



Mineral matter and trace elements in ashes from a high-arsenic lignite fired power plant in Inner Mongolia, China

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ABSTRACT

The distribution, enrichment, and environmental influence of trace elements and mineral matter in fly ash (FA), bottom ash (BA), and flue gas desulfurization gypsum (FGDG) from a high arsenic coal-fired power plant in Inner Mongolia, China were studied by X-ray diffraction spectrometry (XRD), X-ray fluorescence spectrometry (XRF), field scanning electron microscopy equipped with energy dispersive X-ray spectrometry (FSEM-EDX), atomic fluorescence spectroscopy (AFS), and inductively coupled plasma-mass spectrometry (ICP-MS).

The results showed that mineral matter in FA included quartz, mullite, and gypsum, and that in BA included quartz, enstatite, magnetite, and hematite. The arsenic content in the lignites reached 44.4 µg/g, which was much higher than the global average content of lignite. Most of the trace elements in lignite, except for Mo, Sb, and W, had enrichment trends in the fine grade samples. Arsenic is a toxic element enriched in FA compared with the world lignite ash, while there are several elements of environmental concern are depleted elements (Mo, Cd, Ba, and U). The Cs and Hf are also enriched elements in FA, where Hf showed a significant enrichment. The contents of most trace elements in the smaller particles of FA were higher than those in the larger particles, except for Y and Re. The arsenic content in BA was evidently lower than that in FA, which was 24.4 µg/g, and the trace elements enriched in the smaller particles of BA included Li, Zn, Ga, Rb, Mo, Cd, In, Sb, Cs, Nd, Tl, Pb, Bi, and Th. With the exceptions of As, Sr, Cd, and Sb, the amount of trace elements in flue gas desulfurization gypsum was much lower than that in average Chinese soil. Because the fly ash was stacked for a long time, arsenic and several other trace elements leached into soil surrounding the fly ash dump. As a results the elemental content of old fly ash is less than those collected in this study, which indicates the soil in vicinity need to be monitored continuously for their possible environmental effect.

1. Introduction

Coal-fired power plants are major power generators in China, which consume large amounts of coal annually (Yao et al., 2015). Fly ash is the greatest by-product of coal-fired power plants and the amount of which was increasing annual year in China (Gong et al., 2016; Saikia et al., 2015; Yao et al., 2014; Zhang et al., 2010; Zhao et al., 2006). Several trace elements in fly ash are considered as environmentally sensitive elements; if not properly disposed of, fly ash can cause potential hazards to the environment (Choi et al., 2002; Swaine and Goodarzi, 1995; Goodarzi, 2004, 2006a and d; Goodarzi and Huggins, 2001, 2004, 2005a, 2005b; Nyale et al., 2014; Patra et al., 2012; Sushil and Batra, 2006; Yang et al., 2014; Yao et al., 2015; Zhao et al., 2017;

Zhao et al., 2012). Therefore, it is necessary to study the migration and transformation of trace elements in coal ash.

The distribution of trace elements in combustion products largely depends on element volatilization, the modes of occurrence of trace elements in coal, the operation parameters of boilers, and pollution control equipment (Swaine and Goodarzi, 1995; Gentzis and Goodarzi, 2000; Goodarzi 2006a and 2009; Goodarzi and Huggins, 2005a, 2005b; Guedes et al., 2008; Huggins and Goodarzi, 2009; Mardon et al., 2008; Mokhtar et al., 2014; Vassilev and Vassileva, 2007; Xu et al., 2004). Quan et al. (2013) studied the distribution of trace elements in coal and coal-fired products from two power plants in Huainan, China and found that most of the trace elements were present in fly ash, and Fe and Mn enriched bottom ash. Guo et al. (2004) determined the partition arsenic

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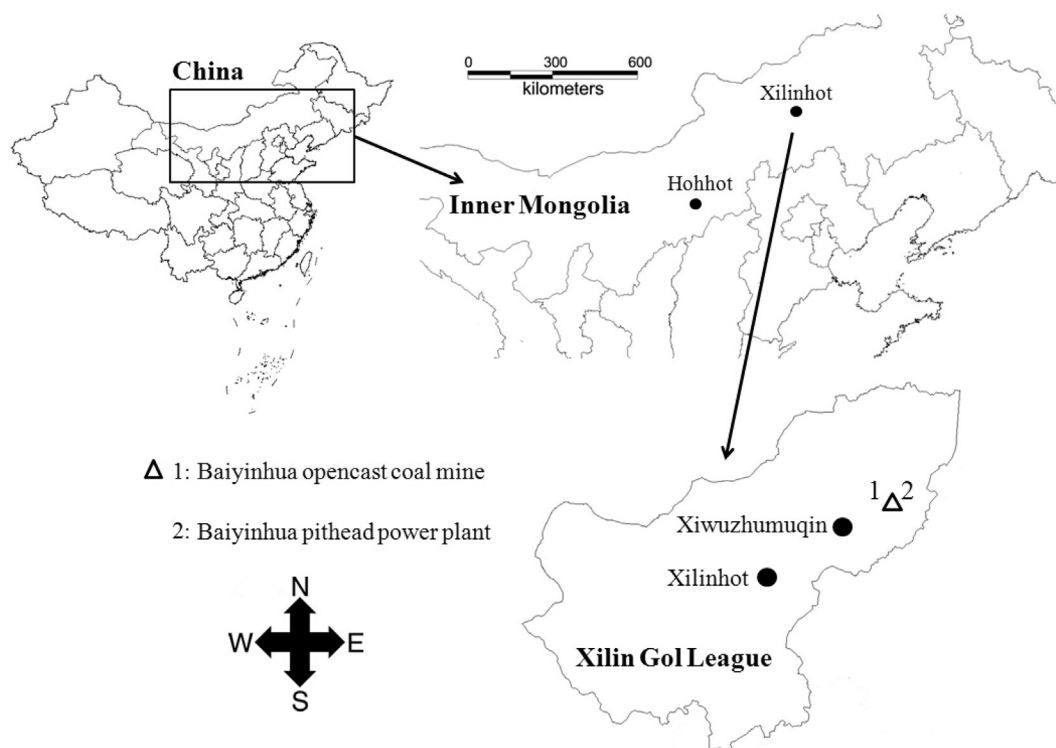


Fig. 1. Sampling locations.

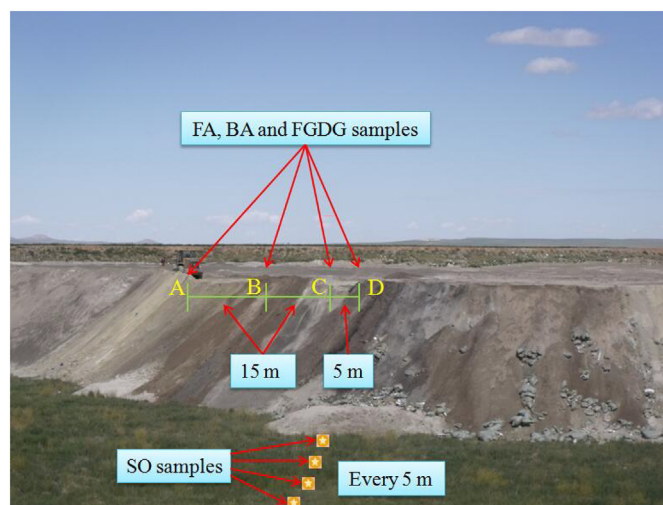


Fig. 2. Sampling sites of the ash dump. FA, fly ash; BA, bottom ash; FGDG, flue gas desulfurization gypsum; SO, soil.

Table 1

Proximate and ultimate analyses of lignites (wt%).

Samples	Proximate analysis				Ultimate analysis (daf)				
	Mad	Vd	Ad	FCd	N	C	H	S	O ^a
RC-1	2.15	46.76	24.71	26.38	1.13	50.24	3.85	0.60	44.18
RC-2	2.70	49.12	18.61	29.58	1.17	54.36	4.37	0.38	39.72
RC-3	11.46	43.48	23.44	21.62	0.89	45.04	3.89	0.67	49.52
RC-4	13.50	47.70	8.37	30.43	1.23	55.85	4.78	0.57	37.57

RC: lignite; M: moisture; ad: as received; V: volatile matter; d: dry base; A: ash; FC: fixed carbon; daf: dry ash-free basis.

^a By difference.

in bottom ash, gas, and fly ash in a coal-fired power plant in China and found that the As distribution ratios were 0.53%, 2.16%, and 84.6%, respectively. The results of Goodarzi and Huggins (2005a) and Goodarzi (2006a) indicated that the concentration As increased progressively from feed coal to bottom ash and then fly ash. Goodarzi (2006c) showed that concentration of As in fly ash from Canadian power plants increased from the large particle (PM > 10) towards PM 10 and 2.5, at rate 0.18/0.37/1.27 (g/h).

Fly ash is considered to be a potential source of release for many environmentally sensitive elements (Goodarzi, 2006a, 2006b, 2006c; Goodarzi and Sanei, 2009). Reservoir or landfill with fly ash may be in contact with aqueous environments, which may cause a considerable amount of trace elements to leach out. Several elements can potentially harm the environment, even at very low concentrations, such as V, Cr, Ni, Cd, and Pb (Swaine and Goodarzi, 1995; Goodarzi, 2006a; Goodarzi and Huggins, 2001, 2005a, 2005b; Patra et al., 2012). Nyale et al. (2014) studied the leaching behaviors and geochemical distributions of several trace elements in a coal ash dump and found that As, Zn, Pb, Ni, Mo, Cr, and Cu were easily leached from the coal ash dump, which would posed a threat to surface water, groundwater, soil, and food production. Neupane and Donahoe (2013) analyzed the leaching characteristics of As, Co, Cr, Ni, Sb, Se, Ti, V, and Zn through leaching experiments, and the results showed that an unlined landfill with fly ash can potentially pollute the environment. Arsenic is a toxic element in coal, it can be released into the atmosphere during coal combustion and enriched in coal-fired by-product, which would be leached into the soil and water, and do harm to the environment and people's health eventually (Goodarzi, 2006a; Goodarzi and Huggins, 2005a; Goodarzi and Huggins, 2001; Goodarzi et al., 2008; Huggins and Goodarzi, 2009; Linak et al., 2011; Zhang et al., 2002; Zhao et al., 2008).

Inner Mongolia is one of the main areas of coal production in China, and the local coal-fired power plants discharge a large amount of fly ash every year. In this work, high-arsenic lignites were collected from the Baiyinhua (BYH) opencast coal mine, and high-arsenic fly ash (FA), bottom ash (BA), and flue gas desulfurization gypsum (FGDG) samples were collected from the Baiyinhua pithead power plant coal ash dump

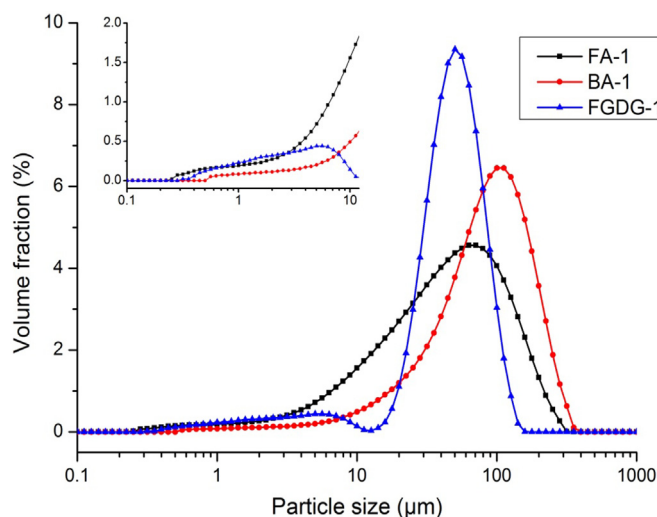


Fig. 3. Particle sizes of samples.

in Inner Mongolia, China. Moreover, soil samples (SO) around the coal ash dump were collected to investigate the environmental influence of piled-up coal ashes. The particle sizes, minerals, major elements, and trace elements in the collected samples were determined in order to analyze the enrichment characteristics and environmental influence of trace elements.

2. Samples and methods

2.1. Sampling locations

The Baiyinhua opencast coal mine is located in the area of Xiwuzhumuqin, and it is one of the ten largest coal mines in Inner Mongolia, China. Its coal reserves reach approximate 14 billion tonnes (Shang et al., 2007). The BYH opencast coal mine includes four mines (mines #1, #2, #3, and #4), and a pithead power plant has been established which combusts lignite from mine #4. The lignite combustion products (FA, BA, and FGDG) generated by the power plant are mainly transported to a special coal ash dump for centralized stacking, where many coal ashes have been stacked for more than five years. To study the distribution and environmental influence of trace elements, lignites, fly ash, bottom ash, flue gas desulfurization gypsum, and the soil samples near the ash dump were systematically collected. The locations of the BYH opencast coal mine and the BYH pithead power plant are shown in Fig. 1.

2.2. Sampling sites from the ash dump

In total, seventeen samples were systematically collected, including four lignite samples, which were collected from the four coal mines at the BYH opencast coal mine. Four FA samples, two BA samples, and two FGDG samples were collected at different sites in the ash dump. Moreover, four soil samples (SO) were collected at the positions by different distances to the edge of ash heap. The sampling sites at the ash dump are shown in Fig. 2.

FA-1 was fresh fly ash (site A), and FA-2, FA-3, and FA-4 were collected at positions that were a certain distance from FA-1: 15 m (site B), 30 m (site C), and 35 m (site D), respectively, which were stacked for some time. BA-1 was fresh bottom ash, and BA-2 was collected at a position that was 30 m from BA-1 (site C), which has stored for some time. FGDG-1 was fresh flue gas desulfurization gypsum, and FGDG-2 was collected at a position 15 m from FGDG-1 (site B). SO-1 was collected at the edge of the ash dump, and SO-2, SO-3, and SO-4 were each collected at positions that were 5 m from the previous site. This ash

dump has been used for about five years, and the coal-fired by-products were stacked from back to front and bottom to top of the ash dump. It can be estimated the approximate stacking time based on the sampling position. Site A is the new emission samples, therefore according to the sampling sites, the site B is the samples stacked for about half a year, the site C is the samples stacked for about one year, and the site D is the samples that had been stacked for more than one year.

All FA samples were sieved into four particle size ranges ($< 150 \mu\text{m}$, $< 75 \mu\text{m}$, $< 38 \mu\text{m}$, and $< 15 \mu\text{m}$). And the lignite samples, BA samples, FGDG samples, and SO samples were sieved into three particle size ranges ($< 150 \mu\text{m}$, $< 75 \mu\text{m}$, and $< 38 \mu\text{m}$). The ferrospheres were separated from FA-1, FA-3, and FA-4 and are defined as ferrospheres-1 (FE-1), ferrospheres-3 (FE-3), and ferrospheres-4 (FE-4), respectively.

2.3. Analytical techniques

The TGA2000 industrial analyzer was used for the approximate analysis of lignites, and an EL-2 elemental analyzer was used for the ultimate analysis of lignites. Major oxides were determined by an X-ray fluorescence spectrometry (XRF) analyzer. The mineralogy of the samples were characterized by X-ray diffraction (XRD), which was used by the X'Pert PRO diffractometer; the accelerating voltage and current were set to 40 KV and 40 mA, respectively, the scanning step was 0.026° , and the interval was 2θ between 10° – 90° . Moreover, a quantitative analysis of minerals was performed on the samples using a reference intensity method (Chung, 1974a, 1974b, 1975; Zhao et al., 2010). Environmental scanning electron microscopy (ESEM) equipped with energy dispersive X-ray spectrometer (EDX) was applied to investigate the mineralogical and morphological characteristics of the samples, and an ESEM-EDX analyzer was employed using the Quanta 200. The laser particle size analyzer used was the Master Min, which is produced by the Malvern company in Britain. The inductively coupled plasma mass spectrometry (ICP-MS) was used for determination of trace elements after digestion. Likewise, a double-channel hydride generation atomic fluorescence photometer (AFS) was used to determine the contents of As after the samples were digested. The digestion method followed the Chinese national standards (GB/T 14506.30-2010), which have been described in previous work (Gong et al., 2016).

3. Results and discussion

3.1. Proximate and ultimate analyses of lignites

The proximate and ultimate analyses of the BYH lignites from the four coal mines are shown in Table 1. The results indicate that BYH lignites are high-volatile lignites, with medium ash and low sulfur contents (8.37–24.71% ash and 0.38–0.67% S).

3.2. Distribution of particle size

The distribution of trace elements may vary greatly in coal-fired by-products with different particle size, therefore, it is necessary to understand the particle size of the samples (Dai et al., 2010; Goodarzi, 2006c; Hower et al., 1999). The particle sizes of FA, BA, and FGDG are shown in Fig. 3. The particle size of FA is mainly concentrated from 1 to $300 \mu\text{m}$, and the peak particle size occurs at $80 \mu\text{m}$. However, a small number of particles are 0.2 – $1 \mu\text{m}$, and these FA samples should be given more attention, due to that fine-grained fly ash is more likely enriched with various harmful substances, such as trace element and organic pollutant, and the fine particles are more easily inhaled, which can cause greater harm to the people's health (Goodarzi, 2006c). The particle size of BA is mainly distributed from 10 to $400 \mu\text{m}$, and there are clearly fewer particles smaller than $10 \mu\text{m}$ compared to those in FA, especially for particles $< 1 \mu\text{m}$. The particle sizes in the FGDG sample show a bimodal distribution, where the majority fall in the range of

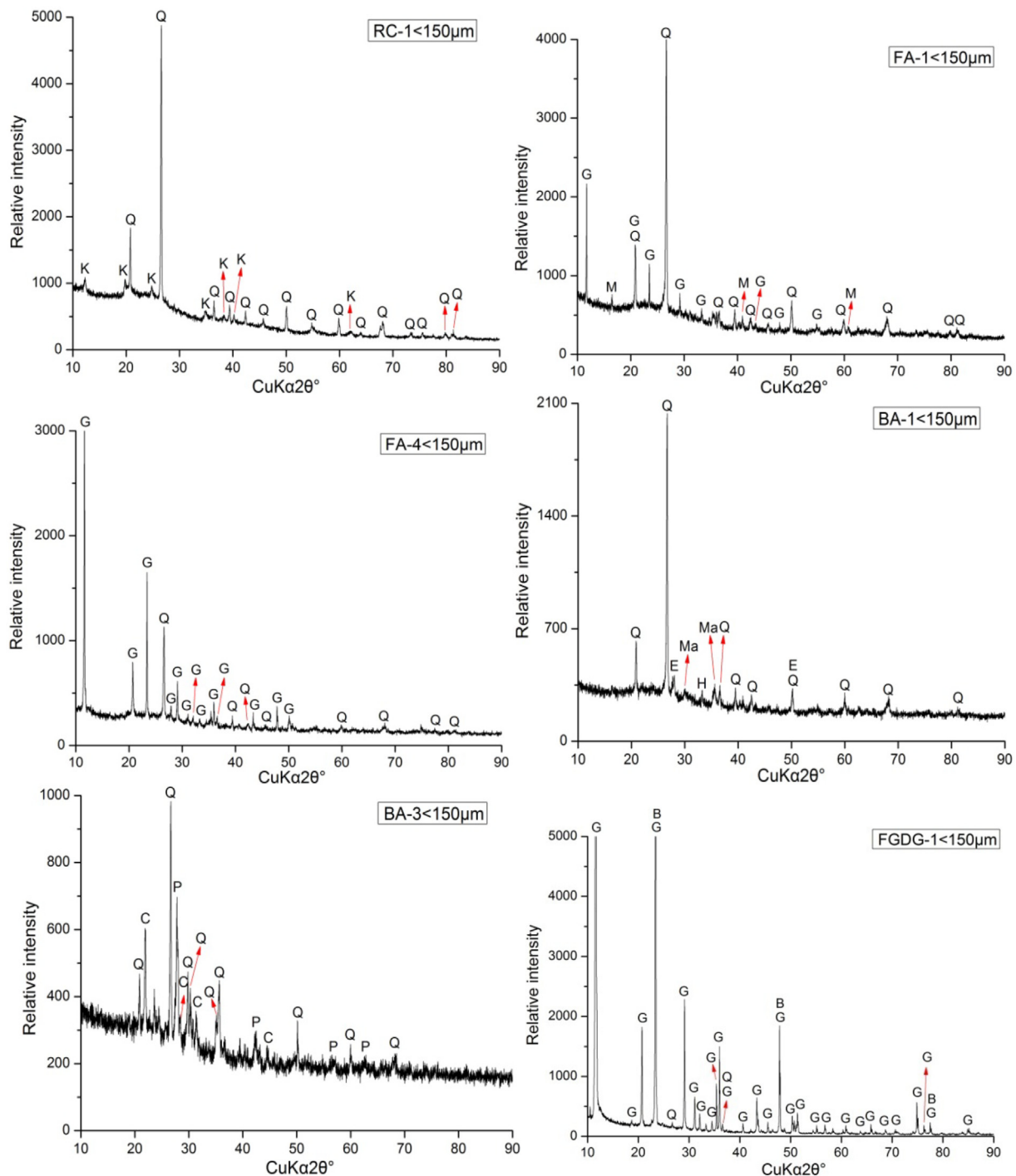


Fig. 4. XRD patterns of the FA, BA, and FGDG samples. Q: quartz; K: kaolinite; M: mullite; G: gypsum; E: enstatite; P: pyroxene; B: brushite; C: cristobalite; Ma: magnetite; H: hematite.

10–150 μm , and the peak also occurs in this range (50 μm). A small portion of FGDG particles are distributed from 0.3–10 μm , with a peak at approximately 6 μm . The number of small particles (< 10 μm) in FGDG are greater than that in BA.

3.3. Mineralogy

Mineral matters in samples of lignit, bottom ash, fly ash, and flue gas desulfurization gypsum were determined using XRD (Fig. 4, Table 2). The main minerals in lignite mainly were quartz and kaolinite. In FA-1, minerals were quartz, mullite, and enstatite, where quartz

accounted for 71%, and the mass fractions of mullite and enstatite comprised 14% and 16% of the total, respectively. FA-4 contained a large amount of gypsum (61%), which probably because gypsum was intermixed in the fly ash after long-term storage. BA-1 which derived from the boiler of the power plant, mainly contained quartz (60%), enstatite (18%), a small amount of magnetite (10%) and hematite (2%). In addition, BA-3 which derived from the small-scale boiler, contained quartz (46%), pyroxene (43%) and cristobalite (11%). This indicated that there were differences in BA minerals from the large-scale and small-scale boilers, and BA in the power plant contained iron-bearing minerals, while the cristobalite was generated in BA from the small-

Table 2
Minerals in the FA, BA, and FGDG samples (%).

Minerals	RC-1	FA-1	FA-4	BA-1	BA-3	FGDG-1
Quartz	85	75	39	60	46	1
Kaolinite	15					
Mullite	–	16	–	–	–	–
Gypsum	–	10	61	–	–	96
Enstatite	–	–	–	18	–	–
Pyroxene	–	–	–	–	43	–
Brushite	–	–	–	–	–	3
Cristobalite	–	–	–	–	11	–
Magnetite	–	–	–	10	–	–
Hematite	–	–	–	2	–	–

–: undetermined.

scale boiler. The FGDG primarily contained gypsum (up to 96%), and small amounts of quartz (1%) and brushite (3%).

3.4. Morphology

3.4.1. Microstructure of the samples

The FA particles (FA-1) were mainly spherical, with sizes generally < 50 μm and primarily < 10 μm , indicating that there was a large number of fine particles in the fly ash (Fig. 5A). FA also contained blocky-shaped particles. There are also some dendritic metallic deposit on the surface of glassy spheres (Fig. 5A) (Goodarzi, 2006b-d). FA-4 is the fly ash that was stacked for the longest time, which also contained several FGDG particles, and the surfaces of the larger particles were

adhered to many fine sphere particles due to long-term storage (Fig. 5B). Most of the BA had irregular porous particles, and there were also several irregular smooth particles. Several spherical Fe-bearing particles were observed, with sizes > 100 μm (Fig. 5C); the particles in the BA (using XRD) were probably magnetite and hematite (Goodarzi, 2006d). The FGDG particles were mainly blocky-shaped particles, with sizes of 100 μm and thicknesses of approximately 10 μm . Many well-crystallized FGDG particles were observed as diamond-shaped particles (Fig. 5D).

3.4.2. Energy spectrum analysis of the FA and BA samples

Several FA particles are analyzed by ESEM-EDX, and the results are shown in Fig. 6. The particle in Fig. 6A is an Fe-bearing aluminosilicate, the elements in which are Fe, Si, and Al, with small amounts of Ca, Mg, K, S, and P. The particle surface is attached to many 2 μm iron particles, which are observed with the backscattered electron image (Fig. 6A). It is speculated that the Fe matrix in the pulverized coal is first melted at a high temperature and condensed on the surface of aluminosilicate particles to form fine iron particles during coal combustion. Aluminosilicates are usually spherical particles, with surfaces that often have several pores (Fig. 6B). Most FA particles are aluminosilicate particles that formed during the fusion-coalescence process in coal combustion (Goodarzi, 2006a-d; Tian et al., 2016; Wang et al., 2008; Yan et al., 2001). The main elements of aluminosilicate particles in FA are Si and Al, with small amounts of Na, Mg, and K. Quartz particles are mainly composed of Si and O, with small amounts of Al, Ca, and S. The surface of quartz is smooth; it is an angular, blocky-shaped particle, with a size of approximately 15 μm (Fig. 6C).

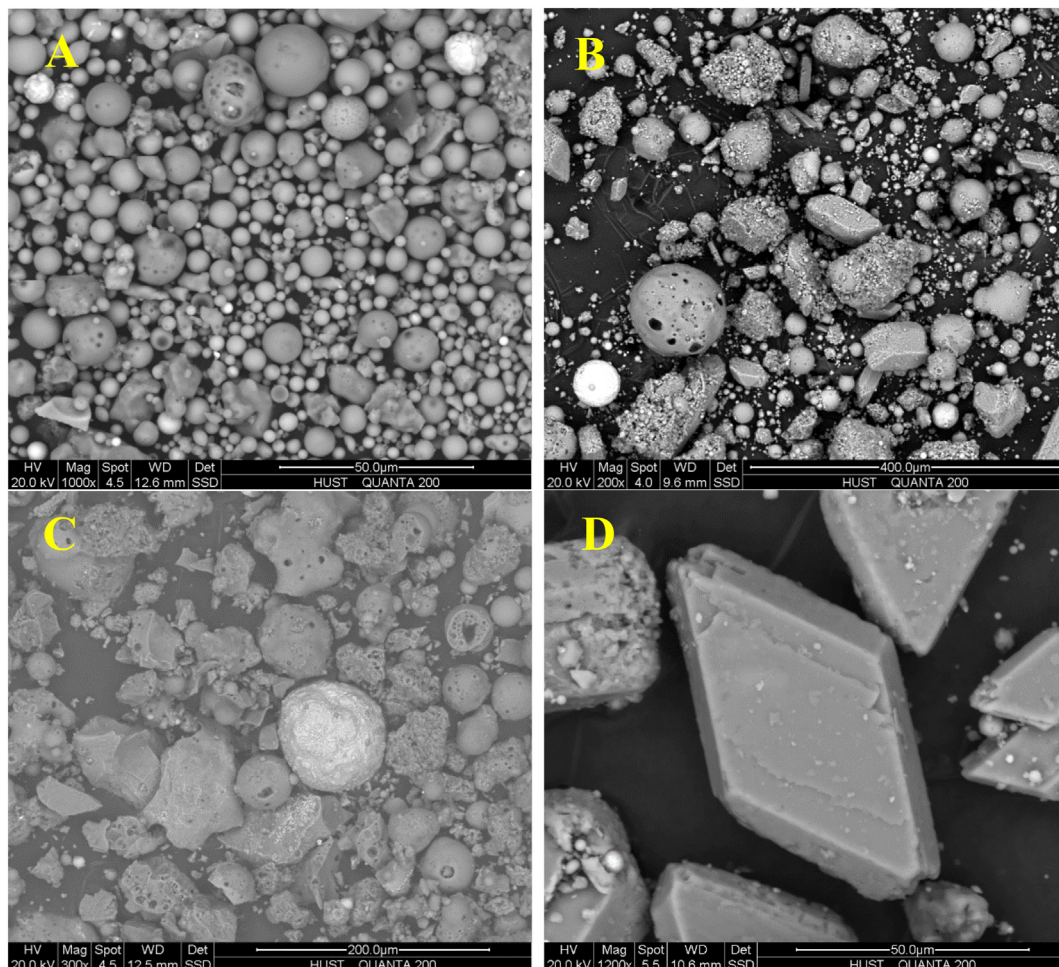


Fig. 5. Micrographs of the samples. A: FA-1; B: FA-4; C: BA-1; D: FGDG-1 (panels A–C are backscattered electron images; panel D is a secondary electron image).

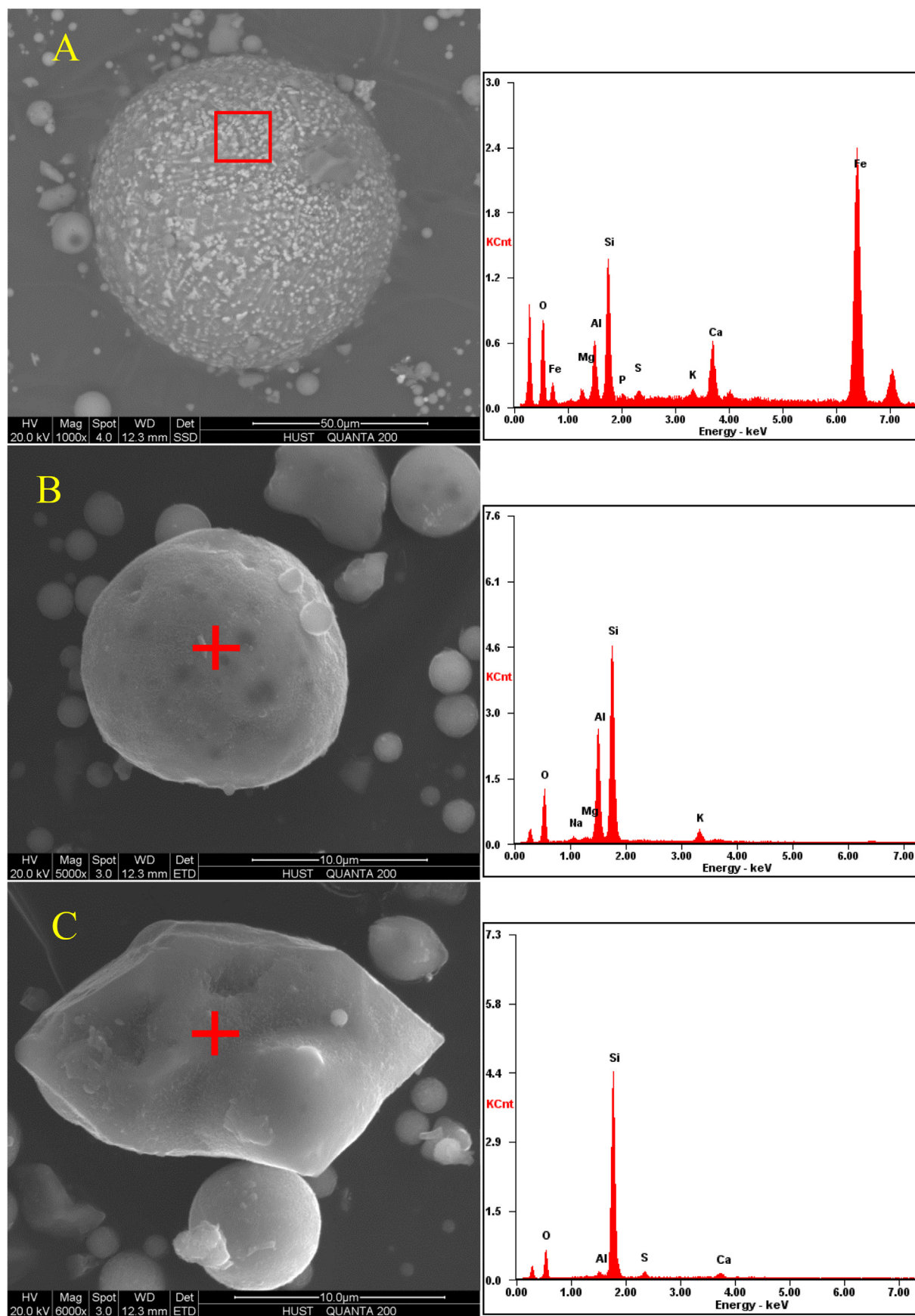


Fig. 6. ESEM-EDX of FA particles. A: Fe-bearing aluminosilicate particle; B: aluminosilicate particle; C: quartz particle (panel A is a backscattered electron image; panels B and C are secondary electron images).

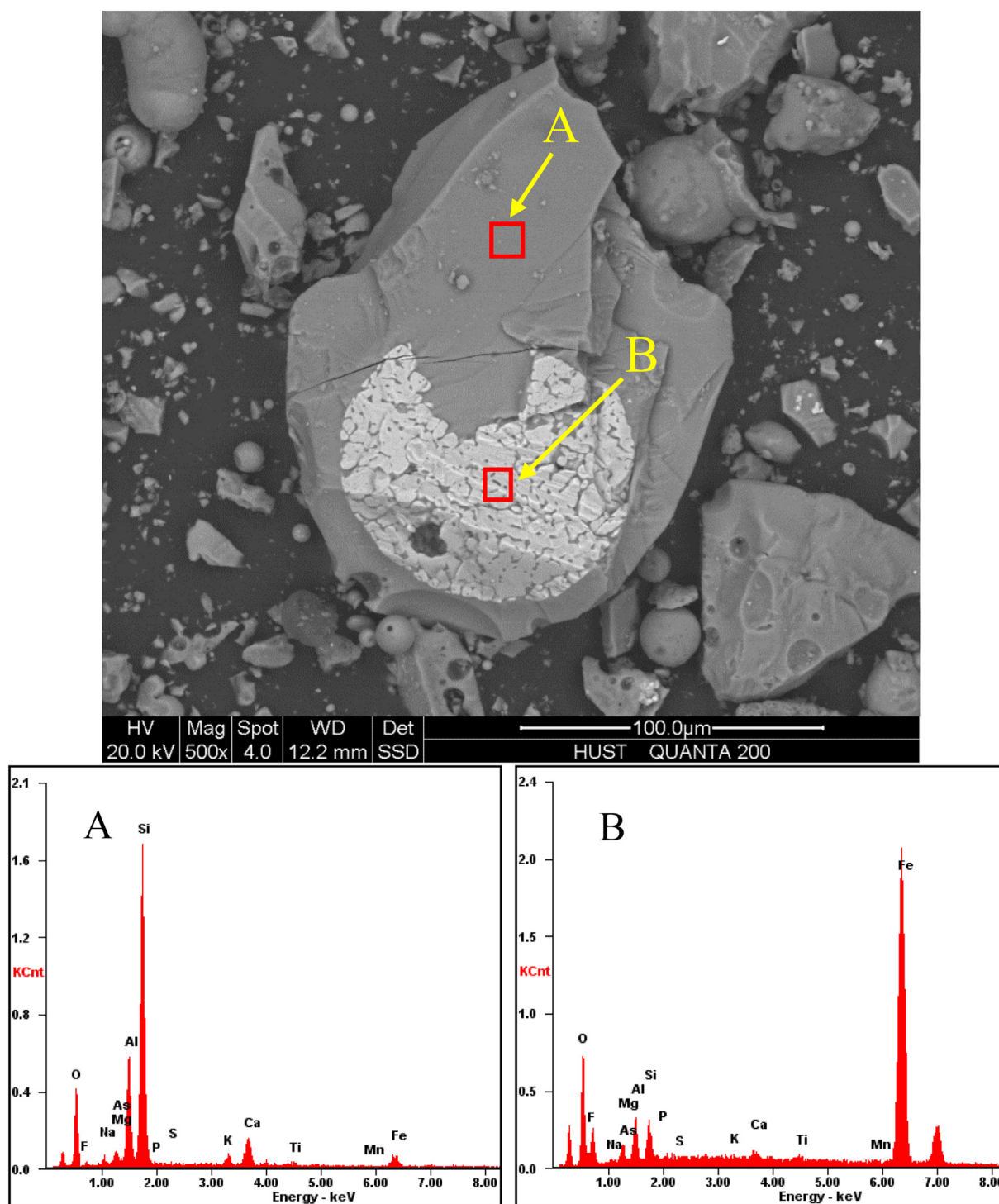


Fig. 7. ESEM-EDX of the BA particle. A: aluminosilicate mineral; B: Fe-bearing mineral (backscattered electron image).

Table 3

Chemical compositions of the ash and soil samples (Wt%).

Samples	Al ₂ O ₃	SiO ₂	SO ₃	K ₂ O	CaO	TiO ₂	Fe ₂ O ₃	P ₂ O ₅
FA-1	19.52	64	2.13	1.38	7.77	1.04	4.03	–
FA-4	12.74	65.33	7.09	1.02	11.39	0.71	1.64	–
BA-1	17.83	67.85	0.47	1.7	5.25	1.02	5.74	–
FGDG-1	2.65	3.81	55.98	–	36.4	–	0.26	1.06
FGDG-2	1.69	3.92	56.85	–	36.28	–	0.22	1.03
SO-1	24.99	65.9	–	2.2	1.47	0.91	4.44	–

–: undetermined.

The BA particle are mostly an aluminosilicate mineral (Fig. 7A) that is surrounded by an iron particle (Fig. 7B). The elements in aluminosilicates are mainly Si, Al, and O, while those in iron particles are mainly Fe and O, with small amounts of Na, Mg, K, Ca, and Ti. These results indicate that the included minerals in lignite melt first at high temperatures and merge during the BA formation, and the formation of aluminosilicate particles is accompanied by the formation of Fe-bearing minerals (Goodarzi, 2006b-d; Guedes et al., 2008; Yang et al., 2014). Most BA particles has many pores, which is due to the gas released from inside of the molten minerals during the rapid cooling process.

Table 4
Concentrations of trace elements in lignites under different particle sizes (µg/g).

Elements	RC-1			RC-2			RC-3			RC-4		
	< 150 µm	< 75 µm	< 38 µm	< 150 µm	< 75 µm	< 38 µm	< 150 µm	< 75 µm	< 38 µm	< 150 µm	< 75 µm	< 38 µm
As	26.9	27.7	31.5	11.5	12.7	12.8	6.22	42.4	42.2	44.4	15.8	15.7
Cd	0.136	0.162	0.173	0.132	0.152	0.269	0.115	0.11	0.16	0.145	0.172	0.169
Cr	18.6	28.8	33.7	26.6	26	44.1	39.5	33.9	43.6	25	31.6	31.1
Cu	62	68.9	71.3	65.9	20.4	94.2	10.8	61.1	69.6	78.5	81.5	73.9
Mo	1.59	1.66	1.48	2.89	2.61	2.77	2.76	2.57	2.97	1.2	1.21	0.977
Ni	5.38	6.28	6.89	8.17	7.89	9.66	36.3	35.7	45.1	11.7	11.1	12.6
Pb	5.87	6.31	9.32	6.7	7.07	10.8	5.37	6.48	9.59	10.6	12	14.6
V	25.2	30.7	30.3	38.2	39.3	42.3	26.3	23.8	29.9	46.4	47.7	41.3
Zn	30	38.2	35.4	28.1	18	35.7	27.3	39.5	52	27.3	33.7	39.4
Ba	272	331	368	260	232	294	200	203	278	195	221	205
Co	4.55	4.26	4.55	4.25	4.3	4.57	5.37	5.28	6.61	5.05	5.24	4.75
U	0.654	0.846	1.04	0.793	0.859	1.04	0.752	0.738	0.996	0.937	0.913	0.837
Th	2.68	3.68	3.95	3.11	3.38	4.47	3.59	3.3	4.96	3.08	3.41	3.59
Li	20.7	32.2	38	9.44	9.9	14.1	7.36	7.32	11.8	2.91	3.43	3.95
Be	1.11	1.21	1.18	1.06	1.12	1.24	1.35	1.19	1.31	0.846	0.916	0.82
Sc	3.97	4.84	5.26	5.33	5.98	6.51	3.59	3.77	4.49	4.41	4.86	4.28
Ga	4.25	5.6	5.41	4.89	5.09	5.96	5.25	4.41	5.58	4.26	4.62	3.93
Rb	16.1	25	26.6	15.8	16.7	25.6	13.3	14.3	25.9	8.78	9.76	11
Sr	179	205	224	153	144	157	115	112	124	72.5	72.9	62
Y	9.74	12.2	14.1	12.2	12.2	14.8	8.25	7.81	11.2	6.67	7.17	6.67
Zr	42.3	57.3	62.4	59.5	48.4	62.9	52.3	46	54	54.6	55.9	46.5
Nb	2.61	2.71	3.17	2.52	2.86	3.54	3.54	2.77	3.74	4.43	4.15	2.99
In	0.039	0.063	0.061	0.045	0.045	0.064	0.037	0.04	0.046	0.042	0.057	0.047
Sb	2.59	2.55	1.78	3.6	2.98	2.78	1.57	1.39	1.38	2.87	2.86	2.53
Cs	4.21	5.54	6.28	3.56	4.13	5.86	3.55	3.59	5.87	1.39	1.61	1.81
La	14.6	19.8	22.1	13	15.5	20.9	12.6	12.7	23.5	8.06	8.78	9.81
Ce	17.6	22.3	24.7	20.9	20.4	25.7	15.4	16.5	26.3	18.4	19.7	19.3
Pr	2.53	3.55	3.64	2.45	2.5	3.26	2.16	2.22	3.26	1.35	1.72	1.88
Nd	10.7	12.4	13.8	11.7	11.6	14.1	8.54	8.73	12.8	5.88	6.82	6.79
Sm	1.95	2.77	2.9	2.18	2.23	2.96	1.55	1.62	2.54	1.33	1.56	1.62
Eu	0.539	0.628	0.664	0.529	0.664	0.744	0.584	0.372	0.477	0.288	0.284	0.344
Gd	1.79	2.49	2.61	2.18	2.17	2.87	1.55	1.46	2.24	1.29	1.41	1.49
Tb	0.362	0.515	0.525	0.445	0.414	0.552	0.287	0.281	0.449	0.265	0.261	0.284
Dy	1.88	2.08	2.3	2.13	2.12	2.59	1.43	1.35	1.9	1.28	1.27	1.31
Ho	0.381	0.438	0.471	0.38	0.433	0.544	0.287	0.266	0.372	0.259	0.252	0.248
Er	0.91	1.27	1.25	1.18	1.17	1.45	0.887	0.795	1.13	0.809	0.782	0.797
Tm	0.227	0.627	0.297	0.193	0.262	0.306	0.187	0.202	0.844	0.21	0.163	0.185
Yb	1.1	1.34	1.54	1.16	1.38	1.6	0.977	0.849	1.16	0.874	0.969	0.864
Lu	0.158	0.225	0.227	0.184	0.185	0.236	0.158	0.137	0.187	0.122	0.126	0.135
Hf	1.34	1.6	1.88	1.42	1.46	1.86	1.05	0.966	1.39	1.24	1.16	1.05
Ta	0.227	0.28	0.292	0.255	0.256	0.329	0.254	0.226	0.342	0.177	0.193	0.162
W	1.92	1.84	1.59	0.673	0.742	0.793	1.89	1.83	1.71	0.713	0.656	0.571
Re	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl	< dl
Tl	0.173	0.239	0.27	0.145	0.146	0.197	0.321	0.27	0.366	0.075	0.087	0.082
Bi	0.08	0.106	0.107	0.104	0.099	0.128	0.085	0.093	0.152	0.122	0.124	0.131

< dl: under detection limit; RC-1: lignite of mine #1; RC-2: lignite of mine #2; RC-3: lignite of mine #3; RC-4: lignite of mine #4.

3.5. Major elements

The FA samples mainly consisted of the Al_2O_3 , SiO_2 , CaO , and Fe_2O_3 (Table 3). Fly ash can be classified into different groups by the ‘inter-section’ of major oxides (Goodarzi, 2006d). The $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{TiO}_2$, $\text{CaO} + \text{K}_2\text{O}$, and $\text{Fe}_2\text{O}_3 + \text{P}_2\text{O}_5 + \text{SO}_3$ account for 84.56%, 9.15%, and 6.16%, respectively, it can be drawn that the high-arsenic fly ash is sialic. FA-4 is fly ash stacked for a period of times, and its Ca content is greater than that of FA-1, which is because some FGDG was mixed into FA-4. The chemical composition of BA is similar to that of FA, except for elements S and K. The main elements in the FGDG samples are Ca and S, and the contents of Al and Si are very low. There is approximately 1% of P_2O_5 in FGDG; based on the XRD analysis, the phosphorus element in FGDG is in the form of brushite. The SO sample mainly contains Al_2O_3 and SiO_2 , with small amounts of K_2O , CaO , TiO_2 , and Fe_2O_3 .

3.6. Trace elements

Goodarzi (2002) classified the trace elements into three groups according to its environmental importance as follows:

- Elements of environmental concern: As, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Se, V, and Zn.
- Elements of environmental interest: Ba, B, Co, Mn, U, Th, F, Br, and Cl.
- Other elements.

The concentrations of trace element in samples were determined and shown in Tables 4–9. In order to understand the enrichment characteristics of trace elements in samples, the concentration of the trace elements are compared with the reference value to obtain the concentration coefficients (CC) (Ketris and Yudovich, 2009; Chi and Yan, 2007). The CC formula is determined as following:

$$\text{CC} = \text{element content in samples} / \text{reference value}$$

where: the reference value for coal is the average concentrations of global lignite; the reference value for FA is the average content in global lignite ash; the reference value for BA, FGDG, and soil is the average content of Chinese soil.

To study the effect of particle size on concentration variations in trace elements in lignites, the relative enrichment factor (RE) was

Table 5
Concentrations of trace elements in fly ashes under different particle sizes ($\mu\text{g/g}$).

Elements	FA-1				FA-2				FA-3				FA-4			
	< 150 μm	< 75 μm	< 38 μm	< 15 μm	< 150 μm	< 75 μm	< 38 μm	< 15 μm	< 150 μm	< 75 μm	< 38 μm	< 15 μm	< 150 μm	< 75 μm	< 38 μm	< 15 μm
As	116	111	146	143	181	138	161	192	110	122	139	183	88.6	86.8	123	141
Cd	0.544	0.562	0.674	0.777	0.532	0.512	0.624	0.725	0.528	0.536	0.648	0.712	0.386	0.444	0.541	0.666
Cr	66.4	61.5	68.5	99.4	62.4	61.9	64.1	72.4	61.8	62.3	59	166	46	54.3	56.2	132
Cu	66.1	62.8	74.5	77.1	65	63.3	69.9	73.5	59.2	61.9	64.1	78.6	43.3	48.4	56.4	68
Mo	3.72	3.55	3.74	4.5	3.87	3.84	4.03	4.33	4.02	4.23	3.97	4.78	2.08	2.9	2.49	2.92
Ni	47.5	48.3	55.2	64.3	48	47.2	50.9	57.7	42.7	45	46.2	73.2	31.4	41.5	44	70.1
Pb	36.4	36.1	38.9	47.9	34.7	35.5	37.9	42.3	32.4	36.4	36.4	73.2	31.4	28.2	36.7	40.3
V	109	101	112	115	103	101	104	105	94.5	97.7	95.2	99.4	72.4	79	88.8	93.1
Zn	130	130	163	193	146	125	156	178	122	122	154	186	117	104	133	179
Ba	608	577	648	655	592	581	623	640	565	580	590	593	433	472	525	590
Co	24.8	22.7	25.3	29	24.1	22.1	25.9	25	21	22.8	23.7	25.8	16.9	20.1	23.3	26.4
U	5.86	5.62	6.08	5.87	7.14	6.38	6.54	6.64	6.54	6.1	5.43	5.57	3.82	4.41	4.67	4.93
Th	18.6	18.7	17.9	18.2	18.3	17.5	18.7	16.8	17.5	17.4	16.5	16	14.7	15.5	17.3	16.6
Li	49.3	46.4	52.4	53.2	49.3	47.7	51.4	51.4	45.7	47.5	47	48.4	37.5	39.2	45.1	50
Be	3.34	3.16	3.46	3.49	3.2	3.17	3.33	3.25	2.82	3.07	2.8	2.95	2.34	2.42	2.78	3.05
Sc	21	19.1	22.2	22.3	19.4	19.3	20.3	19.6	18.1	18.9	18.7	20.1	13.4	14.7	16.3	18.2
Ga	25	23.8	30	35.5	24.2	24.7	28.6	32.5	23.1	23.9	28.7	32.8	18.3	19.2	25.9	31.4
Rb	69.3	64.2	72.5	75.6	68.4	65.2	70.2	71.6	65.9	66.8	65.6	71	51.8	52.6	63.4	71
Sr	389	371	381	373	376	362	364	352	365	395	373	366	281	299	292	326
Y	31.2	30.4	31.4	31.9	29.8	29.2	30.6	29.2	27.6	29.2	27.9	29.2	31.8	32.4	32.8	28.7
Zr	5.37	5.19	5.63	5.55	5.3	5.21	5.66	5.32	5.16	5.57	5.09	5.47	4.33	4.39	5.12	5.67
Nb	17.6	16.8	19.3	19	16.9	15.7	17.7	16.4	15.8	17.3	16	16.3	12.7	13.5	15.4	16.5
In	0.093	0.088	0.116	0.141	0.089	0.088	0.11	0.131	0.083	0.088	0.107	0.122	0.065	0.077	0.097	0.118
Sb	9.3	8.39	11.3	13.1	8.75	8.54	11.9	12.1	9.34	9.43	11.6	12.1	7.27	7.82	10.5	9.86
Cs	20.2	18.7	21.9	23.1	18.5	18.6	21.4	21.2	19.2	19.2	21	21.1	14.9	15.3	19.3	22
La	37.8	36.5	39.7	37.4	37.2	34.7	36.2	35.7	34.9	36	34	35	27.7	27.3	31.1	34.4
Ce	86.4	83.9	92.9	102	86.7	81.5	88.9	84.5	80.2	85.9	82.2	84.8	63.3	67.5	73.7	81.9
Pr	13.2	12	13.2	13.4	11.7	11.7	12.7	11.8	11.8	12	12.1	11.7	8.63	8.99	10.4	12.4
Nd	47.6	46.9	49.2	51.2	47.2	44.3	47.3	45.2	44.1	46.3	43	43.9	34.4	36.1	41.3	46
Sm	9.04	8.51	8.93	9.3	8.49	8.21	8.32	8.58	7.89	8.43	7.68	8.29	6.26	6.71	7.46	8.37
Eu	1.93	1.73	1.96	1.95	1.79	1.72	1.8	1.79	1.69	1.78	1.71	1.65	1.38	1.45	1.5	1.69
Gd	7.86	7.27	7.81	8.27	7.45	7.46	7.58	7.65	7.03	7.19	7	7.03	5.45	6.09	6.6	7.07
Tb	1.33	1.25	1.37	1.38	1.28	1.26	1.24	1.24	1.17	1.27	1.19	1.19	0.955	1	1.13	1.24
Dy	7.15	6.63	7.33	7.43	6.59	6.65	7.11	7.13	6.71	6.62	6.33	6.56	4.97	5.29	6.06	6.73
Ho	1.4	1.32	1.48	1.47	1.34	1.33	1.36	1.4	1.27	1.31	1.28	1.24	0.964	1.09	1.23	1.31
Er	3.79	3.71	3.89	3.89	3.44	3.5	3.76	3.68	3.45	3.52	3.34	3.5	2.59	2.89	3.1	3.37
Tm	0.61	0.594	0.649	0.642	0.566	0.568	0.61	0.615	0.574	0.578	0.577	0.584	0.429	0.468	0.525	0.541
Yb	4.21	3.94	4.41	4.45	3.97	3.86	4.17	4.27	3.98	4.04	3.89	3.96	2.97	3.3	3.68	3.87
Lu	0.607	0.57	0.623	0.608	0.568	0.57	0.61	0.588	0.559	0.58	0.556	0.564	0.421	0.462	0.527	0.555
Hf	170	164	174	175	163	166	175	167	159	171	154	165	135	140	153	175
Ta	1.29	1.2	1.4	1.34	1.19	1.22	1.26	1.21	1.21	1.21	1.21	1.21	0.911	0.991	1.11	1.24
W	3.46	3.22	3.82	10.5	3.55	3.35	3.51	3.56	3.36	3.47	3.96	3.84	2.82	2.96	3.23	3.67
Re	0.006	0.006	0.006	0.007	0.011	0.007	0.009	0.016	0.013	0.012	0.009	0.012	0.004	0.002	0.005	0.003
Tl	0.984	0.945	1.13	1.45	0.914	0.952	1.22	1.29	0.84	0.914	1	1.18	0.69	0.761	0.992	1.26
Bi	0.416	0.403	0.5	0.638	0.407	0.383	0.51	0.581	0.395	0.418	0.459	0.563	0.289	0.293	0.424	0.514

Table 6
Concentrations of trace elements in ferrospheres ($\mu\text{g/g}$).

Elements	FE-1	FE-3	FE-4
As	84.8	69.1	53.2
Cd	0.264	0.241	0.248
Cr	129	154	144
Cu	98.6	87.6	93.1
Mo	3.38	2.99	3.16
Ni	94.4	88.5	90
Pb	7.28	6.81	6.63
V	66.6	57	54.3
Zn	117	119	160
Ba	339	654	316
Co	33.7	28.3	26.6
U	3.1	2.88	2.43
Th	3.52	2.78	3.69
Li	14.5	14.1	15.7
Be	1.53	1.36	1.31
Sc	8.92	7.31	7.39
Ga	22.4	21.3	20
Rb	7.67	8.74	11
Sr	146	149	125
Y	15.3	15	17.6
Zr	37.4	41.9	37.6
Nb	1.18	1.3	0.996
In	0.072	0.074	0.058
Sb	1.48	1.79	1.94
Cs	1.55	1.57	2.77
La	13.7	13	14.7
Ce	26.3	25.3	29.7
Pr	3.53	3.45	3.6
Nd	14.7	12.3	14
Sm	3.1	2.68	3.14
Eu	0.649	0.568	0.767
Gd	3.18	2.91	3.63
Tb	0.517	0.43	0.581
Dy	2.97	2.25	3.02
Ho	0.538	0.449	0.616
Er	1.59	1.48	1.78
Tm	0.271	0.219	0.264
Yb	1.62	1.22	1.63
Lu	0.233	0.213	0.221
Hf	1.23	1.13	1.17
Ta	0.009	0.004	0.007
W	0.95	1.11	0.509
Re	0.01	0.008	0.03
Tl	0.094	0.071	0.076
Bi	0.213	0.231	0.163

introduced in this work (Meij and te Winkel, 2007). RE was defined as the content of trace elements in samples of smaller particle sizes (< 75 or 38 or $15\mu\text{m}$) comparing to that in samples $< 150\mu\text{m}$. The RE formula is defined as follows:

RE = element content in samples < 75 or 38 or $15\mu\text{m}$ /that in sample $< 150\mu\text{m}$.

3.6.1. Distribution of trace elements in high-arsenic lignites

The concentrations of the trace elements in lignites with different particle sizes ($< 150\mu\text{m}$, $< 75\mu\text{m}$, and $< 38\mu\text{m}$) from the four coal mines (#1 (RC-1), #2 (RC-2), #3 (RC-3), and #4 (RC-4)) are shown in Table 4, and the CC results of RC are shown in Fig. 8. The CC between 2–5 indicates that the element is slightly enriched, while $\text{CC} < 0.5$ indicates it is depleted element (Dai et al., 2014; Dai et al., 2017).

(1) Elements of environmental concern and interest.

Several elements of environmental concern, i.e., As, Cr, Cu, Ni, and V, in the four coals probably are slightly enriched in the lignites ($2 < \text{CC} < 5$), while Cd is depleted elements. Cu is enriched in RC-1, RC-2, and RC-4, and the element Ni shows only a slight enrichment in RC-3. The U is significantly depleted in all four samples ($0.5 < \text{CC}$). The RE of Cr, Pb, and U exceed 1.5; however, Mo in RC-1 decrease with

an increase in particle size. The RE of Cu, and As in RC-3 $< 38\mu\text{m}$ are very high (6.44, 6.78, and 4.5, respectively). Ba, Co, U, and Th are the elements of environmental interest (Goodarzi, 2002), and they are close to the average of global lignite.

(2) Other elements

Li, In, Sb, and Cs are slightly enriched in the raw coal, while Pr, Gd, Tl, and Bi are depleted elements. Except of the elements of environmental concern and interest, It can be found that most of the trace elements in the lignites are more likely to be enriched in small particles in the samples. Most of the trace elements in RC-1 $< 75\mu\text{m}$ and $38\mu\text{m}$ have greater contents than those in RC-1 $< 150\mu\text{m}$. Several elements in RC-4 have an increasing tendency in finer samples, but the contents of other elements in finer samples may be lower than those in FA $< 150\mu\text{m}$, including Be, Sc, Ga, Sr, Zr, Nb, Sb, Er, Tm, Hf, Ta, and W. In addition, elements Nb and Sb concentrations decrease as the particle size increases.

3.6.2. Distribution of trace elements in high-arsenic fly ashes

The concentrations of trace elements in fly ashes at different particle sizes ($< 150\mu\text{m}$, $< 75\mu\text{m}$, $< 38\mu\text{m}$, and $< 15\mu\text{m}$) were determined and are shown in Table 5, and the CC results are shown in Fig. 9. Moreover, the concentrations in smaller FA samples ($< 75\mu\text{m}$, $< 38\mu\text{m}$, and $< 15\mu\text{m}$) were compared with those in fly ash $< 150\mu\text{m}$ to calculate the relative enrichment factor of FA, which can reveal the distributions of trace elements in FA with different particle sizes.

(1) Elements of environmental concern and interest

As is slight enriched element in fly ash ($0.5 < \text{CC} < 2$), Mo, Cd, Ba, and U are depleted elements. The RE of Cr and Zn in new fly ash are approximate to or exceeding 1.5, which have high RE in fly ash, while in older fly ash (FA-4), the As, Cr, Cd, Cu, Co, Ni, Pb, and Zn have the high RE, and it is more than that in new emission fly ash. Dai et al. (2010) studied the trace elements in six high-alumina fly ashes of various sizes, the elements As, Cd, Cu, Pb, Se, V, Zn, Co, and F increased as the particle size decreased. Hower et al. (1999) believed that As and Zn were concentrated in fine particles; Ni and Cr were associated with the spinel minerals.

(2) Other elements

Most trace elements in high-arsenic fly ash have contents similar to the average values for global lignite ash; while Cs show slight enrichment, and Hf shows significant enrichment ($\text{CC} > 5$), with a concentration coefficient > 20 . The RE of FA-1 $< 15\mu\text{m}$ are the greatest among the four particle sizes, and the RE close to or exceeding 1.5 include In, W, Tl, and Bi. The contents of Th, and U in FA-1 samples of different sizes change little or show a fluctuation. The majority of trace elements in FA-2 are enriched in smaller particles; however, there are several elements fluctuations that vary with particle sizes, including Be, Sc, Sr, Y, Zr, Nb, La, Ce, Pr, Nd, Eu, Tb, Er, Lu, Hf, Ta, and Th.

By comparing the element contents in fly ash that is stacked for a longer period of time (FA-3 and FA-4) with those in fly ash stacked for a shorter period of time (FA-1 and FA-2), it can be seen that most of the trace elements in FA-3 and FA-4 have lower contents than those in FA-1 and FA-2. It is demonstrated that a certain amount of trace elements in FA may leach out after FA is piled up in the environment for a long time. In particular, most of the trace elements in FA-4 is the lowest contents, and there are two reasons: (1) the stacking time for FA-4 is the longest among the four FA samples, the oxidation, surface leaching and long term degradation are why the elemental concentration varies after samples stacked in the ash dump the elements (Goodarzi and Huggins, 2001), and (2) small amounts of bottom ash and flue gas desulfurization gypsum are mixed in FA-4.

Table 7
Concentrations of trace elements in bottom ashes under different particle sizes ($\mu\text{g/g}$).

Elements	BA-1			BA-2			BA-3
	< 150 μm	< 75 μm	< 38 μm	< 150 μm	< 75 μm	< 38 μm	< 75 μm
As	24.4	34.8	52.5	19	25.5	32.6	21.3
Cd	0.188	0.211	0.249	0.151	0.209	0.233	0.079
Cr	57	61.2	60.9	59.6	60.8	55.8	67.8
Cu	50.5	54.3	54.5	62.3	58.3	62.9	95.4
Mo	2.42	2.49	2.03	1.97	2.21	1.89	6.74
Ni	41.1	43.2	37.8	43	42.7	41.5	97.8
Pb	14.7	17	20.6	16	18.7	20.5	1.52
V	99.9	99.6	85.8	104	102	96.1	125
Zn	63.2	68.2	70	65.5	72.2	78	61.4
Ba	571	609	542	679	668	637	1595
Co	20.5	22.4	18.6	20.7	20.4	19.1	38
U	6.74	6.62	5.93	3.95	3.95	3.33	5.67
Th	15.8	16.5	16.4	16.1	17	18.1	16.5
Li	43.8	45.8	46.3	41.5	44.4	46.9	32.1
Be	3.12	3.1	2.7	3.03	3.07	2.86	3.48
Sc	19	18.9	16.4	20	21.1	18.2	21.5
Ga	14.3	15.4	15.6	16.4	17.1	18.3	12.1
Rb	59.5	61.6	60.8	66	71.8	77.9	64.3
Sr	407	369	328	447	407	384	789
Y	30.7	30.4	29.8	31.8	31.4	32.9	34
Zr	5.5	5.38	5.14	5.93	6.12	5.93	5.21
Nb	18.3	17.4	15.1	17.5	17.3	16.6	16.6
In	0.043	0.051	0.05	0.047	0.055	0.057	0.045
Sb	3.65	3.85	4.07	3.8	4.67	4.78	4.76
Cs	16.6	16.5	17	17.1	18.2	21.1	15.8
La	35.4	37.1	34.6	35.7	37.1	36.1	41.1
Ce	83	89.4	82	88.4	88.3	90.5	97.9
Pr	12.8	12.3	11.2	12.3	12.5	12.2	14.1
Nd	41.2	46.8	43.1	47	47.9	47.2	53.4
Sm	8.68	8.31	7.92	8.57	9.13	9.03	10.3
Eu	1.76	1.82	1.6	2	1.88	1.8	2.49
Gd	7.25	7.47	6.79	7.35	8.12	7.32	9.02
Tb	1.24	1.23	1.13	1.25	1.3	1.23	1.6
Dy	6.6	6.95	6.12	7.11	7.2	6.93	8.64
Ho	1.33	1.3	1.25	1.41	1.45	1.37	1.69
Er	3.47	3.51	3.2	3.6	3.72	3.55	4.27
Tm	0.572	0.606	0.506	0.588	0.628	0.576	0.711
Yb	4.09	4.08	3.63	4.5	4.41	4.2	4.94
Lu	0.564	0.598	0.487	0.628	0.612	0.587	0.678
Hf	170	173	154	172	186	175	149
Ta	1.27	1.22	1.11	1.22	1.28	1.18	1.01
W	2.76	2.88	2.33	2.57	2.63	2.66	2.55
Re	< dl	0.002	< dl	< 0.002	< dl	< dl	0.005
Tl	0.372	0.437	0.491	0.384	0.439	0.509	0.041
Bi	0.133	0.142	0.187	0.16	0.136	0.17	0.011

< dl: under detection limit.

Ferrospheres are one of the important components in fly ash, but it is still lacking for the geochemical characteristics (Yang et al., 2014). Therefore, the contents of trace elements in the ferrospheres of fly ashes (FE-1, FE-3, and FE-4) were determined and are shown in Table 6. The results showed that trace elements in the ferrospheres of fly ash were significantly lower than those in the original fly ash. The trace elements close to the global average of lignite ash include some elements of environmental concern: As, Cr, Cu, Ni, Zn, Ba, Co, and several other elements: Ga, In, and Cs. Many element contents in FE-1 are slightly greater than those in FE-3 and FE-4, including Be, Sc, V, Co, Ni, Cu, Ga, As, Mo, Cd, Nd, Tm, Lu, Hf, Ta, Tl, Pb, and U, which is probably due to the migration of elements from the ferrospheres after FA was stacked in the ash dump.

3.6.3. Distribution of trace elements in bottom ash

The contents of trace elements in BA-1 and BA-2 at different particle sizes (< 150 μm , < 75 μm , and < 38 μm) were determined, and the results are shown in Table 7. The trace elements in bottom ash may cause pollution in soil if they are not properly disposed; therefore, in

order to understand the potential hazards of bottom ash, the contents of trace elements in bottom ash < 150 μm were compared with the average content in Chinese soil to calculate the CC of bottom ash (Chi and Yan, 2007), as shown in Fig. 10.

(1) Elements of environmental concern and interest

All the elements of environmental concern and interest, except for U, in fresh bottom ash (BA-1) are lower than the fresh fly ash (FA), especially, the As and Cd are much lower in BA compared with FA. Goodarzi (2006a, 2009) studied the combustion residues from the Canadian power plants, and found that As, Cd, Pb, and Zn were not enriched in BA. The concentration of As, Cd, Cu, Mo, and U in BA-1 are twice higher than the average concentration of Chinese soil, while Cr, Ni, Pb, V, Zn, Ba, Co, and Th are close to that of Chinese soil. The amounts of As, Cd, Cu, Pb, and Zn in BA-1 increase as the particle size decreasing, while elements Ni, V, and Co decrease as particle size decreasing.

Table 8

Concentrations of trace elements in flue gas desulfurization gypsum under different particle sizes ($\mu\text{g/g}$).

Elements	FGDG-1			FGDG-2		
	< 150 μm	< 75 μm	< 38 μm	< 150 μm	< 75 μm	< 38 μm
As	13.5	10.5	19.3	36.9	40.7	86.6
Cd	0.276	0.248	0.505	0.321	0.341	0.443
Cr	4.96	4.7	9.92	17.6	18.2	30
Cu	6.5	32.3	41.5	52	53.2	53.2
Mo	0.337	0.324	0.587	1.02	1.03	1.88
Ni	8.95	8.92	10.1	15.1	15.8	19.8
Pb	2.74	2.12	47.6	5.02	5.92	9.08
V	3.63	3.36	9.42	23.9	23.9	41.3
Zn	23.8	21.6	39.1	47	41	58.8
Ba	24.9	26	50.9	128	122	241
Co	1.07	1.01	2.02	5.16	5.15	8.05
U	0.51	0.537	1	1.58	1.6	2.52
Th	0.443	0.346	0.823	2.33	2.58	3.86
Li	3.34	2.87	6.24	5.94	6.42	9.1
Be	0.121	0.134	0.247	0.627	0.702	1.07
Sc	1.18	1.17	1.92	4.24	4.62	7.37
Ga	1.24	0.955	2.39	4.28	4.79	7.97
Rb	4.36	3.53	7.83	7.92	9.37	12
Sr	159	153	146	191	203	212
Y	5.85	5.43	9.62	9.47	9.68	15.8
Zr	4.65	3.76	8.22	21.1	21.2	36.3
Nb	0.252	0.184	0.294	0.98	1.17	1.27
In	0.026	0.023	0.128	0.035	0.044	0.062
Sb	0.595	0.478	0.839	0.967	1.14	1.26
Cs	0.767	0.621	1.2	1.51	1.8	2.14
La	3.57	3.52	7.18	9.03	9.38	15.2
Ce	3.07	2.89	6.3	14.6	15.6	25.9
Pr	0.694	0.724	1.42	2.13	2.26	3.94
Nd	3.01	2.94	5.8	8.61	9.49	14.6
Sm	0.569	0.521	1.13	1.66	1.8	2.86
Eu	0.132	0.139	0.249	0.374	0.406	0.663
Gd	0.629	0.612	1.18	1.61	1.76	2.66
Tb	0.104	0.103	0.18	0.264	0.28	0.421
Dy	0.556	0.52	0.984	1.38	1.48	2.29
Ho	0.115	0.107	0.199	0.268	0.293	0.425
Er	0.315	0.328	0.567	0.775	0.839	1.3
Tm	0.074	0.046	0.079	0.121	0.123	0.181
Yb	0.326	0.281	0.466	0.754	0.745	1.24
Lu	0.041	0.038	0.066	0.101	0.108	0.183
Hf	0.147	0.124	0.256	0.623	0.717	1.07
Ta	0.015	< dl	0.003	0.003	0.003	0.004
W	0.198	0.158	0.258	0.46	0.483	0.712
Re	0.004	0.004	0.007	0.003	0.003	0.006
Tl	0.05	0.039	0.075	0.042	0.048	0.066
Bi	0.04	0.039	0.083	0.103	0.125	0.192

< dl: under detection limit.

(2) Other elements

The results show that most elements in bottom ash are close to those in average Chinese soil, where only a few elements are enriched and depleted. It is worth noting that Hf in BA-1 and BA-2 is significantly enriched, with a CC > 20, and only Zr and Bi are depleted. Most element contents in BA increase as the particle size decreases, however, there are also several elements in BA-1 that decrease as particle size decreasing, including Be, Sc, Sr, Zr, Nb, Pr, Sm, Tb, Ho, and Yb; the elements that fluctuated with particle size include Y, In, Ce, Nd, Eu, Gd, Dy, Er, Tm, Lu, Hf, and W.

It can be seen that most element contents in bottom ash increase as the particle size decreases. Comparing the fresh bottom ash of BA-1 with the long-time stacking bottom ash of BA-2, it can be found that many elements in BA-1 have higher than those in BA-2, indicating that the elements in bottom ash may leach out after stacking in the environment.

3.6.4. Distribution of trace elements in flue gas desulfurization gypsum

The contents of trace elements in FGDG-1 and FGDG-2 are shown in Table 8. Similar to bottom ash, the CC were calculated and are shown in Fig. 11, respectively.

(1) Elements of environmental concern and interest

Only element Cd is a slightly enriched element, and As is close to the average concentration of Chinese soil. FGDG-2 is the flue gas desulfurization gypsum that was stacked for a period of time, elements Mo, Ni, Zn, and U are close to the Chinese average soil and the slightly enriched elements include As, Cd, and Cu. Therefore, the elements of environmental concern and interest in the older FGDG are more than that in new emission FA. It can be concluded that the concentrations of trace elements in FGDG-2 are higher than those in FGDG-1 for two reasons: (1) FGDG adsorbs the elements leached from FA or BA; (2) FGDG is mixed with small amounts of FA or BA.

(2) Other elements

The results showed that most of the trace elements in FGDG were depleted. In the fresh FGDG-1, Sr and Sb are close to the average value in Chinese soil. By comparing the element contents in FGDG for different particle sizes, it is obvious that the majority of relative enrichment factors for FGDG are > 1, and most REs for FGDG < 38 μm are > 1.5, indicating that elements are more likely to be enriched in small particles of FGDG. By comparing the element contents in FGDG for different particle sizes, it is obvious that the majority of relative enrichment factors for FGDG are > 1, and most RE for FGDG < 38 μm are > 1.5, indicating that elements are more likely to be enriched in small particles of FGDG.

3.7. Impact on environment

Many elements emitted into the environment from coal-fired power plant are mainly in the by-product of coal combustion. Goodarzi (2006a) found that the emission rate from the stack of the elements of environmental concern, i.e., As, Cd, Hg, Ni and Pb, in a Canadian power plant were only 0.85%, 1.42%, 56.5%, 0.30% and 0.38%, respectively. Therefore, the environmental impact of coal-fired by-product should be investigated. The concentrations of trace element are shown in Table 9, and the CC results are shown in Fig. 12.

- (1) Elements of environmental concern and interest Cd and Zn are the enriched elements in soil. The As content in SO-1 is higher than that in SO-2 and SO-4, and it can be seen that the As levels are greater in soils that are closer to the ash dump. Cd, Cu, Mo, Ni, Pb, Co, U, and Th in SO-1 < 150 μm are lower than those in SO-1 < 75 μm , and all elements of environmental concern and interest in SO-1 < 38 μm have greater amounts than SO-1 < 75 μm . Similarly, there are higher contents for most trace elements in SO-2, SO-3, and SO-4. Goodarzi (2009) indicated that the toxic element (As, Cr, Ni, and Pb) may only be leached in acid environment (HCl), their environmental impact was relatively low. However, the pH value of environment are changing under some conditions, which can cause the mobility of elements. Our previous work also found that As, Cd, Cr, Cu, Mo, Ni, Pb, V, Zn, Ba, Co, U, and Th can leach out after environmental stacking of a fly ash in Inner Mongolia, China (Gong et al., 2016).
- (2) Other elements

Most of elements in the soils around the ash dump are very close to the Chinese soil averages, and only several elements show slight enrichment, and only several elements show slight enrichment, including

Table 9Concentrations of trace elements in soils under different particle sizes ($\mu\text{g/g}$).

Elements	SO-1			SO-2			SO-3			SO-4		
	< 150 μm	< 75 μm	< 38 μm	< 150 μm	< 75 μm	< 38 μm	< 150 μm	< 75 μm	< 38 μm	< 150 μm	< 75 μm	< 38 μm
As	14.3	14.7	14.3	13.6	12.6	12.5	19	19.6	20.2	13.4	13.5	17.3
Cd	0.244	0.228	0.284	0.253	0.275	0.307	0.218	0.238	0.287	0.251	0.282	0.296
Cr	80.6	55.2	65.3	52	61.8	61.7	62.5	62.4	63.1	62.8	73.2	66.4
Cu	23	24.9	27.4	19.6	24.5	24	22.9	23.7	25.2	21.3	26.9	24.7
Mo	1.07	1.2	1.28	1.03	1.17	1.21	1.16	1.18	1.27	0.987	1.16	1.03
Ni	23	23.5	24.5	20.1	25	20.9	22.4	23.2	24.6	21.1	23.9	20.5
Pb	30.2	31.4	37.4	27.7	32.1	32.7	26.1	28.5	31.1	26	32.1	29.9
V	87.2	75.4	85.4	65.7	77.1	78.7	72	80.5	79.7	70.3	80.1	72.4
Zn	154	110	135	114	126	136	107	113	128	109	133	128
Ba	525	459	526	493	576	600	539	578	608	491	553	566
Co	15.8	17.8	18.4	18.3	19.5	18.8	18.2	17.9	18.9	16.3	20.8	20.4
U	2.83	2.57	3.16	2.52	2.94	2.89	2.42	2.59	3.05	2.47	3.1	2.77
Th	14.8	14.6	18.3	15.4	17.6	15	13.9	15.9	16.5	13.2	16.9	16.1
Li	53.5	52.2	62	48.7	60.6	60.7	47.3	47.7	54.8	50.3	60	61.9
Be	2.52	2.45	2.77	2.11	2.45	2.5	2.25	2.36	2.61	2.17	2.59	2.5
Sc	12.9	13	15.8	13.1	14.4	15	12.2	12.2	13.8	13.2	15.7	14.3
Ga	25.4	25.7	28.7	22.7	31.3	28.3	23.4	24.1	25.3	24.4	27.4	26.8
Rb	88.4	87.7	93.9	79.5	94.6	87	83.7	81.2	85.7	83.7	92	85.3
Sr	153	148	158	155	192	185	165	182	171	174	201	181
Y	25.7	26.2	33.1	25.7	30.2	32.9	24.1	26.5	31.3	24.3	29.5	29.4
Zr	6.25	7.18	8.15	6.22	7.89	8.01	5.97	6.37	7.7	6.46	7.81	7.64
Nb	15.2	16.9	19.7	13.3	16.6	16.7	13	15.7	16.6	13.2	16.7	15.1
In	0.07	0.083	0.089	0.078	0.086	0.087	0.071	0.082	0.087	0.077	0.087	0.085
Sb	2.22	2.3	2.49	2.38	2.33	2.41	2.22	2.19	2.21	2.22	2.34	2.45
Cs	18.4	21.8	25.2	22.1	27.5	25.6	20.9	21.5	24.8	22	24.6	25.2
La	31.6	29.8	35	28.2	32.2	33.6	28.8	29.9	31.2	28.1	31.2	32.6
Ce	86.8	75.8	87.7	66.4	81.8	77.4	66.9	69.8	78.9	69	82.2	76.8
Pr	9.6	10.5	12.2	9.02	11.3	11.1	9.06	9.59	11.2	8.65	11.6	11.5
Nd	37.2	39.3	45.6	36.5	39.9	43	34.8	35.8	41	32.9	40.2	40.4
Sm	6.9	7.02	8.76	6.31	7.68	7.68	6.58	6.62	7.23	6.58	7.83	7.26
Eu	1.2	1.32	1.55	1.19	1.34	1.42	1.28	1.28	1.37	1.14	1.54	1.33
Gd	5.59	6.29	6.77	5.4	5.58	6.53	5.12	5.49	6.02	5.08	6.08	5.9
Tb	0.92	1.07	1.23	0.879	1.03	1.03	0.889	0.981	1.07	0.852	1.01	1.01
Dy	5.05	5.16	5.93	4.66	5.65	5.69	4.89	5.19	5.91	4.64	5.93	5.47
Ho	0.935	1.05	1.17	0.881	1.06	1.09	0.929	0.923	0.997	0.913	1.05	1.08
Er	2.38	2.64	2.98	2.37	2.65	2.66	2.4	2.55	2.78	2.23	2.78	2.66
Tm	0.384	0.428	0.466	0.395	0.451	0.444	0.386	0.395	0.445	0.371	0.431	0.439
Yb	3.69	3.05	3.51	2.62	3.11	3.2	2.64	2.76	3.26	2.82	3.49	3.24
Lu	0.367	0.386	0.479	0.353	0.428	0.432	0.369	0.385	0.424	0.376	0.438	0.42
Hf	231	240	248	184	233	245	195	197	231	197	241	242
Ta	1.05	1.19	1.37	0.971	1.25	1.18	0.966	1.05	1.18	1.03	1.19	1.15
W	4.44	4.97	5.83	4.69	6.44	6.76	7.31	8.03	9.47	5.2	6.54	6.64
Re	< dl	< dl	< dl	0.002	< dl	< dl	< dl	< dl	< dl	< dl	0.002	< dl
Tl	0.59	0.639	0.717	0.596	0.691	0.656	0.613	0.637	0.652	0.59	0.687	0.683
Bi	0.249	0.251	0.312	0.262	0.288	0.338	0.25	0.271	0.304	0.274	0.289	0.296

< dl: under detection limit.

Sb, Cs, and W. The Hf shows significant enrichment, with a $CC > 30$. Most elements in SO < 150 μm are lower than smaller soil samples, including Li, Be, Sc, Ga, Pb, Sr, Y, Zr, Nb, In, Sb, Cs, La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Lu, Hf, Ta, W, Tl, and Bi.

Trace elements enriched or depleted in the soil are almost the same as those enriched in fly ash or bottom ash; the enrichment characteristics of soil samples are similar to those of fly ash and bottom ash, indicating that trace elements in fly ash and bottom ash that are stacked in the ash dump can leach into the soil around the ash dump. The ash dump affects the soil in two ways: the trace elements in the ash leach into the soil, and the ash is mixed into the soil directly by wind. These results show that the contents of most elements in soil increase with the decrease in particle size, indicating that the trace elements are enriched in finer particles. In addition, there are also some elements that are enriched in larger particles for several soil samples, such as a few elements in SO-2 and SO-3 < 38 μm are less than those in SO-2 and SO-3 < 75 μm , and most elements in SO-4 < 38 μm have contents less than those in SO-4 < 75 μm ; this is probably because the closer the soil is to the ash dump, the more susceptible ash is to soil.

4. Conclusions

This work studied the mineral and geochemical characteristics of several high-arsenic lignites and high-arsenic lignite ashes in Inner Mongolia, China. The environmental influence of ash dump was also discussed. The conclusions are shown as follows: (1) the main minerals in fly ashes are quartz, mullite, and enstatite, of which quartz accounts for 71%, while mullite and enstatite are relatively rare (14% and 16%, respectively). The minerals in bottom ash are quartz (60%), enstatite (18%), magnetite (10%), and hematite (2%). The flue gas desulfurization gypsum contains mainly gypsum (96%) and small amounts of quartz (1%) and phosphates (3%); (2) The trace elements that slightly enriched in high-arsenic lignites include Li, V, Cr, Ni, In, Cu, As, Sb and Cs. Most trace element contents in fly ash are close to the global average lignite ash contents (ash basis), where As, Cs, and Hf are enriched elements, and Be, Mo, Cd, Gd, Dy, Ho, Tm, Lu, Tl, Bi, U, and Zr are depleted elements. Trace elements may leach out from the ashes after stacking in the environment; (3) Some elements in the soil around the ash dump show obvious enrichment, including Zn, Cd, Sb, Cs, Hf, and

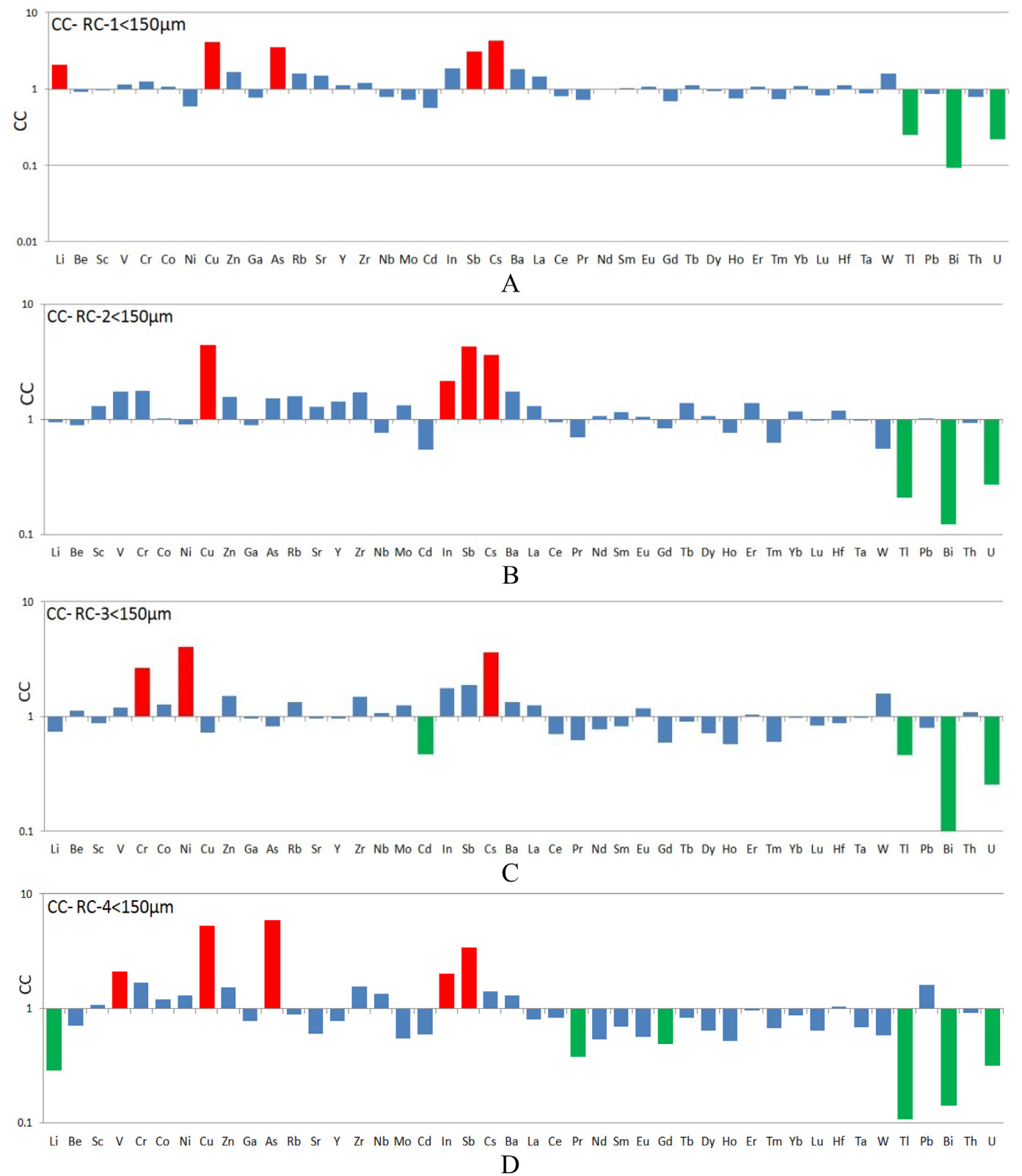
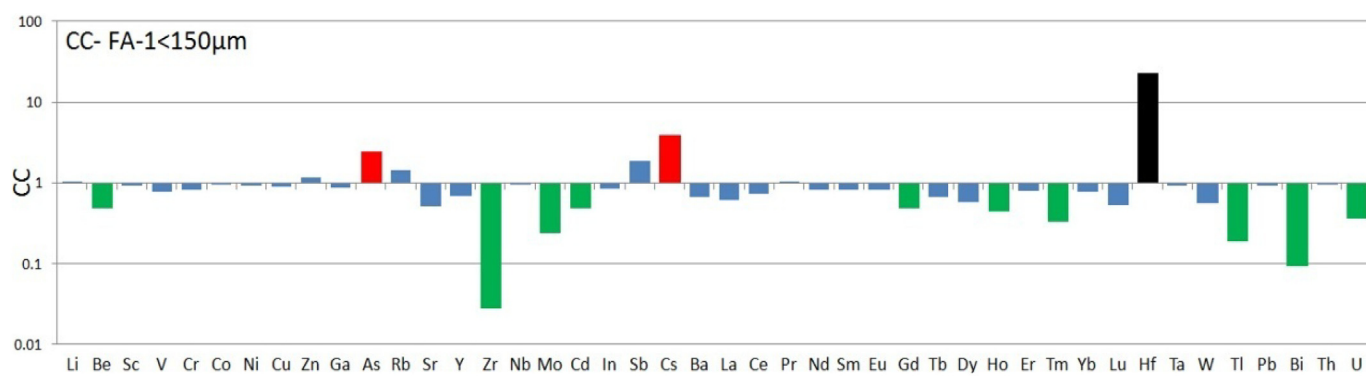


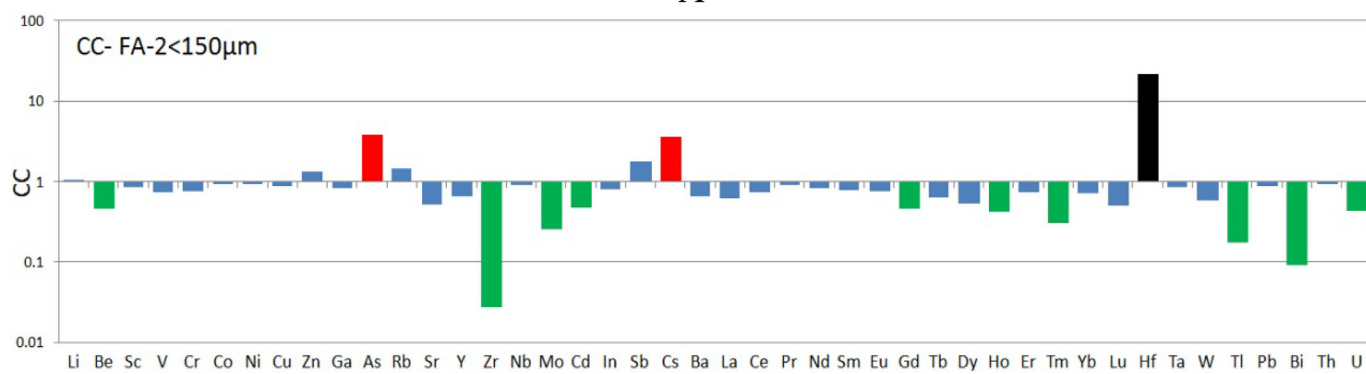
Fig. 8. Concentration coefficients of lignites.

W, among which Hf is significantly enriched, with a concentration coefficient > 30. The As content in the soil near the ash dump is much higher than the average content in Chinese soil. The enrichment

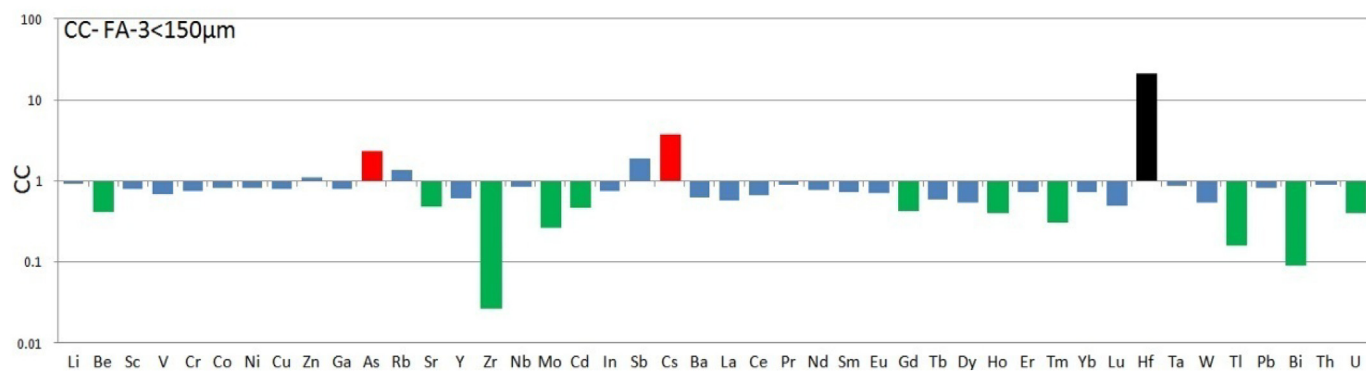
tendency of trace elements in soil is similar to those for fly ash and bottom ash, and the trace elements in ashes can leach into the soil around the ash dump.



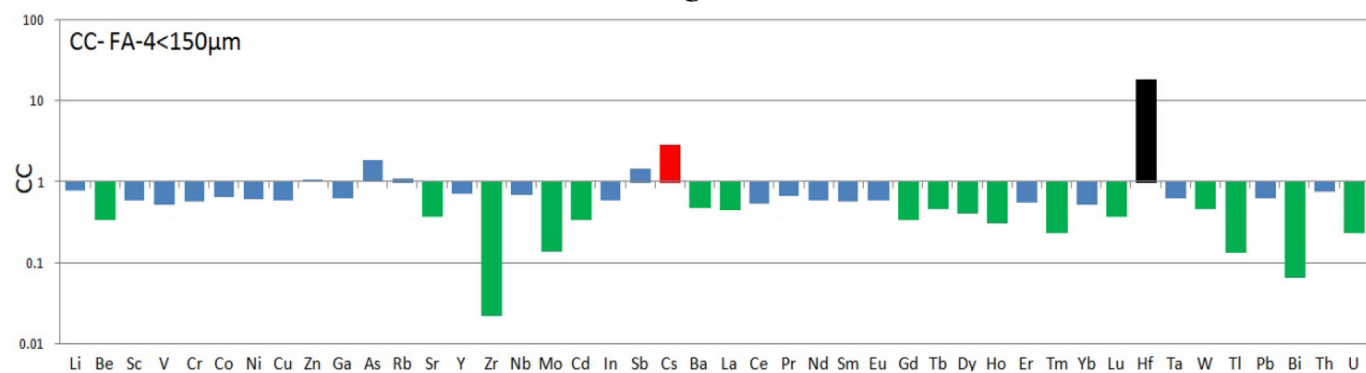
A



B

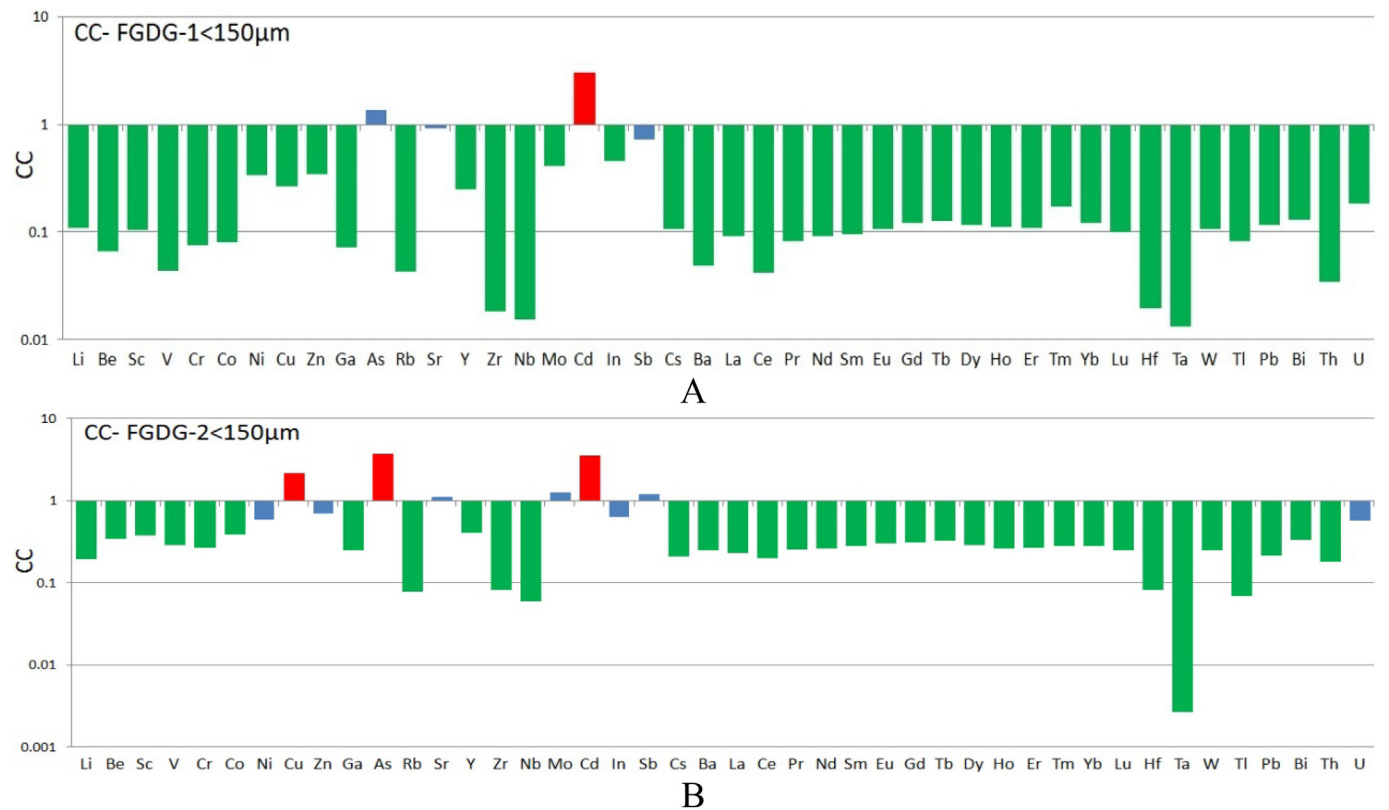
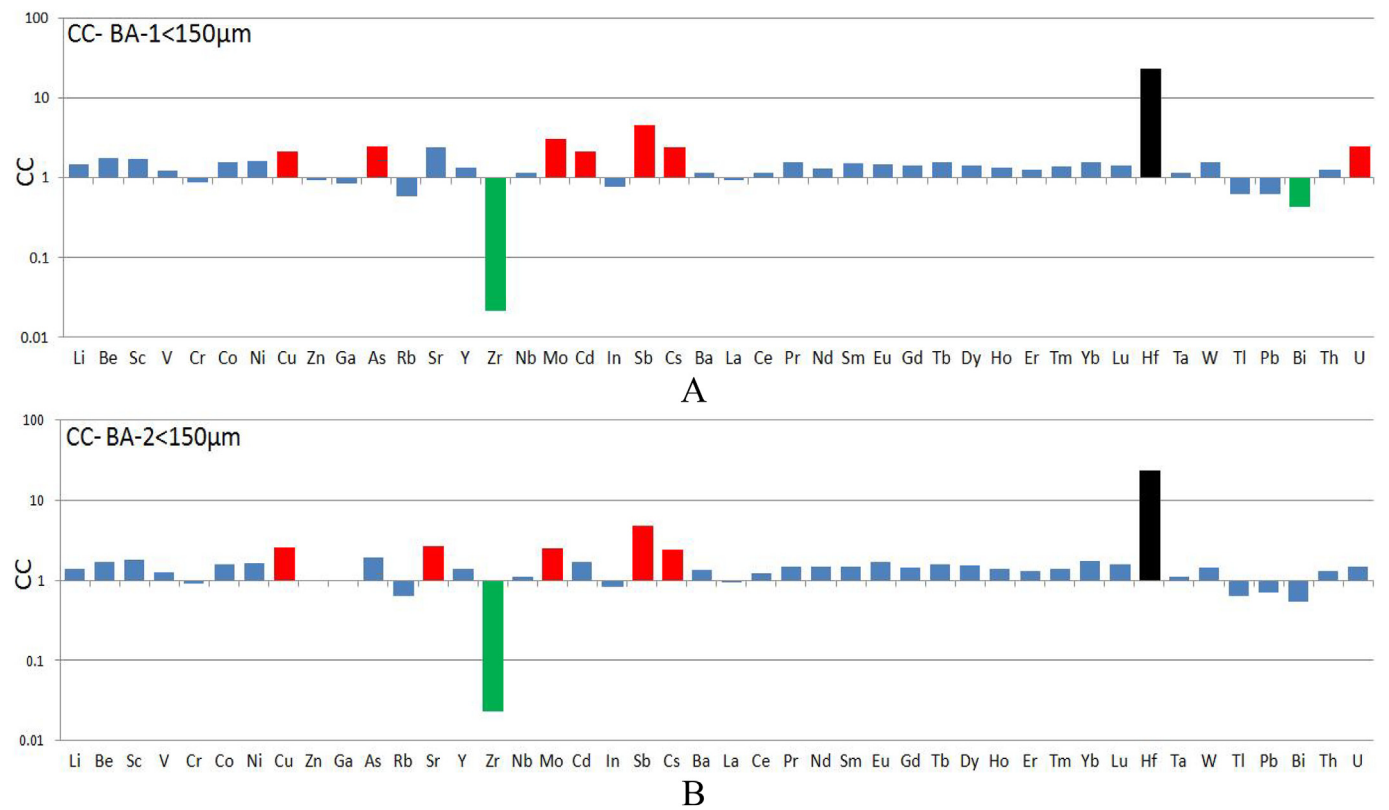


C



D

Fig. 9. Concentration coefficients of fly ashes.



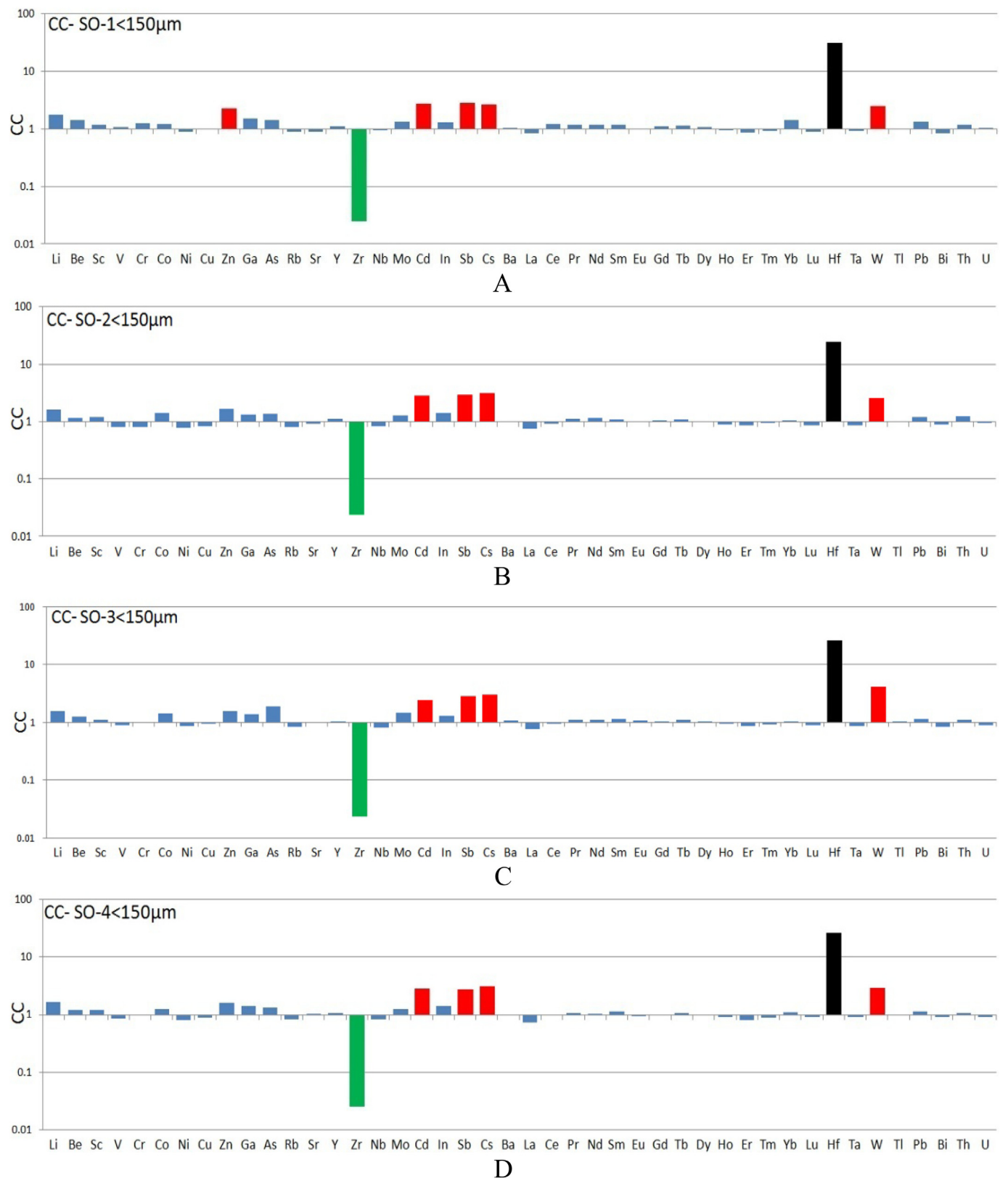


Fig. 12. Concentration coefficients of soils.

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