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Distribution and modes of occurrence of uranium in coals of Eastern Yunnan, China

Xin Wang[1] · Qiyan Feng[2] · Qingjun Meng[2] · Fei Liu[1] · Qihang Cao[1] · Guijian Liu[3]

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Abstract Uranium is an environmentally hazardous element, and is commonly present at trace levels (2.4 µg/g for world coals) in coal deposits. However, selected coal deposits could be highly enriched in uranium. In this study, 15 coal samples were collected from Eastern Yunnan coal deposits, China, aiming to characterize the distribution and the occurrence of uranium in those coals. In studied samples, uranium content varied from 0.36 to 8.28 µg/g, with an average value of 3.76 µg/g. Generally, uranium content in coals from northern coal mines (3.02 ± 2.44 µg/g, n = 5) were lower than it in southern coal mines (4.13 ± 2.30 µg/g, n = 10). Uranium in coal samples showed no obvious correlation with total sulfur, whereas was positively correlated with ash yield. The results of sequential chemical extraction procedure confirm that organic-bound is the dominant occurrence of uranium. The slight enrichment of uranium in studied coals was probably attributed to sedimentation processes, hydrological conditions and tectonic structure of the coal deposits.

Keywords Coal · Uranium · Distribution · Eastern Yunnan · China

1 Introduction

Uranium (U) is a naturally radioactive element. Its abundance in Earth's crust is calculated to be 2.5 µg/g and 0.93 µg/g for upper and lower curst, respectively (Wedepohl 1995). High levels of uranium and its radioactive daughters have been observed in several coal deposits (Ahmed et al. 2020; Duan et al. 2018). For example, the Drama lignite deposit in Northern Greece (Megalovasilis et al. 2013) and Yunnan Late Permian coal deposit, China, have been observed to have uranium content exceeding industrial uranium grade (200 mg/g) (Huang et al. 2012). High-U coals have attracted much attention for industrial utilization (Dai et al. 2016, 2017, 2018). At the beginning of the 'nuclear era' (1960s), intensive efforts had been made to explore U-bearing coals as a uranium source. However, with the discovery of other high-quality raw uranium resources, the interest in U-bearing coals diminished. Nevertheless, U-bearing coals and their combustion products can still potentially be future unconventional nuclear sources (Lauer et al. 2017). During coal combustion, uranium can be concentrated by several folds in the coal combustion products such as slag and fly ash (Li et al. 2020; Kumar et al. 2020; Duan et al. 2019; Chen et al. 2017a, b). At present, uranium in coals is mainly treated as a potentially radioactive contaminate in environment (Talan et al. 2020; Chen 2014; Fujak et al. 2013).

Huang et al. (2012), Dai et al. (2012) and Chen et al. (2017a, b) have reviewed the abundance and distribution of uranium in Chinese coals. Chinese U-rich coal deposits are marginally distributed and usually have low reserves (Yang 2007). Mean uranium content in Chinese coals is estimated

✉ Guijian Liu
lgj@ustc.edu.cn

[1] College of Life Sciences, Huaibei Normal University, Huaibei 235000, China

[2] School of Environment Science and Spatial Informatics, China University of Mining and Technology, Xuzhou 221116, China

[3] CAS Key Laboratory of Crust-Mantle Materials and the Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China

to be 2.43 μg/g (Dai et al. 2012), comparable to its abundance in world coals (2.4 μg/g, Ketris and Yudovich 2009). Uranium is found to be mainly associated with organic matter in low rank coals, while with inorganic minerals in high rank coals (Finkelman et al. 2018).

The coal deposits in Eastern Yunnan, China, have been identified to enrich high levels of uranium (Huang et al. 2012; Wang et al. 2015). However, the enrichment magnitude and extent of uranium are not well studied. In this study, uranium in coal deposits of this area is re-analyzed. This study focuses on the distribution and modes of occurrence of uranium, and the geological controls on the uranium enrichment. This work represents an effort to generalize an extensive knowledge on the geochemistry of radioactive elements in coals.

2 Geological setting and sampling

Eastern Yunnan Province is located at the southern part of the Yangzi craton. This region is composed of intermediate massif and suture zones, and has a quite complicated geological setting due to the existence of extensive faulting (Li et al. 2011). The frequent eruptions of volcanic ashes during geological ages and the alternating marine and terrestrial facies could have led to the geochemical and mineralogical anomalies of coal (Miao 2013). In addition to coal deposits, this area is also abundant in metal resources, such as gold, antimony, germanium, gallium, arsenic, mercury, and thallium. Main coal-forming periods in Eastern Yunnan are Early Carboniferous, Late Permian, Late Triassic and Neogene (Tao et al. 2011). Different ranks of coals, from bituminous to anthracite, have been developed. The frequent eruptions of volcanos during coal-forming periods and the alternated marine and terrestrial facies have been suggested to be the main reasons causing the geochemical and mineralogical anomalies of regional coals (Miao 2013).

In order to better represent the uranium distribution with respect to coal resources and ranks, 15 coal samples from 14 active coal mines (Maoergou, Fukanglu, Kelang, Changsheng, Xujiayuan, Shizhuang, Chaoyang, Meitanchong, Xingying, Xiao chongchong, Tuobai, Xiaoheiqing, Xiaoshishan and Gongqing) in Eastern Yunnan were sampled (Fig. 1 and Table 1) according to Chinese Standard for Sampling of Coal Seam (GB/T 482–2008). The Zhaotong region including sample ZT-1 – ZT-5 was in the north of our sampling area, the Qujing region including sample QJ-1 – QJ-7 and Kunming region, Honghezhou region were in the south of our sampling area. Upon collection, all samples were stored in polyethylene bags to preclude contamination and weathering.

3 Analyses

Coal samples were air-dried, crushed and passed through a 100-mesh sieve before analysis. Proximate analysis was performed according to Chinese standards (GB/T 212–2008; GB/T 476–2001). The total sulfur and different sulfur forms were determined following Chinese standards (GB/T 214–2007) respectively. Powdered samples were digested using mixed acids before uranium and thorium concentration measurement by inductively coupled-plasma mass spectrometry (ICP-MS). The detection limit of the ICP-MS method for uranium is 0.01 μg/g, and the standard deviation for replicate sample digests is less than 10%.

Sequential chemical extraction was one of the most common quantitative methods adopted to determine the modes of occurrence of trace elements in geological and environmental research. The sequential extraction procedure was originally developed to analyze soil samples (Tessier et al. 1979). Afterwards, it had been widely used to study the chemical activity and speciation of diverse materials (Pan et al. 2019). Variants of sequential extraction like BCR-701, the SCEP reported by Dai et al. (2012), and sequential extractions with a float-sink step as described by Davidson (2000) and Huggins FE. (2002) were designed for scientific studies involving the contraposition of multiple elements. Thus, a six-step extraction procedure (shown in Fig. 2) was designed and applied to divide the chemical species into the following six types: water-leachable, ion-exchangeable, organic-bound, carbonate-bound, silicate-bound, and sulfide-bound.

4 Results and discussion

4.1 Coal characterization

The ash yields of studied coals ranged from 4.71% to 32.12% (mean = 17.59%, n = 15), and total sulfur content varied between 0.25% and 4.15% (mean = 1.67%, n = 15), majority of the samples were classified as low ash and low to medium sulfur coal. The volatile matter values were generally low for most coals varying from 5.67% to 23.28%, with the exception of one sample (KM) of the value up to 56.34%. According to the proximate analysis, the studied coal samples were classified into different ranks: lignite, bituminous and anthracite (Table 1).

4.2 Distribution of uranium in coals

The highest uranium content was observed in Xiaochongchong mine (ML-1) being 8.28 μg/g, whereas the lowest uranium content (0.36 μg/g) was found in Changsheng

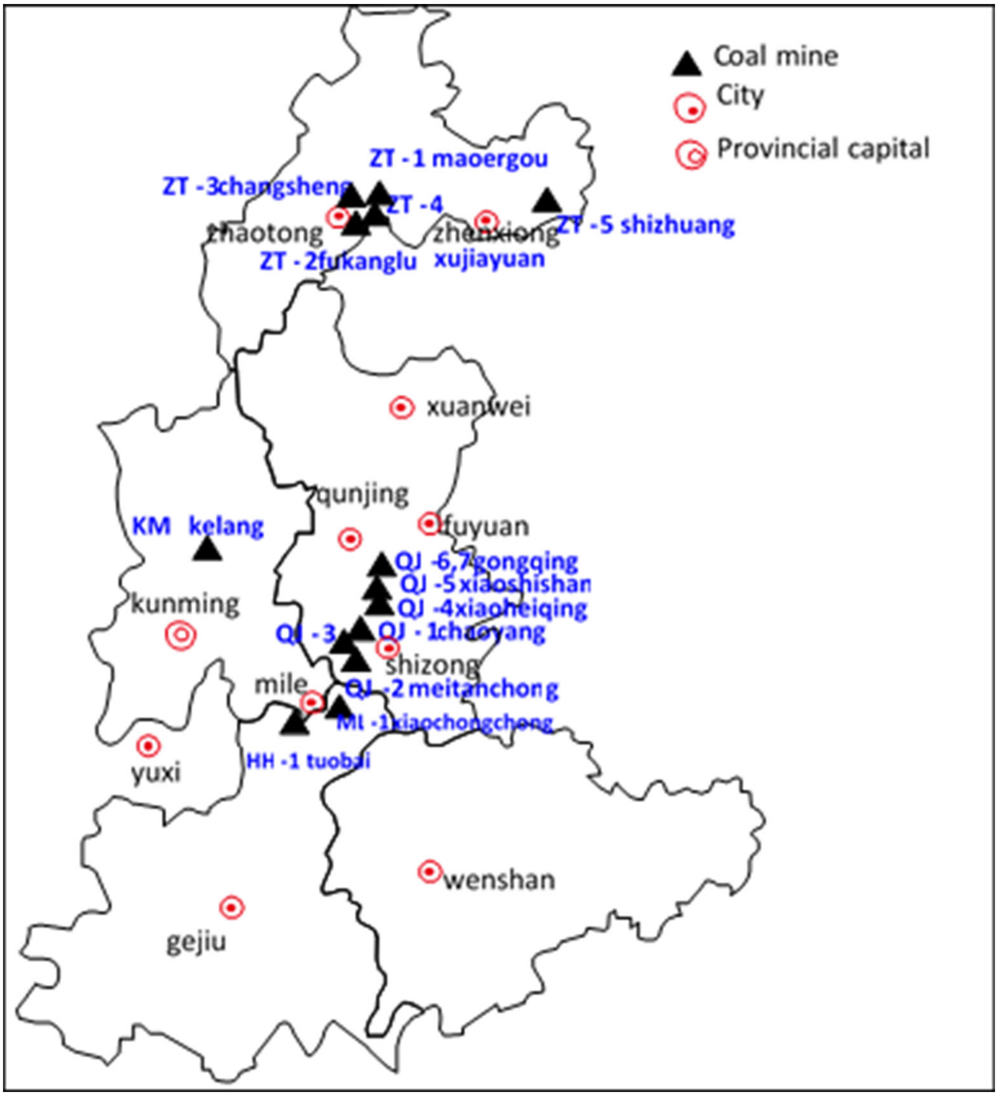


Fig. 1 Sampling locations of the coalmines in Eastern Yunnan, China (ZT: zhaotong, HH: honghezhou, KM: kunming, ML: mile, QJ: qujing)

mine (ZT-3) (Table 1, Fig. 1). The average uranium content in studied coals was 3.76 μg/g, slightly higher than that in Chinese coals (2.71 μg/g) (Ren et al. 1999) and world coals (2.4 μg/g) (Ketris and Yudovich 2009), but significantly lower than that in coals from southwestern China (23.23 μg/g) (Huang et al. 2012). As can be seen from Fig. 1, coals from northern coal mines (i.e., Zhaotong region, ZT-1 – ZT-5) had a mean uranium value of 3.02 μg/g, which was lower than those from southern mines (i.e., Qujing region, QJ-1 – QJ-7; Kunming, KM; Mile, ML-1; Honghezhou, HH-1) with a mean uranium content of 4.13 μg/g. This regional difference in uranium content might be due to the difference in coal ranks. Coal samples from northern mines were anthracite coals in rank, in contrast to the samples from southern mines were lignite and bituminous(GB/T 5751–2009). It had been found that uranium was prone to affiliate with humic acids in coal precursors, which was abundant in low rank coals and depleted in high rank coals (Zhang et al. 2018).

In studied coals, lignite from Kelang mine had a uranium content of 7.32 μg/g, significantly higher than the mean value of uranium in the world lignite (1.51 μg/g) (Bouska and Pesek 1999). The average uranium content in nine bituminous samples (3.78 μg/g) was slightly higher than that in the rest five anthracites (3.02 μg/g). Boxplots provided a quick, visual summary about the distribution of uranium in bituminous and anthracites in the Fig. 3. It showed that the median of uranium in the coals of two ranks were nearly eaual and were all higher than the mean value in world coals (Ketris and Yudovich 2009). Therefore, the disribution of uranium in studied coals of different ranks showed no apparent patterns.

The concentration coefficient was defined as the ratio of element concentration in common coals to that in the world

Table 1 Proximate analysis (%), sulfur (%), uranium (μg/g) and radioactive element ratio (U/Th) of the Eastern Yunnan coals

Sample	Coal mine	Coal bed	Coal rank	GA*	Proximate analysis			$S_{t,d}$	$S_{p,d}$	$S_{s,d}$	$S_{o,d}$	U	U/Th
					M_{ad}	A_d	V_{daf}						
ZT-1	Maoergou	M4	Anthracite	CWF*	1.20	29.24	12.66	4.00	3.65	0.18	0.17	5.50	0.57
ZT-2	Fukanglu	M2	Anthracite	CWF*	0.70	12.72	10.64	1.95	1.15	0.04	0.76	5.00	0.85
KM	Kelang	M8	Lignite	NEO*	9.97	15.46	56.34	4.05	0.94	0.11	3.00	7.32	2.70
ZT-3	Changsheng	C5	Anthracite	CWF*	0.61	4.71	5.67	0.83				0.36	0.34
ZT-4	Xujiayuan	C5	Anthracite	CWF*	0.47	9.47	7.00	1.68	1.40	0.03	0.25	0.54	0.28
ZT-5	Shizhuang	mix	Anthracite	PLF*	0.82	32.12	12.42	3.22	2.83	0.06	0.33	3.72	0.49
QJ-1	Chaoyang	C9	Bituminous	PXF*	0.58	11.23	20.07	0.19				3.14	0.43
QJ-2	Meitanchong	C9	Bituminous	PLF*	0.75	10.03	19.90	0.74				3.86	0.58
QJ-3	Xingying	C9	Bituminous	PLF*	0.62	14.04	19.96	0.46				1.54	0.45
ML-1	Xiaochong chong	C3	Bituminous	PLF*	0.86	27.03	22.91	4.15	3.41	0.30	0.44	8.28	0.95
HH-1	Tuobai	C10	Bituminous	PLF*	0.55	22.81	17.99	0.48				4.76	0.57
QJ-4	Xiaoheiqing	C21	Bituminous	PLF*	0.51	18.88	16.21	1.74	1.21	0.04	0.49	1.64	0.28
QJ-5	Xiaoshishan	C15	Bituminous	PLF*	0.81	25.39	23.28	0.31				4.56	0.50
QJ-6	Gongqing	C16	Bituminous	PLF*	0.69	10.85	22.64	0.25				4.48	0.60
QJ-7	Gongqing	C18	Bituminous	PLF*	0.75	19.81	20.48	0.93				1.74	0.46
YN*	Eastern Yunnan coalfield											3.76[a]	
	North-eastern Yunnan coal											27.7[b]	
WC*			All									2.4[c]	
			Lignite									2.9[c]	
			Anthracite									1.9[c]	

*GA** Geological age, *M* moisture, *A* ash yield, *V* volatile matter, S_t total sulfur, S_p pyritic sulfur, S_s Sulfate sulfur, S_o organic sulfur, *ad* air-dry basis, *d* dry basis, *daf* dry and ash-free basis, *CWF** Carboniferous Wanshoushan Formation, *NEO** Neogene, *PLP** Permian Longtan Formation, *PXP** Permian Xuanwei Formation, *YN** Yunnan coal

[a]From Huang W.H., [b]From Xi W.S., WC* world coal, [c]From Ketris and Yudovich

coals (Avino et al. 2003). Figure 4 showed that only ZT-3 and ZT-4 had concentration coefficients (CC) less than 0.5, indicating a depletion of uranium in these coals (Dai et al. 2014b). Uranium in other samples, including ZT-1, ZT-2, KM and ML-1, was slightly enriched with CCs of 2–5. Uranium content in the remaining coals was close to the average of world coals (0.5 < CC < 2).

4.3 Modes of occurrence of uranium in coals

With respect to the sulfur content in coals (GB/T 15, 224.2–2010), the studied coal samples can be classified into four groups: high-sulfur (> 3%), medium-sulfur (1.5%–2%), low-sulfur (0.5%–1%) and super low-sulfur coal (< 0.5%). The studied coals were characterized as 27% high-sulfur coal, 20% medium-sulfur coal, 20% low-sulfur coal and 33% super low-sulfur coal. Different sulfur forms were determined in medium and high sulfur coal samples (Table 1). High portion of pyritic sulfur (58%–91%) was observed in the coals except one sample (KM), of which 74% of sulfur was in organic form (Fig. 5).

Sulfur forms in coals throughout the world were highly variable, depending on various geologic conditions (Chou 1997). Previous researchers had carried out detailed studies on the distribution and occurrence of sulfur in various high-sulfur coals (Chou 2012; Dai et al. 2013a, b, 2014a, b). The geochemical anomalies, mineralogy, isotopic compositions, and petrology of high-organic-sulfur coals were also extensively reported (Dai et al. 2014a, b). Dai et al. (2008) studied a super high-organic-sulfur (SHOS) Late Permian coal from Yanshan Coalfield, southwestern China, which an evidence of volcanic ash components was found.

Figure 6 indicated that uranium content had no obvious relation with total sulfur in the coal samples ($n = 15$, $r = 0.43$, $p > 0.05$). However, uranium content in medium–high sulfur coal exhibits significant positive correlation with total sulfur as seen in Fig. 7 ($n = 7$, $r = 0.93$, $p < 0.01$). According to Table 1, sulfur in coals was dominantly pyritic sulfur except lignite coal (KM). Uranium sometimes created its own mineral forms on the surface of pyrite. Pyrites in such case act as a reducing agent. Previous data including 37 coal samples from other

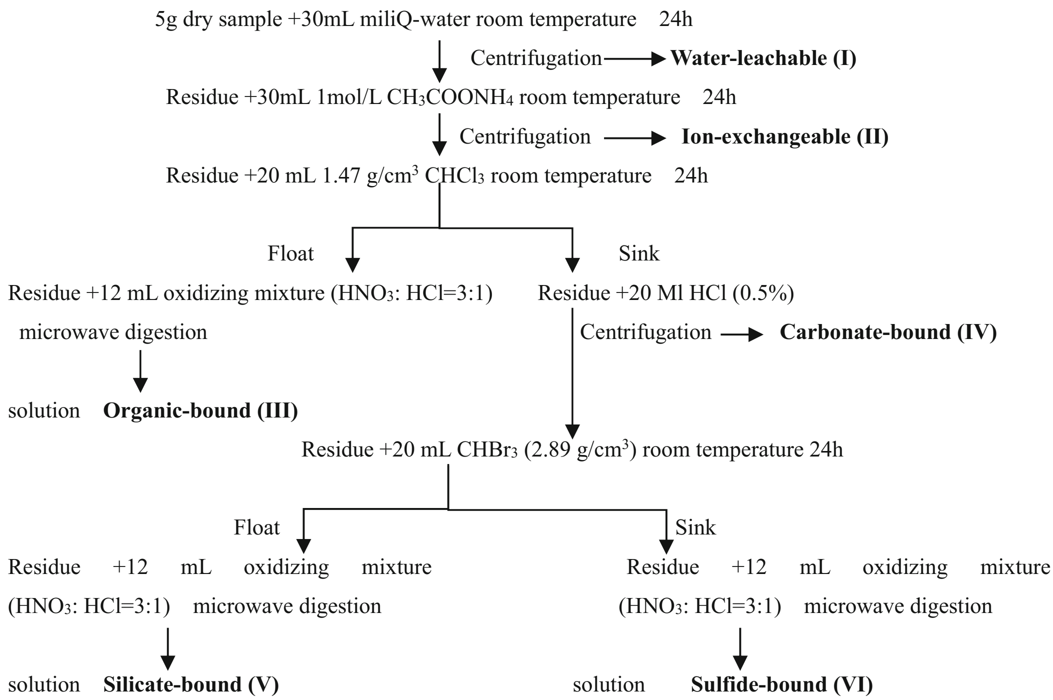


Fig. 2 Diagram of sequential chemical extraction procedure

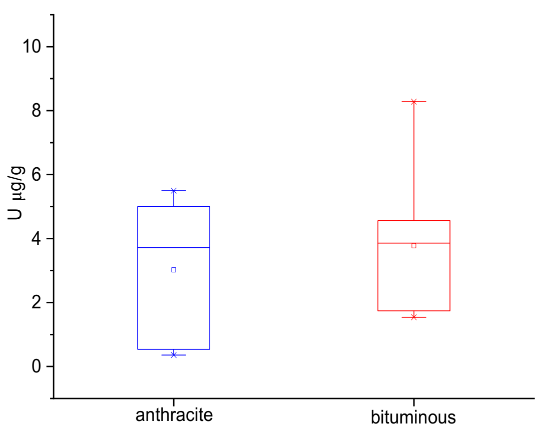


Fig. 3 Distribution of uranium in coals of different ranks in Eastern Yunnan

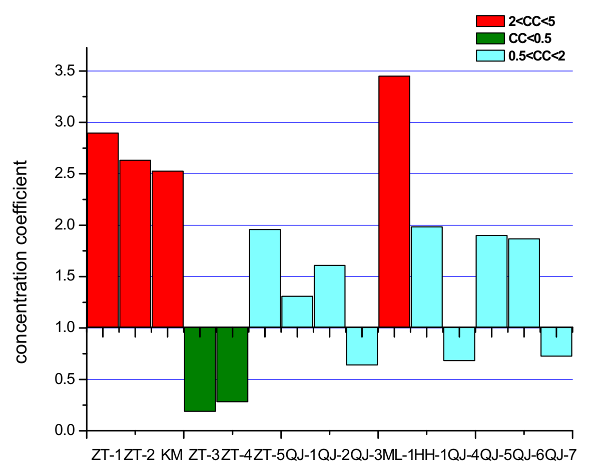


Fig. 4 Concentration coefficients (CC) of uranium in coals of Eastern Yunnan, normalized by average content in the world coals (Ketris and Yudovich, 2009)

coal mines in Yunnan Province were compiled together with samples in present study to analyze the relationship between uranium and sulfur (Fig. 8 and Fig. 9) (Tang 2019; Duan 2017; Zhao 2017). It was shown in Fig. 8 that uranium content had no significant relation with total sulfur in coals ($n = 52$, $r = 0.51$, $p < 0.01$). Meanwhile, results shown in Fig. 9 indicated that uranium was not strongly correlated with sulfur in compiled medium–high sulfur coals ($n = 39$, $r = 0.58$, $p < 0.01$) as it found in collected samples of this study ($n = 7$, $r = 0.93$, $p < 0.01$). Therein, 21 coal samples from Ganhe mine were high organic sulfur coals (Zhao 2017), while sulfur in the collected samples of this study was dominantly pyritic sulfur. Thus correlation of sulfur and uranium content in medium–high sulfur coals was complicated.

4.4 Association of uranium with ash yield

The relationship between trace elements and ash yield had been widely reported to deduce the modes of occurrences of selected elements (Dai et al. 2005; Finkelman 1994). It was generally considered that most trace elements in coal were associated with mineral matters (Gentzis and Goodarzi 1997). The ash yield of studied coal samples ranged

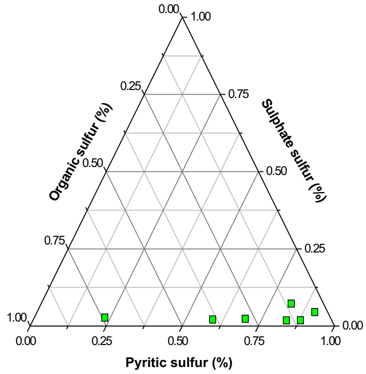


Fig. 5 Ternary diagram of different sulfur forms in medium–high coals

from 4.71% to 32.12%, with an average value of 17.59%. According to the classification on basis of ash yield (GB/T 15, 224.1–2010) (high ash, > 29%; medium ash, 16.01%–29%; low ash, 10.01%–16% and super low ash, < 10%), high-ash coal and super low-ash coal accounted for 13% each, medium-ash coal is 33%, the rest was low-ash coal with 40%. Vassilev et al. (1997) suggested that if there was a positive correlation between studied element in coal and ash yield, this element might be introduced into coal basin by exogenic geological process. Figure 10 showed that uranium in the studied coals had a positive correlation with ash yield ($n = 15$, $r = 0.53$, $p < 0.05$), thus uranium might be contributed to exogenic geological processes during the coal-forming period, probably due to the water input from the rocks adjacent to coal deposits.

However, on basis of the compiled coals samples, Fig. 11 showed that uranium in Yunnan coals was weakly correlated with ash yield ($n = 52$, $r = 0.18$, $p > 0.05$). Therefore, modes of occurrence of uranium in coals were complicated (Finkelman et al. 2018).

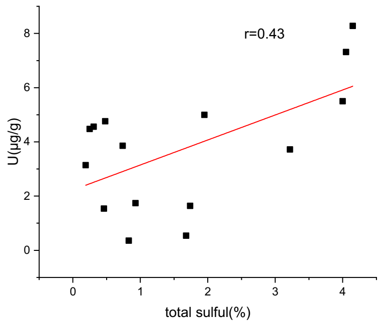


Fig. 6 Correlation between U content and total sulfur in present study

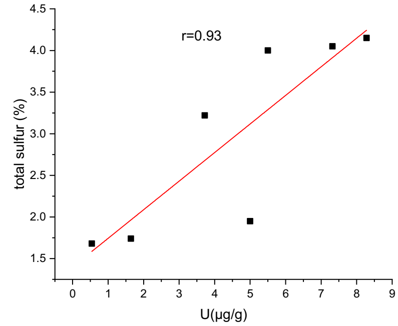


Fig. 7 Variation of the U contents with total sulfur in medium–high sulfur coals

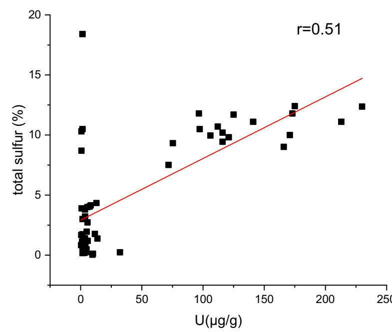


Fig. 8 Correlation between U content and total sulfur in other coals from Yunnan

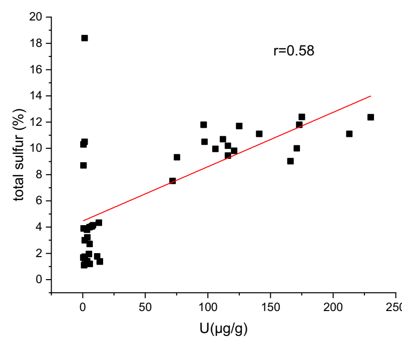


Fig. 9 Correlation between U contents with total sulfur in medium–high sulfur coals from other coal mines in Yunnan

4.5 Results of sequential chemical extraction procedures

To ascertain modes of occurrence of uranium in coals, sequential chemical extraction procedure (SCEP) was performed. These tests measured the fractional amount of U in various density fractions that was soluble in a series of increasingly aggressive solvents and acids. Figure 12 showed the experimental results for 7 samples. The uranium recovery during the SCEP for these coals was 99.17% to 105.07%.

The organic-bound U dominated in KM, QJ-6, ZT-1 and ZT-4, accounting for 72.02%, 51.97%, 38.5% and 37.36%

of the total U, respectively. The ion-exchangeable U was the second most abundant form, accounting for 17.91%, 16.40% and 20.76% in KM, QJ-6, ZT-4, respectively. However, the second most abundant form of ZT-1 was silicate-bound U, accounting for 26.15%. Unlike samples KM, QJ-6, ZT-1 and ZT-4, silicate-bound U dominated in QJ-3, QJ-7 and ZT-3, respectively accounting for 42.79%, 40.38% and 38.38%, respectively. Besides, organic-bound U was the second most abundant form in these coals.

Generally, the main form of uranium was organic-bound, followed by silicate-bound, of which the ratio was much lower than the predominant form. Other modes of uranium such as the water-leachable, ion-exchangeable, carbonate-bound, and sulfide-bound were also present however amounted to about 27% of the total uranium. In consistent with our findings, on basis of the results of SEM–EDX analysis and the sequential solvent extraction experiments, Dai et al. (2008) and Yang (2009) concluded that U in the M9 coal from Yanshan Coalfield (Eastern Yunnan) was mainly associated with silicate minerals and organic matter. In the same area, Liu et al. (2015) also found U was mainly dominated by organic association (occupying 78.2%). Besides, U in Yantang coals from Xuanwei was present as inorganic minerals as confirmed by SEM–EDX analysis (Shao et al. 2015).

Finkleman R.B. et al. (2018) stated that uranium was associated with the silicates (40%) and insoluble phases such as zircon (30%) in the bituminous coals, but bound to the organics (55%) in the low rank coals. Seredin and Finkelman (2008), Arbuzov SI (2012) and Dai et al. (2015) indicated that most of uranium in uranium-bearing coals around the world was bound to organic matter, while uranium in coals with low content below the crustal abundance (clarke) were mainly characterized by the mineral form of occurrence (Yudovich et al. 2002), which was consistent with the results of this study.

4.6 The genesis of uranium enrichment

Uranium was much more soluble than thorium in oxidizing surficial and ground waters, leading to a wide uranium

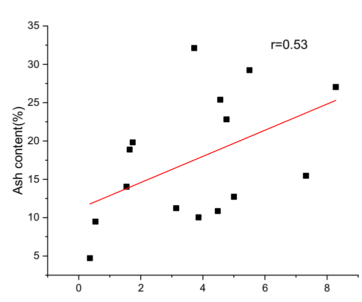


Fig. 10 Correlation between U content and ash yield in present study

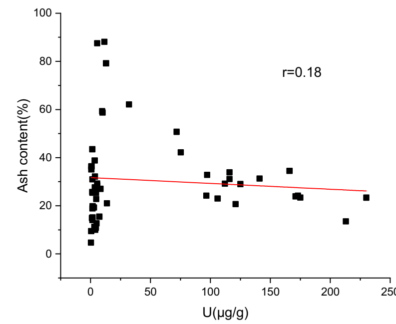
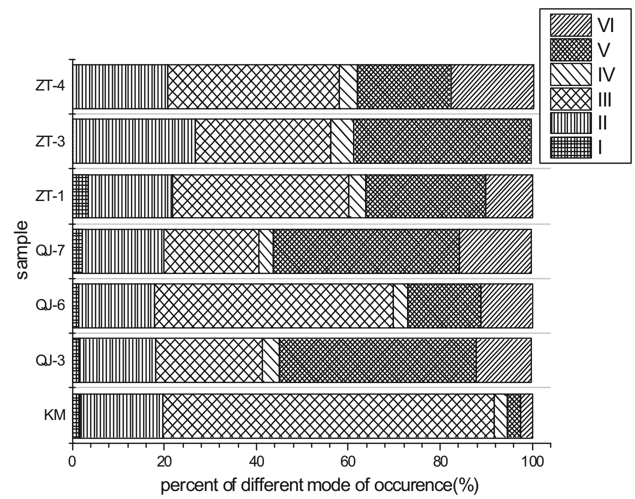


Fig. 11 Correlation between U content and ash yield in other coals from Yunnan



I: water-leachable, II: ion-exchangeable,

III: organic-bound, IV: carbonate-bound, V:

silicate-bound, VI: sulfide-bound.

Fig. 12 Results of sequential chemical extraction procedures

content range and, hence, a more variable U/Th value in sediments (Hwang and Moon. 2018). Jones and Manning (1994) had reported that the ratio of U/Th less than 0.75 indicated an oxidation environment, while between 0.75 and 1.25 demonstrated a reductive environment. The ratio of U/Th in most of the studied coals in Eastern Yunnan was less than 0.75 (Table 1), indicating that the sedimentary environment was dominated by oxidation environment. Under this condition, uranium was easily oxidized to uranyl ion that was readily soluble and thus active in supergenesis. The uranyl ion can be re-fractioned in surface or ground water (Liu and Cao 1987). Therefore, uranium in studied coals probably derived from water entering the peat swamp during peat formation stages and the ground water interact with the coal seams during/after coalification. However, two samples with U/Th ratios were higher than 0.75 (KM and ML-1). This might be due to the occurrence of sulfate reducing bacteria in the peat swamp, which could reduce the dissolved U^{6+} ions into un-dissolved U^{4+} irons (Wang et al. 2010). Uranium in water had been thought to be captured by the organic matter in the following ways (Huang et al. 2012). Firstly, the uranyl ion in the water was reduced by organic matter or by the H_2S gas generated by organic matter and precipitate in the peat swamp (Finkelman et al. 1994). Secondly, an exchange reaction could take place between H^+ ions in humic acids and uranyl ions in solution, and then precipitated in coal seams. Thirdly, the uranyl ion could be captured by coal through surface absorption or static electrical -absorption (Zhang 1988).

The enrichment of uranium in coals of Eastern Yunnan could be attributed to the coal forming environment of Eastern Yunnan, which was affected by the facture zone of Mile-Shizong in the southern part of the Yangzi land mass (Miao 2013). As a result, frequent eruptions of volcanic ashes during geological ages and the alternating marine and terrestrial facies could account for the geochemical and mineralogical anomalies of coal (Luo and Zhang 2013). Under these environmental conditions, the evaporation of the seawater probably resulted in the increase of uranium content in coal-bearing sediments.

5 Conclusions

Coals sampled from Eastern Yunnan, China, were slightly enriched in uranium compared to the mean values in China and the world coals. Uranium content in studied coals (lignite, bituminous and anthracites) varies from 0.36 to 8.28 μg/g. Mean uranium value of coals from northern coal mines is lower than those from southern mines. While the distribution pattern of uranium in studied coals of bituminous and anthracite was not apparent.

A good correlation between the uranium and sulfur content was observed in medium–high sulfur coals. In addition, uranium also showed a good correlation with ash yield, so the uranium could be introduced by exogenic geological process during the coal-forming period, probably due to the water input from the rocks adjacent to coal deposits. Results from sequential chemical extraction procedures showed that the organic-bound was the dominant mode of uranium. The ratio of U/Th in most of the studied coals indicated that the sedimentary environment was dominated by oxidation environment. Therefore, uranium in studied coals probably derived from water entering the peat swamp during peat formation stages and the ground water interact with the coal seams during/after coalification. Furthermore, the mechanisms of uranium enrichment in coals of Eastern Yunnan deserved further research.

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