DETERMINATION OF NATURAL RADIOACTIVITY, ^{222}Rn AND ^{220}Rn EXHALATION RATES AND RADIATION HAZARDS OF FLY ASH AND FLY ASH BRICK USED IN BAOTOU, CHINA

by

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Natural radioactivity levels, ^{222}Rn and ^{220}Rn exhalation rates and radiation hazards of fly ash and fly ash brick used in Baotou, China were determined. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in fly ash samples ranged from 38.81 to 93.73, 40.34 to 135.17, and 66.92 to 290.86 Bq/kg with an average of 76.52, 109.95, and 170.72 Bq/kg, respectively; while in fly ash brick samples, these radionuclides ranged from 42.43 to 71.60, 76.65 to 208.37, and 94.32 to 489.42 Bq/kg with an average of 53.83, 101.93, and 266.48 Bq/kg, respectively. The exhalation rates of ^{222}Rn and ^{220}Rn in all determined samples were in the range of 1.13-20.50 and 15.60-113.00 mBq/m^{2}s, respectively. The calculated results of the radium equivalent activity, external hazard index, internal hazard index, indoor annual effective dose and outdoor annual effective dose indicated that fly ashes and fly ash bricks collected from some brick factories of Baotou would pose excessive radiation risks to inhabitants and that they are not suitable for use in building construction. The natural radioactivity level of fly ash and fly ash brick needs to be constantly monitored considering the radiation safety of the local residents.

Key words: natural radioactivity, radon exhalation rate, radiation hazard, fly ash, building material

INTRODUCTION

Coal, formed through complicated geological processes (*e. g*. petrification and coalification), is an abundant natural energy source and an important fossil fuel. It is widely used to produce electricity. The increasing demand for electricity generation for industrial development and human living standards worldwide is met by combustion of fossil fuels [1]. China largely depends on coal for its energy needs, which contribute more than 70 % of the total power generated in China [2]. Large quantities of fly ash are expelled from coal-fired thermal power plants along with coal combustion [3]. Fly ash is a by-product or combustion residue of coal burning at electric utility plants and is collected by electrostatic or mechanical precipitation of dust-like particles from the fuel gases of furnaces fired with coal. Like other natural materials, coal contains natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K, which may be concentrated and enriched in fly ash during coal combustion owing to its smaller size and the larger surface area [4]. Since fly ash contains elevated natural radionuclides compared to coal, it is considered as a potential source of radiation exposure to man.

Natural radionuclides in fly ash may pose radiation risks externally due to their gamma-ray emissions and internally due to radon and its progenies that emit alpha particles when fly ash is used in building materials or piled on the ground [4-6]. Radon (Rn), a naturally radioactive inert gas, has three isotopes, *i. e*. ^{222}Rn, ^{220}Rn, and ^{219}Rn. Their half-lives are 3.82 days, 55.6 s, and 3.96 s, respectively. Amongst these three isotopes, ^{222}Rn has caused more concerns due to its relatively long half-life. However, recent long-term surveys of indoor ^{220}Rn and its progenies showed that doses from the ^{220}Rn series should no longer be considered as negligible [7]. ^{222}Rn is derived from the radioactive decay of ^{226}Ra, a decay element in the uranium series, while ^{220}Rn is derived from the radioactive decay of ^{232}Th [8]. The sources of ^{222}Rn and ^{220}Rn in indoor air mainly depend on the natural radionuclides in the foundation soil/rock, building materials, domestic water and fossil fuels [9].

Natural radioactivity and associated radiation hazard of fly ash have become a subject of worldwide interest in recent years because of the diverse use of fly ash in the manufacture of cement, clay ash bricks, ash

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bricks/blocks, cellular concrete blocks, asbestos products, as a replacement for sand and cement in building materials, for filling of underground cavities, in construction of roads/rail embankments/reinforced earth walls and mine filling [1-3,10-16]. The relative research in China, however, is limited, especially the ^{222}Rn and ^{220}Rn exhalation rates of fly ash and the building materials made from fly ash. In China, coals used in coal-fired thermal power plants are found to have a high ash contents, resulting in the production of large amount of fly ash, which is mainly used to produce cement, brick, and blocks. The main objectives of the present work were to determine the activity concentrations of natural radionuclides, ^{222}Rn and ^{220}Rn exhalation rates of fly ash and fly ash brick used in Baotou, China, and to assess their radiological hazards to local inhabitants. The results could offer basic information for the safe use and management of these materials.

MATERIALS AND METHODS

Sample collection and preparation

To know the natural radioactivity, ^{222}Rn and ^{220}Rn exhalation rates and radiation hazards of fly ash (FA) and fly ash brick (FAB), FA samples (numbered FA1 to FA4), and FAB samples (numbered FAB1 to FAB4) were collected from four main brick factories of Baotou. At each brick factory, 4-6 FA samples and 10-20 FAB samples were collected randomly. All collected samples were kept in cleaned and numbered polyethylene bags and transported to the laboratory. The FAB samples were crushed and ground into a fine powder. The ground FAB samples and FA samples were dried in a temperature-controlled furnace at 110 ºC for 12-24 h to remove moisture. Finally, the dried samples were weighted and stored in gas-tight, radon impermeable, cylindrical polyethylene plastic containers (7.0 cm height and 6.5 cm diameter). These containers were hermetically sealed to prevent the escape of gaseous ^{222}Rn and ^{220}Rn from the samples and kept in a laboratory for more than 30 days to ensure radioactive equilibrium between ^{226}Ra and ^{232}Th and their decay products [17].

Radioactivity measurement

The specific activities of ^{226}Ra, ^{232}Th and ^{40}K in all samples were measured by a 3 × 3 inch NaI(Tl) gamma ray spectrometric system with >8 % energy resolution (^{137}Cs 661.6 keV). The detector, placed in a lead shielding room with a thickness of 10.5 cm, height of 38 cm and inner diameter of 21 cm, was coupled to a 1024 microcomputer multi-channel pulse height analyzer and the system was calibrated for the gamma energy range 50 keV to 3.2 MeV. The activity of ^{226}Ra was determined by the photopeaks of ^{214}Bi at 609.3 and 1764.5 keV, and the activity of ^{232}Th was measured through the photopeaks of ^{212}Pb at 238.6 keV and ^{208}Tl at 2614 keV [18]. ^{40}K activity was determined directly by its own gamma ray at 1460.8 keV. All samples were counted for 5 h. Each sample was counted twice before an average value was calculated.

Exhalation rate measurement of ^{222}Rn and ^{220}Rn

The ^{222}Rn and ^{220}Rn exhalation rates of fly ash and fly ash brick samples were measured with an electrostatic radon sampler-2 (ERS-2) made by the Tracerlab company of Germany [19]. The ERS-2 operates with an alpha spectroscopy detector and multi-channel analyzer with 256 channels. When measuring, ERS-2 was placed on the surface of the measured sample with a silicone-sealing ring to avoid air leakage. Then the diffusion mode operation was selected and high voltage was turned on to reach the stable HV 500. Each sample was counted for 10 minutes and 12 times, and the ^{222}Rn and ^{220}Rn exhalation rate in the sample was calculated by the ERSEval software [20, 21].

RESULTS AND DISCUSSION

Specific activity

The minimum, maximum and mean of the specific activity values of ^{226}Ra, ^{232}Th, and ^{40}K in fly ash (FA) and fly ash brick (FAB) samples are shown in tab. 1. As listed in tab. 1, the specific activities of ^{226}Ra,

Table 1. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in fly ash and fly ash brick from Baotou, China

Sample No.	Activity concentration [Bqkg^{-1}]								
	^{226}Ra			^{232}Th			^{40}K		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
FA1	38.31	40.67	39.73	40.34	50.83	50.17	66.92	82.11	74.66
FA2	80.99	91.91	87.09	126.18	135.17	129.65	100.35	169.7	136.30
FA3	85.71	93.73	89.92	124.53	132.35	128.61	143.19	290.86	165.62
FA4	64.05	68.93	65.69	89.89	92.62	91.66	261.24	290.86	275.49
FAB1	42.43	56.30	48.97	77.12	93.03	82.40	263.01	393.61	308.51
FAB2	44.51	71.60	51.63	76.65	91.08	84.50	183.84	331.44	223.03
FAB3	56.20	62.93	61.09	77.74	113.63	84.98	341.35	489.42	373.17
FAB4	51.66	67.75	59.13	167.22	208.37	180.58	94.32	186.66	132.21

^{232}Th, and ^{40}K in fly ash samples ranged from 38.81 to 93.73, 40.34 to 135.17, and 66.92 to 290.86 Bq/kg with an average of 76.52, 109.95, and 170.72 Bq/kg, respectively. The concentrations of ^{226}Ra and ^{40}K in all investigated fly ash samples were less than the typical activity concentrations of ^{226}Ra and ^{40}K in fly ash (180 and 650 Bq/kg for ^{226}Ra and ^{40}K, respectively) of European Union countries [22]. The concentrations of ^{232}Th in fly ash samples from two brick factories were less than, while in the samples from another two brick factories they were higher than the typical activity concentration of ^{232}Th in fly ash (100 Bq/kg) of European Union countries [22]. ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations in FAB samples varied from 42.43 to 71.60, 76.65 to 208.37, and 94.32 to 489.42 Bq/kg with an average of 53.83, 101.93, and 266.48 Bq/kg, respectively. The mean values of the ^{226}Ra and ^{232}Th activity concentration in fly ash brick exceeded that of the worldwide population-weighted average value for soil (32 and 45 Bq/kg for ^{226}Ra and ^{232}Th, respectively), whereas the mean concentration of ^{40}K in fly ash brick was less than the worldwide population-weighted value for soil (420 Bq/kg) [6]. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in all fly ash and fly ash brick samples were in the range of Chinese soil values [6]. As can be seen from tab. 1, there was no corresponding correlation in ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations between fly ash brick and fly ash from the same brick factories. This may be related to the fly ash adding and other raw materials during the fly ash brick production. Fly ash brick is made from fly ash, lime, gypsum, aggregate and water. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in fly ash brick depend on the natural radioactivity level of the aforementioned raw materials and their proportions.

Table 2 presents the comparison of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations in fly ash used in Baotou with other areas [2, 10, 16, 19, 23]. The average value of ^{226}Ra in fly ash used in Baotou are is less than the other compared areas, whereas the average value of ^{40}K in fly ash used in Baotou are is lower than values reported for the fly ash from Xiangyang and Turkey, and close to the values of Xijiao and Baqiao. The average concentration of ^{232}Th in fly ash from Baotou is close to Xianyang and Turkey and higher than the values of Xijiao and Baqiao. The differences of ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations among the compared fly ashes are mainly related to their raw coal types and their natural radioactivity level [12].

Radon exhalation rate

^{222}Rn and ^{220}Rn exhalation rates (minimum, maximum and mean) in the determined samples are shown in tab. 3 and tab. 4, respectively. The ^{222}Rn exhalation rate values in fly ash and fly ash brick samples ranged from 2.03 to 17.40 and 1.13 to 20.50 mBq/m^2/s with an average of 5.75 and 6.36 mBq/m^2/s, respectively (tab. 3), while the ^{220}Rn exhalation rate values in fly ash and fly ash brick samples varied from 15.60 to 113.00 and 30.50 to 110.00 mBq/m^2/s with an average of 62.08 and 67.82 mBq/m^2/s, respectively (tab. 4). The values of the ^{222}Rn exhalation rate in all samples are lower than the values of the ^{220}Rn exhalation rate, which is consistent with other research [9]. The aver-

Table 2. Comparison of activity concentrations [Bqkg^{-1}] in fly ash with other areas

Area	^{226}Ra			^{232}Th			^{40}K			References
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	
Greece	273	1377	–	41	65	–	143	661	–	[23]
Greece	794	1028	–	52	57	–	403	516	–	[10]
Turkey	17	2720	360.3	9.0	696.0	101.7	12	2914	517	[16]
Xijiao, China	46.4	148.0	69.5	59.3	153.9	79.3	123.3	343.0	233.0	[2]
Xiangyang, China	90.3	1799.4	440.5	59.9	145.6	110.3	309.0	906.3	510.1	[19]
Baqiao, China	60.5	131.81	83.2	61.5	164.6	87.4	155.9	316.1	234.8	[2]
Baotou, China	38.31	91.91	58.44	40.34	137.17	103.79	66.92	489.42	244.27	Present study

Table 3. ^{222}Rn exhalation rate ER_i and ^{222}Rn concentrations C calculated for different air exchange rates (λ_v)

Sample No.	ER_i [mBqm^{-2}s^{-1}]			C [Bqm^{-3}]											
	Min	Max	Mean	$\lambda_v = 0$ [h^{-1}]			$\lambda_v = 0.2$ [h^{-1}]			$\lambda_v = 0.5$ [h^{-1}]			$\lambda_v = 1.2$ [h^{-1}]		
				Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
FA1	3.05	7.42	5.24	2908.61	7076.03	4992.32	105.81	257.40	181.60	43.25	105.22	74.23	18.18	44.23	31.21
FA2	2.39	6.71	5.08	2279.21	6398.94	4847.68	82.91	232.77	176.34	33.89	95.15	72.08	14.25	40.00	30.30
FA3	6.45	17.40	9.02	5407.15	16593.38	8599.47	196.69	603.61	312.82	80.40	246.74	127.87	33.80	103.73	53.76
FA4	2.03	5.10	3.65	1935.89	4863.58	3483.18	70.42	176.92	126.71	28.79	72.32	51.79	12.10	30.40	21.77
FAB1	1.89	16.60	7.05	1802.38	15830.46	6722.23	65.56	575.86	244.53	26.80	235.39	99.96	11.27	98.96	42.02
FAB2	2.03	11.60	5.24	1935.89	11062.25	4993.42	70.42	402.41	181.64	28.79	164.49	74.25	12.10	69.15	31.22
FAB3	1.13	20.50	6.93	1077.62	19549.67	6611.60	39.20	711.15	240.51	16.02	290.69	98.31	6.74	122.21	41.33
FAB4	2.84	11.00	6.22	2708.34	10490.07	5932.61	98.52	381.59	215.81	40.27	155.98	88.22	16.93	65.58	37.09

Table 4. ^{220}Rn exhalation rate ER_i and ^{220}Rn concentrations C calculated for different air exchange rates (λ_v

Sample No.	ER_i [mBqm^{-2}s^{-1}]			C [Bqm^{-3}]											
	Min	Max	Mean	λ_v = 0 [h^{-1}]			λ_v = 0.2 [h^{-1}]			λ_v = 0.5 [h^{-1}]			λ_v = 1.2 [h^{-1}]		
				Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
FA1	15.60	38.30	26.50	2.50	6.15	4.32	2.49	6.12	4.31	2.48	6.08	4.28	2.44	5.99	4.21
FA2	24.10	51.70	36.20	3.87	8.30	5.81	3.85	8.26	5.78	3.82	8.20	5.74	3.77	8.08	5.66
FA3	50.20	113.00	69.40	8.06	18.13	11.14	8.02	18.05	11.09	7.97	17.93	11.01	7.85	17.66	10.85
FA4	25.50	45.30	30.45	4.09	7.27	4.89	4.07	7.24	4.86	4.05	7.19	4.83	3.99	7.08	4.76
FAB1	53.10	110.00	80.84	8.52	17.65	12.97	8.48	17.57	12.91	8.43	17.46	12.83	8.30	17.19	12.63
FAB2	41.80	101.00	82.55	6.71	21.34	13.25	6.68	21.25	13.19	6.63	21.11	13.10	6.53	20.79	12.90
FAB3	30.50	90.70	56.80	4.89	14.55	9.11	4.87	14.49	9.07	4.84	14.39	9.01	4.77	14.17	8.88
FAB4	39.70	77.90	51.08	6.15	12.50	8.20	6.12	12.44	8.16	6.08	12.36	8.11	5.99	12.17	7.98

age ^{222}Rn exhalation rate values in fly ash samples are close to fly ash brick samples, while the average ^{220}Rn exhalation rate values in most fly ash samples are lower than fly ash brick samples. The ^{222}Rn exhalation rate of fly ash and other building materials was extensively reported in literature, while the work on the ^{220}Rn exhalation rate of building materials is limited. Table 5 shows the comparison of the ^{222}Rn exhalation rate of fly ash from Baotou and other areas. As can be seen from tab. 5, the ^{222}Rn exhalation rate values of fly ash from Baotou are close to which that of fly ash from Xiangyang [19], and are significantly higher than which that of fly ash from four different thermal power stations situated in two different states of India [1, 4, 5]. The ^{222}Rn and ^{220}Rn exhalation rates in the samples are influenced by the grain size of the sample, ambient temperature, atmospheric pressure, humidity, and the activity concentrations of ^{226}Ra and ^{232}Th in the sample [7, 15, 24].

Assessment of the radiation hazard

In the present study, to assess the radiation hazard of fly ash and fly ash brick used in building construction or piled on the ground, the radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), indoor air absorbed dose rate (D_{in}), annual effective dose (AED_{in}), indoor ^{222}Rn/ ^{220}Rn exposure dose (E_i, $i = ^{222}$Rn, ^{220}Rn), outdoor air absorbed dose rate (D_{out}), and annual effective dose (AED_{out}) were calculated.

Table 5. A comparison of the ^{222}Rn exhalation rate in fly ash of Baotou and other areas

Area	^{222}Rn exhalation rate [mBqm^{-2}s^{-1}]			Reference
	Min	Max	Mean	
Baotou, China	2.03	17.40	5.75	Present study
Xiangyang, China	1.31	20.61	5.86	[19]
Dadri, Uttar Pradesh, India	0.02	0.07	0.04	[1]
Durgapur, West Bengal, India	0.10	0.13	0.11	[5]
Kolaghat, West Bengal, India	0.31	0.43	0.36	[4]
Kasimpur, Uttar Pradesh, India	0.15	0.41	0.30	[4]

The radium equivalent activity (Ra_{eq}), assuming that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq/kg of ^{40}K can produce the same gamma ray dose, is a single index to describe the radiation hazard from different radionuclide mixtures in a material [25]. It is calculated using the following relation [26]

$$Ra_{eq} \quad C_{Ra} \quad 1.43C_{Th} \quad 0.077C_{k} \qquad (1)$$

where C_{Ra}, C_{Th}, and C_k are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively. The calculated values of Ra_{eq} for all samples are shown in fig. 1. As shown in fig. 1, Ra_{eq} values ranged from 116.39 to 369.90 Bq/kg, which are below the recommended limit of 370 Bq/kg [6].

To limit the external gamma radiation dose from building materials to 1.5 mSv, the external hazard index (H_{ex}) is defined by the following equation [26]

$$H_{ex} \quad \frac{C_{Ra}}{370} \quad \frac{C_{Th}}{259} \quad \frac{C_{k}}{4810} \qquad (2)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively. The value of H_{ex} must be less than unity for the radiation hazard to be negligible. As it is shown in fig. 2, the H_{ex} values of the studied samples are less than unity.

In addition to the external radiation hazard, radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon

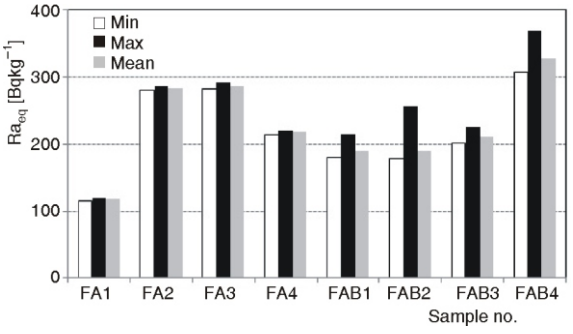

Figure 1. Calculated values of radium equivalent activity (Ra_{eq}) in the studied samples

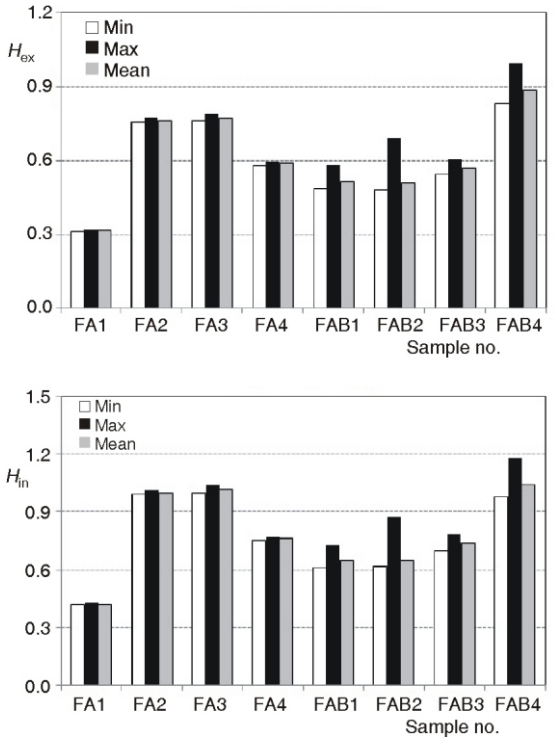

Figure 2. Calculated values of the external hazard index (H_{ex}) and internal hazard index (H_{in}) in the studied samples

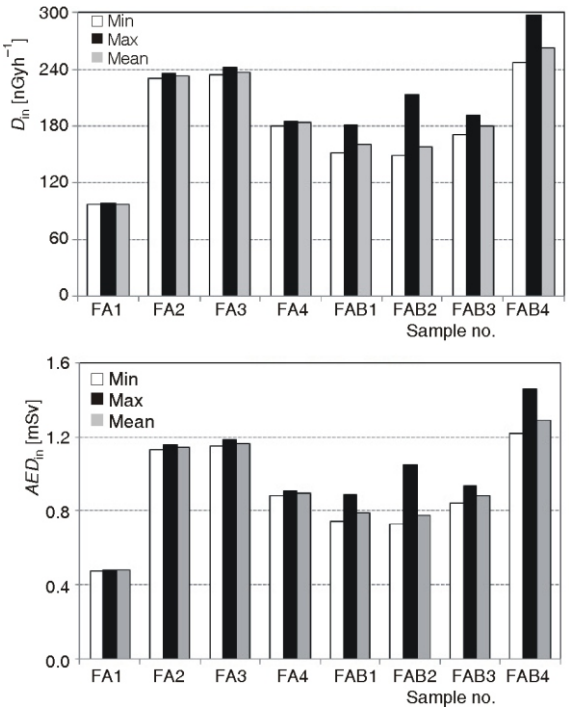

Figure 3. Calculated values of the indoor air absorbed dose rate (D_{in}) and the corresponding annual effective dose (AED_{in}) in the studied samples

and its decay products quantified by the internal hazard index (H_{in}) which is defined by the equation [26]

$$H_{in} \quad \frac{C_{Ra}}{185} \quad \frac{C_{Th}}{259} \quad \frac{C_k}{4810} \tag{3}$$

where C_{Ra}, C_{Th}, and C_k are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively. For the safe use of a material in the construction of dwellings, H_{in} should be less than unity. As can be seen from fig. 2, the H_{in} values of FA2, FA3, and FAB4 samples are close to unity. The large H_{in} values for these samples are mainly due to these samples having an elevated ^{226}Ra and/or ^{232}Th activity concentration.

The absorbed dose rate of door air due to gamma ray emission from the radionuclides in the building materials was evaluated. According to the report of the European Commission, the indoor air absorbed dose rate (D_{in}) is determined by the equation [22]

$$D_{in} \quad 0.92C_{Ra} \quad 1.1C_{Th} \quad 0.08C_k \tag{4}$$

where C_{Ra}, C_{Th}, and C_k are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg, respectively. The unit of D_{in} is [nGyh^{-1}]. Figure 3 shows the estimated D_{in} values for the investigated samples, ranging from 96.98 to 297.43 nGy/h with an average of 189.24 nGy/h. They were significantly higher than the world population-weighted average indoor absorbed gamma dose rate of 84 nGy/h [6]. The D_{in} values for most samples (97 %) were higher than the average indoor gamma radiation dose rate of China, 99 nGy/h, [6] and Baotou 99.0 nGy/h [27]. Considering the conversion coefficient 0.7 Sv/Gy and the indoor occupancy factor 0.8 proposed by the United Nations Scientific Committee on Effects of Atomic Radiation [6], the corresponding annual effective dose AED_{in} due to gamma ray emission of ^{226}Ra, ^{232}Th, and ^{40}K from the investigated samples ranged from 0.476 to 1.459 mSv with an average of 0.923 mSv (fig. 3), which were higher than the worldwide average of 0.41 mSv for the indoor annual effective dose rate [6]. The AED in values of FA2, FA3 and FAB4 were also higher than the recommended limit of 1 mSv [22].

Indoor ^{222}Rn and ^{220}Rn air concentration originating from the investigated samples can be estimated by the equation [7]

$$C_i \quad \frac{ER_i S}{(\lambda \quad \lambda_v)V}, \; i \quad ^{222}\text{Rn}, \; ^{220}\text{Rn} \tag{5}$$

where C_i is the indoor ^{222}Rn or ^{220}Rn air concentration [Bqm^{-3}] released from the investigated building materials (*i. e.* fly ash and fly ash brick), ER_i is the ^{222}Rn or ^{220}Rn exhalation rate [mBqm^{-2}s^{-1}], S [m^2] – the area of the material exhaling ^{222}Rn or ^{220}Rn, V [m^3] – the room volume and λ [s^{-1}] is the room air exchange rate, and λ [s^{-1}] – the ^{222}Rn or ^{220}Rn decay constant. In the calculation, the concentration of ^{222}Rn and ^{220}Rn from the building material was assessed by assuming the room was a cavity with $S/V = 2.0$ m^{-1} [1, 4, 5, 19]. In a typical dwelling, the λ_v ranges from 0.20 to 1.20 h^{-1} with 0.5 h^{-1}, as average [6]. Different air exchange rates ($\lambda_v = 0$, 0.2, 0.5, 1.2 h^{-1}) were considered in this work

and the results are presented in tabs. 3 and 4. The difference in behavior of ^{222}Rn (tab. 3) and ^{220}Rn (tab. 4) with respect to the air exhalation rate was observed. Table 3 shows that the ^{222}Rn concentration obviously decreased with the increasing of the air exchange rate, whereas there was no effect on the ^{222}Rn concentration with the change of the air exchange rate (tab. 4), which is due to the ^{220}Rn decay constant being more than the air exchange rate.

The indoor annual exposure effective dose (E_i) of ^{222}Rn or ^{220}Rn released from the investigated materials for inhabitants was calculated by the equation [6]

$$E_i \quad EEC_i \; O \; F_i \; C_i \quad i \quad ^{222}\text{Rn}, \; ^{220}\text{Rn} \qquad (6)$$

where C_i is the indoor air concentration of ^{222}Rn or ^{220}Rn [Bqm^{-3}] released from the investigated materials, F_i – the equilibrium factor, *i. e*. typically 0.4 for ^{222}Rn indoor and 0.1 for ^{220}Rn indoor [28], EEC_i represents the recommended value to convert the ^{222}Rn or ^{220}Rn equilibrium-equivalent concentration to the population effective dose, *i. e*. 9 [nSv/Bqh^{-1}m^{-3}] for ^{222}Rn, 40 [nSv/Bqh^{-1}m^{-3}] for ^{220}Rn [6, 7]. O represents the occupancy factor expressed as the number of hours which the inhabitant spends in the building in one year, in further calculations taken to be 7008 h (80 %). The indoor annual exposure effective dose of ^{222}Rn and ^{220}Rn released from the investigated samples, calculated based on different air exchange rates, is presented in tabs. 6 and 7, respectively. Table 6 indicates that in the worst case when there is no ventilation ($\lambda_v = 0$) the mean values of the indoor annual exposure effective dose due to ^{222}Rn released from all investigated samples ranged from 87.88 to 216.95 mSv, which are 4.4 to 10.8 times the upper limit (20 mSv) recommended by ICRP-65 [29]. The indoor annual exposure effective dose values of ^{222}Rn for the investigated samples obviously decreased with the air exchange rate increasing (tab. 6) and the mean values of the indoor annual exposure effective dose of ^{222}Rn for the samples at the average air exchange rate ($\lambda_v = 0.5$) varied from 1.31 to 3.23 mSv.

Table 7 shows that there was no significant variation in the annual exposure effective dose values of ^{220}Rn released from the samples under different air exchange rates. The mean values of the indoor annual exposure effective dose due to ^{220}Rn released from the investigated samples at the average air exchange rate ($\lambda_v = 0.5$) ranged from 119.89 to 367.24 μSv, which were ~10 % of the mean values (1.31 to 3.23 mSv) of the indoor annual exposure effective dose of ^{222}Rn at the average air exchange rate.

An attempt was also made in the study to evaluate the gamma radiation from the ash pond or fly ash brick factory. Conversion factors were used to transform specific activities, C_{Ra}, C_{Th} and C_K of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in the air. The absorbed dose rate at 1 m above the ground (nGy/h per Bq/kg) was calculated by using the following eq. [30]

$$D_{out} \quad 0.461C_{Ra} \quad 0.623C_{Th} \quad 0.0414C_k \qquad (7)$$

where C_{Ra}, C_{Th} and C_k are the activities of ^{226}Ra, 232Th and ^{40}K in [Bqkg^{-1}], respectively. The calculated results present show that the outdoor air absorbed dose

Table 6. The estimated values of the indoor annual exposure effective dose of ^{222}Rn [mSv] under different air exchange rates

Sample No.	$\lambda_v = 0$ [h^{-1}]			$\lambda_v = 0.2$ [h^{-1}]			$\lambda_v = 0.5$ [h^{-1}]			$\lambda_v = 1.2$ [h^{-1}]		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
FA1	73.38	178.52	125.95	2.67	6.49	4.58	1.09	2.65	1.87	0.46	1.12	0.79
FA2	57.50	161.44	122.30	2.09	5.87	4.45	0.86	2.40	1.82	0.36	1.01	0.76
FA3	136.42	418.63	216.95	4.96	15.23	7.89	2.03	6.22	3.23	0.85	2.62	1.36
FA4	48.84	122.70	87.88	1.78	4.46	3.20	0.73	1.82	1.31	0.31	0.77	0.55
FAB1	45.47	399.38	169.59	1.65	14.53	6.17	0.68	5.94	2.52	0.28	2.50	1.06
FAB2	48.84	279.09	125.98	1.78	10.15	4.58	0.73	4.15	1.87	0.31	1.74	0.79
FAB3	27.19	493.21	166.80	0.99	17.94	6.07	0.40	7.33	2.48	0.17	3.08	1.04
FAB4	68.33	264.65	149.67	2.49	9.63	5.44	1.02	3.94	2.23	0.43	1.65	0.94

Table 7. The estimated values of the indoor annual exposure effective dose of ^{220}Rn [μSv] under different air exchange rates

Sample No.	$\lambda_v = 0$ [h^{-1}]			$\lambda_v = 0.2$ [h^{-1}]			$\lambda_v = 0.5$ [h^{-1}]			$\lambda_v = 1.2$ [h^{-1}]		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
FA1	70.17	172.28	121.22	69.86	171.51	120.69	69.40	170.38	119.89	68.34	167.79	118.07
FA2	108.40	232.55	162.83	107.92	231.52	162.11	107.21	229.99	161.04	105.58	226.50	158.59
FA3	225.81	508.29	312.17	224.80	506.03	310.78	223.32	502.69	308.73	219.92	495.05	304.04
FA4	114.70	203.76	136.97	114.19	202.86	136.36	113.44	201.52	135.46	111.71	198.46	133.40
FAB1	238.85	494.79	363.63	237.79	492.60	362.01	236.22	489.34	359.62	232.63	481.90	354.16
FAB2	188.02	598.25	371.34	187.19	595.59	369.69	185.95	591.66	367.24	183.12	582.67	361.66
FAB3	137.19	407.98	255.49	136.58	406.17	254.36	135.68	403.48	252.68	133.62	397.35	248.84
FAB4	172.28	350.40	229.76	171.51	348.85	228.74	170.38	346.54	227.23	167.79	341.28	223.78

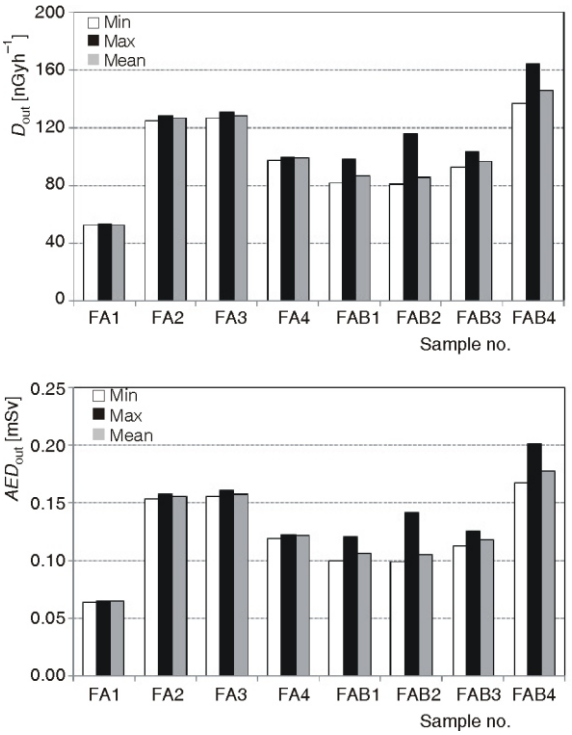

Figure 4. Calculated values of the outdoor air absorbed dose rate (D_{out}) and the corresponding annual effective dose (AED_{out}) in the studied samples

rate (D_{out}, nGy/h) ranged from 52.29 to 163.90 nGy/h with an average of 102.57 nGy/h (fig. 4), which were close to or significantly higher than the population-weighted average value of the global primordial radiation of 59 nGy/h [6]. The D_{out} values for most samples (97 %) were higher than the average outdoor natural gamma radiation dose rate of China (62.0 nGy/h) [6] and Baotou (61.8 nGy/h) [27]. The corresponding annual effective dose rate (AED_{out}), considering the conversion coefficient from the gamma absorbed dose in air to the effective dose (0.7 Sv/Gy) and outdoor occupancy factor (0.2) proposed by UNSCEAR [1], varied from 0.064 to 0.201 mSv with an average of 0.125 mSv (fig. 4). The AED_{out} values in most samples (97 %) were higher than the worldwide average (0.07 mSv) for the outdoor annual effective dose rate [6].

CONCLUSIONS

Radioactivity levels, ^{222}Rn and ^{220}Rn exhalation rates and radiation hazards of fly ash and fly ash brick used in Baotou, China, have been determined in this work. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in all investigated samples are in the range of Chinese soil values. No corresponding correlations were observed in the ^{226}Ra, ^{232}Th, and ^{40}K activity concentrations between fly ash brick and fly ash from the same investigation site. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in fly ash brick depend on the natural radioactivity levels of the raw materials and their usages. The exhalation rate values of ^{222}Rn and ^{220}Rn in all investigated samples ranged from 1.13-20.50 and 15.60-113.00 mBqm^{-2}s^{-1}, respectively. The ^{222}Rn exhalation rate values in all samples are lower than the ^{220}Rn exhalation rate values. The radium equivalent activity and the external hazard index values of all samples are lower than the recommended value of 370 Bq/kg and unity, respectively. Due to containing higher ^{226}Ra and/or ^{232}Th concentrations, the internal hazard index and the indoor annual effective dose rate values of some fly ash and fly ash brick samples were larger than unity and 1 mSv, respectively. The outdoor annual effective dose rate values in most samples were higher than the worldwide average value for the outdoor annual effective dose rate. The contribution of the investigated samples to the indoor air ^{222}Rn concentration obviously decreases with the indoor air exchange rate increasing, whereas no similar phenomenon was found for the indoor air ^{220}Rn concentration. The indoor annual exposure effective dose due to ^{220}Rn released from the investigated samples is much lower than for ^{222}Rn. The fly ash and fly ash brick collected from some brick factories of Baotou would pose an excessive radiation risk to inhabitants and they are not suitable for use in building construction. The management of usage of fly ash and fly ash brick, and their radioactivity level measurement should be strengthened by the local government and environmental protection agency.

AUTHORS' CONTRIBUTIONS

All samples were collected by X. Lu and X. Zhang. Experiments were finished by Y. Li under supervision and guidelines of X. Lu. All authors discussed the results. Figures were prepared by X. Zhang. The manuscript was written by Y. Li and reviewed by X. Lu.

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ОДРЕЂИВАЊЕ ПРИРОДНЕ РАДИОАКТИВНОСТИ, ЈАЧИНЕ ЕКСХАЛАЦИЈЕ ^{222}Rn И ^{220}Rn И РАДИЈАЦИОНОГ РИЗИКА ОД ЛЕТЕЋЕГ ПЕПЕЛА ГРАЂЕВИНСКОГ МАТЕРИЈАЛА КОРИШЋЕНОГ У БАОТОУ У КИНИ

У овом раду одређени су нивои природне радиоактивности јачине ексхалације ^{222}Rn и ^{220}Rn и радијациони ризици од летећег пепела и летећег пепела грађевинског материјала коришћеног у Баотоу у Кини. Концентрације активности ^{226}Ra, ^{232}Th, и ^{40}K у узорцима летећег пепела биле су у опсезима 38.81-93.73 Bq/kg, 40.34-135.17 Bq/kg, и 66.92-290.86 Bq/kg, са средњим вредностима од 76.52 Bq/kg, 109.95 Bq/kg и 170.72 Bq/kg, респективно; док су у узорцима грађевинског материјала са летећим пепелом концентрације активности за ове радионуклиде износиле 42.43-71.60 Bq/kg, 76.65-208.37 Bq/kg и 94.32-489.42 Bq/kg, са средњим вредностима од 53.83 Bq/kg, 101.93 Bq/kg и 266.48 Bq/kg, респективно. Јачине ексхалације ^{222}Rn и ^{220}Rn у свим узорцима кретале су се у распону 1.13-20.50 mBq/m^{2}s и 15.60-113.00 mBq/m^{2}s, респективно. Израчунате вредности еквивалентне активности радијума, индекса спољашњег ризика, индекса унутрашњег ризика, годишња ефективне дозе у затвореном простору и годишње ефективне дозе у животној средини, указују да летећи пепео и грађевински материјал са летећим пепелом из неких фабрика у Баотоу представаљају значајан радијациони ризик за становништво и да нису погодни за примену у градњи зграда. Ниво природне радиоактивности летећег пепела и грађевинског материјала са летећим пепелом морају бити под сталним мониторингом ради радијационе безбедности локалног становништва.

Кључне речи: природна радиоактивност, јачина ексхалације радона, радијациони ризик, летећи пепео, грађевински материјал