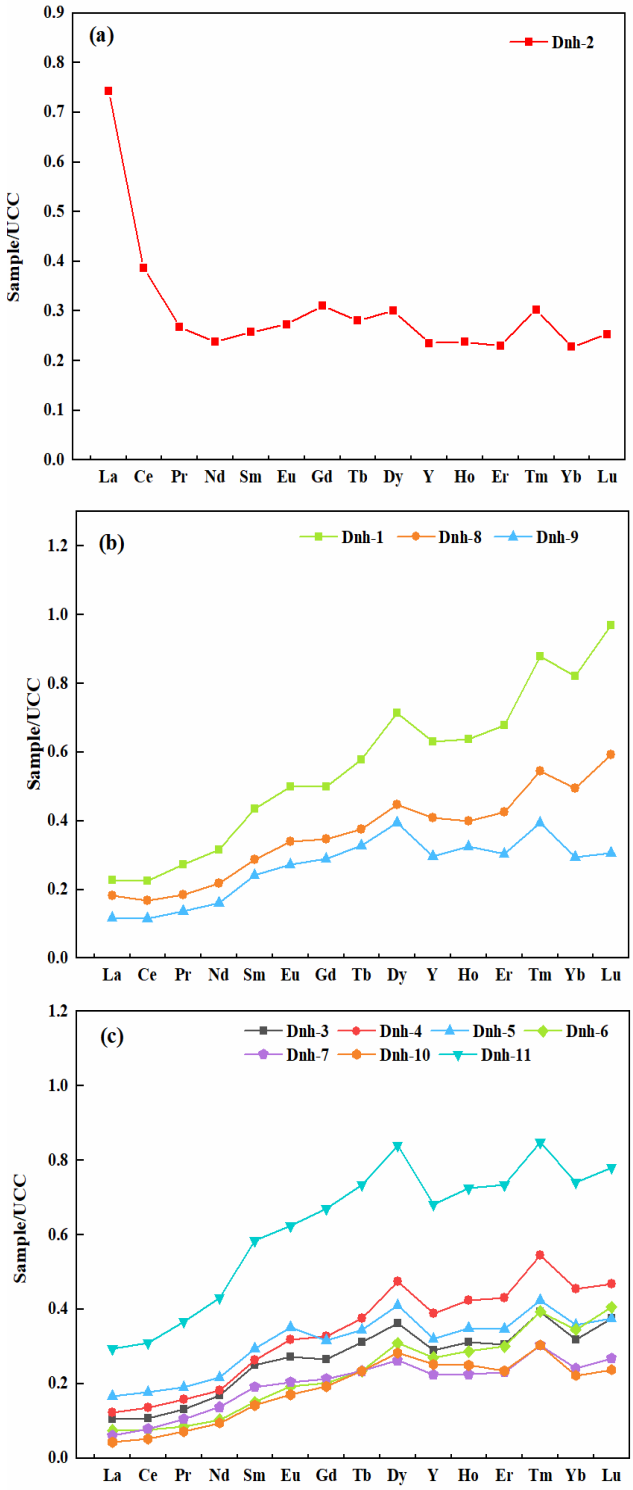


Figure 8. UCC-normalized REY distribution patterns of the Dananhu samples. (**a**) REY distribution pattern of Dnh-2; (**b**) REY distribution pattern of Dnh-1, Dnh-8 and Dnh-9; (**c**) REY distribution pattern of the rest samples.

5. Discussion

5.1. Evaluation of Critical Metal Elements

Table 4 calculated the average concentration of critical metal elements in the Dananhu coal ash. The average concentration of Li_2O in the Dananhu coal ash was 96.23 μg/g, which is higher than the world average (66 μg/g) [40] but much lower than the cut-off grade of

lithium recovery in coal (0.08%) [46]. Therefore, the lithium in the Dananhu coal cannot be used as an alternative resource.

The average concentrations of Ga and Ge in the Dananhu coal ash were 49.67 μg/g and 2.24 μg/g (on an ash basis, Table 4). Although the Ga content in raw coal was relatively low (3.53%, CC = 0.64, Table S1), the low ash yield of the Dananhu coal (8.17%, Table 1) makes the Ga content in the Dananhu coal ash close to the cut-off grade of Ga in coal ash (50 μg/g) [11,46], which has potential economic recovery value. However, the average content of Ge (2.24 μg/g) is far lower than the cut-off grade value of Ge in coal ash proposed by Dai et al. [46], which is 300 μg/g, so the Ge in the Dananhu coal has no potential economic recovery value.

The average content of $(Nb, Ta)_2O_5$ in the Dananhu coal ash was 67.10 μg/g, although it was slightly lower than the cut-off grade of the weathered Crust-type Nb (Ta) ore deposit (80–100 μg/g) [47]. However, the $(Nb, Ta)_2O_5$ concentrations (151.86 μg and 119.37 μg/g) of samples Dnh-5 and Dnh-11 reached the critical value of the weathering crust-type Nb (Ta) deposits. The $(Zr, Hf)_2O_5$ content in the Dananhu coal ash was 645.84 μg/g, which exceeded the marginal grade of the Coast Sand type Zr ore deposit (400–600 μg/g) [47]. Therefore, the Dananhu coal is considered a potential source of Nb (Ta) and Zr(Hf) ore deposits. Seredin and Dai [43] suggested that REY oxides (REO) and an outlook coefficient (Coutl, the ratio of the relative amount of critical REY (Nd, Tb, Eu, Dy, Y, and Er) for excessive REY (Ce, Tm, Ho, Yb, and Lu) is higher than 1000 μg/g and 0.7, respectively, and REY has a potential economic recovery value [48]. The relationship between REO and Coutl of the Dananhu coal is shown in Figure 9. The outlook coefficient was higher than expected at 0.7 for almost all samples. However, due to the low ash yield and the REY concentration, the REY of Dananhu coal is unpromising. In summary, the critical metal elements of Nb (Ta), Zr (Hf), and Ga in the combustion products (coal ash) of No. 22 coal in Dananhu Coal have potential economic recovery value.

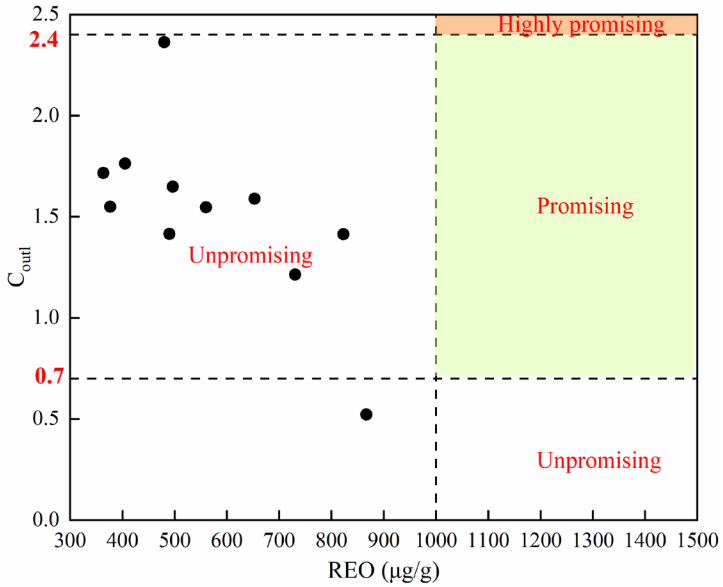


Figure 9. Evaluation of the economic potential of REY of the Dananhu coal.

5.2. Sedimentary Source Region

The Al_2O_3/TiO_2 ratio is a reliable parameter for determining the origin of sediments (coal seams) [9,45,49]. When $8 > Al_2O_3/TiO_2 > 3$, the rock type of the source is mafic igneous, while the rock type of the source is felsic igneous with $70 > Al_2O_3/TiO_2 > 21$. And those with Al_2O_3/TiO_2 between 8 and 21 are intermediate igneous [49]. The average value of the Al_2O_3/TiO_2 ratio of the Dananhu coals was 20.10 (ranging from 10.57 to 40.12), indicating that the sediment source of the Dananhu coals is felsic igneous rocks and intermediate igneous rocks (Figure 10a).

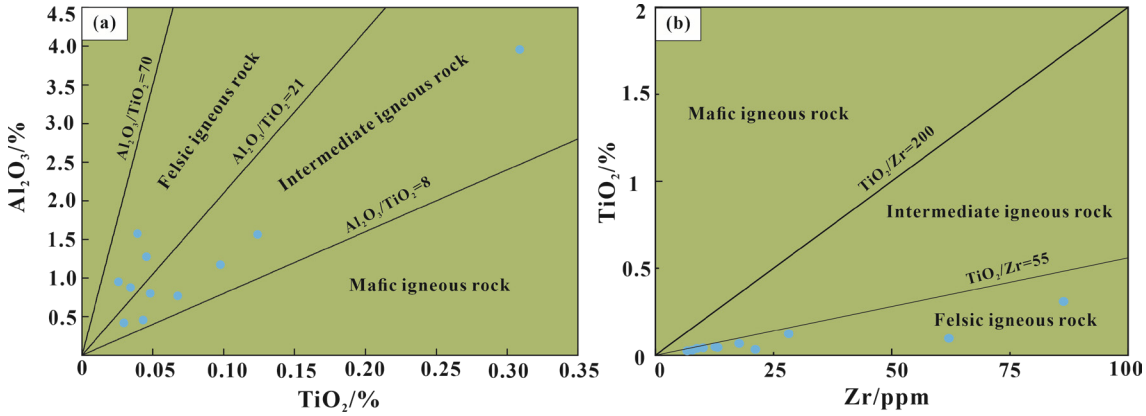


Figure 10. Sediment source of the No. 22 coals samples of the Dananhu Mine. (**a**) relationship between Al_2O_3 and TiO_2; **(b)** relationship between TiO_2 and Zr.

TiO_2/Zr ratios could also be used to indicate the origin of sediments. Generally, TiO_2/Zr ratios that are greater than 200 indicate mafic igneous rocks, while TiO_2/Zr ratios below 55 indicate felsic igneous rocks. Moreover, when $199 > TiO_2/Zr > 55$, it represents intermediate igneous rocks [49–51]. As shown in Figure 10b, the sedimentary sources of the Dannanhu coals are mainly felsic igneous and intermediate igneous rocks. The Dananhu depression has an overall paleogeographic pattern of high in the northeast and low in the southwest. Previous studies have suggested that the Jurassic sediment source mainly came from the north [14,15,18,19]. Wei et al. [41] suggested the sediment sources of the Sandaoling Mine, which is adjacent to the Dananhu Mine, come from the Paleozoic intermediate and felsic igneous rocks in Harlik Mountain and Eastern Bogda Mountain. Harlik Mountain and Eastern Bogda Mountain in the northeastern Turpan-Hami Basin may be potential sediment provenances (Figure 1b). In order to investigate the sediment source of the Dannanhu coals, the Al_2O_3/TiO_2 and Sr/Y of the Dannanhu coals were compared with those of the Paleozoic igneous rocks from the Harlik Mountain and Eastern Bogda Mountain in the Turpan-Hami Basin (Figure 11) [41,52–57]. The results show that the sediment source of the Dananhu coals is the felsic igneous and intermediate igneous rocks from Harlik Mountain and Eastern Bogda Mountain (Figure 11). Paleozoic dacite, andesite, and a few granite are the detrital sources of the No. 22 coal seam.

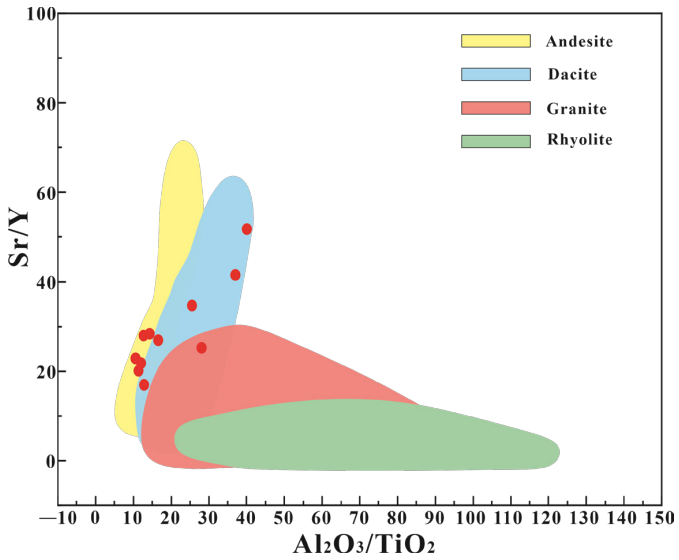


Figure 11. The diagrams between Sr/Y and Al_2O_3/TiO_2 of the No. 22 coal (Andesite, Dacite, Granite and Rhyolite data from [52–57]).

5.3. Depositional Environment

The Sr/Cu ratio is often widely used to judge the degree of dryness to the humidity of the climate. When the Sr/Cu ratio is greater than 10, it indicates a dry and hot climate; when the Sr/Cu ratio is between 1 and 10, it indicates a warm and humid climate [58]. The Sr/Cu ratio of the Dananhu coals varies greatly (9.78~40.42), with an average value of 19.51, indicating that the climate is mainly dry and hot (Figure 12a).

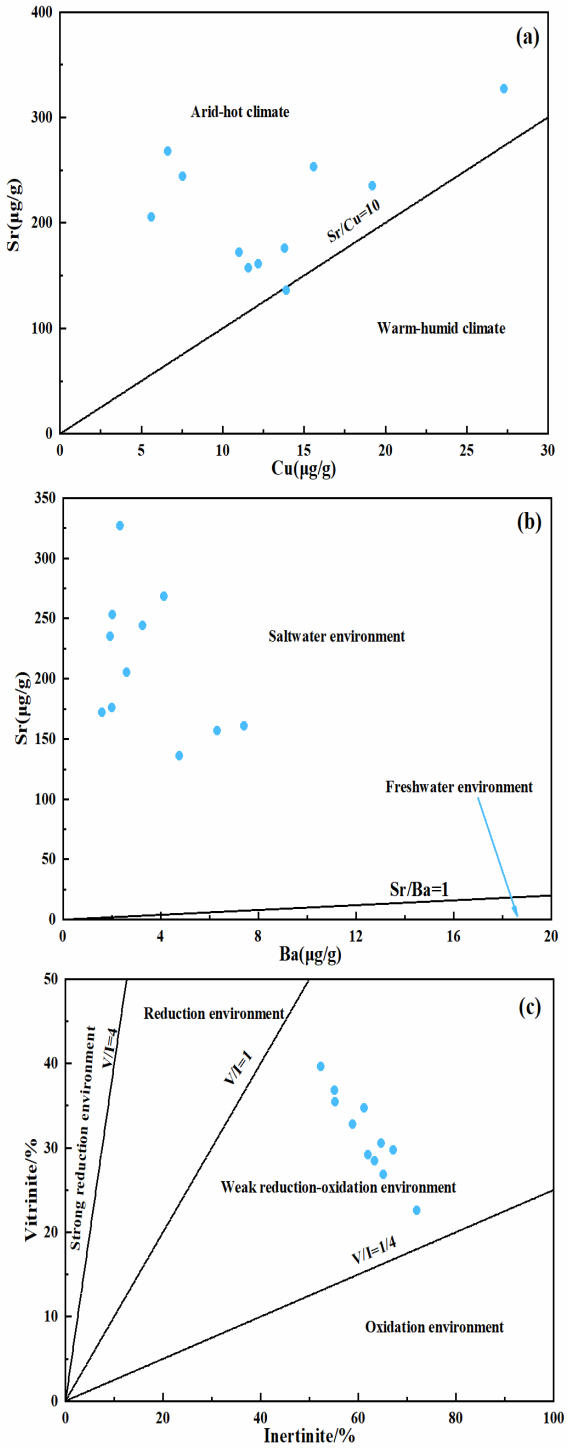


Figure 12. Depositional environment of the Dananhu samples: (**a**) Sr and Cu; (**b**) Sr and Ba; (**c**) Vitrinite and inertinite.

Sr is more easily enriched in sulfate-rich waters than Ba, so Sr/Ba can represent the degree to which peat swamps are affected by seawater [59]. Generally, a Sr/Ba >1 indicates that peat swamps are affected by seawater [60,61].

In the inland sedimentary system that is not affected by seawater, the continuous hot and arid environment will cause the evaporation of the peat swamp water to be greater than the recharge, increasing water salinity and thus leading to a higher Sr/Ba [62]; therefore, in inland sedimentary systems, high Sr/Ba values can be used to indicate the salinity of peat swamps water. Generally, a Sr/Ba > 1 indicates a saltwater environment or a hot and dry climate; on the contrary, a freshwater environment or a warm and humid climate [62–65]. The high Sr/Ba ratio (average 79.48) and low total sulfur content (average 0.18%, Table 1) indicate that the Dananhu coals are affected by the high salinity of the lake water (Figure 12b).

Vitrinite is formed by plant gelation, and a high vitrinite content usually indicates a deep water-covered reduction environment. At the same time, inertinite is generally the product of oxidation, so a high inertinite content usually means a shallow, dry oxidation environment [66,67]. Therefore, V/I can directly reflect the degree of water-covered swamps and the climate of wet and dry conditions. When V/I > 4, the peat swamps are a strong reduction environment; when 1 < V/I < 4, it is a reduction environment; when 1/4 < V/I <1, it is a weak reduction-oxidation environment; when V/I < 1/4, it is an oxidation environment [68]. The average value of V/I of the Dananhu coals is 0.52 (from 0.31 to 0.76), which is in a weak reduction-oxidation environment (Figure 12c).

5.4. Reasons for Low Elemental Contents in the Dananhu Coals

The content of trace elements of the No. 22 coal in the Dananhu Mine is low, especially Rb, Cs, Ba, Tl, Bi, and Ge (CC < 0.5, Table 3, Figure 6). Similarly, only Sr and Ba of the No. 4 coal in the nearby Sandaoling Mine are normal, and other elements are depleted (compared with world-hard coal) [41]. Identifying the causes of the low content of critical metal elements of the Dananhu coals is crucial for a thorough understanding of the associated elements in coals from northwestern China.

In general, the water properties of peat swamps (pH, Eh, H_2S, etc.) can produce certain geochemical barriers and affect the enrichment of critical metal elements in coal [69]. The No. 22 coal was mainly formed in a dry, hot, high salinity, and weak reduction-oxidation environment (Figure 11); on the one hand, the dry and hot climate leads to the decrease in weathering intensity in the sediment source area and the decrease in terrigenous detritus material input into peat swamps (ash yield of the No. 22 coal is 8.17%), while most of the critical metal elements (Li, Ga, Ge, Zr, Nb, Hf, and Ta) mainly occur in minerals [4]. Less input of terrigenous clastic material reduces the content of critical metal elements in the source. On the other hand, although the high salinity changes the water properties of the peat swamps (pH, Eh, and H_2S), making it more favorable for the enrichment of critical metal elements (Figure 13a), the weak reduction-oxidation environment is not favorable for the enrichment of critical metal elements (Figure 13b). In summary, the low terrigenous clastic material input and weak reduction-oxidation environment led to the low concentration of critical metal elements of the No. 22 coal.

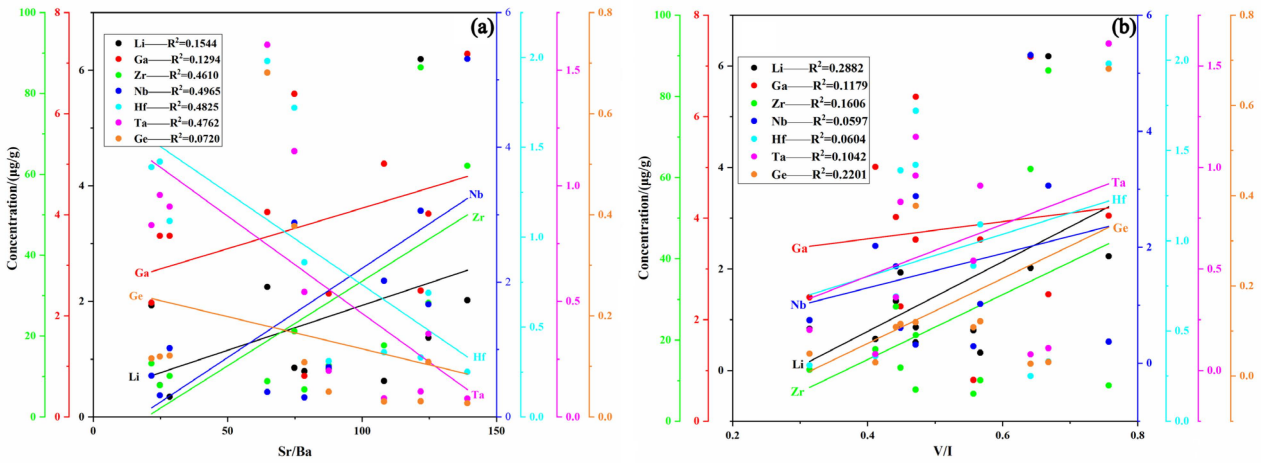


Figure 13. Relationship between depositional environment and critical metal elements of Dananhu coal samples: (**a**) Relationship between V/I and concentration of critical metal elements (Li, Ga, Ge, Zr, Nb, Hf, Ta); (**b**) The relationship between Sr/Ba and the concentration of critical metal elements (Li, Ga, Ge, Zr, Nb, Hf, Ta).

6. Conclusions

1. The Dananhu coals are characterized by a low ash yield, low total sulfur content, and high volatile yield. The inertinite is the most abundant maceral, followed by vitrinite, and the liptinite is less. Quartz, kaolinite, and illite are the main minerals. In addition, there are small amounts of calcite, chalcopyrite, anatase, and zircon.
2. Compared with the world's low-rank coals, Ni, Co, Mo, and Ta are slightly enriched, and Li, Rb, Cs, Ba, Tl, Bi, Ge, and other elements are depleted. The content of other trace elements is equivalent to the average values of the world's low-rank coals. The REY of the Dananhu coals shows high fractionation, mainly H-type- and M-H-type enrichment. The concentration of critical metals (Li, Ga, Ge, Zr, Nb, Hf, Ta, etc.) in the coal ash of the Dananhu coals is heterogeneous. The content of Ga, Zr (Hf), and Nb (Ta) in the coal ash has reached the cut-off grade of industrial mining, which has the potential to become a substitute for rare metal resources, while Li, Ge, and REY elements have no potential economic mining value.
3. The sediment sources of the Dananhu coal mainly come from the Paleozoic dacite, andesite, and a small amount of granite from Harlik Mountain and Eastern Bogda Mountain in the Turpan-Hami Basin. The Dananhu coals are generally in a dry and hot depositional environment with high salinity and weak reduction oxidation. Low source input and a weak reduction-oxidation environment have led to a low concentration of critical metal elements in the Dananhu coals.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/min14080767/s1, Table S1: the trace element content of the Dananhu coal.

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