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Occurrence and Environmental Features of Heavy Metal Elements of Coal from Coalfield in the Southeast Shanxi

Duan-Le Li, Dong-Min Wang*, Yi-Ren Wang, Cai-Fu Ren and Ning Yuan

School of Chemical and Environmental Engineering, China University of Mining and Technology, Beijing 100083, PR China

Abstract

The coal was sampled from No.15 coal seam in coalfield of southeast Shanxi. The Hg, As, Se, Cr and Pb occurrence in coal seam and environmental features of the heavy metal elements of coal were studied by X-ray fluorescence spectrometry (ARL Quant 'X), X-ray diffraction (XRD) and inductively couple-plasma mass spectrometry (ICP-MS). Environmental features, such as heavy metal element concentration, distribution and occurrence mode, etc., were studied by systematic determination of element concentration, inorganic element and mineral composition in coal. The results showed that the coal from No.15 coal seam had the mercury (Hg) concentration between 0.44 and 0.268 µg/g, and average Hg concentration of 0.11 µg/g; arsenic (As) concentration between 0.31 and 0.91 µg/g, 0.61 µg/g on average; selenium (Se) concentration between 0.94 and 3.56 µg/g, 2.45 µg/g on average; chromium (Cr) concentration between 9.54 and 24.58 µg/g, 17.06 µg/g on average; lead (Pb) concentration between 10.23 and 23.56 µg/g, 18.13 µg/g on average. The comparison with the Clark value showed Se was significantly enriched, As and Cr were lost, and Hg and Pb were normal. Compared with the coal from other counties and regions both at home and abroad, as was lost and other elements were normal. The elements irregularly distributed vertically. In spite of different element concentrations in adjacent coal seams, no significant enrichment occurred. The target elements mainly occurred in the form of inorganic matters except As. Hg was mainly concentrated in pyrite and clay minerals. Se, Cr and Pb were mainly concentrated in kaolinite, and Se was mainly concentrated in illite. Because of different occurrences or different forms in one mineral, the Hg, As, Se, Cr, and Pb loss rates were 99.43%, 85.91%, 77.89%, 73.81%, and 68.85% at 815°C, respectively.

Keywords: Coal ash; Heavy metal elements; Morphological distribution; Environmental features; Loss rate

Introduction

During coal combustion, heavy metal elements in coal, such as Hg, Se, As, Cr, Pb, etc., migrate into air or concentrate in coal ash, leading to serious harm to human health [1-6]. PECH and Swaine et al. [7,8], expressly listed the heavy metal elements Hg, Se, As, Cr and Pb in coal as key monitored elements. Although coal contains low harmful heavy metal element concentration, usually below 0.5 µg/g, it is the main artificial source of harmful heavy metal elements in air because of large coal consumption [9], and these heavy metal elements in coal ash bring countless harm to environment and society. Thus, it is greatly important to study the environmental features of coal, such as heavy metal element concentration, occurrence, etc. and microelements migration and enrichment during combustion [9-13]. Qinshui basin, located in southeast of Shanxi (north latitude: 35°~38°, east longitude: 111°00′~113°50′), is a key coal production place in Northern China, and has rich coal reserves. Although no pollution event about release of harmful heavy metal elements from coal is available in Shanxi, it is found during research that some sensitive elements can be enriched in some coal seams [14], resulting in potential environmental hazards. Herein, No.15 coal seam in the Coalfield in Southeast Shanxi was investigated in this paper. Five elements, Hg, As, Se, Cr and Pb, with big environmental impact were selected to analyze their concentration and morphologic distribution characteristics in the coal ash, study their migration laws and environmental features, in consideration of coal seam petralogy and geochemical composition analysis, obtain their loss rate at 850°C, and further analyze their migration and enrichment behaviors in coal ash during coal combustion.

Materials and Methods

Sample collection and preparation

According to national standard GB/T 482-2008, 20 samples were collected from the coal seam with thickness between 16 and 22 cm by the continuous channel sampling method. 1# and 2# samples were from the roof, and 19# and 20# samples were from the floor. 3#~18# samples were from the coal seam, with the mass of about 1.5 kg. All the samples were aired, split, ground to 200 mesh (75 µm), and then stored in sealed brown wide mouth bottles in a cool place against sunlight for later use.

Methods

According to GB/T 212-2008, Proximate analysis of coal, the coal samples were industrially tested. The test results are listed in Table 1. It could be known from Table 1 that 9#, 10# and 17# samples contained gangue in coal seam, and had the high temperature ash (HTA) output ratio of 44.53%, 42.67% and 46.21%, respectively. The coal samples were completely calcined at 815°C according to GB/T 30725-2014 in order to determine macroelements in HTA by XRF. The sulphur concentration was determined by a coulomb sulphur analyzer and converted to SO_3 concentration.

Raw coal had low inorganic mineral concentration, which was directly determined by XRD. The minerals with the concentration below 3.0% were difficultly detected. The coal samples were calcined in pure oxygen atmosphere in a tubular furnace for a long time at 370°C, at which organic components could be effectively removed from raw coal without

***Corresponding author:** Dong-Min Wang, School of Chemical and Environmental Engineering, China University of Mining and Technology, Beijing 100083 PR China, Tel: +8615901245174; E-mail: wangdongmin-2008@163.com

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NO.	Proximate analysis				Major-element oxides concentration									
	M_{ad}	A_d	V_{daf}	FC_{ad}	Al_2O_3	SiO_2	Fe_2O_3	K_2O	CaO	Na_2O	MgO	P_2O_5	TiO_2	SO_3
1	1.21	93.24	16.24	9.26	34.64	56.80	0.81	2.99	0.80	1.37	1.77	0.05	0.88	1.16
2	1.13	72.16	13.21	4.21	34.62	55.16	4.64	2.27	0.59	1.05	1.45	0.27	1.27	2.36
3	0.95	16.4	11.24	0.95	33.11	54.68	0.94	2.11	3.64	1.03	1.42	0.14	0.97	1.39
4	0.96	8.42	11.21	0.61	34.57	53.93	1.27	2.14	3.66	1.05	1.44	0.11	3.33	1.45
5	0.97	8.44	10.31	0.84	34.77	55.24	1.48	2.15	1.03	1.05	1.45	0.20	1.00	1.57
6	0.89	8.46	11.25	0.74	34.50	55.14	1.23	2.65	7.24	1.05	1.45	0.01	1.40	1.32
7	0.92	8.24	11.17	0.91	34.06	53.78	1.40	2.22	4.90	1.14	1.63	0.17	1.78	1.43
8	0.96	8.31	11.26	0.82	34.38	47.82	3.76	2.13	0.40	1.04	1.43	1.18	1.41	1.88
9	0.88	44.53	15.3	3.07	32.88	50.89	1.10	2.46	1.78	1.06	1.46	0.04	1.49	1.26
10	0.94	42.67	14.21	2.15	35.11	41.55	1.14	2.17	0.41	1.06	1.46	0.06	1.25	1.48
11	0.92	8.16	11.23	0.76	34.85	55.44	1.17	2.16	0.86	1.06	1.45	0.07	1.44	1.50
12	0.97	7.52	11.15	0.72	33.92	55.86	1.28	2.10	5.03	1.03	1.41	0.12	1.82	1.36
13	0.94	7.41	11.14	0.35	33.82	55.30	2.10	2.09	2.72	1.02	1.41	0.04	1.41	1.60
14	0.93	6.72	11.18	0.86	32.29	54.98	4.90	2.00	7.72	0.98	1.35	0.81	0.19	1.48
15	0.98	7.53	11.35	0.42	31.76	50.43	1.67	1.96	9.10	0.96	1.32	0.29	1.12	1.38
16	0.96	8.51	11.24	0.38	30.15	55.67	1.18	1.86	16.88	0.91	1.26	0.08	1.35	1.50
17	0.95	46.21	14.52	4.28	37.86	55.47	1.06	0.88	1.20	1.07	1.47	0.03	1.07	1.13
18	0.93	8.42	11.26	0.91	25.78	54.28	2.63	0.79	22.87	0.78	1.08	1.56	1.29	1.67
19	1.05	67.18	12.76	6.24	34.67	56.49	3.61	2.14	0.11	1.05	1.45	0.69	1.61	1.52
20	1.21	87.61	18.42	7.18	34.99	55.53	1.16	2.16	0.97	1.06	1.46	0.07	1.07	1.39

Table 1: Test Results Unit: wt%.

impact on most inorganic minerals [15]. Corundum international standard was added to the low temperature ash (LTA). After that, the samples were scanned by D8 Advance X-ray diffractometer made by Germany Bruker to obtain the XRD spectra, and the minerals in LTA were qualitatively identified by Highscore software based on PDF-2007 database. Figure 1 shows the XRD spectra and main minerals of the samples from the floor. Various minerals were accurately determined by XRD spectra fitting using RockJack system, with the degree of fitting of 0.1583%. Generally, the degree of fitting below 0.2% showed that the fitting result was relatively reliable [16]. The mineral concentrations were converted to raw coal-based concentrations by LTA output ratio. The results are summarized in Table 2.

The raw coal samples were digested by ERHOS1 microwave digestion system made by Italy Milestone. The digestion system was $HNO_3/HF+H_3BO_3$. 0.1000 g of raw coal sample was weighed to a PTFE tank, following by addition of 7 ml of 70% HNO_3 and 1.5 ml of 40% HF. The obtained sample was heated to 80°C, 140°C and 210°C, i.e., three-stage temperature rise, and primary digested at 210°C for 10 min before cooled to room temperature. 10 ml of 5% H_3BO_3 was added. The sampled was secondarily digested in a microwave digestion system at 150°C before 8 ml of 40% HNO_3. 2% HNO_3 was added till volume. The clear solution was diluted and determined by ICP-MS. 0.0500 g of HTA sample was added to a PTFE tank. 2 ml of HF and 10 ml of HNO_3 were added before the sample was kept still for 2 h. The sample was stirred until it was completely dissolved. After the revomal of acid, the solution was diluted and determined by ICP-MS. According to the blank and standard sample test data, the quantitative analysis curves for element standards were plotted. According to NIST-1632b and GBW07406, quantitative calculation was performed for the sample. The measured target element concentrations are presented in Table 3.

Hg element was tested by DMA-80 direct mercury analyzer in such a principle that the solid sample was automatically dried and thermally decomposed in oxygen atmosphere; the decomposition product was catalytically reduced to Hg atoms in a catalysis tube; Hg vapor was completely amalgamated in an amalgamation tube; decomposed at high temperature; the carrier gas entrains Hg vapor to an absorption

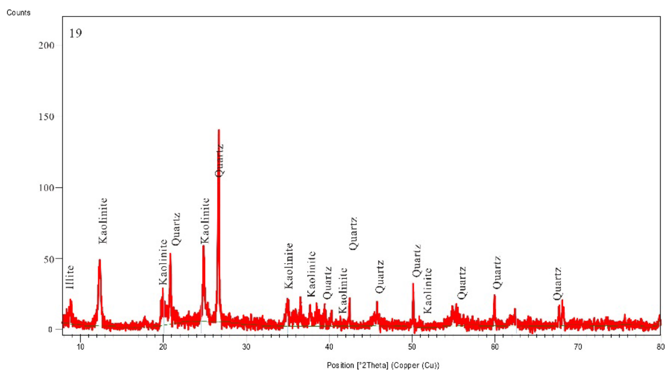

Figure 1: XRD spectra and main minerals of the LTA from the floor.

cell with single wave long path atomic absorption spectrophotometer, where absorption peak height and area of Hg were determined at 253.7 nm. To avoid sample boat pollution and impact of Hg residue in the spectrophotometer during determination, a blank test (without sample) was performed before the sample was tested. When the absorbency was below 0.003 and stable, the actual sample could be tested. 0.0500 g of raw coal sample and HTA sample were put in a quartz boat, which was baked at 650°C in a muffle for 30 min before use in order to eliminate background effect, respectively. The measured Hg concentrations are listed in Table 3.

Results and Discussion

Heavy metal element concentration

The coal from 15# coal seam in the Coalfield in Southeast Shanxi had Hg element concentration between 0.044 and 0.268 μg/g, 0.11 μg/g on average; As element concentration between 0.31 and 0.91 μg/g, 0.61 μg/g on average; Se element concentration between 0.94 and 3.56 μg/g, 2.45 μg/g on average; Cr element concentration between 9.54 and 24.58 μg/g, 17.06 μg/g on average; Pb element concentration between 10.23 and 23.56 μg/g, 18.13 μg/g on average. The average heavy metal element

Sample No.	LTA output ratio	Kaolinite	Illite	Montmorillonite	Quartz	Calcite	Dolomite	Pyrite	White Mica	Siderite	Anatase
1	94.56	48	21	6.1	18.7	0.2					0.4
2	88.24	34	26	4.7	22.1	1.1					0.3
3	31.26	17	7	1.7	4.1			0.6		0.2	0.7
4	26.21	10	9	1.9	3.4	0.7	0.4				0.6
5	26.75	11	11	1.2	1.2	1.2			0.7		0.2
6	26.53	13	6	0.4	6.3			0.4			0.4
7	25.64	9	12	2.1	1.8	0.7					
8	25.84	12	4	2.4	4.6	1.6	0.8		0.1		0.3
9	58.46	32	14	3.4	6.8	1.2		0.6		0.4	0.1
10	56.48	28	13	1.5	8.4	2.5		1.6		0.3	1.2
11	31.56	20	7	1.5	2.1	0.2	0.1				0.7
12	24.68	10	6	0.5	7.4	0.1			0.2		0.5
13	21.42	8	10	1.7	1.5	0.1					0.1
14	16.22	10	2	1.2	2.5	0.3					0.2
15	10.51	5	2	0.8	1.2	1.3	0.1				0.1
16	19.54	13	1	1.8	3.4	0.1					0.2
17	53.27	27	16	1.6	7.5	0.3		0.1		0.2	0.6
18	24.16	12	4	0.8	6.2	0.5	0.3				0.4
19	75.26	26	33	1.6	13.4	1					0.3
20	85.29	42	30	1.2	10.5	0.8	0.2		0.1		0.5

Table 2: Mineral Analysis. Unit: %.

	Raw coal					HTA				
No.	Hg	As	Se	Cr	Pb	Hg	As	Se	Cr	Pb
1	0.044	0.41	0.94	23.60	17.29	0.003	0.28	0.46	15.2	15.26
2	0.065	0.52	1.16	20.52	18.41	-	0.25	1.29	13.12	19.66
3	0.177	0.31	2.83	16.23	22.54	0.004	0.21	2.62	22.52	24.64
4	0.071	0.46	2.74	18.24	19.48	-	0.22	2.56	23.21	23.02
5	0.089	0.72	2.76	17.51	16.52	-	0.3	2.59	22.25	18.75
6	0.104	0.88	2.18	13.16	14.36	0.007	0.35	2.5	21.33	19.68
7	0.102	0.64	2.57	13.35	12.02	0.002	0.27	2.51	21.1	21.96
8	0.085	0.63	2.94	12.46	18.94	-	0.23	2.59	19.26	22.66
9	0.174	0.36	3.31	24.58	26.15	-	0.22	2.67	23.24	28.95
10	0.268	0.55	3.56	22.34	23.56	-	0.22	2.63	22.41	26.88
11	0.131	0.89	2.94	18.55	19.20	0.004	0.33	2.55	20.2	24.16
12	0.092	0.76	2.62	17.52	17.61	0.001	0.29	2.54	19.13	23.85
13	0.101	0.72	2.27	9.86	16.55	-	0.25	2.54	18.29	22.56
14	0.082	0.78	2.00	11.77	10.23	-	0.28	2.53	19.81	19.84
15	0.085	0.70	2.13	9.54	10.56	0.001	0.28	2.55	19.11	20.26
16	0.093	0.42	2.41	16.79	19.42	-	0.23	2.52	21.21	23.25
17	0.126	0.91	2.94	20.11	21.84	0.006	0.36	2.6	26.52	25.27
18	0.098	0.50	2.52	17.01	18.10	0.002	0.21	2.58	24.87	24.24
19	0.099	0.56	2.01	19.28	20.49	-	0.22	1.31	14.02	26.34
20	0.074	0.44	2.24	18.73	19.26	0.001	0.22	1.29	12.56	15.88

Table 3: Heavy metal element concentrations in raw coal and ash (μg/g).

concentrations and Clark value in coal seam in Zhang Village Mine, the heavy metal element concentrations of the coals from China, USA and other countries and regions are comparatively listed in Table 4 in order to study the heavy metal elements enrichment in 15# coal seam. In comparison of the concentrations and Clark values of five heavy metal elements in coal seam in Zhang Village Mine, Hg, As, Se, Cr, and Pb element enrichment coefficients were 1.38, 0.34, 49.00, 0.17, and 1.45 μg/g, respectively. The analysis showed that for earth crust, As and Cr were lost; Hg and Pb concentrations are normal; Se was enriched. In comparison of target element concentrations in the coals from China, USA and other countries and regions, Hg, Se, Cr, and Pb concentrations were normal, and as was lost in 15# coal seam in the Coalfield in Southeast Shanxi.

Vertical distribution characteristics of sensitive heavy metal elements

The component sources were very distinct in different phases during peat accumulation. The vertical distribution of heavy metal element concentrations was varied in the coal seam. For example, the undeformed coals from different layers of Yanzhou Coalfield displayed different element concentrations, such as Cu, Zn, Pb, etc. [17] When the coal bearing formation underwent tectogenetic movement and

Countries/Regions	Hg	As	Se	Cr	Pb
Southeast Shanxi	0.11	0.61	2.45	17.06	18.13
Clark value	0.08	1.8	0.05	100	12.5
Global	0.10	5	3	20	25
China	0.22	4.7	2	16	14
USA	0.17	24.0	2.47	15	11.0
Australia	0.10	1.5	12	12	10~15
Ukraine	0.02	80	1.8	15	10
India	-	-	-	35	10
Northern China	0.17	3	6	16	20
Huainan-Huaibei mining area	-	5.53	5.5	34.47	15.81
Enrichment coefficient	1.38	0.34	49.00	0.17	1.45

Table 4: Average heavy metal element concentration of coal from Southeast Shanxi and other countries/regions (μg/g).

deterioration and its associated kinetic heat effect, the heavy metal elements in it would migrate. Li Yunbo found during his study on Coalfield in Northern Anhui Province that Pb, Cr, etc. glomerated with deformation of deformed coals [14]. This study focused on different layers in one coal seam, with consistent undeformed macrographic ingredient structure, but many layers of baulk in the coal seam, and complicated heavy metal element distribution in the coal seam.

Figure 2 shows the vertical distribution of five harmful metal elements concentrations in 15# coal seam in the Coalfield in Southeast Shanxi. Different layers in one coal seam had significantly different heavy metal element concentrations. Hg element was significantly lost in the roof and floor, but significantly enriched in the baulk layer, and Hg concentration in each coal seam was not very different; As element was slightly lost in the roof and floor, its concentration was high in the ingredient layer in addition to the baulk layer. For example, as concentration reached 0.88 μg/g in 6# sample; Se element concentration was significantly lost in the roof, not very different in other layers, and high, 3.59 μg/g, in 10# sample from baulk layer; Cr element was enriched in the coal seam roof, and its concentration was high in the baulk layer. For example, Cr element concentration was 24.58 and 2234 μg/g in 9# and 10# samples, respectively; Pb element concentration was high in the baulk layer, but it was not different in other layers. In spite of significant difference in some element concentrations in the adjacent layers, such as Hg concentration of 0.268 and 0.131 μg/g in 10# and 11# samples, and as concentration of 0.42 and 0.91 μg/g in 16# and 17# samples, they were not significantly enriched. There was no law of Hg, As, Se, Cr and Pb distribution in each layer in 15# coal seam.

Occurrence analysis

It is very difficult to study the occurrence modes of heavy metal elements because of very low heavy metal element concentrations in coal and different heavy metal element occurrences in the coal from different origins or in different coal forming periods. The methods for research on the occurrence modes of heavy metal elements mainly include sequential chemical extraction process, electron microscope and energy spectrum analysis, etc., and correlation between micro element and inorganic element or minerals. Different regions had different micro element occurrence. For example, Finkelman found during sink float experiment, combustion experiment and mineralogy that Hg mainly occurred in inorganic minerals, and Se was related to organic substances, and believed that Se could occurred in pyrite in place of sulphur and Pb mainly occurred in sulfide or sulfide-related minerals, and usually in galenite (PbS) [18,19]. Dvornikov found during his study on the coal from Donbass in Ukraine that most Hg occurred in the form of soluble solid in pyrite [20]. Zhang et al. proposed during their

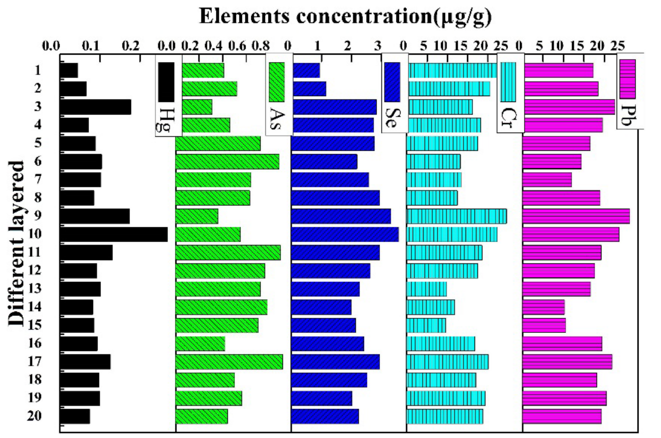

Figure 2: Distribution of heavy metal elements in different layers.

study on Hg distribution characteristics in main associated minerals in the coal seam in Southwest Guizhou that clay mineral contained Hg, and calcite from hydrothermal origin contained Hg, etc. [21]; Wang found during his sinkfloat experiment that Cr related to organic matter and clay [22]; Zhao found during his study on Shanxi Coalfield that Pb mainly entered the organic matters in the coal and crystal lattices in clay minerals when the coal contained no Pb sulfide [23]; Dai et al. found that Se and Pb in the coal from Ha'er Wusu open-cast mainly occurred in clausthalite; Se, Pb and Hg related to Pb mineral Zhungeer Coalfield, and As and Hg in dias coal mainly occurred in getchellite [24-26]. Thus, micro element occurrence was very complicated.

As an indirect statistical method to study the element occurrence in coal, multivariate statistical analysis has been extensively used in research in recent years [3,27-29]. According to the correlation coefficients among elements in coal, ash and macroelements, the element occurrence in coal was identified by this method in consideration of macroelement occurrence in coal.

Figure 3 shows the relationship between five heavy metal element concentrations in coal seams and ash output ratio. As shown in Figure 3, except As, the other element concentrations are positively correlated with ash output ratio. It can be concluded that Hg, Cr, Se, and Pb in coal seams mainly occurred in the form of inorganic matters.

Table 5 listed the correlation coefficients between inorganic macroelements and between inorganic macroelement and target element in coal. The inorganic elements Al, Si, K, Na, and Mg had the correlation coefficient between 0.72 and 0.98, indicative of high correlativity, and mainly occurred in the form of aluminosilicate (clay mineral) in coal; the correlation coefficient between S and Fe reached 0.71, suggesting that they mainly occurred in the form of pyrite, which was found from the mineral analysis results of 3, 6, 9, and 17# samples (Table 2).

The XRD analysis of LTA showed that (Table 2), 15# coal seam in Southeast Shanxi had major minerals including clay mineral (kaolinite, illite and montmorillonite) and quartz, and minor minerals including calcite, dolomite, pyrite, white mica, siderite and anatase. The clay had the highest mineral concentration and relative concentration above 70% generally, and it mainly included kaolinite and illite. To more systematically understand five harmful micro elements and relationship between different mineral, the correlation between Hg, Cr, Se, Pb in the form of inorganic matters and main inorganic minerals were analyzed.

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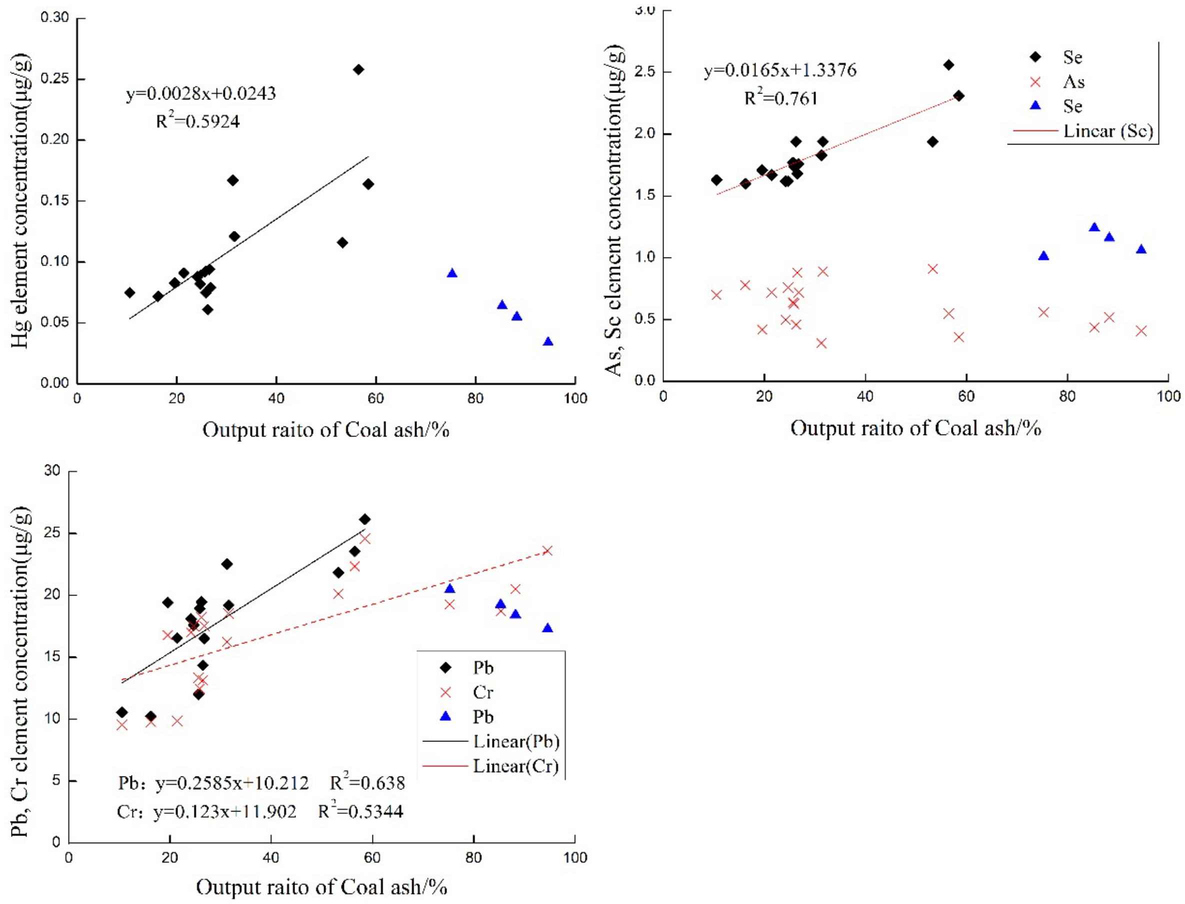

Figure 3: Correlation between heavy metal element concentration and output ratio of coal ash.

It could be seen from Table 5 that Hg was not only highly correlative with S and Fe, but also significantly correlative with main elements in clay mineral, Al, Si, K, and Na, so it can be concluded that Hg closely related to pyrite, and very closely related to clay mineral; although the correlation coefficient between Hg concentration and P concentration reached 0.90, but no phosphonium-containing mineral, such as apatite, cryptolite, xenotime, etc., was founded. Figure 4 shows that Hg was very positively correlative with pyrite, and Hg concentration closely related to kaolinite unless otherwise in coal seam roof and floor (peat), with the correlation coefficient as high as 0.77. Figure 5 shows energy dispersive spectra under a scanning electron microscope. Hg element was founded in pyrite and clay mineral, indicating that Hg closely related to pyrite and clay mineral from the angle of energy dispersive spectra.

It could be known from Table 5 that Cr, Se and Pb were not significantly correlative with macroelements, for example, the correlation coefficients between them and Al, Si, K, and Na were 0.41~0.59, indicating a weak correlation. Figure 4 shows Pb and Cr elements were significantly correlative with kaolinite, with their correlation coefficients of 0.49 and 0.77, respectively; and not significantly correlative with the other minerals; Se element closely related to kaolinite unless otherwise in the roof and floor (peat), and was correlative with illite, with the correlation coefficients of 0.84 and 0.65, respectively. Se, Cr and Pb mainly occurred in kaolinite, and Se also occurred in illite. The correlation coefficient between Hg and Se was 0.72, and Se was weakly correlative with Al, Si, K, and Na, so it was concluded that Hg and Se coexisted possibly in clay mineral. It was found from the energy dispersive spectra under a scanning electron microscope that Hg and Se exited in clay mineral (Figure 5), indicating that Se occurred in clay mineral from the angle of energy dispersive spectra.

Heavy metal element loss rate

Five heavy metal elements were volatile, so they easily entered air during coal combustion and led to air pollution and human health hazards [11,12]. To further understand their potential hazards, HTA (850°C) was tested on its element concentration. Five elements concentrations in HTA are listed in Table 3. The loss rates of five elements were calculated (Figure 6). It could be known from Figure 6 that Hg loss rate was the highest, 99.43% on average, and higher in each layer; As, Se, Cr and Pb loss rates were 85.91%, 77.89%, 73.81% and 68.85%, respectively; As and Se loss rates were very large, above 90%, in 4~8# and 11~16# samples (Figure 7).

Except from As, the target elements closely related to clay mineral. Hg main occurred in pyrite. Different heavy metal elements occurred very differently, and had significantly different loss rate at the same temperature. During coal combustion, minerals in coal were converted under heating. For example, kaolinite always converted to metakaolin, akerite and mullite at about 600°C, 900°C and 1200°C, respectively; in this process, water was lost, mother crystal structure was destroyed and chemical bonds were broken, resulting in release of the microelements in the adsorption state or in crystal interfaces [15]; pyrite was converted to hematite or magnetite at about 500°C, whereas calcite, siderite decomposed into calcium oxide and iron oxide at 800°C [30], so

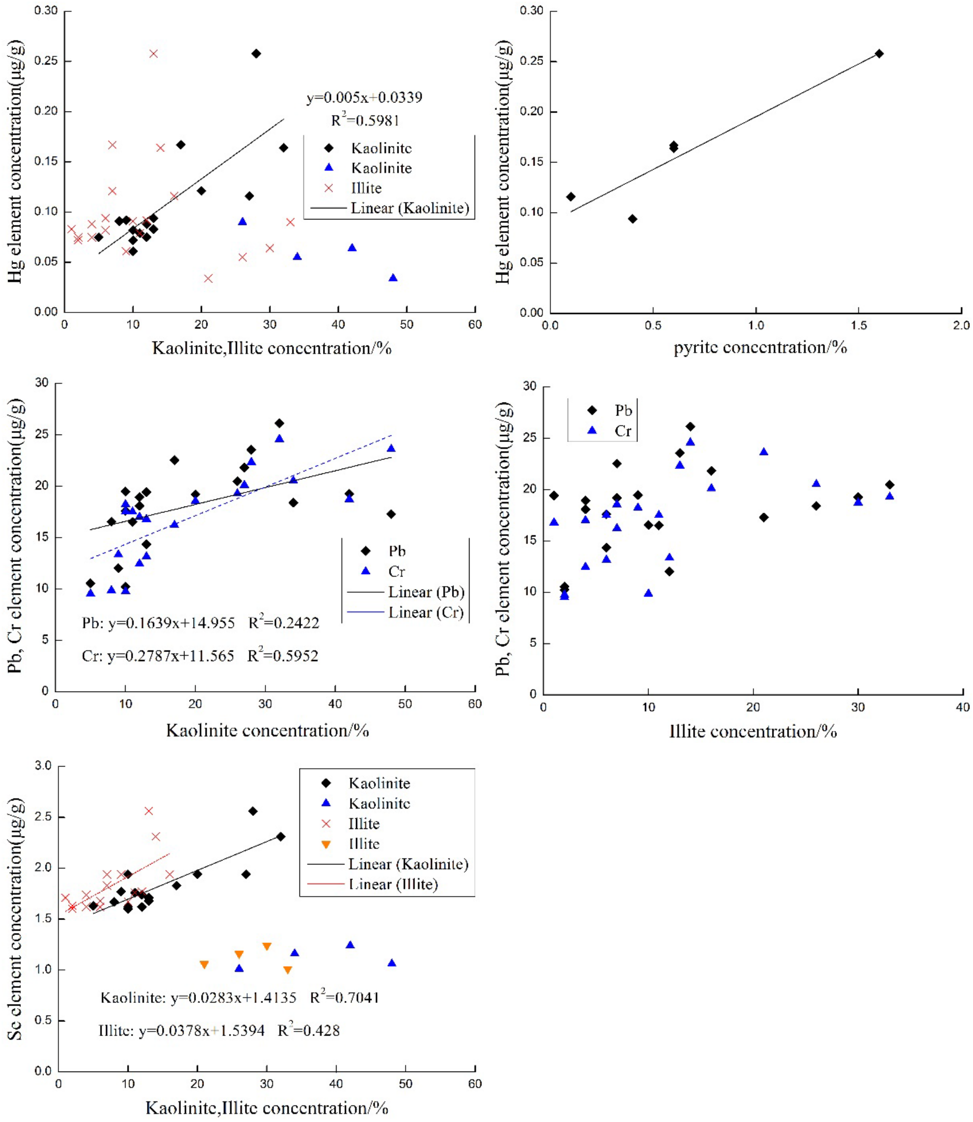

Figure 4: Correlation between mineral concentration and heavy metal element concentration.

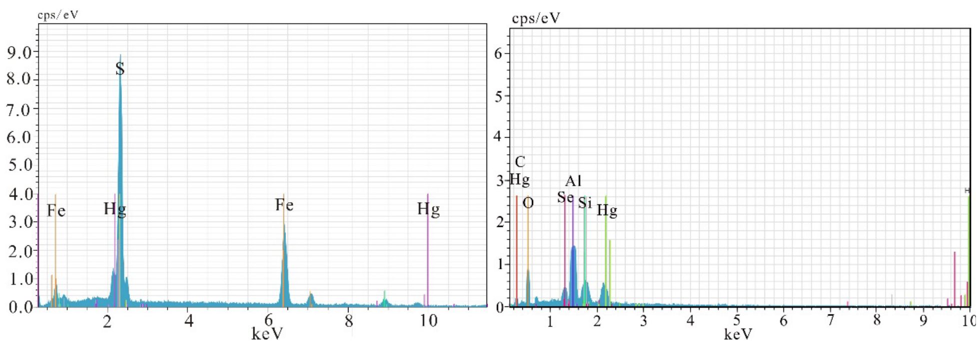

Figure 5: Elements in minerals from coal by SEM-EDX.

quick breakage of chemical bonds significantly accelerated release of isomorphous micro elements in mineral crystal [15]. Chemical bonds in pyrite were broken at low temperature (500°C), and Hg existing in crystal interfaces and stagnant zone and in the isomorphous form was completely released, so Hg had higher loss rate; chemical bonds of clay mineral were broken at higher temperature [15]; clay mineral always

	S	Fe	Si	Al	Na	Ca	Mg	K	P	Ti	Hg	Cr	As	Se	Pb
S	1														
Fe	0.71	1													
Si	-0.52	-0.62	1												
Al	-0.35	-0.58	0.94	1											
Na	-0.61	-0.65	0.73	0.72	1										
Ca	0.41	0.58	-0.76	-0.74	-0.74	1									
Mg	-0.58	-0.67	0.78	0.78	0.98	-0.77	1								
K	-0.74	-0.68	0.83	0.75	0.83	-0.76	0.83	1							
P	0.57	0.77	-0.63	-0.34	-0.52	0.55	-0.55	-0.67	1						
Ti	0.10	-0.02	0.08	-0.09	-0.03	-0.14	0.14	-0.07	-0.12	1					
Hg	0.65	0.77	0.63	0.63	0.62	0.58	0.60	0.66	0.90	0.25	1				
Cr	0.38	0.27	0.44	0.44	0.43	0.23	0.41	0.30	0.42	-0.27	0.29	1			
As	-0.36	-0.24	0.26	0.22	0.15	-0.22	0.12	0.20	-0.38	0.18	-0.09	-0.43	1		
Se	0.51	0.23	0.57	0.59	0.56	0.22	0.54	0.57	0.53	0.26	0.72	0.08	-0.08	1	
Pb	0.45	0.25	0.44	0.46	0.42	0.27	0.43	0.46	0.52	-0.30	0.56	0.76	-0.46	0.47	1

Table 5: Correlation coefficients between inorganic macroelements and between inorganic macroelements and heavy metal elements in coal.

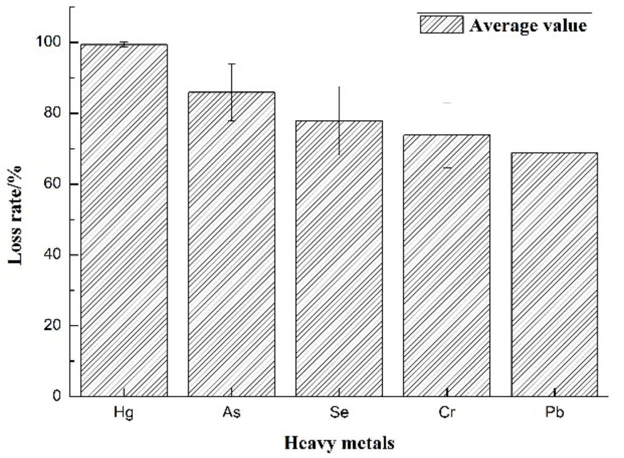

Figure 6: Average loss rate of each heavy metal element. .

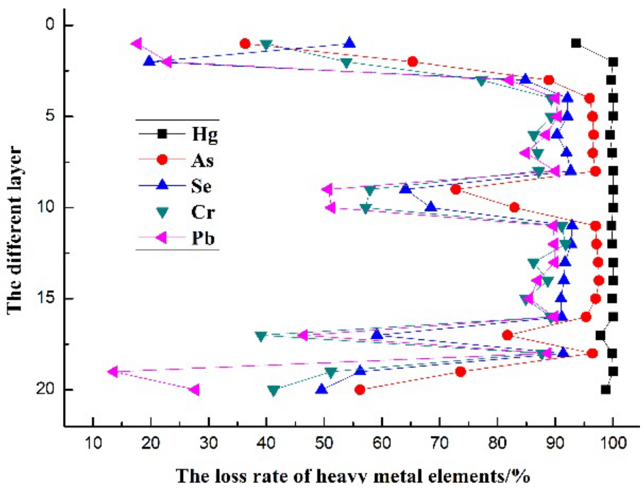

Figure 7: Heavy metal element loss rate in each layer.

was converted to metakaolin and akerite at about 800°C [15], and its chemical bonds were partially broken to mainly release the micro elements in the adsorption state or in crystal interfaces and minorly release isomorphous micro elements, so its loss rate was relatively low.

Conclusions

(1) 15# coal seam in Coalfield in Southeast Shanxi had Hg element concentration between 0.044~0.268 μg/g, 0.11 μg/g on average; As element concentration between 0.31~0.91 μg/g, 0.61 μg/g on average; Se element concentration between 0.94~3.56 μg/g, 2.45 μg/g on average; Cr element concentration between 9.54~24.58 μg/g, 17.06 μg/g on average; Pb element concentration between 10.23~23.56 μg/g, 18.13 μg/g on average. Compared with Clark value, Se was enriched very much, As and Cr were lost, and Hg and Pb concentrations were normal in the coal from 15# coal seam in Coalfield in Southeast Shanxi. Compared with the element concentrations in coals from other regions both at home and abroad, Hg, Se, Cr, and Pb concentrations were normal and as was lost in 15# coal seam in Coalfield in Southeast Shanxi.

(2) Five heavy metal elements concentrations were significantly different in the coal seam cross section, significantly high in the baulk layer, and vertically irregular; in spite of significant difference in element concentrations between adjacent layers, no significant enrichment occurred.

(3) Correlation between five heavy metal elements and ash and inorganic elements, mineral concentrations in LTA and SEM-EDX spectra of mineral particles showed that Hg closely related to pyrite and clay mineral; Se, Cr and Pb mainly occurred in clay mineral kaolinite, and Se also occurred in illite.

(4) Five heavy metals occurred in different minerals or existing in the adsorption state, in crystal interfaces and stagnant zone and in the isomorphous form one mineral at different proportions. After treatment at high temperature, the average Hg, As, Se, Cr and Pb loss rates were 99.43%, 5.91%, 77.89%, 73.81% and 68.85%, respectively.

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