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Concentration and distribution of sixty-one elements in coals from DPR Korea

Jun Hu [a,b,*], Baoshan Zheng [a], Robert B. Finkelman [c], Binbin Wang [a],
Mingshi Wang [a,b], Shehong Li [a], Daishe Wu [d]

[a] State Key Lab of Environmental Geochemistry, Institute of Geochemistry, CAS, Guiyang, Guizhou 550002, China
[b] Graduate School of CAS, Beijing 100039, China
[c] US Geological Survey, Mail Stop 956, Reston, VA 20192, USA
[d] The College of Environmental Science and Enginnering at Nanchang University, Nanchang, Jiangxi 330029, China

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Abstract

Fifty coal samples (28 anthracite and 22 lignites) were collected from both main and small coal mines in DPR Korea prioritized by resource distribution and coal production. The concentrations of 61 elements in 50 coal samples were determined by several multielement and element-specific techniques, including inductively coupled plasma atomic emission spectrometry (ICP-AES), and inductively coupled plasma mass spectrometry (ICP-MS), ion chromatogram (IC), cold-vapor atomic absorption spectrometry (CV-AAS), and hydride generation atomic absorption spectrometry (HGAAS). The ranges, arithmetic means and geometric means of concentrations of these elements are presented. A comparison with crustal abundances (Clarke values) shows that some potentially hazardous elements in the coals of DPR Korea are highly enriched Li, B, S, Cl, Zn, As, Se, Cd, Sn, Sb, W, Te, Hg, Ag, Pb, and La, Ce, Dy, Tm, Ge, Mo, Cs, Tl, Bi, Th and U are moderately enriched. A comparison of ranges and means of elemental concentrations in DPR Korea, Chinese, and world coals shows the ranges of most elements in DPR Korea coals are very close to the ranges of world coals. Arithmetic means of most elements in DPR Korea coals are close to that of American coals. Most elements arithmetic means are higher in Jurassic and Paleogene coals than coals of other ages. In DPR Korea coals, only seven elements in early Permian coals are higher than other periods: Li, Zn, Se, Cd, Hg, Pb, and Bi. Only five elements B, As, Sr, Mo, W in Neogene coals have arithmetic means higher than others. SiO_2 and Al_2O_3 in ashes are more than 70% except six samples. The correlation between ash yields and major elements from high to low is in the order of $Si > Al > Ti > K > Mg > Fe > Na > Ca > P > S$. Most elements have high positive correlation with ash ($r > 0.5$) and show high inorganic affinity.

Keywords: DPR Korea; Coals; Elements; Concentration; Distribution

1. Introduction

Coal is one of the most complex organic rocks in nature. It is reported that 86 elements, including 12 major elements and 74 trace elements, are detected in coals by modern analytical techniques [1]. In special geological circumstances, some elements in coal can be enriched to industrial standards and can be extracted for use, such as gallium, germanium, uranium, vanadium, etc. In addition, with the increasing use of coal, the growing impact on the environment and human health from potentially hazardous trace elements released and transformed in the course of coal exploitation, coal cleaning, coal transportation, coal pick and combustion becomes a great concern. Impacts occur in the Southeast Asia, such as acid rain, endemic arsenosis and fluorosis and so on [2–9]. Many countries have established relevant standards of water and air quality to limit the contents of As, Cl, Pb, Hg, Cd, Se, Mn, Ni, Cu, Zn, F, Cr, Sb, Co, Mo, Be, V, Tl, Th, U, and Ag [10]. Swaine and Goodarzi [11] listed 24 elements that can harm the environment. Coals are the most important sources of these elements. Therefore, information about the concentration and distribution of these elements in coal is urgently needed in coal utilization and environmental assessments [7]. In the 1960s, some major coal producing countries started to do regional and nationwide investigations about the concentration and distribution of trace elements in coals [12]. For example, since

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* Corresponding author. Address: State Key Lab of Environmental Geochemistry, Institute of Geochemistry, CAS, Guanshui Road 43, Guiyang, Guizhou 550002, China. Tel.: +86 851 5891373; fax: +86 851 5891609.
E-mail address: hujun79@126.com (J. Hu).

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the 1970s, the United States systematically investigated trace element distribution in coals from large coal fields and main coal strata [13]. The US Geological Survey (USGS) issued the CD-ROM with their US coal quality database (version 2.0) in 1998. The CD-ROM contains the coal quality data of 7430 coal samples, with about 136 parameters recorded for each coal sample [14]. In China, researchers started to publish coal data in the 1980s [1,15–19], but so far have not produced one integral report of coal quality data.

In DPR Korea, the information about the concentration and distribution of the elements in coal is lacking in the public sector. In this paper, 50 coal samples from coal mines in DPR Korea were studied and analyzed for 61 elements. Concentrations, ranges, arithmetic means, and geometric means of these elements are presented.

2. Status and distribution of coal resources in DPR Korea

DPR Korea lacks domestic petroleum and natural gas reserves. The quantity of petroleum imports is limited for economic reasons. DPR Korea relies on two domestic sources of commercial energy—coal and hydropower—for most of its energy needs. In 2001, coal accounted for about 86% of the country's primary energy consumption [20–22]. There are about 19 billion tons of proven coal resources, 70% of which is anthracite—about 12 billion tons are distributed north of South Pyongan Province [23]. Main anthracite producing areas, such as Sunchon, Kaechon, Tokchon, Pukchang, Jiktong, etc., are in South Pyongan Province. Lignite is mainly distributed in areas of North Hamgyong Province and the Anju region of South Pyongan Province. The developing coal mines are distributed in the northeast of Hamgyong Province and in South Pyongan Province linked with the capital Pyongyang [20]. In DPR Korea, the main coal forming geological ages include early Permian (P_1), early Jurassic (J_1), late Jurassic (J_3), Paleogene (Pg_3) and Neogene (N_1) [24].

3. Samples and analytical procedures

3.1. Samples studied

The coal-sampling program was designed according to the distribution of coal resources and production. A total of 50 coal samples were collected from the main and small mines in DPR Korea, and sample locations are shown in Fig. 1. The samples were collected from six provinces and one municipality: three ones from Pyongyang, 24 from South Pyongan Province, two from North Pyongan Province, two from Chagang Province, 13 from North Hamgyong Province (seven from northern part, six from southern part), five from South Hamgyong Province, and one from North Hwanghae Province. The coal-forming geological ages, systems, series, coal fields, districts, coal mines, depositional environment, coal ranks, and numbers of samples, are listed in Table 1. All samples were collected and stored in plastic bags to prevent contamination and weathering. In the laboratory, all the samples were pulverized for determining the contents of the 61 elements.

3.2. Analytical procedures

Concentrations of the 61 elements in all 50 samples were determined by a combination of multielement and element-specific techniques in US Geological Survey (USGS) laboratory. The lower temperature (525 centigrades) of the USGS ashing procedure is recommended for trace element determinations using inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS). ICP-AES involved a sinter digestion to determine the major components of Si, Al, Ca, Mg, K, Fe, Ti, P, and the trace elements B, Ba, Zr, plus an acid digestion to determine the concentrations of Na, Be, Co, Cr, Cu, Li, Mn, Ni, Sc, Sr, Th, V, Y, Zn. ICP-MS involved the same acid digestion process to determine the trace elements Ag, As, Bi, Cd, Cs, Ga, Ge, Mo, Nb, Pb, Rb, Sb, Sn, Te, Tl, U and a sinter digestion to determine the concentrations of Ce, Dy, Er, Eu, Gd, Hf, Ho, La, Nd, Pr, Sm, Ta, Tb, Tm, W, Yb. Mercury, Se and Cl are determined on whole coals by cold-vapor atomic absorption spectrometry analysis, hydride generation atomic absorption spectrometry and ion chromatogram, respectively. Sulfur is determined on the ash (ICP-AES), reported as SO_3 and total sulfur by a LECO apparatus (LECO uses direct combustion and infrared detection).

4. Results and discussion

All the elemental concentrations are reported on a remnant moisture (as-determined) basis. This basis was chosen by USGS for previous US data reported by Bragg et al. [14]. As-determined data represents the values obtained at the particular moisture level in the sample at the time of analysis. The average remnant moisture for the DPR Korea coals is 4.0% with a maximum of 14.2%. The average remnant moisture for early Permian coals is 1.1% with a maximum of 2.0%, for Jurassic coals is 1.8% with a maximum of 3.0%, and for Tertiary coals is 8.8% with a maximum of 14.2%. In general, the values of element concentrations of Permian and Jurassic coals reported in this paper are less 2% than those same values reported on a dry basis, but the values of Tertiary coals reported in this paper are less than 10% those same values reported on a dry basis.

4.1. Concentrations of 61 elements in 50 coal samples

The ranges, arithmetic means, geometric means, standard deviations, Clarke value, and enrichment factor (EF) for 50 elements are given in Table 2. In Table 2, the EF value was calculated from the arithmetic mean (EF, formula (1)) [10].

$$\mathrm{EF} = (A_i/B_{\mathrm{Sc}})/(C_i/D_{\mathrm{Sc}}) \tag{1}$$

where

A_i, the concentration mean of element i in DPR Korea coals;
B_{Sc}, the concentration mean of scandium in DPR Korea coals;

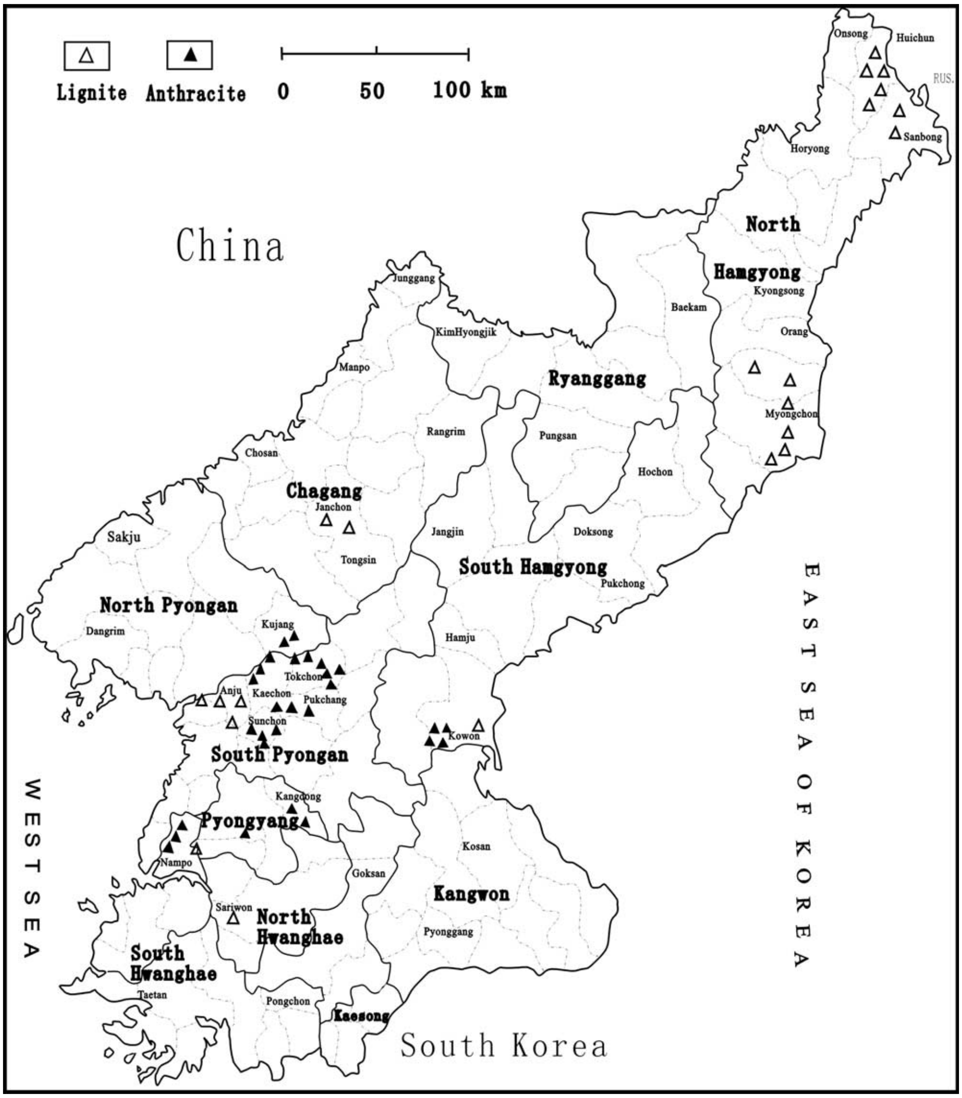

Fig. 1. Major districts and provinces in DPR Korea and locations of coal samples in this study.

C_i, the Clarke value of element i;
D_{Sc}, the Clarke value of scandium.

In comparison with the crustal average (Clarke value), we conclude that some potentially hazardous elements are highly enriched Li (11×), B (20×), S (102×), Cl (14×), Zn (5.2×), As (36×), Se (208×), Cd (12×), Sn (11×), Sb (8.9×), W (9.8×), Te(18×), Hg (20×), Ag (19×) and Pb (8.7×), and La, Ce, Dy, Tm, Ge, Mo, Cs, Tl, Bi, Th and U (2–5×) are moderately enriched.

4.2. Comparison of element concentrations in DPR Korea, Chinese, American, and World coals

Table 3 gives a comparison of DPR Korea coals with American coals. It shows that the geometric means of the elements in DPR Korea coals are very close to that in American coals except these elements (Li, P, Cl, Mn, Rb, Zr, Ag, Sn, La, Ce, Sm, Hf, Tl, Sm, Ta, W, Pb), and the maximum of elements in DPR Korea coals are much less than the maximum of corresponding elements in American coals. The ranges of most elements in DPR Korea coals are close to the ranges of corresponding elements in World coals except several elements (Cu, As, Rb, Cd, Sn, Hg, Pb, Bi). But it should be noted that the number of coal samples in this paper (50) is much less than the number used for US coals (about 7400). So it can only be said that in the 50 coal samples from DPR Korea no elements with the abnormally high concentrations analyzed are found. More researches on DPR Korea coals should be done in future.

In order to compare the arithmetic means of the elements in DPR Korea coals with China, American and World coals, we introduce the max-min relative difference of the means (MMRDM, formula (2)).

$$\mathrm{MMRDM(a–b)} = [1 - \mathrm{Min}(A_{ai}, B_{bi})/\mathrm{Max}(A_{ai}, B_{bi})] \times 100\% \quad (2)$$

Table 1
Coal-forming ages, systems, series, coal fields/districts, coal mines, depositional environment, coal rank, coal thickness in meters, and numbers of coal samples in this study [24]

Coal (ages)	Systems	Series	Coal fields/districts	Coal mine locations	Environment	Coal ranks (thickness)	No. of samples (samples identifier)
Early Permian	Pyongan	Sadong	Southern Phyongnam coal field (Pyongyang, Kangso)	Samsin, Kangdog Hukryong, Kangso	Terrigenous clastic	Anthracite (3–5, 1–2, 1–1.5)	6 (1–6)
	Pyongan	Sadong	Northern Phyongnam coal field (Sunchon, Kaechon, Pukchang, Tokchon, Kujang)	Jiktong, Ryongdae, Jajak, Ramion, Tukjiang, Hoean, Jangan, Tokchon, Namyang, Ryongsu, Ryongmun Songnam	Terrigenous clastic	Anthracite (5, 2, 15)	18 (7–24)
	Pyongan	Sadong	Kowon-Munchon coal field (Kowon)	Sudong, Jangdong	Terrigenous clastic	Anthracite	4 (25–28)
Late Jurassic	Taedong	Jung-am	Jonchon	Jonchon	Terrigenous clastic	Lignite (2–2.5)	2 (29–30)
Early Jurassic	Taedong	Songnimsan	Kangso	Taebosan	Terrigenous clastic, With volcanic intercalations	Lignite (0.1–0.2, 0.4–0.5)	1(31)
Paleogene	Anju	Ryongrim	Anju	Sosa, Ryongrim, Chili, Taeyang	Terrigenous clastic	Lignite	4 (32–35)
		Pongsan	Sariwon	Sariwon	Terrigenous clastic	Lignite (thin)	1 (36)
Neogene	Hamgyong	Onsong	Downstream of river Tuma	Kogonwon-Ryongbuk, Taesan	Lacustrine-boggy	Lignite	7 (37–43)
	Hamgyong	Myongchon	Kilju-Myongchon	Hwasong, Pyongban, Myongchon	Marine	Lignite	5 (44–48)
	Hamgyong	Kocham	Kocham	Kocham	Terrigenous	Lignitev	1 (49)
	Hamgyong	Jinsuri	Kumya	Kumya	Terrigenous	Lignite	1 (50)

where

a, country a; b, country b;
$A_{\mathrm{a}i}$, the arithmetic mean of element i in country a;
$B_{\mathrm{b}i}$, the arithmetic mean of element i in country b.

For all 61 elements calculated the MMRDM values between DPR Korea and Chinese coals, and American coals, and World coals, respectively. Elements with MMRDM values greater than 50% ($A_{\mathrm{a}i}$ or $B_{\mathrm{b}i}$ is greater than twice $B_{\mathrm{b}i}$ or $A_{\mathrm{a}i}$) are presented in Table 4.

Table 4 shows that the arithmetic means of most elements in DPR Korea coals are close to the arithmetic means of the corresponding elements in American coals. There are only 11 elements with MMRDM more than 50%: Tl, Ge, Sn, W, Li, Sb, Mo, Nb, Cs, Pb, and Hg. Of the 11 elements, Tl has the highest MMRDM (79%), based on an arithmetic mean in DPR Korea coals of 0.25 and 1.2 ppm in American coals. However, Tl is close to the arithmetic mean (0.4 ppm) in Chinese coals, within the range of World. In comparison with the arithmetic means of World coals, the following 13 elements with MMRDM values are more than 50%: Hg, Cs, Sb, Rb, Cl, Ge, Mo, As, Li, Sn, Nd, Ce, and Y. Of the 13 elements, there are eight elements in DPR Korea coals have arithmetic means higher than the corresponding elements in World coals. For example, the arithmetic mean of Hg in DPR Korea coals is 0.36 ppm, twice that in Chinese coals (0.15 ppm) or American coals (0.17 ppm), and 30 times that in world coals (0.012 ppm). Thus, the MMRDM of Hg in World coals and DPR Korea coals is 97%. Comparison of the arithmetic means of DPR Korea coals and Chinese coals shows 19 elements with MMRDM are more than 50%. The concentration of the element Te with the largest MMRDM (97%) is 0.08 ppm in DPR Korea, which is 35 times lower than Chinese coals (2.8 ppm).

4.3. Concentration of elements in coals of different geological age

In DPR Korea, there are five main coal-forming geological ages: early Permian (P_1), Neogene (N_1), Paleogene (Pg_3), early Jurassic (J_1) and late Jurassic (J_3). Early Permian coals are anthracite, and comprises approximately 70% of total proven coal resources. The Tertiary coals (N_1 and Pg_3) are lignite with large reserves. The coal resources of Jurassic ages are much less comparatively speaking, so only three samples were collected and analyzed.

In Table 5, arithmetic means and selected geometric means of elements in the 50 coal samples are presented by different coal-forming geological ages. Geometric means for the Paleogene and Jurassic coals were not included because the number of samples with these ages (5 and 3, respectively) was small. A comparison of the 61 elements arithmetic means of different coal-forming ages indicates that arithmetic means of early Permian coals were lower for all elements except Li, Zn, Se, Cd, Hg, Pb, Ge, Cl and Bi. Arithmetic means of only five elements B, As, Sr, Mo, W are higher in Neogene coals. The arithmetic means of most elements in Paleogene are very close to that in Jurassic coals except for several elements (B, Na, Si, S, K, Ca, Cr, Hf, Rb, Sr, Zr, Sn, Cs, Ba, W). Most elements

Table 2
Concentrations and statistical results of 62 elements in DPR Korea coals, values are in parts per million (ppm) except major elements in ash where noted as percent (%)

Element	Range	Arithmetic mean	Geometric mean	Standard deviations	Clarke value[a]	EF	No. of samples
Na (%)	0–0.5	0.1	0.1	0.1	2.36	0.2	50
Mg (%)	0.01–0.9	0.2	0.2	0.2	2.33	0.4	50
Al (%)	0.4–8	2.4	1.9	1.6	8.23	1.3	50
Si (%)	1–17	4.6	3.6	3.6	28.15	0.7	50
P (%)	0.01–0.2	0.03	0.02	0.04	0.105	1.3	50
S (%)	0.1–4	0.6	0.4	0.7	0.026	102	50
K (%)	0.03–2	0.4	0.3	0.4	2.09	0.9	50
Ca (%)	0.01–2	0.4	0.2	0.5	4.15	0.4	50
Ti (%)	0.03–0.4	0.1	0.1	0.07	0.57	0.9	50
Fe (%)	0.2–3	0.8	0.6	0.7	5.63	0.7	50
La	3–53	14.5	12	9.7	30	2.2	50
Ce	5–102	27.2	22.6	18.2	60	2	50
Pr	0.6–10	2.9	2.5	1.9	8.2	1.6	50
Nd	2–38	11.1	9.4	7.0	28	1.8	50
Sm	0.6–7	2.3	1.9	1.3	6.0	1.7	50
Eu	0.1–1	0.5	0.4	0.3	1.2	1.8	50
Gd	0.4–4	1.4	1.2	0.8	5.4	1.2	50
Tb	0.1–0.8	0.3	0.3	0.2	0.9	1.5	50
Dy	0.6–5	2	1.7	1.1	3.0	2.9	50
Ho	0.1–0.8	0.4	0.3	0.2	1.2	1.4	50
Er	0.3–2	1.1	1.0	0.6	2.8	1.7	50
Tm	0.1–0.6	0.3	0.2	0.1	0.48	2.5	50
Yb	0.3–2	1	0.9	0.5	3.0	1.5	50
Y	2.9–22	7.2	6.2	4.4	33	1	50
Sc	2–12	4.9	4.2	2.8	22	1	50
Li	2–190	49.2	28.6	47.4	20	11	50
Be	0.3–4	1.2	1.0	0.8	2.8	1.9	50
B	4–218	44.7	23.3	55.1	10	20	47
Cl	200–1000	400	400	200	130	14	40
V	7–93	32.2	25.8	22.5	135	1.1	50
Cr	4–80	17.8	13.8	14.9	100	0.8	50
Mn	4–395	78.4	51.2	76.6	950	0.4	50
Co	0.9–29	6.5	4.9	5.4	25	1.2	50
Ni	2–53	13.8	10.6	10.6	75	0.8	50
Cu	0.9–176	17.8	11.9	24.9	55	1.4	49
Zn	3–2960	81.0	17.9	416	70	5.2	50
Ga	2–18	6.3	5.5	3.6	15	1.9	50
Ge	0.06–28	1.4	0.6	3.9	1.5	4.2	50
As	1–183	14.6	7.7	26.2	1.8	36	50
Se	0.1–22	2.3	1.0	3.6	0.05	208	47
Rb	1–153	27.1	16.7	31.8	90	1.3	50
Sr	24–510	132	97.1	112	375	1.6	50
Zr	9–132	40.2	33.7	25.6	165	1.1	50
Nb	0.6–8	2.4	2.0	1.6	20	0.5	50
Mo	0.2–8	1.3	0.8	1.5	1.5	3.8	50
Ag	0.2–0.4	0.3	0.3	0.1	0.07	19	3
Cd	0.01–20	0.5	0.1	2.9	0.2	12	48
Sn	0.5–87	4.8	2.5	12.2	2	11	49
Sb	0.08–2	0.4	0.3	0.3	0.2	8.9	50
Te	0.01–0.3	0.1	0.7	0.05	0.02	18	49
Cs	0.2–11	2.5	1.7	2.3	3	3.7	50
Ba	10–812	165	106	169	425	1.7	50
Hf	0.3–4	1.1	0.9	0.8	3	1.7	50
Ta	0.07–0.8	0.3	0.3	0.2	2	0.6	42
W	0.3–26	3.3	2.0	4.2	1.5	9.8	50
Hg	0.02–3	0.4	0.2	0.1	0.08	20	50
Tl	0.01–1	0.3	0.2	0.2	0.45	2.5	50
Pb	1.37–259	24.4	12.9	44.6	12.5	8.7	50
Bi	0.01–3	0.2	0.1	0.4	0.17	4.7	48
Th	0.6–20	5.1	4.1	3.7	9.6	2.4	49
U	0.2–7	1.4	1.1	1.3	2.7	2.4	50

All the elemental concentrations are reported on a whole coal remnant moisture (as-determined) basis. Bold for rare earth elements (REEs). nd means no data EF is the enrichment factor. Standard deviation and EF are all calculated by arithmetic mean.

Table 3
Comparison of concentrations of elements in DPR Korea coals, Chinese coals, American coals, and World coals, values are in parts per million (ppm), except major elements in ash where noted as percent (%)

Element	DPR Korea coal				Chinese coal[a]			USA Coal[b]			World coal	
	Range	AM	GM	No.	Range	AM	No.	Max	AM	GM	Range[c]	AM[d]
Na (%)	0–0.5	0.1	0.1	50	0.002–0.46[e]	0.08[e]	126	1.4	0.08	0.04	nd	0.02
Mg (%)	0.01–0.9	0.2	0.2	50	0.05–3.97[e]	0.42[e]	45	1.5	0.11	0.07	nd	0.02
Al (%)	0.4–8	2.4	1.9	50	0.10–7.11[e]	1.94[e]	56	10.6	1.5	1.1	nd	1.0
Si (%)	1–17	4.6	3.6	50	nd	nd	nd	(13.0)	(2.4)	nd	nd	nd
P (%)	0.01–0.2	0.03	0.02	50	0.001–0.1	0.02	1770	5.8	0.043	0.002	0.001–0.3	nd
S (%)	0.1–4	0.6	0.4	50	nd	nd	nd	(3.0)	(2.17)	nd	nd	nd
K (%)	0.03–2	0.4	0.3	50	0.01–2.89[e]	0.33[e]	131	2.0	0.18	0.10	nd	0.01
Ca (%)	0.01–2	0.4	0.2	50	0.17–4.82[e]	1.31[e]	45	72	0.46	0.23	nd	1.0
Ti (%)	0.03–0.4	0.1	0.1	50	0.001–0.42	0.052	831	0.74	0.08	0.06	0.001–0.2	nd
Fe (%)	0.2–3	0.8	0.6	50	0.07–4.48[e]	1.21[e]	127	24	1.3	0.75	nd	1.0
La	3–53	14.5	12	50	0.21–118[e]	26.1[e]	126	300	12	3.9	1–40	10
Ce	5–102	27.2	22.6	50	2.35–225[e]	49.8[e]	126	700	21	5.1	2–70	11.5
Pr	0.6–10	2.9	2.5	50	0.15–28.2[f]	3.8[f]	110	(65.0)	(4.81)	nd	1–10	nd
Nd	2–38	11.1	9.4	50	0.06–88.7[e]	22.1[e]	127	230	9.5	nd	3–30	4.7
Sm	0.6–7	2.3	1.9	50	0.08–19.3[e]	4.09[e]	126	18	1.7	0.35	0.5–6	1.6
Eu	0.1–1	0.5	0.4	50	0.02–2.54[e]	0.72[e]	126	4.8	0.40	0.12	0.1–2	0.7
Gd	0.4–4	1.4	1.2	50	0.26–19.3[f]	3.4[f]	110	(21.0)	(1.50)	nd	0.4–4	nd
Tb	0.1–0.8	0.3	0.3	50	0.03–2.4[e]	0.58[e]	126	3.9	0.30	nd	0.1–1	0.3
Dy	0.6–5	2	1.7	50	0.27–25.1[f]	3.14[f]	110	(28.0)	(1.49)	nd	0.5–4	nd
Ho	0.1–0.8	0.4	0.3	50	0.06–6.5[f]	0.73[f]	110	(12.0)	(0.47)	nd	0.1–2	nd
Er	0.3–2	1.1	1.0	50	0.13–19.5[f]	2.1[f]	110	(11.0)	(0.63)	nd	0.5–3	nd
Tm	0.1–0.6	0.3	0.2	50	0.02–3.7[f]	0.34[f]	110	(5.1)	(0.28)	nd	nd	nd
Yb	0.3–2	1	0.9	50	0.05–20.2[e]	1.78[e]	127	20	0.95	nd	0.3–3	0.5
Y	2.9–22	7.2	6.2	50	0.5–22	8	806	170	8.5	6.6	2–50	15[c]
Sc	2–12	4.9	4.2	50	0.5–12	3	1129	100	4.20	3.00	1–10	5
Li	2–190	49.2	28.6	50	0.5–37	14	354	370	16	9.2	1–80	20[c]
Be	0.3–4	1.2	1.0	50	0.1–0.6	2	1123	330	2.2	1.3	0.1–15	1.5[c]
B	4–218	44.7	23.3	47	10–250	63	884	1700	49	30	5–400	75
Cl	200–1000	400	400	40	50–500	220	280	8800	614	79	50–2000	1000
V	7–93	32.2	25.8	50	2–100	21	1141	370	22	17	2–100	25
Cr	4–80	17.8	13.8	50	2–50	12	1410	250	15	10	0.5–60	10
Mn	4–395	78.4	51.2	50	4–109	77	1070	2500	43	19	5–300	50
Co	0.9–29	6.5	4.9	50	1–20	7	1405	500	6.1	3.7	0.5–30	5.0
Ni	2–53	13.8	10.6	50	2–65	14	1236	340	14	9.0	0.5–50	15
Cu	0.9–176	17.8	11.9	49	1–50	13	1218	280	16	12	0.5–50	15
Zn	3–2960	81.0	17.9	50	2–106	35	1352	19000	53	13	5–300	50
Ga	2–18	6.3	5.5	50	1–20	9	2428	45	5.7	4.5	1–20	7
Ge	0.06–28	1.4	0.6	50	0.5–10	4	3084	780	5.7	0.59	0.5–50	6[c]
As	1–183	14.6	7.7	50	0.4–10	5	1915	2200	24	6.5	0.5–80	5.0
Se	0.1–22	2.3	1.0	47	0.1–11	2	1315	150	2.8	1.8	0.2–10	3.0
Rb	1–153	27.1	16.7	50	1–30	8	404	140	21	0.62	2–50	5
Sr	24–510	132	97.1	50	27–300	136	509	2800	130	90	15–500	130
Zr	9–132	40.2	33.7	50	20–150	52	411	700	27	19	5–200	30
Nb	0.6–8	2.4	2.0	50	1–97	14	66	70	2.0	1.0	1–20	nd
Mo	0.2–8	1.3	0.8	50	1–15	4	271	280	3.3	1.2	0.1–10	5.0
Ag	0.2–0.4	0.3	0.3	3	0.2–1	0.5	99	19	(<0.1)	0.01	0.02–2	<0.1[c]
Cd	0.01–20	0.5	0.1	48	0.01–3	0.2	1201	170	0.47	0.02	0.1–3	0.3
Sn	0.5–87	4.8	2.5	49	0.4–5	2	105	140	1.3	0.001	1–10	2[c]
Sb	0.08–2	0.4	0.3	50	0.1–10	2	446	35	1.2	0.61	0.05–10	3.0
Te	0.01–0.3	0.1	0.7	49	0.09–20	2.8	20	nd	(<0.1)	nd	nd	nd
Cs	0.2–11	2.5	1.7	50	0.1–0.3	1	314	15	1.1	0.7	0.3–5	0.2
Ba	10–812	165	106	50	13–400	82	549	22000	170	93	20–1000	120
Hf	0.3–4	1.1	0.9	50	0.01–9	2.4	372	18	0.73	0.04	0.4–5	nd
Ta	0.07–0.8	0.3	0.3	42	0.06–4	0.7	357	1.7	0.22	0.02	0.1–1	0.3
W	0.3–26	3.3	2.0	50	0.1–9	2	378	400	1.0	0.10	0.5–5	2
Hg	0.02–3	0.4	0.2	50	0.01–1	0.15	1466	10	0.17	0.10	0.02–0.1	0.012
Tl	0.01–1	0.3	0.2	50	0.1–1	0.4	768	52	1.2	<0.01	<0.2–1	nd
Pb	1.37–259	24.4	12.9	50	10–47	13	1280	1900	11	5.0	2–80	25

Table 3 (*continued*)

Element	DPR Korea coal				Chinese coal[a]			USA Coal[b]			World coal	
	Range	AM	GM	No.	Range	AM	No.	Max	AM	GM	Range[c]	AM[d]
Bi	0.01–3	0.2	0.1	48	0.1–1.4	0.8	65	nd	(<1.0)	nd	<0.05	nd
Th	0.6–20	5.1	4.1	49	0.5–15	6	442	79	3.2	1.7	0.5–10	6.3
U	0.2–7	1.4	1.1	50	0.5–10	3	621	1300	2.1	1.1	0.5–10	1

nd means no data. Bold for rare earth elements (REEs). Max, maximum of concentrations; AM, arithmetic mean; GM, geometric mean; No, number of samples.

[a] From Zhao et al. (2002) [18].

[b] From Finkelman (1993) [27].

[c] From Swaine (1990) [28], data for most coals in world.

[d] From Valkovic (1983) [29]. Data in () were calculated by the data from USGS CD-ROM (7430 samples) [14].

[e] From Ren et al. (1999) [10].

[f] From Zhao et al. (2002) [25].

arithmetic means are much higher in Jurassic and Paleogene coals: Al, K, Ti, Sc, V, Cr, Mn, Fe, Co, Ni, Ga, Rb, Nb, Cs, Sn, Ba, La, Ce, Hf, Nd and Th. The geometric means of elements in Permian coals are very close to that in Neogene coals exclude these elements: Li, B, S, Ca, V, Se, Sr, Ba and Pb.

From Fig. 1 and Table 1, the Permian coals are distributed in South Pyongan Province around the capital Pyongyang. The Neogene coals are mainly distributed in the areas of North Hamgyong Province, and the Paleogene coals are distributed in the Anju region on the coast of the west sea of Korea. Two of the three Jurassic coal samples were collected from Jonchon region in Chagang province, one was from Kangso district. So, some trends of the element contents varying in different areas can be known from the above analysis of different ages, and we did not try to analyze it again.

4.4. Main ash compositions

Table 6 reports the ash yields and the arithmetic means of main chemical composition in 50 coals samples with different geological ages. The ash yields in DPR Korea coals range from 4 to 56%, and the arithmetic mean is 18%. The ash yields vary with different ages. In Paleogene, Neogene, Jurassic and early Permian, the ranges of ash yields are 11–52%, 4–44%, 22–56%, 5–30% and their arithmetic means are 28, 18, 42, 14%, respectively. From the analysis, the maximum of ashes yield in Permian coals is 30%, but the minimum of ashes yield in Jurrassic coals is 22%. Four high ash coal samples with the ash yields more than 40% were collected from Jonchon, Ryongrim and Ryongbuk regions. In conclusion, most of DPR Korea coals have low-medium ash yield (8–25%),, and the ash yields of anthracite are mainly distributed in 8–20%, but the ash yields of lignite have no rule from 4 to 56%.

In 50 coal samples, the contents of SiO_2 and Al_2O_3 in ashes are more than 70% except six samples, which indicates that clay minerals and quartz are very rich in the mineral components of DPR Korea coals. The contents of Fe_2O_3 in ashes range from 1 to 13% except one sample collected from Sariwon district with the very high content (36%). Fe_2O_3 is relatively enriched in coal ashes, especially in Paleogene coals. CaO and SO_3 range from 0.1 to 20%, 0.2 to 17%, and they are richer in Paleogene and Neogene coals. K_2O and Na_2O contents in the ashes range from 0.5 to 6%, 0.1 to 6%, and are richer in Jurassic coals, Neogene coals, respectively. MgO, TiO and P_2O_5 range from 0.4 to 5%, 0.5 to 6%, and 0.04 to 3%, respectively.

4.5. Affinity of the elements

Table 7 presents the correlation between ash yields and major elements and selected elements combinations. The relationship between ash yields and major elements from high to low is in the order: Si (0.97), Al (0.95), Ti (0.91), K (0.85), Mg (0.78), Fe (0.66), Na (0.35), Ca (0.29), P (0.09), S (−0.01). There is a very good correlation ($r = 0.98$) between ash yields and combination of Si and Al, which may indicate that Al and Si are mainly associated with aluminosilicate minerals in DPR Korea coals. Relatively high correlation for Ti and ash ($r = 0.91$) suggest the existence of titanium oxides in all coal samples. The high correlation for K with ash, Si, Al may suggest the existence of K-bearing clay minerals. The relatively low correlation for Na, Ca and ash may be that Na, Ca bearing minerals has much difference in different areas. There is no correlation between phosphorus and sulphur with ash yields. The further discussion will be carried out after the analysis of minerals in DPR Korea coals.

Table 4
The elements with MMRDM values (>50%) between DPR Korea and Chinese, American, and world coals

Countries	Two large MMRDM	The elements: MMRDM > 50% (ordered by the values from big to small)
Korea-Chinese	Te (97%), Nb (83%)	**Sb**, Bi, Li, Rb, **Mo**, **As**, Ge, **Cd**, Cs, Ta, **Hg**, **Sn**, **Zn**, Ti, Hf, **U**, **Ba**
Korea-American	**TI** (79%), Ge (75)	**Sn**, W, Li, Sb, Mo, Nb, Cs, **Pb**, **Hg**
Korea-World	**Hg** (97%), Cs (92%)	**Sb**, Rb, **Cl**, Ge, **Mo**, **As**, Li, **Sn**, Nd, Ce, Y

Bold words are the elements that may cause environmental harm (Swaine, 1995) [11]. Underlined elements indicate higher arithmetic means in DPR Korea coals than in other country coals.

Table 5
Arithmetic and geometric means of element of coals with different coal-forming ages in DPR Korea

Element	Early Permian (P_1)			Neogene (N_1)			Paleogene(Pg_3)		Jurassic(J_1, J_3)	
	AM	GM	No.	AM	GM	No.	AM	No.	AM	No.
Na (%)	0.09	0.07	28	0.2	0.1	14	0.05	5	0.2	3
Mg (%)	0.2	0.1	28	0.2	0.2	14	0.4	5	0.4	3
Al (%)	1.8	1.7	28	2.4	1.9	14	3.8	5	4.9	3
Si (%)	3.3	3.2	28	4.3	3.4	14	6.5	5	12.7	3
P (%)	0.03	0.02	28	0.03	0.01	14	0.02	5	0.02	3
S (%)	0.4	0.3	28	0.8	0.6	14	1.4	5	0.5	3
K (%)	0.4	0.3	28	0.2	0.1	14	0.6	5	1.6	3
Ca (%)	0.15	0.07	28	0.8	0.6	14	0.9	5	0.3	3
Ti (%)	0.1	0.1	28	0.1	0.1	14	0.2	5	0.3	3
Fe (%)	0.6	0.5	28	0.7	0.5	14	1.9	5	1.6	3
La	12.0	10.9	28	12.0	10.2	14	28.8	5	26.4	3
Ce	21.8	19.8	28	24.2	20.6	14	52.5	5	48.8	3
Pr	2.4	2.2	28	2.6	2.0	14	5.3	5	5.6	3
Nd	9.0	8.1	28	10.3	8.8	14	19.8	5	20.6	3
Sm	1.8	1.7	28	2.2	1.9	14	3.8	5	4.0	3
Eu	0.4	0.3	28	0.5	0.5	14	0.8	5	0.8	3
Gd	1.2	1.1	28	1.4	1.2	14	2.2	5	2.4	3
Tb	0.3	0.2	28	0.3	0.3	14	0.4	5	0.5	3
Dy	1.8	1.6	28	1.92	1.6	14	2.7	5	3.1	3
Ho	0.3	0.3	28	0.4	0.3	14	0.5	5	0.6	3
Er	1.0	0.9	28	1.1	0.9	14	1.4	5	1.6	3
Tm	0.2	0.2	28	0.3	0.2	14	0.3	5	0.4	3
Yb	0.9	0.8	28	1.0	0.9	14	1.2	5	1.5	3
Y	5.1	4.7	28	8.9	7.8	14	13.1	5	9.2	3
Sc	3.8	3.5	28	5.8	4.9	14	7.2	5	7.7	3
Li	72.8	51.4	28	13.5	1.1	14	26.3	5	33.7	3
Be	0.9	0.9	28	1.3	1.0	14	2.1	5	2.0	3
B	12.0	10.2	25	94.0	71.9	14	38.0	5	98.6	3
Cl	500	400	28	200	200	8	300	5	400	1
V	20.8	18.8	28	42.6	34.1	14	50.1	5	59.8	3
Cr	12.0	10.9	28	16.7	13.5	14	35.5	5	48.2	3
Mn	61.9	42.1	28	86.3	50.1	14	139	5	93.9	3
Co	5.1	3.8	28	6.8	5.8	14	11.5	5	10.3	3
Ni	11.2	9.01	27	11.4	9.11	14	25.1	5	30.4	3
Cu	18.6	10.6	28	13.3	10.9	14	21.9	5	24.7	3
Zn	122	17.2	28	16.4	11.8	14	44.7	5	57.9	3
Ga	5.1	4.7	28	6.4	5.6	14	9.8	5	11.8	3
Ge	0.5	0.4	28	3.3	1.4	14	1.1	5	1.8	3
As	12.5	9.4	28	19.4	5.0	14	13.8	5	13.3	3
Se	3.7	2.5	28	0.2	0.2	11	0.4	5	1.0	3
Rb	21.6	18.9	28	12	7.3	14	50.6	5	109	3
Sr	110	77.8	28	184	155	14	158	5	51.5	3
Zr	34.5	31.4	28	42.4	33.6	14	35.3	5	92.7	3
Nb	2.0	1.9	28	1.8	1.6	14	4.2	5	5.6	3
Mo	0.6	0.5	28	2.4	1.7	14	1.7	5	1.2	3
Ag	0.3	0.3	3	Nd	nd	nd	nd	nd	nd	nd
Cd	0.8	0.1	28	0.07	0.05	13	0.1	4	0.2	3
Sn	2.7	2.2	28	3.5	2.4	13	20.6	5	3.7	3
Sb	0.3	0.3	28	0.4	0.3	14	0.7	5	0.7	3
Te	0.09	0.07	28	0.05	0.04	13	0.1	5	0.1	3
Cs	1.9	1.6	28	2.1	1.2	14	3.9	5	7.3	3
Ba	70.0	56.9	28	253	198	14	404	5	255	3
Hf	1.0	0.9	28	1.2	0.9	14	1.1	5	2.9	3
Ta	0.3	0.2	25	0.2	0.2	10	0.5	4	0.6	3
W	2.5	1.7	28	5.3	3.4	14	1.2	5	4.7	3
Hg	0.5	0.4	28	0.07	0.05	10	0.1	5	0.08	3
Tl	0.2	0.2	28	0.12	0.09	14	0.6	5	0.6	3
Pb	34.2	19.5	28	6.1	4.8	14	21.2	5	22.4	3
Bi	0.3	0.1	28	0.06	0.05	12	0.05	5	0.1	3
Th	4.2	3.8	27	3.9	3.2	14	10.4	5	8.6	3
U	1.3	1.0	28	1.4	1.0	14	1.8	5	2.2	3

Values are in parts per million (ppm), except main elements in ash where noted as percent (%). Bold for rare earth elements (REEs). nd means no data. AM, arithmetic mean; GM, geometric mean; No, number of samples.

Table 6
The arithmetic mean of main ash composition in 50 coal samples with different coal-forming periods. All values are percent (%)

Ages	Ash	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	TiO_2	K_2O	Na_2O	P_2O_5
Pg_3 (5)	28	44	24	13	6	2	7	1	2	0.2	0.2
N_1 (14)	18	48	24	6	7	2	8	1	1	1.6	0.5
$J_{1,3}$ (3)	42	64	22	6	1	2	1	1	4	0.6	0.1
P_1 (28)	14	55	25	6	2	2	2	1	3	0.9	0.4
Total(50)	18	52	24	7	3	2	4	1	3	1.0	0.4

Table 7
The correlation coefficients between ash yield and the content of each major elements or selected elements combination

	Si	Al	Ti	K	Mg	Fe	Na	Ca	P	S	Si+Al	Na+K	Ca+Mg	Fe+S
Ash	0.97	0.95	0.91	0.85	0.78	0.66	0.35	0.29	0.09	−0.01	0.98	0.87	0.47	0.37
Si		0.92	0.88	0.87	0.68	0.57	0.30	0.12	0.02	−0.10	0.99	0.88	0.31	0.26
Al			0.92	0.78	0.68	0.55	0.30	0.24	0.14	−0.04	0.96	0.79	0.40	0.29
Ti				0.80	0.67	0.52	0.29	0.24	0.12	−0.08	0.91	0.81	0.40	0.24
K					0.62	0.53	0.20	−0.03	0.16	0.09	0.85	0.98	0.16	0.25
Mg						0.73	0.34	0.56	0.03	0.05	0.69	0.65	0.75	0.44
Fe							0.21	0.40	0.00	0.49	0.57	0.54	0.54	0.86
Na								0.22	−0.07	−0.04	0.31	0.39	0.28	0.10
Ca									0.14	0.17	0.16	0.01	0.97	0.33
P										−0.09	0.06	0.14	0.12	−0.05
S											−0.08	−0.09	0.15	0.87

Table 8 lists the elements affinity from correlation coefficients between element contents and ash yields and selected major elements. All elements are classified to four groups according to their correlation coefficients with ash yield. The first group has a very high positive correlation with ash yields ($r_{ash} > 0.7$: La,Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Sc, Hf, Ta, Cr, Th, V, Zn, Al, K, Mg, Ti, Zr, Cs, Ga, Nb, Rb, Si). These elements have high inorganic affinity. All determined rare earth elements (except Y) have high correlation with ash yields, in addition, light REEs have higher correlation (r from 0.81 to 0.88) than heavy REEs (r from 0.72 to 0.82). Most of these elements have high positive correlation coefficient with aluminosilicate minerals ($r_{Si+Al} > 0.7$). The second group includes five elements (Fe, Co, Ni, Tl, Y) with medium positive correlation with ash yields varying from 0.51 to 0.69, which have prevailing inorganic affinity. The third group includes the elements with less correlation with ash yield (from 0.21 to 0.5): Na, Be, Mn, Ca, B, Ba, Mo, Sb, Sn, Te, U. The fourth group includes the elements with the lowest correlation with ash yields below the statistically significant value ($r < 0.20$): Cl, Se, Hg, Cu, Li, Sr, As, Bi, Cd, Ge, Pb, W, S, P. These elements are all chalcophile elements except Li and W. Three elements (Cl, Se, Hg) have relatively high negative correlation with ash yield (−0.12, −0.16, and −0.28, respectively).

5. Conclusions

1. In this paper, the ranges, and some statistic results of concentrations of 61 elements in 50 coal samples from DPR Korea are presented. The means of concentrations of these 61 elements are also presented by coal-forming age.
2. A comparison with published crustal abundances indicates that DPR Korea coals are highly enriched in Li, B, S, Cl, Zn, As, Se, Cd, Sn, Sb, W, Ag, Te, Hg and Pb, and are moderately enriched in La, Ce, Dy, Tm, Mo, Cs, Tl, Bi, Th and U.

Table 8
Element affinities deduced from Pearson's correlation coefficients between the content of each element in coals and ash yields, and the content of Si+Al, Ca+Mg, Ca+Mg, respectively

Element affinity	Elements
$r_{ash} > 0.7$	La,Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Sc, Hf, Ta, Cr, Th, V, Zn, Al, K, Mg, Ti, Zr, Cs, Ga, Nb, Rb, Si
$r_{ash} > 0.5$	Fe, Co, Ni, Tl, Y
$r_{ash} > 0.2$	Na, Be, Mn, Ca, B, Ba, Mo, Sb, Sn, Te, U
$r_{ash} < 0.2$	Cl (−0.12), Se (−0.16), Hg (−0.28), Cu, Li, Sr, As, Bi, Cd, Ge, Pb, W, S, P
$r_{Si+Al} > 0.7$	La,Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Sc, Hf, Cr, Th, V, Zn, K, Ti, Zr, Cs, Ga, Nb, Rb
$r_{Si+Al} > 0.5$	Ni, Y, Fe, Mg, Tl
$r_{Si+Al} > 0.2$	Na, Be, Co, Cu, Mn, Ba, Sb, Sn, Te, U
$r_{Ca+Mg} > 0.7$	No elements
$r_{Ca+Mg} > 0.5$	Mn, V, Y, Fe, B, Ba,
$r_{Ca+Mg} > 0.2$	La,Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Na, Be, Co, Cr, Ni, Sc, Sr, Th, Zn, Al, Ce, Si, Ti, Zr, Cs, Ga, Mo, Nb, Rb, Sn, U
$r_{Fe+S} > 0.7$	No elements
$r_{Fe+S} > 0.5$	Co, Ni, Y Sb
$r_{Fe+S} > 0.2$	La,Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Be, Cr, Mn, Sc, Th, V, Zn, Al, Ca, Yb, Si, K, Mg, Ti, Ba, As, Cs, Ga, Mo, Rb, Sn, Tl, U

3. Comparisons with Chinese coals, American coals, and World coals show that the ranges of most elements in DPR Korea coals are close to that of corresponding elements in World coals except several elements (Cu, As, Rb, Cd, Sn, Hg, Pb, Bi). Arithmetic means of most elements in DPR Korea coals are close to those of American coals exclude the elements with RWs more than 50%: Tl, Ge, Sn, W, Li, Sb, Mo, Nb, Cs, Pb, and Hg. There are no elements with abnormally high contents discovered in the 50 coal samples from DPR Korea.
4. Arithmetic means of only several elements in early Permian coals are higher than other ages: Li, Zn, Se, Cd, Hg, Pb, and Bi. Only five elements B, As, Sr, Mo, W in Neogene coals have higher arithmetic means than others. Most elements arithmetic means are much higher in Jurassic and Paleogene coals.
5. Clay minerals and quartz can be very rich in the mineral components of DPR Korea coals. SiO_2 and Al_2O_3 in ashes are more than 70% except six samples. The relationship between ash yields and major elements from high to low is in the order of $Si > Al > Ti > K > Mg > Fe > Na > Ca > P > S$.
6. Most elements have high positive correlation with ash ($r > 0.5$) and show high inorganic affinity.

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