Geochemical characteristics of No. 6 coal from Nanyangpo Mine, Datong coalfield, north China: Emphasis on the influence of hydrothermal solutions

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Abstract
This paper discusses the volcanic influence on the mineralogical and elemental geochemistry of No. 6 coal from Nanyangpo Mine in the Datong coalfield, north China. Sixteen samples (14 coal and 2 partings) were analyzed by different geochemical methods. The results revealed that the No. 6 coal is a medium–high volatility, highly calorific, bituminous coal with a low-ash and medium-sulfur content. The minerals mainly consisted of kaolinite, calcite, dolomite, and pyrite. In addition, traces of apatite, sphalerite, and anatase were also found. Calcite, dolomite, and pyrite in the coal samples were mainly derived from epigenetic hydrothermal solutions, whereas kaolinite and apatite originated from volcanic ash. Harmful trace elements including Pb, Cu, Mo, Tl, and Hg in the No. 6 coal samples were higher than those of common global coals. These harmful elements mainly occurred in sulfide minerals, which were probably influenced by hydrothermal inputs.

Keywords
Datong coalfield, hydrothermal solutions, mineralogy, harmful trace elements
Introduction

Global coal consumption grew by 1.4% in 2018, which was twice the average growth rate in the previous 10 years. Consumption growth was led by India and China (BP, 2019). Furthermore, coal utilization will continue to play a leading role in energy consumption in the next decades (Qin et al., 2015). Along with the massive consumption of coal, the processes such as storage, transportation, combustion, processing, and utilization have resulted in many harmful trace elements (e.g. Cu, Pb, Hg, Mo, and F) being discharged into the atmosphere, soil, and water (Baruah and Khare, 2010; U.S. Environmental Protection Agency, 2005; Xiao et al., 2016). The discharged toxic elements during coal processing would have potential adverse effects to the environment and human health (Yuan et al., 2018). For example, a high content of lead in ambient environment could cause children to suffer from a variety of health effects, including hyperactivity, lethargy, hearing, or memory loss (Yu et al., 2001). Moreover, Arroyo et al. (2018) found that a higher trace metals content in ambient air could accelerate aerosol aging, which causes air pollution. Because of the hazards to the environment and humans, the deleterious constituents and elements are required to be removed or minimized before utilization of coal using suitable techniques (Rajak et al., 2019; Singh et al., 2015).

The Datong coalfield is one of the most important coal-producing areas in China, with a total coal reserve of 71.8 billion tons (Kuang et al., 2019). In recent years, the annual coal output of the entire coalfield has been approximately 70 million tons. The produced coal is mainly transported to the Beijing-Tianjin-Hebei region. Some studies have been undertaken on the coal from the Datong coalfield; for example, Liu et al. reported on the concentration of trace elements in the No. 2 coal seam (Liu et al., 2013), and Yuan et al. (2018) reported the mineralogical compositions, geochemical characteristics, and origin of the trace elements in the No. 4 coal seam. Furthermore, hydrothermal solutions could affect the safety, productivity, and economic viability of coal mines (Qin et al., 2015); there are numerous studies that reported the effects of hydrothermal solutions on the organic matter of coal (Dutcher et al., 1966; Meyers and Simonneit, 1999; Ward et al., 1989). Information about the effects of hydrothermal intrusions on the inorganic constituents in coal may provide insights into the resource potential of coal and coke affected by the intrusion. However, little has been reported regarding the impact of hydrothermal solutions on inorganic constituents in the coal. Moreover, there is an absence of geochemical data for No. 6 coal, especially the occurrence and origin of harmful trace elements.

In this study, samples were collected from Nanyangpo Mine, which is located in the southern region of the Datong coalfield. Geochemical methods such as polarizing microscopy, scanning electron microscopy (SEM) in conjunction with an energy-dispersive X-ray spectrometer (EDS), inductively coupled plasma mass spectrometry (ICP-MS), X-ray diffraction (XRD), and X-ray fluorescence (XRF) were used to investigate the geochemical characteristics of the No. 6 coal seam in Nanyangpo Mine. The mineralogical characteristics and the occurrence and distribution of trace elements, especially harmful elements (e.g. Pb, Cu, Mo, Tl, and Hg), in this No. 6 coal, are discussed. The new data provide evidence for the occurrence of harmful elements in No. 6 coal, which were probably influenced by volcanic activities and hydrothermal inputs and provide a guiding significance for the comprehensive utilization of various types of coal in the Datong coalfield.
Geological setting

The Datong coalfield is located in the northern Shanxi Province, north China, between longitudes 112° 30′ E–113° 15′ E and latitudes 39° 35′ N–40° 12′ N (Figure 1(a)). The area is a Carboniferous (C)–Permian (P)–Jurassic (J) coal-bearing basin (Yang et al., 2017). The coal-forming environment of C–P coal in the Datong coalfield was a humid-reducing environment, and the sedimentary facies of C–P coal is marine-terrigenous facies, whereas the J coal was mostly deposited in a continental lake environment. There are two different magma intrusions in the Datong coalfield (Liu et al., 2013; Ying et al., 2007). The lamprophyre was formed during the Middle and Late Triassic ~23 billion years ago, and the diabase was formed during the late Early Cretaceous (~101 Ma) (Zhao et al., 2002). A series of reverse faults was developed on the southern and southeastern margins of the basin including the Qingciyao Fault, Wangjiayuan Fault, Meiyukou Fault, and Emaokou Fault from north to south. These faults probably provided channels for hydrothermal injections into the coal-bearing deposits (Yuan et al., 2018, 2019).

Nanyangpo Mine is located in the southwest region of the Datong coalfield and has an area of 7.58 km². The mineable coal seams are Nos. 2, 3, 5, 6, and 8. No. 6 is located with the Taiyuan Formation and has an average thickness of ~3 m. The roof of the No. 6 coal seam mainly comprises carbonaceous mudstones and some siltstone, whereas the floor is mainly siltstones.

Sampling and analytical procedures

**Sampling**

According to GB/T 482–2008 (2008) and in appreciation of the in situ mining situation, 16 bench samples of No. 6 coal from Nanyangpo Mine, consisting of six partings (NYP-2,
NYP-G1, NYP-G2, NYP-12, NYP-13, and NYP-14) and 10 coal benches (NYP-1, NYP-3, NYP-4, NYP-5, NYP-6, NYP-7, NYP-8, NYP-9, NYP-10, and NYP-11) with a total thickness of 1.6 m, were collected from incremental locations: the bottom to the top of the mine (Figure 1(b)). Each bench sample was cut over an area measuring 10 cm wide by 10 cm deep. All collected samples were immediately stored in plastic bags to minimize contamination and oxidation. Samples were freeze-dried and divided into two equal parts. One part was used for the analyses and the other part was stored as a reserve. For the petrographic and geochemical analysis, all samples were crushed to pass 80 and 200 mesh, respectively.

**Proximate analysis**

The proximate and related analyses including moisture, ash yield, volatile matter, total sulfur, and gross calorific values of the coals were based on ASTM standards D3173-11 (ASTM, 2011a), D3174-11 (ASTM, 2011b), D3175-11 (ASTM, 2011c), and D2492-02 (ASTM, 2007), and Chinese standards GB/T 15224.3–2010 (GB/T, 2010a), respectively.

**Mineralogical analysis**

Mineralogical compositions were determined by optical microscopy (Leica DM 2500P microscope by Leica Microsystems, Solms, Germany) and XRD (Rigaku, Japan). Low-temperature ashing (LTA, Quorum Technologies, UK) of coal was performed on an EMITECH K1050X plasma asher below 200°C (75 W power). XRD analysis on the resultant LTA samples was performed using a D/max-2200/PC powder diffractometer with Ni-filtered Cu-Kα radiation and a scintillation detector. The XRD patterns were recorded over a 2θ interval of 5–70° with a step size of 0.01°.

The 80-mesh size samples were used to analyze both the mineralogical composition and distribution. SEM (HITACHI UHR FE-SEM, SU8220, Japan)-EDS and optical microscopy were used to study the occurrence and distribution characteristics of the minerals. The gold-plating coating was performed on the sample before the analysis of samples by SEM-EDS.

**Elemental analysis**

Samples were crushed and ground to pass through a 200 mesh (75 μm) for analysis. Each coal or non-coal sample was high-temperature ashed at 815°C, and loss on ignition was calculated. XRF (ARL9800 XRF, Thermo Fisher Scientific, USA) was used to determine the oxides of major elements (SiO₂, TiO₂, Al₂O₃, Fe₂O₃, MgO, CaO, MnO, Na₂O, K₂O, and P₂O₅). Uniquant (version 5.46), a “standard-less” methodology based on a set of fundamental parameters and unique algorithms, was used for matrix correction and calibration of the XRF data. Trace elements, including rare earth elements and yttrium (REY), except for As, Se, Hg, and F, were determined via ICP-MS (X series II) in a pulse counting mode (three points per peak) The ICP-MS analyses were performed on raw coal samples. For the ICP-MS analysis, microwave digestion of an approximate 40-mg sample was weighed into Poly Tetra Fluoro Ethylene (PTFE) vessels; 2 mL of HF (50%) + 5 mL of HNO₃ (65%) + 2 mL of H₂O₂ (30%) were added, and microwave digestion was performed for 75 min at a temperature of 200°C. This solution was then transferred into 125-mL fluorinated ethylene propylene (FEP) bottles that were filled with 100 g of deionized water (Zhao
et al., 2012). Mercury was determined using a Milestone DMA-80 Hg analyzer (Milestone, Sorisole, Italy).

Results

Coal characteristics

Table 1 provides the results of proximate analysis of the No. 6 coal samples. The results showed that the samples contained low-ash and medium-sulfur contents, were highly-calorific, and of a medium-high volatility based on the Chinese standards GB/T 15224.1–2010 (coals with an ash yield range from 5.01% to 10.00% are low-ash coal) (GB/T, 2010b), GB/T 15224.2–2010 (coal with a total sulfur content <2.00% and >1.51% are defined as medium-sulfur coal) (GB/T, 2010c), GB/T 15224.3–2010 (according Chinese National standards high calorific coal can contribute calorific value 24.01–27.00 MJ/kg) (GB/T, 2010a), and MT/T849–2000 (≤10.00% for super-low-volatility coal, 10.01%–20.00% for low-volatility coal, 20.01%–28.00% for medium-volatility coal, 28.01%–37.00% for medium-high-volatility coal, 37.01%–50.00% for high-volatility coal, and >50.00% for super-high-volatility coal) (MT/T, 2010), respectively.

Minerals in the coal samples

The XRD data (Figure 2) of the LTA and the results of optical microscopy observations showed that the No. 6 coal samples were mainly composed of clay minerals (kaolinite), carbonate minerals (calcite, dolomite), and sulfide minerals (pyrite, sphalerite), along with trace amounts of anatase and apatite.

Clay minerals. The clay minerals in No. 6 coal samples mainly occurred as vermicular (Figure 3(a)) and cell-fillings (Figure 3(b)) with a microgranular surface. This is common

Table 1. Proximate analysis, total sulfur analysis and calorific value analysis of coal sample the from Nanyangpo mine.

| Sample  | \(M_{ad}\) | \(A_{ad}\) | \(V_{daf}\) | \(S_{ad}\) | \(Q_{gr,ad}\) (\(Q_{gr,ad}\)) |
|---------|-------------|-------------|-------------|-------------|---------------------------|
| NYP-1   | 4.51        | 9.35        | 38.98       | 1.11        | 28.60                     |
| NYP-3   | 3.84        | 9.40        | 30.02       | 0.44        | 28.81                     |
| NYP-4   | 4.12        | 7.20        | 29.37       | 0.55        | 29.11                     |
| NYP-5   | 3.51        | 13.05       | 31.15       | 0.61        | 26.95                     |
| NYP-6   | 4.58        | 8.65        | 35.53       | 0.76        | 28.84                     |
| NYP-7   | 3.50        | 13.60       | 36.50       | 4.24        | 26.77                     |
| NYP-8   | 4.22        | 9.40        | 36.90       | 1.64        | 26.52                     |
| NYP-9   | 3.94        | 8.75        | 35.30       | 1.18        | 29.23                     |
| NYP-10  | 3.12        | 15.55       | 36.88       | 2.26        | 28.08                     |
| NYP-11  | 2.64        | 27.95       | 31.63       | 2.39        | 22.15                     |
| NYP- ZHI | 4.39       | 11.10       | 33.47       | 0.77        | 27.36                     |
| NYP-ZH2 | 4.15        | 17.80       | 28.86       | nd          | 27.66                     |
| Ave.    | 3.88        | 12.65       | 33.72       | 1.45        | 27.51                     |

Ave.: average of coal samples; A: ash yield; M: moisture; V: volatile matter; S: total sulfur; \(Q_{gr,ad}\): gross calorific value, on a dry basis; nd: not determined; ad: on air dry basis; daf: on dry and ash free basis.
in many other coals and closely associated coal-bearing strata (Ward, 2016). Clay minerals with this kind of morphology indicate that clay–mineral formation was related to the deposition of the volcanic ash and that it probably took place during early diagenesis (Vejahati et al., 2010). XRD and EDS data revealed that the main identifiable clay mineral in the No. 6 coal samples was kaolinite.

**Carbonate minerals.** Carbonate minerals are commonly formed during the authigenic, epigenetic, and post-coalification stages (Kortenski, 1992). The carbonate minerals in coal usually include calcite and siderite, although some coal also contain large amounts of dolomite, aragonite, and magnesite (Wang et al., 2019; Zhao et al., 2007). According to the SEM-EDS and XRD analyses, the main carbonate minerals identified in the samples were calcite and dolomite.
Calcite. Various forms of calcite were observed via optical microscopy and SEM, such as fragmental structure (Figure 4(a)), cell-filled, vein-filled (Figure 4(b)), calcite, sphalerite, and dolomite symbiosis. Generally, square-flake calcite was formed during the syn-sedimentary stage, whereas cell-filling and vein-filling calcites were formed during the epigenetic stage, and a few parts were formed within sediments (Sun et al., 2010).

Dolomite. Numerous dolomites were found in the No. 6 coal samples and most of them occurred as lumps (Figure 4(c)) that were densely distributed (Figure 4(d)). The origin of these kinds of dolomites is often associated with the invasion of hydrothermal solutions. Zhao et al. (2014) summarized that the occurrence of dolomite in metamorphic coal and bituminous coal has mostly been influenced by hydrothermal intrusions.

Sulfide minerals. XRD patterns revealed that the main identifiable sulfide mineral in the samples was pyrite, which is one of the most common sulfides in coal (Liu et al., 2009). Pyrite was mostly found as lumps, cell-filled (Figure 5(a)), fissure-filled (Figure 5(b)), and as a trace of disseminated pyrite. Agglomerated pyrite exhibited a rough surface and irregular shape with a diameter of up to 200 μm. Fissure-filled pyrite was generally filled in cells such as in the fusinite cell. Fissure-filled pyrite mainly co-occurred with calcite and dolomite, and usually filled in cracks or cut through the organic components. It is generally believed that agglomerated pyrite forms during the early diagenetic stage, whereas cell-filled pyrite forms during the late diagenetic stage, and fissure-filled pyrite forms during the epigenetic stage (Altschuler et al., 1983; Vejahati et al., 2010).

![Figure 4. Carbonate minerals in coal. (a) Optical photomicrographs of calcite (detrital calcite); (b) optical photomicrographs of calcite (vein filled); (c) SEM image (secondary electron excitation) of dolomite, pyrite and calcite; (d) SEM image (secondary electron excitation) of dolomite (densely distributed) and pyrite.](image_url)
Other minerals. Traces of anatase, sphalerite, and apatite were also found via SEM-EDS. Most anatases were of a regular polygon shape with a smooth surface, and generally occurred as fine-grained anatase within the kaolinite (Figure 6(a)). Anatases were an important carrier of Ti in the No. 6 coal samples. Volcanic anatase has been observed in altered mafic volcanic ash (Sun and Puttmann, 1996).

Apatite was rare in the studied coal and was often below the detection limit of the XRD analysis; hence it could only be observed using SEM. The apatite in the samples mostly existed as individual particles and represented the most important carrier of P (Figure 6(b)), thus suggesting that the formation of the No. 6 coal seam was closely related to volcanic activity.

In addition, traces of sphalerite were observed via SEM-EDS, which revealed that sphalerite was fissure filled (Figure 6(c)) and usually in symbiosis with calcite.

Major elements

The proportion of major oxides (Al$_2$O$_3$, SiO$_2$, Fe$_2$O$_3$, CaO, P$_2$O$_5$, TiO$_2$, MgO, K$_2$O, Na$_2$O, and MnO) in the No. 6 coal samples in comparison to the averages for other Chinese coals is listed in Table 2. This shows that the average content of P$_2$O$_5$ (0.192%) in the samples was much higher than that of Chinese coals (0.092%) (Xu et al., 2011), whereas the average contents of other major elements were lower than those of Chinese coals. The enrichment factor (EF = average elemental concentration ratio of coal samples investigated vs. average of Chinese coals) of P$_2$O$_5$ reached 2.09, which was probably related to the volcanic ash produced by volcanic activity (Ward, 2002). The SiO$_2$/Al$_2$O$_3$ ratio of the No. 6 coal samples (average 0.98) was lower than that of Chinese coals (average 1.42), and may have related to the absence of quartz (Sun et al., 2016).

Harmful trace elements

The average abundance of selected trace elements in the samples was compared to those of global bituminous coal (Ketris and Yudovich, 2009), as presented in Table 3. Some harmful elements (e.g. Pb, Hg, Tl, Cu, and Mo) were enriched in the No. 6 coal samples (concentration coefficient, CC, 2–5), although one sample (NYP-7) was highly enriched.
These harmful elements are significantly related to volcanic activity (Zhang et al., 2013), and the highly enriched sample may indicate the intermittent influence of volcanic activity during or after early diagenesis in the study area.

Rare earth elements and yttrium

In this study, the REY are classified into three groups according to their geochemical characteristics: light (LREY: La, Ce, Pr, Nd, and Sm), medium (MREY: Eu, Gd, Tb, Dy,
Table 2. Concentrations of major oxides in the Nanyangpo coal samples, as well as their comparison with averages for Chinese coals (%).

| Sample  | Al₂O₃ | SiO₂ | Fe₂O₃ | CaO | P₂O₅ | TiO₂ | MgO | K₂O | Na₂O | MnO | SiO₂/Al₂O₃ |
|---------|-------|------|-------|-----|------|------|-----|-----|------|-----|-------------|
| NYP-1   | 2.65  | 2.47 | 0.77  | 2.21| 0.006| 0.28 | 0.08| 0.07| 0.06 | 0.04| 0.0083     |
| NYP-3   | 3.81  | 3.61 | 0.15  | 0.90| 0.238| 0.15 | 0.04| 0.04| 0.004| 0.04| 0.035       |
| NYP-4   | 3.32  | 3.18 | 0.05  | 0.13| 0.285| 0.08 | 0.01| 0.03| 0.003| 0.03| 0.003       |
| NYP-5   | 5.95  | 6.35 | 0.06  | 0.33| 0.083| 0.14 | 0.03| 0.02| 0.004| 0.004| 0.0008      |
| NYP-6   | 3.89  | 3.84 | 0.05  | 0.36| 0.395| 0.07 | 0.02| 0.01| 0.004| 0.015| 0.0010      |
| NYP-7   | 5.32  | 4.76 | 3.43  | 1.17| 0.293| 0.10 | 0.08| 0.03| 0.033| 0.013| 0.0028      |
| NYP-8   | 3.15  | 2.80 | 1.27  | 1.34| 0.459| 0.06 | 0.05| 0.01| 0.005| 0.005| 0.0052      |
| NYP-9   | 3.89  | 3.91 | 0.43  | 0.40| 0.085| 0.09 | 0.07| 0.03| 0.006| 0.004| 0.0040      |
| NYP-10  | 6.07  | 5.94 | 2.31  | 1.33| 0.079| 0.06 | 0.16| 0.03| 0.010| 0.0079| 0.98       |
| NYP-11  | 12.57 | 12.87| 2.74  | 0.92| 0.074| 0.48 | 0.13| 0.063| 0.031| 0.0043| 1.02       |
| NYP-ZH1 | 5.05  | 5.36 | 0.13  | 0.04| 0.005| 0.24 | 0.03| 0.077| 0.009| 0.0002| 1.06       |
| NYP-ZH2 | 8.18  | 8.38 | 0.20  | 0.20| 0.297| 0.30 | 0.03| 0.031| 0.006| 0.0008| 1.02       |
| Ave     | 5.32  | 5.29 | 0.97  | 0.78| 0.192| 0.15 | 0.09| 0.028| 0.007| 0.0033| 0.98       |
| China   | 5.98  | 8.47 | 4.85  | 1.23| 0.092| 0.33 | 0.22| 0.190| 0.160| 0.0150| 1.42       |
| EF      | 0.89  | 0.62 | 0.20  | 0.63| 2.09 | 0.45 | 0.41| 0.15| 0.04 | 0.22 | nd          |

Ave.: average of coal samples; EF: enrichment factor = Ave./China.

Table 3. Concentrations of harmful elements in the Nanyangpo coal samples and parting, as well as their comparison with averages for world bituminous coals (µg/g).

| Sample  | Cu  | Mo | Tl | Pb | Hg |
|---------|-----|----|----|----|----|
| NYP-1   | 3.85| 3.68| 0.17 | 12.34| 0.09|
| NYP-2   | 5.39| 2.4 | 0.04 | 31.45| 0.1 |
| NYP-G1  | 8.65| 1.96| 0.04 | 30.51| 0.05|
| NYP-3   | 3.96| 1.41| 0.03 | 12.42| 0.02|
| NYP-4   | 14.4| 0.933| 0.05 | 19.94| 0.03|
| NYP-5   | 15.22| 0.969| 0.03 | 20.9 | 0.03|
| NYP-G2  | 5.29| 2.59| 0.09 | 15.67| 0.4 |
| NYP-6   | 16.86| 2.08| 0.04 | 10.13| 0.04|
| NYP-7   | 17.96| 5.9 | 5.6 | 80.62| 1.61|
| NYP-8   | 20.53| 2.86| 0.02 | 24.17| 0.1 |
| NYP-9   | 10.37| 2.54| 0.44 | 8.32 | 0.35|
| NYP-10  | 21.78| 4.75| 2.08 | 40.32| 0.97|
| NYP-11  | 28.18| 5.84| 1.49 | 35.04| 0.67|
| NYP-12  | 32.43| 12.2| 0.05 | 35.71| 0.1 |
| NYP-13  | 24.65| 4.87| 0.06 | 28.07| 0.14|
| NYP-14  | 35.18| 11.1| 0.17 | 37.77| 0.63|
| Ave (coal) | 17.05| 3.10| 1.00 | 25.52| 0.39|
| Ave (parting) | 18.99| 5.85| 0.08 | 29.86| 0.24|
| Ave     | 17.78| 4.13| 0.65 | 27.15| 0.33|
| World   | 16   | 2.1 | 0.58 | 9    | 0.1 |
| CC      | 1.11 | 1.97| 1.12 | 3.02 | 3.33|

Ave.: average of coal samples; CC: concentration coefficients = Ave./world.

From Ketris and Yudovich (2009).
and Y), and heavy (HREY: Ho, Er, Tm, Yb, and Lu) (Sun et al., 2015). Furthermore, three enrichment types are identified based on geochemical parameters (Zhao et al., 2017): L-type (light-REY: \( \text{LaN/LuN} > 1 \)), M-type (medium-REY: \( \text{LaN/SmN} < 1, \text{GdN/LuN} > 1 \)), and H-type (heavy REY: \( \text{LaN/LuN} < 1 \)). Additionally, normal type (N-type) of REY distribution patterns represents very weak or no fractionation among the L-, M-, and H-REY (Arbuzov et al., 2019).

The concentrations of total REY (from La to Lu plus Y) in coal samples are averaged at 98.65 µg/g (Table 4), which is higher than that of world bituminous coals (60 µg/g) (Ketris and Yudovich, 2009). In particular, the sample No. NYP-3 is mostly enriched in REY with a total REY content of 266.98 ppm. The REY distribution patterns for the NYP-3, NYP-4, and NYP-6 are different from that of the other samples in No. 6 Coals (Figure 7(a) and (b)). The REY enrichment in the three samples belong to L-type (\( \text{LaN/LuN} = 2.01 \), Table 4) and have higher average REY concentration than the other samples, with a strong positive Ce-anomaly (average of NYP-3, NYP-4, and NYP-6 of \( \text{Ce/Ce}^* = 10.23 \)). The REY enrichment in the other samples falls into H-type (\( \text{LaN/LuN} = 0.56 \), Table 4). The ratio of GdN to GdN* of all the samples varies from 0.73 to 1.40, with an average ratio of 1.10.

**Discussion**

**Minerals in No. 6 coal samples**

**Calcite and dolomite.** Calcite and dolomite in the No. 6 coal samples are thought to be mainly derived from the deposition of epigenetic hydrothermal solutions. In addition, only traces of detrital calcites (Figure 5(a)) were found in the samples. Detrital calcites in coal usually indicate an alkaline environment (Rao and Walsh, 1999; Wang et al., 2016a). Given that calcite can be easily dissolved in an acidic environment, the formation of authigenic kaolinite is favored, which is consistent with the abundant kaolinite in the No. 6 coal samples. As described above, the dolomite that occurs as veins, cell infillings, and fracture infillings in the No. 6 coal can be considered to be precipitated from Fe–Mg–Ca-rich hydrothermal

| Sample | REY  | LREY | MREY | HREY | (La/Lu)N | (La/Sm)N | (Gd/Lu)N | GdN/GdN* | CeN/CeN* |
|--------|------|------|------|------|----------|----------|----------|----------|----------|
| NYP-1  | 24.91| 8.37 | 13.14| 3.40 | 0.03     | 0.07     | 0.46     | 0.73     | 0.01     |
| NYP-3  | 266.98| 232.71| 28.57| 5.70 | 2.17     | 1.82     | 1.49     | 1.40     | 14.57    |
| NYP-4  | 253.15| 215.83| 31.68| 5.65 | 1.85     | 1.19     | 1.78     | 1.36     | 11.78    |
| NYP-5  | 51.37 | 31.15 | 16.25| 3.97 | 0.38     | 1.10     | 0.44     | 0.92     | 0.24     |
| NYP-6  | 155.92| 129.74| 22.67| 3.51 | 2.01     | 1.18     | 2.09     | 1.31     | 4.34     |
| NYP-7  | 50.75 | 37.39 | 11.33| 2.03 | 0.98     | 1.13     | 1.11     | 1.20     | 0.35     |
| NYP-8  | 70.93 | 56.47 | 12.23| 2.23 | 1.41     | 1.56     | 1.18     | 1.25     | 0.82     |
| NYP-9  | 29.22 | 20.23 | 7.35 | 1.64 | 0.57     | 0.92     | 0.74     | 0.96     | 0.10     |
| NYP-10 | 41.62 | 25.28 | 13.67| 2.68 | 0.24     | 0.26     | 1.00     | 0.92     | 0.13     |
| NYP-11 | 41.59 | 26.17 | 12.56| 2.86 | 0.29     | 0.44     | 0.72     | 0.94     | 0.15     |
| AVE    | 98.65| 78.33| 16.95| 3.37 | 0.99     | 1.17     | 1.16     | 1.10     | 1.56     |

**Table 4.** REY parameters of the coal samples from the Nanyangpo mine.

AVE: average of coal samples. \( \text{GdN/GdN}^* = \text{GdN}/(0.33\text{SmN} + 0.67\text{TbN}) \); \( \text{Ce/Ce}^* = 2\text{CeN}/(\text{LaN} + \text{PrN}) \); REY are normalized by Upper Continental Crust (UCC) when \( \text{(La/Lu)}_N, (\text{LaN/SmN})_N, (\text{Gd/Lu})_N, (\text{La/Yb})_N \); Eu/Eu* and Ce/Ce* are calculated.
fluids circulating in the coal seams in the late stage of diagenesis (Karayiğit et al., 2017; Ward, 2002, 2016). The modes of occurrence of the epigenetic dolomite provide further evidence that there may be injection by low temperature hydrothermal fluids into the paleomire (Arbuzov et al., 2019).

Pyrite and sphalerite. As described in the ‘Geological setting’ section, the coal-bearing strata of the Taiyuan Formation were deposited in a marine-terrestrial transitional environment. Seawater may have influenced the mineralogy of the No. 6 coal during the peat-accumulation process. For example, the formation of pyrite (e.g. framboids, euhedral crystals, and massive form) (Figure 4(c) and (d)) is generally induced by seawater during peat accumulation or early diagenesis (Naik et al., 2016; Ward, 2002), and in this study, the presence of pyrite may also suggest the influence of seawater. In addition to seawater, the pyrite in the No. 6 coal samples is also thought to be derived from the deposition of epigenetic hydrothermal solutions. Fracture-filling pyrite (Figure 6(b)) also suggests multiple hydrothermal inputs into the coal. The distribution and modes of sphalerite in the samples also provide strong evidence for hydrothermal intrusions (Dai and Ren, 2007; Gulick, 1998).
Kaolinite. Kaolinite is the most abundant mineral in altered volcanic ashes and similar materials in coal (Staub and Cohen, 1978) and coal-bearing sequences. The kaolinite in the No. 6 coal samples was mainly of an authigenic origin. In addition, a portion of kaolinite was be found as vermicular shape; the occurrence of kaolinite could be an indicator that it was derived from volcanic ash (Dai et al., 2017). A study has reported that such vermicular kaolinite is generally regarded as a typical feature of tonsteins derived from altered volcanic ash layers deposited in peat swamps (Bohor and Triplehorn, 1993).

Other minerals. Apatite were observed in the samples NYP-6, NYP-7, and NYP-8, which is consistent with the relatively high concentration of P₂O₅ listed in Table 2. Apatite in coal is usually affected by volcanic ash. Most of the apatite in the No. 6 coal samples was authigenic, thus indicating that the origin of apatite was probably volcanic ash (Dai et al., 2017; Liu et al., 2013; Ward, 2016; Ward et al., 1996, 2016a). Anatase was closely associated with authigenic kaolinite in the No.6 coal; it indicates that anatase have co-precipitated with the kaolinite-forming material in this study. A previous study by Dai et al. (2015) showed that fine-grained anatase within kaolinite was often associated with acidic hydrothermal solutions.

Harmful elements

Figure 8 shows that the vertical variation trends of environmentally sensitive elements Cu, Mo, Tl, Hg, Pb, and S were very similar, which suggest that these elements probably had the same source region or origin. Figure 8 also suggests that the content of most harmful elements (Cu, Mo, Tl, Hg, Pb, and S) in sample NYP-7 was higher than other samples. Table 3 shows that the average content of Cu, Mo, Tl, Hg, Pb, and S in the No. 6 coal samples from Nanyangpo Mine was higher than those of global bituminous coals. This might indicate that hydrothermal intrusions had a significant influence on the individual
coal layers in the No. 6 coals (Zhang et al., 2013), thus leading to the increased content of harmful elements in the coal from the Nanyangpo Mine (Wang et al., 2016b; Zhang et al., 2013). The significant enrichment of these harmful trace elements may bring potential harm to the environment during transportation, combustion, processing, and utilization of the No. 6 coals from the Nanyangpo Mine.

**Copper.** The Cu concentration in the No. 6 coal samples varied from 3.85 to 36.38 μg/g with a weighted average of 17.78 μg/g (Table 3), which was slightly higher than the average Cu in global bituminous coal (average 16.0 μg/g) (Ketris and Yudovich, 2009).

Cu is one of the harmful trace elements in coal. It can be deduced that Cu mainly occurred in the sulfur minerals based on the positive correlation of the Cu-S yield (R² = 0.7677, Figure 9(a)). Although the average Cu content in the entire coal seam was only slightly higher than the value for global hard bituminous coal (Ketris and Yudovich, 2009), the concentrations of Cu are relatively high in the lower coal samples of the No.6 coal seam (from NYP-7 to NYP-11). Especially, the content of sample NYP-7 reached 36.38 μg/g (Table 3), which was regarded as hydrothermal intrusion of origin, and then the element sedimented to the lower coal benches. However, the higher coal bench samples have fairly low concentration of Cu, as well as the parting layers; it may indicate that the Cu in higher coal bench samples may be derived from the felsic and felsic-intermediate rocks (Zhao et al., 2014).

**Molybdenum.** The average concentration of Mo in the No. 6 coal samples was 4.13 μg/g (sample NYP-7: 5.9 μg/g), which was higher than that of common global bituminous coals (2.1 μg/g) (Ketris and Yudovich, 2009). The high correlation coefficients of Mo-S (0.79, Figure 9(b)) and Mo-Fe₂O₃ (0.90, Figure 9(c)), thus indicating that sulfur minerals (pyrite) were the main carrier of Mo in our samples. As indicated by Sun et al. (2013), high concentrations of Mo that occur through coal seam sections were probably largely derived from hydrothermal solutions during peat accumulation or in the early diagenetic stages. The similar vertical variation trends of Mo and other five harmful elements (Figure 8) provide further evidence of the hydrothermal solutions origin of Mo.

**Mercury.** Hg is a chalcophile element that is toxic and volatile (Kolker et al., 2006). Although the Hg content of coal is generally low, the large consumption of coal in China has resulted in the release of Hg from coal being an important source to the atmosphere (Guo, 2018). The average Hg content of the No. 6 coal samples (0.33 μg/g) was higher than that of common Chinese coal (Ward, 2002, 2016).

The low correlation coefficient between Hg and ash (0.21, Figure 9(d)) along with the high correlation coefficient of Hg and S (0.91, Figure 9(e)) suggest that Hg was mainly associated with pyrite or organic sulfur. Some researchers believe that the high content of Hg in coal relate to an alteration by hydrothermal intrusion (Finkelman et al., 2019; Yang et al., 2016). When the heat from a hydrothermal solution dissipates and the temperature decreases or reaches the surrounding temperature, Hg re-precipitates in solution or interacts with the cooling hydrothermal solution, and is subsequently absorbed by the coal (Sun et al., 2000). The enrichment of Hg in Sample NYP-7 indicates that when the heat dissipated and temperatures dropped to ambient levels, Hg may come from the solutions, which were derived from or had interacted with the cooling solution and then deposited in the coal. Another explanation is that Hg was driven off from the organic component
in coal by solution heat and then redeposited nearby, leading to the enrichment of Hg in sample NYP-7.

**Thallium.** The Tl concentration in the studied samples varied from 0.02 to 5.6 μg/g, with an average of 1.00 μg/g, which was higher than that of global bituminous coals (0.58 μg/g) (Ketris and Yudovich, 2009). The vertical variation trends of Tl and Hg (Figure 8) in the

![Figure 9](image-url)  
**Figure 9.** Correlation coefficients between ash yield and selected elements and between selected trace elements and major elements yield of the Nanyangpo coals.
No. 6 coal seam were almost identical, thus indicating that Hg and Tl may have had the same carrier. Furthermore, the high correlation coefficients between Tl and S ($R^2 = 0.89$; Figure 9(f)) suggest that sulfide containing matters were probably the carrier for both Tl and Hg. As reported by Dai et al. (2013), the high-Tl coals are predominantly attributed to hydrothermal solutions. The highest concentration of Tl was also found in the sample NYP-7 (Table 3); the result also indicates that there was a hydrothermal solution influence.

**Lead.** The average concentration of Pb in the samples was 25.52 $\mu$g/g, which was higher than that of common global bituminous coals (7.8 $\mu$g/g) (Ketris and Yudovich, 2009). Galena and/or some sulfide minerals are the main hosts for Pb in coal (Finkelman et al., 2019). Figure 9(g) shows that the relationships between Pb and Al-Si in the No. 6 coal sample were poor. A CC of 0.11 along with the high correlation coefficient between Pb and Fe$_2$O$_3$ (0.80, Figure 9(h)) suggest that the occurrence of Pb in the No. 6 coal samples may be associated with sulfide minerals such as pyrite and not aluminosilicate minerals. According to the result, the origin of Pb in the No.6 coal seam was associated with hydrothermal solution rather than volcanic ash.

**Abnormal of REY**

The distribution pattern and content of rare earths in samples NYP-3, NYP-4, and NYP-6 are different from other samples. The REY enrichment in the three samples belong to L-type, indicating that the three abnormal samples probably have terrigenous or tuffaceous origin (Arbuzov et al., 2019). Besides, the three samples have a higher total average REY concentration than the other samples, with a strong positive Ce-anomaly (average of NYP-3, NYP-4, and NYP-6 of Ce/Ce* = 10.23). A study by Arbuzov et al. (2019) on the REY in coals suggested that groundwater or hydrothermal leaching of partings can cause a strong positive Ce anomaly both in the partings and the underlying coal benches.

Gadolinium anomalies in coal have rarely been discussed (Zhao et al., 2017). Gd-positive anomalies are mainly controlled by several factors, such as the rocks in the sediment source region of coal basins, the influence of seawater and hydrothermal fluids (Sun et al., 2015). Previous studies suggested that the coals preserved within carbonate successions show weak positive Gd anomalies (Sun et al., 2015). The positive anomalies in Gd of the No. 6 samples (average of all the samples= 1.10) indicate that the formation of rare earth elements probably be influenced by hydrothermal fluid.

**Conclusion**

The No. 6 coal from the Nanyangpo Mine is classified as a low-ash (12.65%), medium-sulfur (1.45%), high-calorific (27.51 MJ/kg), and medium-high volatile (33.72%) coal. Minerals in the No. 6 coal mainly include kaolinite, calcite, dolomite, and pyrite, with trace amounts of anatase, sphalerite, and apatite. Calcite, dolomite, and pyrite in the coal samples studied here are thought to be mainly derived from epigenetic hydrothermal solutions, whereas kaolinite and apatite were probably influenced by volcanic ash.

REY enrichment patterns of the coal benches in the present study are characterized by alternative L-type and H-type. The high consistency of harmful trace elements (Cu, Mo, Tl, Hg, and Pb) in the profiles and the high correlation coefficient between these elements and S
suggest that these harmful elements had the same carrier (e.g. pyrite) in No. 6 coal. And the abnormal REY indicates the influence of hydrothermal solutions.

The influence of hydrothermal intrusion means that the contents of harmful elements (e.g., Cu, Mo, Tl, Hg, Pb, and S) in the No. 6 coal from the Nanyangpo Mine (especially that of the significantly enriched sample NYP-7) are higher than those of global bituminous coal. The EF of P2O5 reached 2.09 and was related to the volcanic ash produced by volcanic activity. The significant enrichment of harmful elements may bring potential harm to the environment during transportation, combustion, processing, and utilization of the No. 6 coals from Nanyangpo Mine.

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