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Review

Abundance, Distribution, and Modes of Occurrence of Uranium in Chinese Coals

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Abstract: Due to its environmental and resource impacts, the geochemistry of uranium in coal is of both academic and practical significance. In order to give a comprehensive summary about the geochemistry of uranium in coals, the abundance, distribution, and modes of occurrence of uranium in Chinese coals were reviewed in this paper. Although some coals from southwestern and northwestern China are significantly enriched in uranium, the common Chinese coals are of a comparable uranium concentration to the world coals. The roof and floor rocks, and parting of coalbeds, or coal benches that are close to the surrounding rock are favorable hosts for uranium in one coalbed. The uranium concentrations in coals of different ages decrease in this order, e.g., Paleogene and Neogene > Late Permian > Late Triassic > Late Carboniferous and Early Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic. Uranium in Chinese coals is mainly associated with organic matter, and is correspondingly enriched in subbituminous coal and lignite.

Keywords: uranium; geochemistry; abundance; distribution; modes of occurrence; Chinese coals

1. Introduction

Uranium is a radioactive element, which is both chemically and radiologically toxic [1]. Uranium connately and ubiquitously occurs in coal. Thus, coal combustion is considered as one source of radioactive material in the environment [2–5]. Though it is controversial, the radiation doses from atmospheric emission of a coal-fired power plant were considered to be greater than those from a nuclear plant of comparable size [6,7]. In 1978, it was reported that as high as 2,975 kg uranium were emitted into the atmosphere from one Chinese coal-fired power plant [8]. Additionally, Chen et al. [9] estimated that about 62.9 tons of uranium were released into the atmosphere from Chinese coal-fired power plants in 2014. Moreover, coal combustion residues derived from coals with the uranium concentrations higher than 10 mg/kg would be associated with radioactivity exceeding the standards for radiation in building materials [10].

Besides the detrimental aspects of uranium in coals, uraniferous coal (with 30–50 mg/kg uranium) has been classified as an unconventional uranium resource [11]. If a coal had a uranium concentration higher than 200 mg/kg, it could be regarded as a resource for industrial extraction [12]. However, Huang et al. [13] suggested that 50 mg/kg in coal was comparable in grade to a low-grade yellowcake deposit. Furthermore, Sun et al. [14] set the benchmark for uranium recovery from coal as low as 40 mg/kg. Factually, uranium production is the first example for critical element utilization from coal and coal ash [15]. Uranium extraction from high-U coals in the United States of America (USA) and the former Union of Soviet Socialist Republics (USSR) had led to the essential acceleration of the establishment of a nuclear industry in both countries during post-WWII years [15–17]. High-U coals have again attracted much attention for industrial utilization [9,17,18]. In addition to coal as the hosted uranium deposit, the host rocks of the coal seams (such as floor [19]) and stone coals [20] also contain high concentrations of uranium and thus have both potential industrial significance and adverse environmental effects.

As a companion paper to Chen et al. [9], the abundance of uranium in common Chinese coals, some abnormally uranium-rich coals, spatial and temporal distribution of uranium in Chinese coals, modes of occurrence of uranium in Chinese coals, and relation of uranium to coal ranks are discussed in detail.

2. Abundance of Uranium in Chinese Coals

2.1. Abundance of Uranium in Chinese Coals

Historically, Chen et al. [21,22], Ren et al. [23], Tang and Huang [24], Tang et al. [25], Ren et al. [26], and Yang [27] reported the uranium abundance of Chinese coals. Based on 1,883 data points, Dai et al. [28] assigned a latest datum of 2.43 mg/kg for uranium abundance of common Chinese coals, which is comparable to that of the world coal (2.40 mg/kg [29]).

A total of 2,670 data points of uranium concentrations in Chinese coals were collected from a previous study [9]. However, in view of the unavailable first-hand data in some papers and the wide use of the data of Dai et al. [28] as backgrounds of trace elements in Chinese coals for geochemical comparison to other coals, 2.43 mg/kg was set as the concentration of common Chinese coals in this paper.

2.2. Significant Enrichment of Uranium in Some Chinese Coals

According to the concentration coefficients (CC: ratio of trace element concentration in targeted coal to the averages of common Chinese coals or world coals), Dai et al. [30] divided the enrichment of trace elements in coal into five types, i.e., abnormal enrichment (CC > 100), significant enrichment (100 > CC > 10), enrichment (10 > CC > 5), slight enrichment (5 > CC > 2), and depletion (0.5 > CC). Based on this suggestion, if the uranium concentration in a coal is higher than 24.3 mg/kg, then it was classified as significant enrichment.

All of the significantly uranium-rich Chinese coals are tabulated in Table 1. Significant uranium enrichment in coals only occurs in Shanxi, Yunnan, Guizhou, Guangxi, Xinjiang, Inner Mongolia, Sichuan, and Chongqing Provinces of China (Table 1), e.g., the Datong coal (averaging 38.2 mg/kg and 28.8 mg/kg uranium [31]), the Dazhai coal (52.5 mg/kg [32] and 56.0 mg/kg [33]), the Yanshan coal (167 mg/kg [34] and 153 mg/kg [35]), the Luquan coals (34.1 mg/kg [36]), the Guiding coal (229 mg/kg [34] and 200 mg/kg [30]), the Puan coal (32.4 mg/kg [37]), the Zhijin coal (49.6 mg/kg [38]), the Heshan coal (10.2 mg/kg to 326 mg/kg [39–42]), the Yishan coal (71.7 mg/kg [43]), the Sawabuqi coal (365 mg/kg [44]), the Yili coal (320 mg/kg [45] and 147 mg/kg [46]), the Shenli coal (25.9 mg/kg [47]), the Shiping coal (39.8 mg/kg [48]), and the Moxinpo coal (376 mg/kg [49]).

Coal with a uranium concentration of higher than 200 mg/kg was regarded as a resource for industrial extraction [12]. Some coals in China with abnormally high uranium concentrations (>243 mg/kg, one hundred times higher than the average of common Chinese coals) are classified as the coal-hosted uranium deposits. Chen et al. [9] summarized that some coal-hosted uranium deposits, i.e., coals from the Yili and Tarim Basins of Xinjiang Province, Bangmai Basin of Yunnan Province, and Mabugang Basin of Guangdong Province.

Table 1. Significantly uranium-rich coals in China.

Coalfields/Provinces	Ranges/Means mg/kg [a]	Coalbeds	Coal Ranks	Coal-Forming Periods	Reference
Datong coalfield/Shanxi	33.0–42.0/38.2	4	n.d. [b]	Early Permian	Wang et al. [31]
Datong coalfield/Shanxi	5.00–92.0/28.8	3, 5, 8	n.d. [b]	Late Carboniferous	Wang et al. [31]
Dazhai Mine/Yunnan	9.56–130/52.5	S1, Z2, X1	Lignite	Neogene	Dai et al. [32]
Dazhai Mine/Yunnan	1.05–640/56.0	n.d.	Lignite	Miocene	Hu et al. [33]
Yanshan coalfield/Yunnan	167–167/167	M9	Low volatile bituminous	Late Permian	Liu et al. [34]
Yanshan coalfield/Yunnan	111–178/153	M9	Semi-anthracite	Late Permian	Dai et al. [35]
Luquan/Yunnan	22.6–47.2/34.1	Thin coal bed	Liptobiolith	Middle Devonian	Dai et al. [36]
Guiding coalfield/Guizhou	192–264/229	M1, M3	High to low volatile bituminous	Late Permian	Liu et al. [34]
Guiding coalfield/Guizhou	67.9–288/200	M1, M3	Bituminous	Late Permian	Dai et al. [30]
Puan coalfield/Guizhou	2.54–133/32.4	1, 2, 8, 11, 17	Semianthracite	Late Permian	Yang [37]
Zhijin coalfield/Guizhou	n.d./49.6	9	Low volatile bituminous	Late Permian	Dai et al. [38]
Heshan coalfield/Guangxi	10.2–176/73.8	3A, 3B, 3C, 4A, 4B	Low volatile bituminous	Late Permian	Shao et al. [39]
Heshan coalfield/Guangxi	12.0–326/69.0	#3, #4	Low volatile bituminous	Late Permian	Zeng et al. [40]
Heshan coalfield/Guangxi	10.2–176/73.8	2, 3, 4	Low volatile bituminous	Late Permian	Shao et al. [41]
Heshan coalfield/Guangxi	12.4–111/56.1	3U, 3L, 4U, 4L	Low volatile bituminous	Late Permian	Dai et al. [42]
Yishan coalfield/Guangxi	35.0–123/71.7	K3, K6, K7	Semianthracite and low volatile bituminous	Late Permian	Dai et al. [43]
Sawabuqi Mine/Xinjiang	210–520/365	M1, M9, M13	Lignite	Early Jurassic	Liu et al. [44]
Yili coalfield/Xinjiang	1.76–7207/320	12, 11, 10	High volatile bituminous coal	Early-Middle Jurassic	Dai et al. [45]
Yili coalfield/Xinjiang	6.89–724/147	11, 12	Lignite	Early Jurassic	Yang et al. [46]
Shengli coalfield/Inner Mongolia	0.42–148/25.9	6–1	Lignite	Early Cretaceous	Qi et al. [47]
Shiping Mine/Sichuan	0.75–155/39.8	C19, C25	Bituminous	Late Permian	Luo and Zheng [48]
Moxinpo coalfield/Chongqing	295–476/376	K1	Medium volatile bituminous	Late Permian	Dai et al. [49]

[a] On whole coal basis, and means are the arithmetical averages; [b] n.d.—no data.

3. Spatial and Temporal Distribution of Uranium in Chinese Coals

3.1. Uranium in Coals from Different Coalfields in China

To illustrate the lateral distribution of uranium in coals from different coalfields in China, all of the collected data were classified according to its coalfields. The five uranium enrichment categories in coal, i.e., abnormal enrichment, significant enrichment, enrichment, slight enrichment, and normal level, were filled in different colors on a China map (Figure 1).

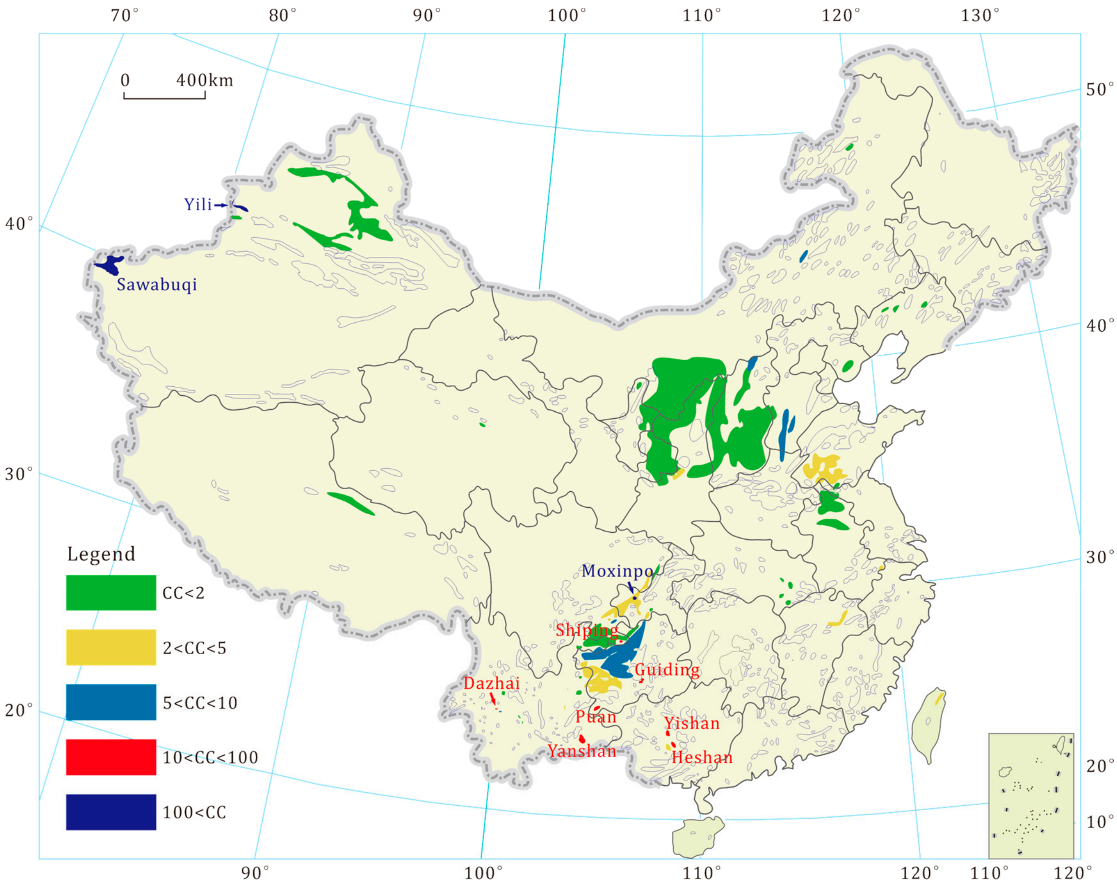

Figure 1. Distribution of uranium in different coalfields of China. Concentration coefficients (CC): ratio of uranium concentration in targeted coal to the average of common Chinese coals (2.43 mg/kg, Dai et al. [18]). Reproduced with permission from Dai et al. [50]; published by Elsevier Science, 2014.

As can be seen from Figure 1, the available data of uranium concentration in coals are limited to few coalfields stretching from the northern China (i.e., Inner Mongolia, Hebei, and Shanxi Provinces) to the southwestern China (Chongqing, Guizhou, and Yunnan Provinces). Almost all of the uranium-rich coals exist in southwestern and northwestern China (Figure 1), i.e., Yunnan, Guizhou, Guangxi, Chongqing, and Xinjiang Provinces.

The enrichment of uranium in coal might result from the weathering of source rocks, volcanic ashes (just the felsic or intermediate volcanic ashes [51]), magmatic intrusion, marine water influence, groundwater, hydrothermal fluids, organic matter, paleoclimate, and geologic conditions of coal-accumulating basins [9]. Note that the factors listed above have different contribution for uranium enrichment. The significant enrichment of uranium in coals was usually caused by exfiltrational and infiltrational solutions, as reported by Seredin and Finkelman [52] and Dai et al. [17]. However, other factors could only cause a slight enrichment of uranium in coals. Groundwater or hydrothermal leaching on the intra-seam non-coal partings or roofs could also lead to the enrichment in coal. The uranium in leachates could then be re-deposited in the underlying organic matter of coals [53–55].

If the prerequisites of sources, pathways, and sinks were occasionally satisfied, and combined with a proper paleoclimate and tectonics condition, the enrichment of uranium could be achieved in a coal (Figure 2). The uranium-rich coals in China are located at the margin of South China Block and Tarim Block, where the source and sink of uranium might simultaneously occur.

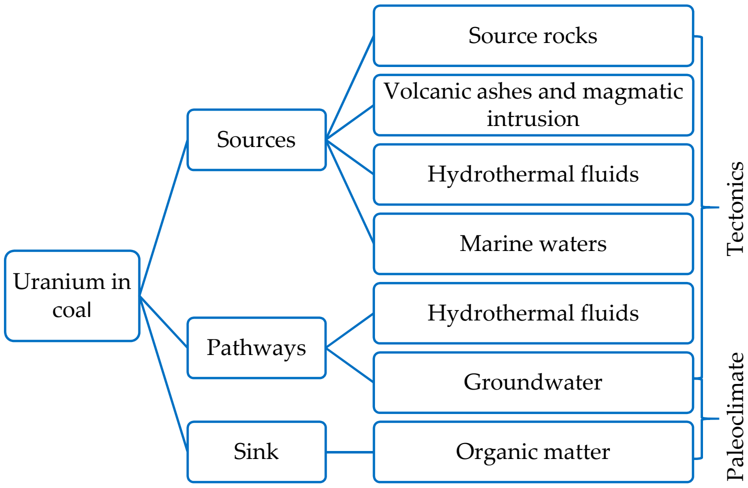

Figure 2. Genetic factors for uranium enrichment in Chinese coals. The schematic diagram is reorganized from Chen et al. [9].

3.2. Variation of Uranium in Certain Coalbed

The vertical variation of trace elements in coalbeds might be an implication for the modes of occurrence, time of emplacement, and origin of elements, and even depositional environment and coalification processes of coal [42,56–58]. Generally, the roof and floor rocks, and parting of coalbed, or coal benches close to the surrounding rock are favorable sites for uranium precipitation, such as the Yanzhou coal [59] and Zaozhuang coal [60] in Shandong Province, the Xishan coal [61] and Antaibao coal [62,63] in Shanxi Province, the Heshan coal [40,42] and Fusui coal [64] in Guangxi Province, and the Leping coal in Jiangxi Province [65].

The vertical profiles of both ash yield and uranium concentration of the Nos. 5 and 6 coals (the Late Permian Longtan Formation) from the Nantong coalfield in Chongqing, southwestern China, are presented in Figure 3. The thicknesses of the two coalbeds are 0.91 m and 0.72 m, respectively. Sandstone and mudstone compose the roofs of the Nos. 5 and 6 coals, respectively. Both of the floor rocks are mudstones. The average contents of volatile matter are 16.82% and 16.23%, suggesting a low volatile bituminous rank according to ASTM Standard D388-12 [66]. Uranium concentrations in both coals indicate slightly increases in bench coal samples adjacent to the roof and floor rocks (Figure 3). Moreover, uranium concentration shows a slightly discrepant trend to the ash yield, indicating a probable organic affinity of uranium and an alternative origin of uranium besides the detrital input. The sharp increase of uranium concentration in the NT-5-4 might be related to the alkaline volcanic ash fall during coal accumulation.

3.3. Uranium in Coals of Different Coal-Forming Periods

Yang et al. [27] ordered the weighted uranium concentrations in coal of six coal-forming periods in China as follows: the Late Permian, Paleogene and Neogene, Late Triassic, Late Carboniferous and Early Permian, Late Jurassic and Early Cretaceous, and Early and Middle Jurassic. However, Huang et al. [13] stated that the variation of an element with geologic times was not as useful as the element's variation by coal region and coalfield.

The data with information about coal-forming periods in the supplementary material of Chen et al. [9] were reorganized. Samples from the Early Carboniferous are unavailable. The relationship of uranium concentration with coal-forming periods is shown in Figure 4. The uranium concentrations present great ranges and show abnormal distribution in coals of individual age, except for the Late Triassic coal (Figure 4). Therefore, the fiftieth percentile was chosen to represent the average uranium concentration of this coal-forming period. The mean uranium concentrations decrease in the following order, i.e., the Paleogene and Neogene > Late Permian > Late Triassic > Late Carboniferous and Early Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic.

In fact, except for the Paleogene and Neogene coals, coals of other five ages show small medians of uranium concentrations. The relationship between uranium concentration and coal-forming periods is just an alternative appearance of its relation to coal-ranks, because uranium is always strongly organically associated, especially for humic and fulvic acids in low rank coals.

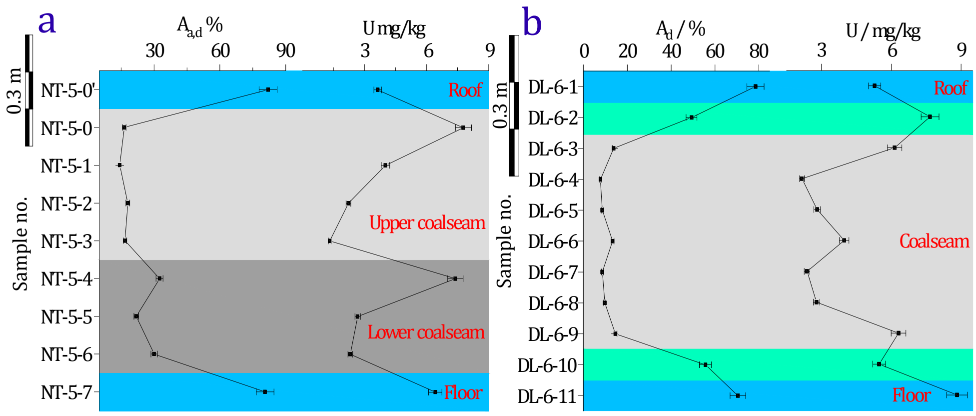

Figure 3. Vertical variation of ash yield and uranium in the Nos. 5 (**a**) and 6 (**b**) coals from the Nantong coalfield in Chongqing, southwestern China. Panel b was cited from Chen et al. [67].

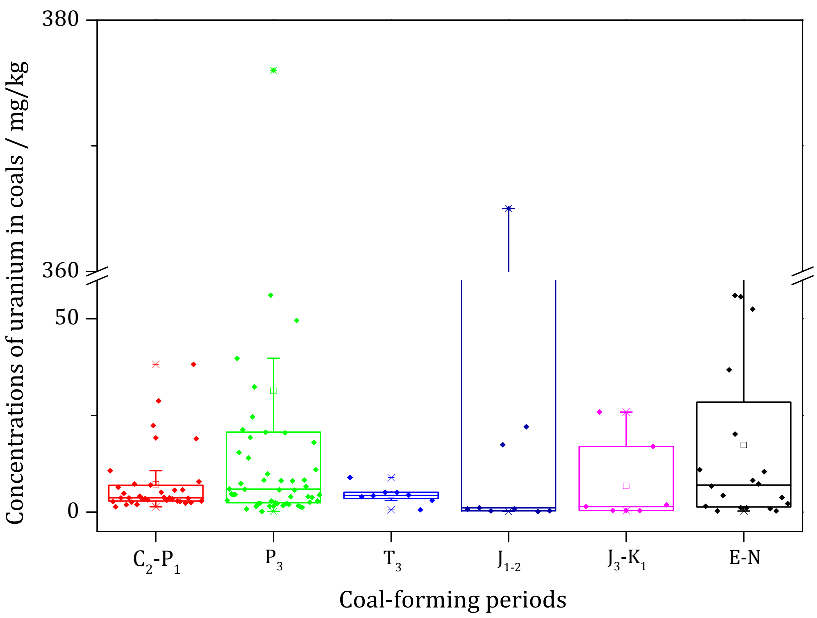

Figure 4. Variation of uranium concentrations in Chinese coals of different coal-forming periods. C_2-P_1: Late Carboniferous and Early Permian; P_3: Late Permian; T_3: Late Triassic; J_{1-2}: Early and Middle Jurassic; J_3-K_1: Late Jurassic and Early Cretaceous; E-N: Paleogene and Neogene.

4. Modes of Occurrence of Uranium in Chinese Coals

The modes of occurrence of uranium in coal is one key factor for extraction, partition, removal, and fate of uranium during coal beneficiation and utilization. In low rank coals, uranium is generally organically associated [13,68–71]. Uranium-bearing minerals, i.e., pitchblende [45,72–75], coffinite [30,45,49,72,73,76], and brannerite [30,76], were identified in coals. However, uranium minerals are always presented in a finely dispersed form, which makes the discrimination between minerals and organic associations very difficult [72].

With respect to the Chinese coals, uranium is associated with: (1) organic matter [30,34,36,39,42,77–83], (2) as physical adsorption by pores and pelitic components [46,84], (3) silicates [34,36,37,47,65,66,77,82,85–93], (4) phosphate minerals [94–96], (5) uranium minerals [30,34,45], and (6) mixed affinity to both organic and inorganic matter [19,35,97]. Overall, the modes of occurrence of uranium in Chinese coals was deduced indirectly, i.e., correlation with ash yield and major element oxide, and sequential chemical extraction, with only a few from direct evidences of SEM-EDS (Scanning electron microscope combined with an X-ray energy dispersive spectrometer) identification. The organic matter and silicates are primarily the hosts of uranium in Chinese coals.

5. Relation of Uranium to Coal Ranks

Uranium in coal is mainly associated with organic matter in low rank coals [13,98]. The decarboxylation from low rank coal to high rank one is an important factor in the mobilization and enrichment of many elements during coalification [99]. Almost all of the uranium-rich coals are lignite [73,75].

Owing to the heterogeneity of source area, facial differences, varied influence of syn-depositional volcanism, the direct comparison of the uranium concentrations in coals of different ranks does not reach correct conclusions [2]. However, the relationship of uranium concentration to coal ranks and coal-forming periods represent the geochemical habit of uranium—organic affinity. During the coal rank elevation from lignite to anthracite, the active functional groups of organic matter lost [100], correspondingly resulted in the release of organically associated elements [101–103], including uranium. Geologically younger and lower ranks coal tends to be enriched in uranium. The subbituminous coal and lignite in China are generally enriched in uranium (Figure 5).

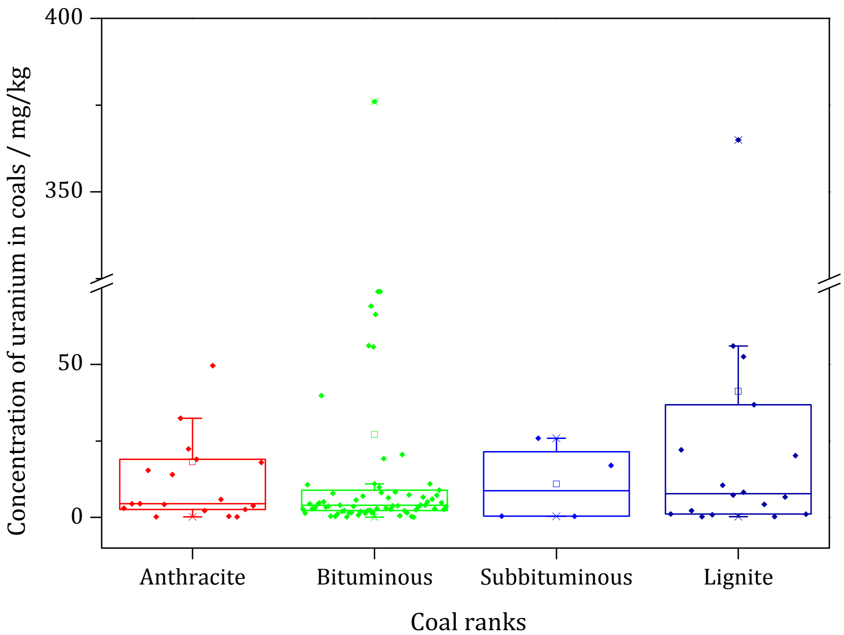

Figure 5. Variation of uranium concentrations in Chinese coals of different ranks.

6. Conclusions

The abundance of uranium in common Chinese coal is 2.43 mg/kg, which is comparable to that of world coals. Significant uranium enrichment only occurs in limited coals from certain local regions of southwestern and northwestern China, i.e., Shanxi, Yunnan, Guizhou, Guangxi, Xinjiang, Inner Mongolia, Sichuan, and Chongqing Provinces, resulting from the satisfaction of source and sink of uranium at the margins of South China and Tarim Blocks.

The roof and floor rocks, and partings of coalbed, or coal benches close to the surrounding rock are favorable sites for uranium in one coalbed. Regarding to the uranium concentrations in coals of different ages, it decrease in this order, e.g., Paleogene and Neogene > Late Permian > Late Triassic > Late Carboniferous and Early Permian > Late Jurassic and Early Cretaceous > Early and Middle Jurassic.

Uranium in Chinese coals is mainly associated with organic matter and silicate minerals. Both the relations of uranium concentration to coal ranks and coal-forming periods actually represent the geochemical habit of uranium—organic affinity. Therefore, the younger and lower the ranks of coal, the more uranium might occur. Correspondingly, the subbituminous coal and lignite in China are generally enriched in uranium.

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