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ORIGINAL ARTICLE

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Measurement of natural radioactivity in sand samples collected from the Baoji Weihe Sands Park, China

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Abstract The activity concentrations and the gamma-absorbed dose rates of the primordial naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K were determined for sand samples collected from the Baoji Weihe Sands Park, China, using γ-ray spectrometry. The natural radioactivity concentration of sand ranges from 10.2 to 38.3 Bq kg^{-1} for ^{226}Ra, 27.0 to 48.8 Bq kg^{-1} for ^{232}Th and 635.8 to 1,126.7 Bq kg^{-1} for ^{40}K with mean values of 22.1, 39.0 and 859.1 Bq kg^{-1}, respectively. The concentrations of these radionuclides are compared with the typical world values and the average activity of Chinese soil. The measured activity concentration of ^{226}Ra and ^{232}Th in sand is lower than the world average while that of ^{40}K is higher. . To evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity, the external hazard index, the absorbed dose rate, and the effective dose rate have been calculated and compared with internationally approved values. The radium equivalent activity values of all sand samples are lower than the limit of 370 Bq kg^{-1}. The values of the external hazard index are less than unity. The mean outdoor air absorbed dose rate is 69.6 nGy h^{-1} and the corresponding outdoor effective dose rate is 0.085 mSv y^{-1}.

Keywords Natural radioactivity · Sand · γ-Ray spectrometry · Gamma dose rate · Sands Park · China

Introduction

Exposure to ionizing radiation is generally undesirable at all levels by the public, although no harmful effects are presently proven for very low exposure (UNSCEAR 1993). Studies on the background to natural radiation are of great importance because it is the main source of exposure for humankind. The natural radionuclides produce environmental dose levels that are the most significant component of the total environmental dose received by man.

Sediment sand of the river was formed through weathering and erosion of rock and soil. Natural radioactivity in soil, sand and rock comes from ^{226}Ra, ^{232}Th and ^{40}K. The study of the distribution of primordial radionuclides allows the understanding of the radiological implication of these elements due to the γ-ray exposure of the body and irradiation of lung tissue from inhalation of radon and its daughters (Alam et al. 1999; Singh et al. 2005; Veiga et al. 2006). In particular, it is important to assess the radiation hazards arising due to the use of soil or sand in the construction of dwellings (Khatibeh et al. 1997; Kumar et al. 2003). Therefore, the assessment of γ-radiation dose from natural sources is of particular importance as natural radiation is the largest contributor to the external dose of the world population (UNSCEAR 2000). These dose rates vary depending upon the concentration of the natural radionuclides—^{226}Ra, ^{232}Th and ^{40}K—present in soil, sand, and rock, which in turn depend upon the local geology of

each region in the world. As these radionuclides are not uniformly distributed, the knowledge of their distribution in soil, sand, and rock play an important role in radiation protection and measurement. Several authors have studied the levels of natural background radiation by in situ measurements or by analysis of radionuclide concentration in soil or sand samples (Ahmad et al. 1997; Alam et al. 1999; Alencar and Freitas 2005; El-Arabi 2005; Fasasi et al. 2003; Freitas and Alencar 2004; González-Chornet and González-Labajo 2004; Malanca et al. 1996; Matiullah et al. 2004; Veiga et al. 2006).

Baiji Weihe Sands Park (China), with an area of about 3 km^2, was built in 2000. Entrance is free to citizens. Most citizens go there to walk and rest there during off duty hours. To determine the radiation hazard in this area, NaI(Tl) γ-ray spectrometer was used to find the concentration of natural radionuclides—^{226}Ra, ^{232}Th and ^{40}K—in sand samples collected from Baoji Weihe Sands Park. The radium equivalent activity, the external hazard index, the absorbed dose rate, and the effective dose rates were calculated and compared with internationally approved values.

Materials and methods

Sample collection and preparation

Sand samples were collected from the tourist sites of Baoji Weihe Sand Park, China. Figure 1 shows the Baoji Weihe Sands Park location and the sampling site locations. At every sampling site, sand samples were collected from the surface layer (0–20 cm depth) of four corners and the center of a square area corresponding to 1 m^2. The five samples were homogenized in situ, and this sand mixture, weighing approximately 1.5 kg, was considered representative of the sampling site. In the laboratory, the samples were crushed, sieved through a 1 mm mesh, and dried for about 24 h in an oven at 110°C (Benke and Kearfott 1999; Veiga et al. 2006). Finally, weighed (1,000 ± 1 g) samples were stored in gas-tight, radon impermeable, trap-shape polyethylene containers (10 cm diameter and 16 cm height, the diameter and depth of trap is 6 cm and 9 cm, respectively). These containers were hermetically sealed to prevent the escape of gaseous ^{222}Rn and ^{220}Rn from the samples and kept aside for about 30 days to ensure radioactive equilibrium (Alencar and Freitas 2005). After that period, the samples were analyzed using gamma spectrometry.

Calibration and measurement by gamma ray spectrometry

The concentration of natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) in the samples was determined using a 5×5 cm NaI(Tl) γ-ray spectrometric system with excel 8% energy resolution (^{137}Cs 661 KeV) and 20% counting efficiency. The detector is maintained in a vertical position in a lead cylindrical shield. The detector was coupled to a 256 multi-channel pulse height analyzer and the system was calibrated for the γ-energy range 80 KeV—3.2 MeV. The energy region for ^{40}K 1.46 MeV γ-rays, ^{226}Ra 1.76 MeV γ-rays (Bi−214) and ^{232}Th 2.61 MeV γ-rays (Tl-208) were chosen as 1.30–1.60, 1.62–2.00 and 2.45–2.90 MeV, respectively. The standard sources for ^{226}Ra and ^{232}Th (in secular equilibrium with ^{228}Th) were prepared using known activity contents and mixing it with the matrix material of phthalic acid powder. To avoid the loss of gaseous daughter products of ^{226}Ra and ^{232}Th that may lead to disturbance in radioactive equilibrium, the prepared standard sources were kept in sealed trap-shape polyethylene containers (10 cm diameter and 16 cm height, the diameter and depth of trap were 6 cm and 9 cm, respectively). Analar grade potassium chloride (KCl) of a known amount of the same geometry was used as the standard source of ^{40}K.

The spectra were collected for each standard material and background over counting times of 400 min each. The spectral windows were adjusted around the 1.46, 1.76 and 2.61 MeV energy peaks. The sample counting time was 400 min. Each sample was counted twice before an average was taken. The characteristics of the γ-ray spectrometer used are given in Table 1. The derivation of ^{226}Ra, ^{232}Th and ^{40}K concentrations of samples was performed in two steps. First, it was necessary to identify the peaks from ^{226}Ra, ^{232}Th and ^{40}K and the energy regions where they are well resolved in the energy spectra of standards and samples. In the second step, as there was some overlap between the γ-ray peaks from potassium, uranium series and thorium series, a stripping technique, described by Chiozzi et al. (2000), was applied to measure the natural radioactivity levels of the samples, making all the required corrections. By this technique, the count rate in each of the three spectral windows is related to the concentrations of the radioactive elements in the standards by the following set of equations (Iqbal et al. 2000)

$$R_{ij} = \sum \left(A_{ik} C_{kj}\right) + B_i, \tag{1}$$

where R_{ij} is the count rate for the ith region of the jth standard, A_{ik} is the calibration coefficient and B_i is the background count rate. Equation 1 can be expressed in matrix form as:

$$[R] = [A][C] + [B], \tag{2}$$

where $[R]$ is a 3×1 matrix of the observed count rates, $[A]$ is a 3×3 matrix of the calibration coefficients, $[C]$ is a 3×1 matrix of the nuclide concentrations and $[B]$ is a 3×1 matrix of the background count rates. Inverting the $[A]$ matrix can solve the above equation:

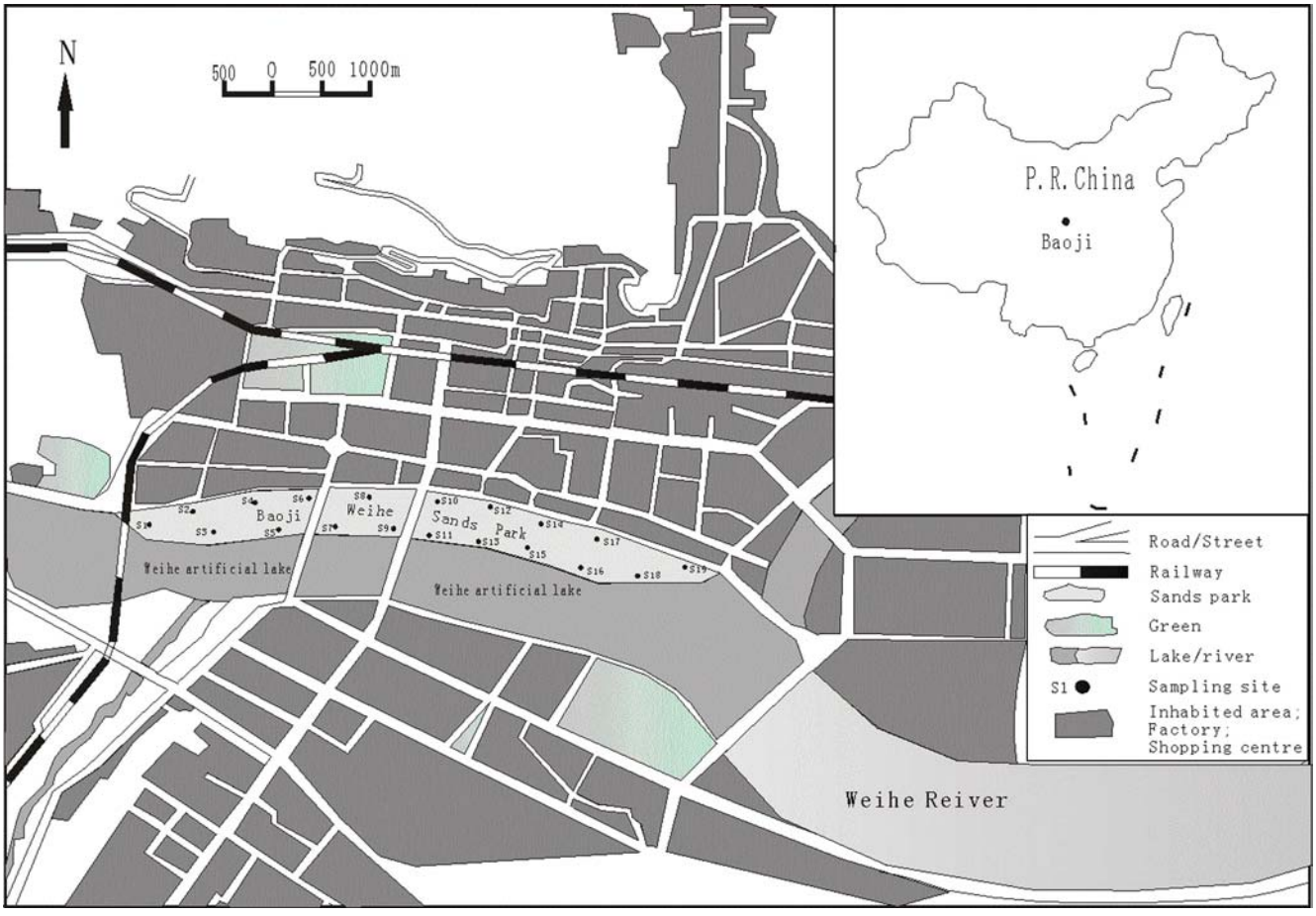

Fig. 1 Location of the study area and the sampling site

$$[C] = \left[A^{-1}\right]\{[R] - [B]\}. \quad (3)$$

The activity mass concentration was calculated using the following relations:

$$C_{\mathrm{K}} = A^{-1}\{R_{\mathrm{K}} - B_{\mathrm{K}}\}, \quad (4)$$

$$C_{\mathrm{Ra}} = A^{-1}\{R_{\mathrm{Ra}} - B_{\mathrm{Ra}}\}, \quad (5)$$

$$C_{\mathrm{Th}} = A^{-1}\{R_{\mathrm{Th}} - B_{\mathrm{Th}}\}. \quad (6)$$

Table 1 Characteristics of radionuclides and γ-ray spectrometer

Radionuclide	$T_{1/2}$(years)	Nuclide	Γ-ray energy (MeV)
^{226}Ra	$1{,}599 \pm 7$	^{214}Bi	1.76
^{232}Th	$1.41 \pm 0.01 \times 10^{10}$	^{208}Tl	2.61
^{40}K	$1.2771 \pm 0.008 \times 10^{9}$		1.46

The matrix $[A]$ in Eq 2 was determined from the known activity concentration of the standard material and the count rates of the specified windows after background subtraction. The inverse matrix A^{-1} was found and used for determination of the unknown activity concentration.

Results and discussion

Activity in sand of Baoji Weihe Sands Park

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in sand samples from Baoji Weihe Sands Park are shown in Table 2. The activity concentrations of ^{226}Ra range from 10.2 to 38.3 Bq kg^{-1} with an average of 22.1 Bq kg^{-1}. The activity concentrations of ^{232}Th range from 27.0 to 48.8 Bq kg^{-1} with an average of 39.0 Bq kg^{-1}. The activity concentrations of ^{40}K range from 635.8 to 1126.7 Bq kg^{-1} with an average of 859.1 Bq kg^{-1}. All the values of the activity per unit mass are in the ranges of the corresponding typical world values (UNSCEAR 1993) which are 50 and 50 Bq kg^{-1} for ^{226}Ra and ^{232}Th, respectively, except for ^{40}K (the corresponding typical

Table 2 Characteristics of the radioactivity in sand samples collected from the Baoji Weihe Sands Park, China

Sample number	Radioactivity concentration (Bq kg^{-1})			Ra_{eq} (Bq kg^{-1})	H_{ex}	Dose rate (nGy h^{-1})	Effective dose rate (mSv y^{-1})
	^{226}Ra	^{232}Th	^{40}K				
S1	24.5	45.8	780.6	150.1	0.41	71.5	0.088
S2	24.1	40.6	733.3	138.6	0.37	66.2	0.081
S3	27.2	36.8	713.4	134.8	0.36	64.5	0.079
S4	35.1	32.9	777.6	142.0	0.38	68.5	0.084
S5	30.4	39.8	730.3	143.5	0.39	68.5	0.084
S6	14.9	34.3	1,126.7	150.7	0.41	74.6	0.091
S7	11.3	30.6	1,094.6	139.3	0.38	69.3	0.085
S8	20.0	30.9	1,090.8	148.2	0.40	73.4	0.090
S9	10.2	31.2	1,075.6	137.6	0.37	68.4	0.084
S10	15.2	45.8	856.5	146.6	0.40	70.4	0.086
S11	14.4	44.7	843.7	143.3	0.39	68.8	0.084
S12	17.8	43.9	857.2	146.6	0.40	70.5	0.086
S13	19.3	42.7	922.8	151.4	0.41	73.2	0.090
S14	16.9	27.1	1,070.2	138.1	0.37	68.8	0.084
S15	19.8	27.0	1,031.9	137.9	0.37	68.5	0.084
S16	26.7	48.5	660.8	146.9	0.40	69.2	0.085
S17	38.3	41.9	635.8	147.2	0.40	69.5	0.085
S18	25.1	48.1	654.9	144.3	0.39	68.0	0.083
S19	28.7	48.8	666.9	149.8	0.40	70.5	0.087
Mean	22.1	39.0	859.1	144.1	0.39	69.6	0.085

world value is 500 Bq kg^{-1}). The average values of ^{226}Ra and ^{232}Th concentrations in Baoji Weihe sands are lower than the average values in soils of China (Wang 2002) and the average values in soils of Shaanxi (Chen et al. 1994), while the average value of ^{40}K in Baoji Weihe sands is higher than the average concentration for Shaanxi soil (Chen et al. 1994) and Chinese soil (Wang 2002). The activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in all sand samples from Baoji Weihe Sands Park are in the range of Chinese soil values.

Dose rate in sand

An attempt has been made in the present study to provide a characteristic of the external primordial γ-radiation. The total air absorbed dose rate (nGy h^{-1}) 1 m above the ground due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) was calculated using the formula (UNSCEAR 2000; Veiga et al. 2006)

$$\mathrm{D}\left(\mathrm{nGyh}^{-1}\right) = 0.0417C_{\mathrm{K}} + 0.462C_{\mathrm{Ra}} + 0.604C_{\mathrm{Th}} \quad (7)$$

Table 2 gives the estimated external gamma dose rate due to natural gamma emitters as measured in the sand. The results show that the absorbed dose rates range from 64.5 to 74.6 nGy h^{-1} with mean value of 69.6 nGy h^{-1}, which is close to the average value of natural gamma radiation dose rate of Baoji (63.0 nGy h^{-1}) (Zhang and Li 1994). All the calculated dose rates were higher than the estimate of average global primordial radiation of 55 nGy h^{-1} and are comparable with the world range (28–120 nGy h^{-1}) (UNSCEAR 1993) and lower than the Chinese average value (81.5 nGy h^{-1}) (Wang 2002). To estimate the annual effective dose rates, the conversion coefficient from the absorbed dose in the air to the effective dose (0.7 Sv Gy^{-1}) and the outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) were used. The effective dose rate was calculated from the formula (Yang et al. 2005):

$$\text{Effective dose rate (mSv per year)} = D\left(\text{nGy h}^{-1}\right) \times 8,760\left(\text{hy}^{-1}\right) \times 0.2 \times 0.7\left(\text{Sv Gy}^{-1}\right) \times 10^{-6} \quad (8)$$

The results of the calculation are presented in Table 2. The effective dose rates in the air varied from 0.079 to 0.091 mSv y^{-1} with an average value of 0.085 mSv y^{-1}. The calculated effective dose rates are lower than the mean value of Shaan effective dose rate from natural gamma radiation (Zhang and Li 1994). Figure 2 shows the relative contributions of ^{226}Ra, ^{232}Th and ^{40}K contents to the absorbed dose rate and the effective dose rate for all sand samples. The average relative contribution by individual components of natural radioactivity is 15% from ^{226}Ra, 34% from ^{232}Th and 51% from ^{40}K. In areas with normal background radiation, the average annual external effective dose rate from the primordial radionuclides is 0.46 mSv y^{-1} (UNSCEAR 1993; Yang et al. 2005). The obtained values of natural radioactivity and γ-absorbed dose rates due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of sand in the air show that Baoji Weihe Sands Park can be regarded as an area with normal natural background radiation (UNSCEAR 1993, 2000).

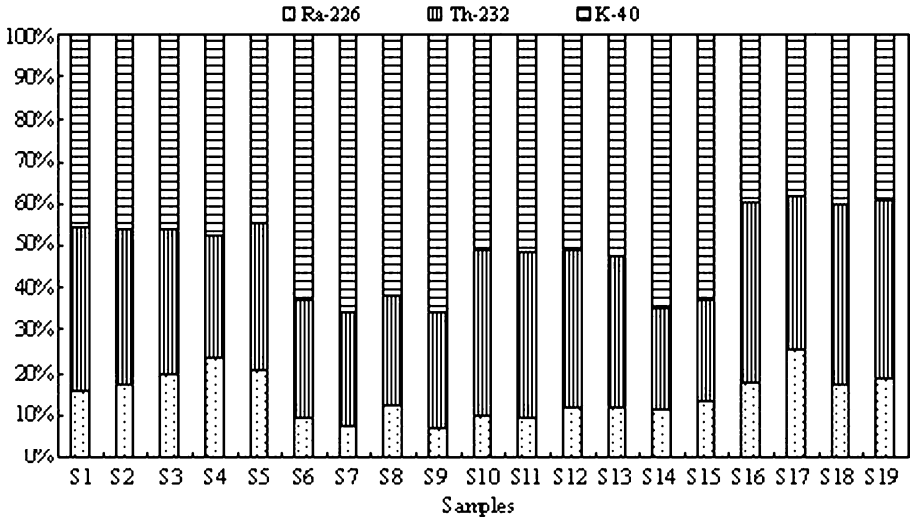

Fig. 2 Relative contributions to the absorbed dose rate and effective dose rate owing to ^{226}Ra, ^{232}Th and ^{40}K for the samples

γ-Ray radiation hazard indexes

As sand from Baoji Weihe River is commonly used as a building material, it is important to assess the γ-Ray radiation hazards of sand to humans. The γ-ray radiation hazards due to the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were assessed by different radiation hazard indices. The most widely used radiation hazard index, radium equivalent activity (Ra_{eq}), can be calculated using the formula as (Beretka and Mathew 1985):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (9)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The values of Ra_{eq} for the studied sand samples are given in Table 2. As can be seen from Table 2, the Ra_{eq} values for the sand samples from Baoji Weihe Sands Park varied from 134.8 to 151.4 Bq kg^{-1}, with the average of 144.1 Bq kg^{-1}. All the Ra_{eq} values of samples are below the internationally accepted value 370 Bq kg^{-1}.

The external hazard index (H_{ex}) is another radiation hazard index defined by Beretka and Mathew (1985) to evaluate the indoor radiation dose rate due to the external exposure to γ-radiation from the natural radionuclides in the construction building materials of dwellings. This index value must be less than unity to keep the radiation hazard insignificant, i.e. the radiation exposure due to the radioactivity from construction materials to be limited to 1.5 mSv y^{-1} based on the formula (Beretka and Mathew 1985):

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (10)$$

where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. For the maximum value of H_{ex} to be less than unity, the maximum value of Ra_{eq} must be less than 370 Bq kg^{-1}. The

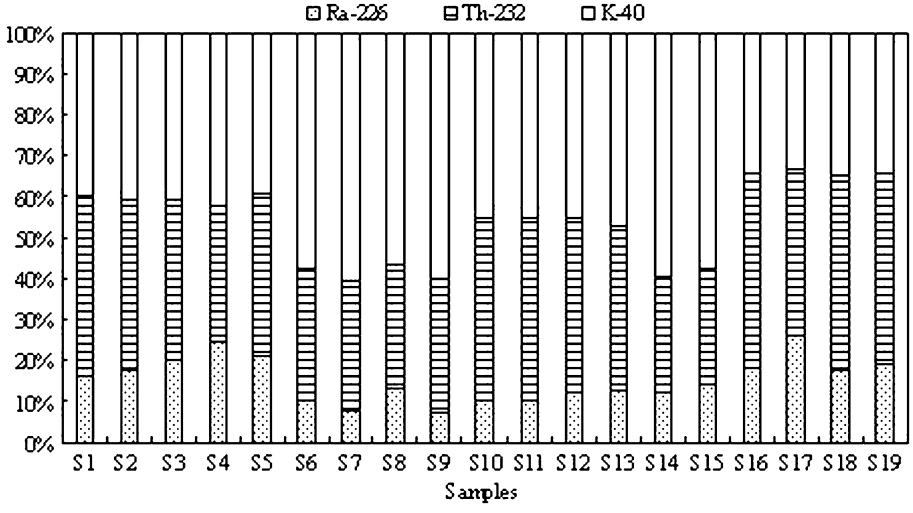

Fig. 3 Relative contributions to the Ra_{eq} and H_{ex} rate owing to ^{226}Ra, ^{232}Th and ^{40}K for the samples

calculated results of H_{ex} for the study samples range from 0.36 to 0.41 with an average of 0.39 as shown in Table 2, values which indeed are less than unity. Figure 3 shows the relative contributions of ^{226}Ra, ^{232}Th and ^{40}K contents to the Ra_{eq} and the H_{ex} for all sand samples. The average relative contribution by individual components of natural radioactivity is 15% from ^{226}Ra, 39% from ^{232}Th and 46% from ^{40}K.

Conclusions

The natural radioactivity of sand from Baoji Weihe Sands Park was measured by using NaI(Tl) γ-ray spectrometer. The results show that the average concentrations of ^{226}Ra and ^{232}Th in the studied sand samples are lower than the average values of soil in China and the soil of Shaanxi, while the average concentration of ^{40}K is higher than the average values for Shaanxi soil and Chinese soil. In Baoji Weihe Sands Park, the absorbed dose rates range from 64.5 to 74.6 nGy h^{-1} with a mean value of 69.6 nGy h^{-1}. All the calculated dose rates were higher than the estimate of average global primordial radiation of 55 nGy h^{-1}. The effective dose rates in the air varied from 0.079 to 0.091 mSv y^{-1} with an average value of 0.085 mSv y^{-1}. The obtained values of natural radioactivity and γ-absorbed dose rates due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of sand in the air show that Baoji Weihe Sands Park can be regarded as an area with normal natural background radiation, and the sand can be safely used in construction.

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