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Radioactivity of Natural Nuclides (^{40}K, ^{238}U, ^{232}Th, ^{226}Ra) in Coals from Eastern Yunnan, China

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Abstract: The naturally occurring primordial radionuclides in coals might exhibit high radioactivity, and can be exported to the surrounding environment during coal combustion. In this study, nine coal samples were collected from eastern Yunnan coal deposits, China, aiming at characterizing the overall radioactivity of some typical nuclides (*i.e.*, ^{40}K, ^{238}U, ^{232}Th, ^{226}Ra) and assessing their ecological impact. The mean activity concentrations of ^{238}U, ^{232}Th, ^{40}K and ^{226}Ra are 63.86 (17.70–92.30 Bq·kg^{-1}), 23.76 (11.10–37.10 Bq·kg^{-1}), 96.84 (30.60–229.30 Bq·kg^{-1}) and 28.09 Bq·kg^{-1} (3.10–61.80 Bq·kg^{-1}), respectively. Both ^{238}U and ^{232}Th have high correlations with ash yield of coals, suggesting their inorganic origins. The overall environmental effect of natural radionuclides in studied coals is considered to be negligible, as assessed by related indexes (*i.e.*, radium equivalent activity, air-adsorbed dose rate, annual effective dose, and external hazard index). However, the absorbed dose rates values are higher than the average value of global primordial radiation and the Chinese natural gamma radiation dose rate.

Keywords: coal; radioactivity; nuclide; Yunnan; China

1. Introduction

Combustion of coals that are enriched in radioactive nuclides, particularly those of U series, possibly magnifies the background levels of radioactive nuclides in the surrounding environment and increases human exposure risks [1–4]. High concentrations of radioactive nuclides in coals have been reported worldwide. For example, Barber and Giorgio (1977) [5] found that the radioactive level of ^{226}Ra in a coal sample from Illinois, USA, could reach up to 1500 Bq·kg^{-1}. Measurement of ^{226}Ra on the lignite originated from Kotili of Xanthi Prefecture, Northern Greece, showed a radioactivity as high as 2600 Bq·kg^{-1}, which is more than one order of magnitude higher than that (110–260 Bq·kg^{-1}) of lignite commonly used in coal-fired power plants [6,7]. High levels of radioactive nuclides in Chinese coals are rarely reported. Jiang *et al.* (1989) [8] reported that the weighted means of ^{226}Ra, ^{232}Th and ^{40}K in Chinese coals are 36 (2–2300 Bq·kg^{-1}), 30 (4–110 Bq·kg^{-1}) and 104 Bq·kg^{-1} (5–750 Bq·kg^{-1}), respectively. Liu *et al.* (2007) [9] calculated a weighted mean of 79.5 ± 45, 73.9 ± 53, 40.3 ± 34 and 152.4 ± 21 Bq·kg^{-1} for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, respectively, from ~1000 samples collected from several Chinese coal mines. However, Dai *et al.* found that high concentration of U occurs in some coals from southwestern China, e.g., Linchang and Yanshan in Yunnan province [10,11] and Guiding in Guizhou province [12].

The coal deposits in eastern Yunnan province, China, have been identified to enrich high levels of natural radionuclides [13–17]. However, their spatial variation, modes of occurrences, and controlling factors are rarely illustrated. In this study, the radioactive nuclides in coal deposits of this area are reassessed, in an effort to generalize a basic knowledge on their potential environmental impacts.

2. Geological Setting

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 complicated geological setting due to extensive development of faulting and folding systems [18–21]. In addition to coal deposits, this area is also abundant in metal resources, such as gold, antimony, germanium, gallium, arsenic, mercury, and thallium. The coal-forming periods in eastern Yunnan include the Early Carboniferous, Late Permian, Late Triassic and Neogene [18,19]. The main coal-bearing strata are the Upper Carboniferous Wanshoushan Formation, the Lower Permian Longtan Formation and the Miocene Xiaolongtan Formation. Different ranks of coal, from gas coal (subbituminous coal) to anthracite, have been developed. The frequent eruptions of volcanoes 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 [22,23].

3. Sampling and Analysis

Nine coal samples from eight active coalmines (Maoergou, Kelang, Changsheng, Xujiayuan, Shizhuang, Xingying, Tuobai and Gongqing) in eastern Yunnan were sampled (Figure 1 and Table 1) according to the Chinese Standard Method GB/T 482-2008 [24]. The samples include anthracite from Zhaotong (coalmines of Maoergou, Changsheng, Xujiayuan and Shizhuang), lignite from Kunming (Kelang coalmine) and coking (bituminous) coals from Qujing (coal mines of Xingying and Gongqing) and Honghezhou (Tuobai coal mine) (Table 1), on basis of GB5751-86 [25]. Each sample was taken by cutting 10-cm wide and 10-cm deep into the coalface to represent the full height or a section of the coal. Upon collection, all samples were stored in polyethylene bags to preclude contamination and weathering.

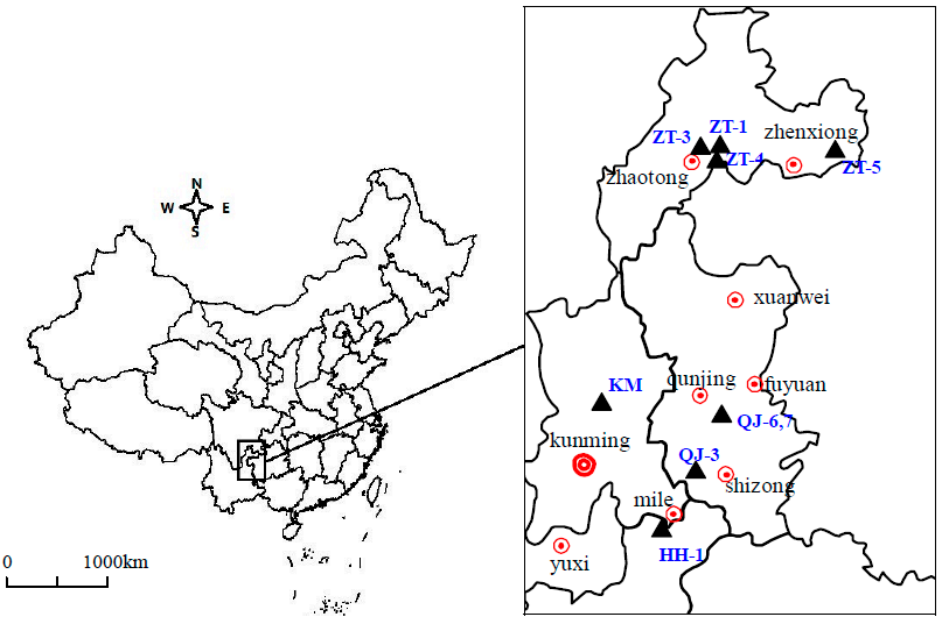

Figure 1. Sampling locations (filled triangle) of the coalmines in eastern Yunnan, China (HH: Honghezhou; KM: Kunming; QJ: Qujing; ZT: Zhaotong).

Table 1. Proximate analysis (%), sulfur (%), radioactive elements (μg/g) of the eastern Yunnan coal.

Sample ID	Coal Mine	Coal Rank	Proximate Analysis			$S_{t,d}$	U	Th	Ra
			M_{ad}	A_d	V_{daf}				
ZT-1	Maoergou	Anthracite	1.20	29.24	12.66	4.00	5.50	9.65	1.87
KM	Kelang	Lignite	9.97	15.46	56.34	1.95	7.32	2.71	2.49
ZT-3	Changsheng	Anthracite	0.61	4.71	5.67	0.83	0.36	1.06	0.12
ZT-4	Xujiayuan	Anthracite	0.47	9.47	7.00	1.68	0.54	1.93	0.18
ZT-5	Shizhuang	Anthracite	0.82	32.12	12.42	3.22	3.72	7.59	1.26
QJ-3	Xingying	Coking coal	0.62	14.04	19.96	0.46	1.54	3.46	0.52
HH-1	Tuobai	Coking coal	0.55	22.81	17.99	4.15	4.76	8.41	1.62
QJ-6	Gongqing	Coking coal	0.69	10.85	22.64	1.74	1.64	7.47	1.52
QJ-7		Coking coal	0.75	19.81	20.48	0.93	1.74	3.78	0.59

M, moisture; A, ash yield; V, volatile matter; S_t, total sulfur; ad, air-dry basis; d, dry basis; daf, dry and ash-free basis.

Coal samples were air-dried, pulverized using a jaw crusher and passed through a 100-mesh sieve before analysis. Proximate analysis (ash yield, volatile matter, moisture) and total sulfur were determined according to ASTM standards [26–29]. Powdered samples were digested using mixed acids (HNO_3:$HClO_4$:HF = 3:1:1) before U and Th measurement by inductively coupled-plasma mass spectrometry (ICP-MS). The method detection limit is 0.002 ng/mL for U and 0.003 ng/mL for Th. The uncertainty of elemental concentration is within ± 10% as evaluated by soil standard GBW 07114 [30] (GBW: national standard substance). The standard deviation for replicate sample digests is less than 10%.

Before radioactive measurement, powdered samples were dried in a temperature controlled furnace at 70 ± 1 °C for 24 h to remove moisture. After cooling down to room temperature, ~500 g of samples were sealed in gas-light, Rn impermeable cylindrical polyethylene containers (Inner Diameter = 40 mm). The samples were sealed and stored for 40 days in order to reach a radioactive equilibrium between ^{226}Ra and its daughters. A GEM60P4-83 high-purity Ge gamma ray spectrometer (ORTEC, Oak Ridge, TN, USA) shielded from background radiation using Pb bricks was employed to measure the most prominent γ-ray energies of the radionuclides in equilibrium with ^{238}U, ^{226}Ra, ^{40}K and ^{232}Th. The gamma spectrometry measurements were made with an energy resolution of 1.95 keV at the 1.332 Mev of ^{60}Co and the relative efficiency of 60%. The activity concentrations of ^{238}U was determined through its daughter product ^{234}Th, ^{226}Ra by its daughter products ^{214}Bi and ^{214}Pb and ^{232}Th by its daughter products ^{228}Ac and ^{208}Tl [31]. The activity concentration of ^{40}K was obtained from its single gamma ray lines of 1460.83 keV. The detector output was connected to a spectroscopy amplifier. The counting time for each sample and background was 10,000 s. The energy and absolute efficiency calibrations of the spectrometers were carried out using calibration sources with energy range covering the studied nuclides. The activity concentrations (*As*) for the natural radionuclides is calculated as:

$$A_S = \frac{N}{\varepsilon P_r M t} \quad (1)$$

where N is the net counting rate for a specific gamma line corrected for background, ε is the detector efficiency of a specific gamma ray, Pr is the absolute transition probability of gamma decay, M is the mass of the sample (kg), and t is counting time (s). The national standard reference materials (GBW04127 [32], GBW04325 [33] and GBW04326 [34]) were used for quality assurance, and the typically obtained values are within 5% of certified values. The statistic uncertainties of activity concentrations for radionuclides are estimated less then 2%.

4. Results and Discussion

4.1. Coal Basic Parameters

The studied coals have ash yields ranging from 4.71% to 32.12% (mean = 17.61%), and total sulfur ranging from 0.46% to 4.15% (mean = 2.11%), classifying as low ash, medium-high sulfur coals. The volatile matter values are generally low for most coal samples, varying from 5.67% to 23.28%, with the exception of one sample (KM) of up to 56.34%. According to the proximate analysis and on the basis of the Chinese Standard GB5751-86 [25], the studied coal samples are classified into different ranks: lignite, coking coal (bituminous coal) and anthracite (Table 1).

4.2. Concentrations of U and Th in Coals

The highest U concentration is observed in Kelang mine (KM) of 7.32 μg/g, whereas the lowest U concentration in Changsheng mine (ZT-3) of 0.36 μg/g (Figure 1). The average U concentration in studied coals is 3.01 μg/g, which is slightly higher than that in Chinese coals (2.71 μg/g) [35] and world coals (2.40 μg/g) [36], but is significantly lower than that in coals from Yunnan, China (7.08 μg/g) reported by Li *et al.* [37]. For Th in coals, the highest concentration is 9.65 μg/g in Maoergou mine (ZT-1), and the lowest concentration is 1.06 μg/g in Changsheng mine (ZT-3). The average Th concentration in studied coals is 5.12 μg/g, which is slightly higher than that in Chinese coals (4.93 μg/g) [35] and world coals (3.3 μg/g) [36], but is still lower than that in coals from Yunnan, China (5.83 μg/g) reported by Li *et al.* [37].

4.3. Levels of Radioactivity in Coals

The radioactivity of these radionuclides in studied coals is summarized in Table 2. The mean radioactivity of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are 63.86, 28.09, 23.76 and 96.84 Bq·kg^{-1}, respectively. The largest contributor to the total radioactivity is ^{40}K for these coal samples.

Table 2. Comparison of radioactivity (Bq·kg^{-1}) of nuclides in coals of eastern Yunnan, China with previous publications.

Sample ID	^{238}U	^{232}Th	^{226}Ra	^{40}K
ZT-1	84.90	36.00	46.40	165.00
KM	68.80	17.00	61.80	229.30
ZT-3	17.70	10.30	3.10	30.60
ZT-4	29.80	15.80	4.50	78.70
ZT-5	92.30	37.10	31.30	134.20
QJ-3	65.10	32.30	13.00	46.10
HH-1	78.20	33.70	40.20	65.00
QJ-6	59.30	11.10	37.80	54.90
QJ-7	78.60	20.50	14.60	67.80
mean	63.86 ± 25	23.76 ± 11	28.09 ± 20	96.84 ± 66
Yunnan [a]	36.60 ± 31	16.50 ± 15	39.20 ± 4	36.10 ± 35
China [a]	64.90 ± 32	37.50 ± 18	49.40 ± 31	106.00 ± 27
Poland [b]	23.50	14.30	18.10	129.90
Australia [c]	25.00	24.00	21.00	75.00

[a] Liu *et al.* (2007) [12]; [b] Bemet *et al.* (2002) [38]; [c] Fardyet *et al.* (1989) [39].

For comparison, the radioactivity of these radionuclides in coals from Yunnan province, China, and other countries are shown in Table 2. The average radioactivity of ^{238}U and ^{40}K in studied coals are 63.86 ± 25 and 96.84 ± 66 Bq·kg^{-1}, respectively. Both values are comparable to those in Chinese coals, but are elevated by a factor of 1.7–2.7 as compared to those previously measured in Yunnan coals [11], Radioactivity of ^{232}Th and ^{226}Ra in both previously measured Yunnan coals and

this study are slightly lower than those in Chinese coals [12]. It is noted that radionuclides in specific coals can reach to rather high levels such as ^{238}U (84.90 Bq·kg^{-1}), ^{232}Th (36.00 Bq·kg^{-1}) and ^{40}K (165.00 Bq·kg^{-1}) in Maoergou Mine (ZT-1), and ^{238}U (68.80 Bq·kg^{-1}), ^{226}Ra (61.80 Bq·kg^{-1}) and ^{40}K (229.30 Bq·kg^{-1}) in Kelang Mine (KM) (Table 2). Australia coals generally have low levels of radioactivity of nuclides, while Poland coals have relatively high radioactivity of ^{40}K [38,39].

4.4. Factors Influencing Radioactivity of Nuclides

Coles *et al*. (1978) [40] identified radionuclides associated with organic matter and sulfides are more easily volatilized than those with affinity to alumino-silicate minerals. The correlation analysis of these radionuclides with coal ash yield is used to determine their preliminary modes of occurrence (Figure 2). As shown in Figure 2, the ^{238}U has a positive correlation with ash yield (R^2 = 0.81), indicating a dominant inorganic affinity. However, the correlation between U and ash yield is insignificant albeit positive (R^2 = 0.24). Uranium can reside in coal matrix in a highly disperse, organic state, or as discrete minerals such as brannerite, uraninite and coffinite, depending on the coal-forming environment [10,40,41]. Seredin and Finkelman (2008) [41] and Dai *et al*. (2014) [10] showed that U in U-bearing coal deposits worldwide is mainly associated with the organic matter, and that only a small proportion of the U occurs in U-bearing minerals. The inferred modes of occurrence for ^{238}U and U by statistic correction method are not consistent in present study, which need further confirmation.

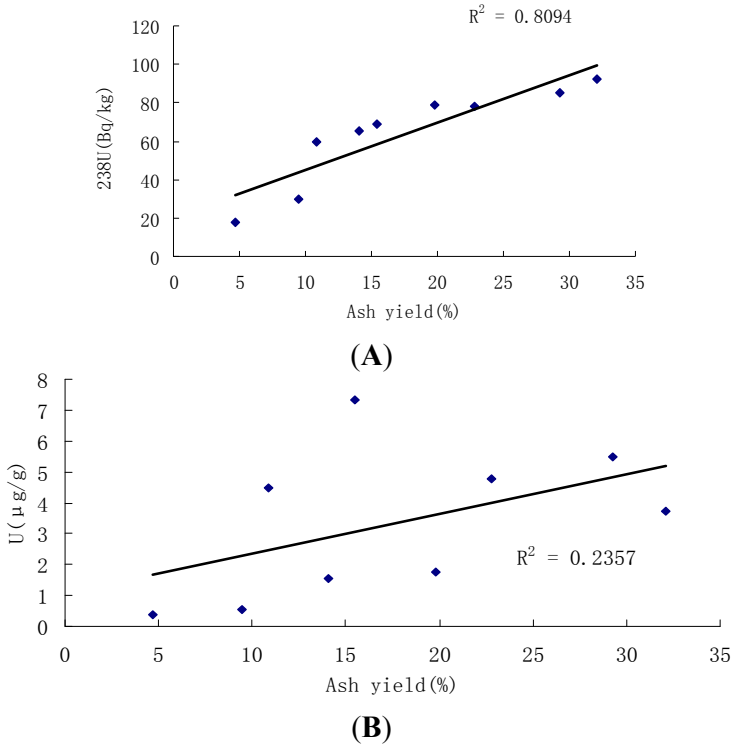

Figure 2. Correlations of ^{238}U (**A**) and U (**B**) with ash yield.

Thorium, Ra and K are commonly considered as refractory elements and associated with alumino-silicate ash of coal [40]. Thorium is generally associated with zircon ($ZrSiO_4$) [42], a highly chemical-resistant mineral. Zircons in coals can have both volcanic and authigenic origins [43,44]. Besides, Th can be adsorbed by clay minerals in coals [45]. The positive correlations (Figure 3) of ^{232}Th radioactivity (R^2 = 0.73) and Th (R^2 = 0.58) with ash yield demonstrate a dominant inorganic affinity of Th. It has been shown that the radioactivity of ^{226}Ra in Indian coal slag is two times higher than that in feed coal [35,36], implying a dominant inorganic Ra state. A highly positive correlation

of Ra with ash yield has been observed in our study (R^2 = 0.50). However, the correlative of ^{226}Ra with ash yield is weak (Figure 4).

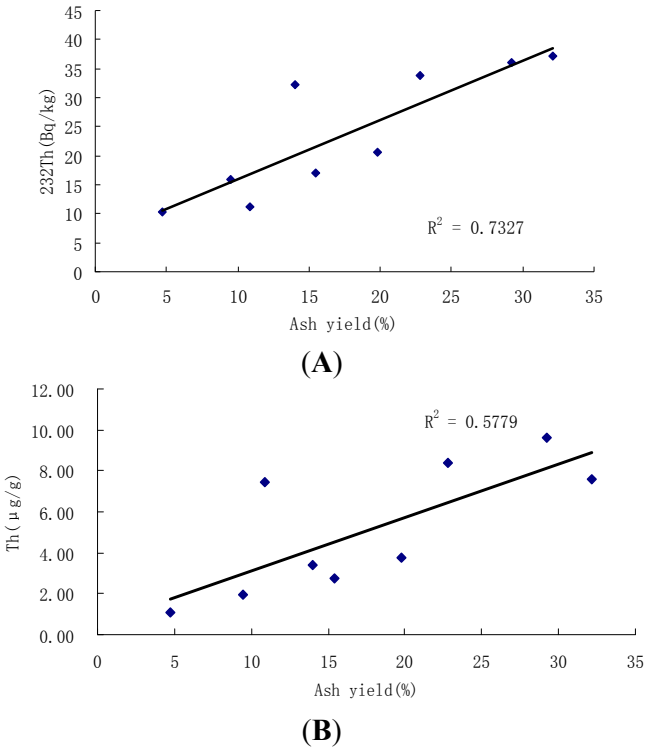

Figure 3. Correlations of ^{232}Th (**A**) and Th (**B**) with ash yield.

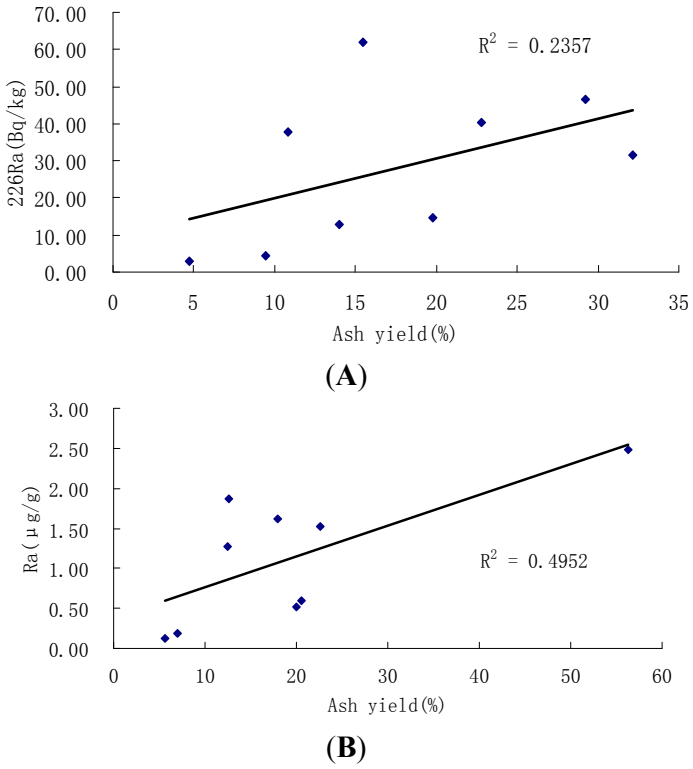

Figure 4. Correlations of ^{226}Ra (**A**) and Ra (**B**) with ash yield.

4.5. Radioactive Impacts of Nuclides

The assessment of the radioactive impacts of nuclides in coals has an important environmental significance, since coal is abundantly used in power generation. Emissions from coal-fired power plants in gaseous and particulate forms containing nuclides probably discharge and accumulate in the surrounding environment, thus causing radiation exposures to the local population [46,47]. Inhalation of fly ash emitted from the stacks and the ingestion of foodstuffs receiving atmospheric deposition are the main pathways to increase human radiation exposure [48].

It has been hypothesized that 370 Bq· kg^{-1} of ^{226}Ra or 259 Bq· kg^{-1} of ^{232}Th or 4810 Bq· kg^{-1} of ^{40}K could produce the same gamma dose rate [31,49]. The Ra equivalent radioactivity, the internal exposure index (I_{Ra}) and the external hazard index (I_r) can be calculated according to Beretka and Mathew (1985) [50] and GB6566-2010 [51] as:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

$$I_{Ra} = \frac{C_{Ra}}{200} \leqslant 1 \quad (3)$$

$$I_r = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leqslant 1 \quad (4)$$

where C_{Ra}, C_{Th} and C_K, are the radioactivity of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The maximum value of Ra_{eq} in construction materials is set to 370 Bq· kg^{-1} for safe use [52], the maximum values of I_{Ra} and I_r must be lower than 1.0 according to GB6566-2010 [48]. The calculated values of Ra_{eq} for studied coals vary from 19.97 to 109.43 Bq· kg^{-1} (Table 3), with an average of 68.84 Bq· kg^{-1}, which is lower than the recommended limit. The calculated values of I_r and I_{Ra} are generally below than 0.3 significantly lower than unity.

Table 3. Radium equivalent activity (*Raeq*), external hazard index (I_r), internal exposure index (I_{Ra}), air absorbed dose rates (D (nGy/h)) and annual effective dose (AED) of radionuclides in coals from eastern Yunnan, China.

Sample ID	*Raeq*	I_r	I_{Ra}	D (nGy/h)	AED
ZT-1	110.59	0.30	0.23	50.06	61.40
KM	103.77	0.28	0.31	48.38	59.33
ZT-3	20.19	0.05	0.02	8.93	10.95
ZT-4	33.15	0.09	0.02	14.90	18.28
ZT-5	94.69	0.26	0.16	42.47	52.08
QJ-3	62.74	0.17	0.07	27.44	33.65
HH-1	93.40	0.25	0.20	41.64	51.06
QJ-6	57.90	0.16	0.19	26.46	32.45
QJ-7	49.14	0.13	0.07	21.95	26.92
mean	69.51	0.19	0.14	31.36	38.46

The total outdoor air absorbed dose rate (nGy/h) due to terrestrial gamma rays at 1 m above the ground is calculated from ^{226}Ra, ^{232}Th and ^{40}K. The conversion factors used to calculate the absorbed dose rates are given as follows [53]:

$$D(nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (5)$$

The calculated absorbed dose rates range from 8.93 to 50.06 nGy/h with a mean value of 31.36 nGy/h (Table 3), which is lower than the average value of global primordial radiation of 59 nGy/h [53] and the Chinese natural gamma radiation dose rate of 63.0 nGy/h [53].

To estimate the annual effective dose rates, the conversion coefficient from the absorbed dose in air to the effective dose (0.7 nGy/h) and the outdoor occupancy factor (20%) recommended

by UNSCEAR (2000) [53] are used. The annual effective dose rate is calculated by the following formula [52]:

$$\text{Effective dose rate } (\mu\text{Sv/year}) = \text{D } (\text{nGy/h}) \times 8760 \text{ (h/year)} \times 0.7 \times 10^6 \mu\text{Sv}/10^9 \text{nGy} \times 0.2 \quad (6)$$

The calculated effective dose rates vary from 10.95 to 61.40 μSv/year with an average value of 38.46 μSv/year.

5. Conclusions

This study provides an initial assessment on the natural radionuclides in coals from eastern Yunnan, China. The mean specific activity concentrations of radionuclides ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are 63.86, 28.09, 23.76 and 96.84 Bq·kg^{-1} in coals, respectively. Radioactivity of ^{238}U and ^{40}K in studied coal are elevated by a factor of 1.7–2.7 as compared to previously measured Yunnan coals. Radioactivity of ^{238}U and ^{40}K in all Yunnan coals is slightly lower than Chinese coals. Radioactivity of ^{238}U and ^{232}Th is closely correlated with coal ash, suggesting their inorganic origins. The environmental effect of natural radionuclides is considered to be negligible as evaluated by indexes of *Raeq*, I_R and I_r. The absorbed dose rates D (nGy/h) values are lower than the average value of global primordial radiation and the Chinese natural gamma radiation dose rate.

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Conflicts of Interest: The authors declare no conflict of interest.

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