

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

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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. SiO₂ and Al₂O₃ 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.

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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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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 Suncheon, Kaecheon, 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].

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

where

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

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