

Specific Activity of ^{226}Ra , ^{232}Th and ^{40}K for Assessment of Radiation Hazards from Building Materials Commonly Used in Upper Egypt

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Abstract: In this paper, the specific activity of natural radionuclides (^{226}Ra , ^{232}Th , and ^{40}K) in some building materials, soil, sand, redbrick, clay brick, limestone, alabaster and marble commonly used in Upper Egypt is presented. Measurements were done by using gamma spectrometry (NaI (TI) 3"x 3"). Concentrations of natural radionuclides (C_{Ra} , C_{Th} and C_K), radium equivalent (Ra_{eq}), external hazard index (H_{ex}), the specific dose rates in door (D) and the annual effective dose (DE) due to gamma radiation from building materials was calculated. Concentrations of natural radionuclides (^{226}Ra and ^{232}Th) are in usual range and below maximal permitted values. The lowest value of (H_{ex}) is 0.15 for sand while the highest one is 0.5 for Redbrick. The ranges of (DE) are between 0.9 and 3.5 μSv^{-1} , it is below maximal permitted values, so that examined materials can be used for construction of new buildings (for interior and external works) as well as for covering of pavements, floors, etc.

Key words: Building materials, activity concentration index, dose rate, annual effective dose

Yukarı Mısır Bölgesinde Yaygın Olarak Kullanılan Yapı Malzemelerindeki Radyasyon Riskinin Değerlendirilmesi Açısından ^{226}Ra , ^{232}Th ve ^{40}K 'ın Özgül Aktivitelerinin Belirlenmesi

Özet: Yukarı Mısır bölgesinde yaygın olarak kullanılan toprak, kum, kıvıllı tuğla, killi tuğla, kireç taşı, kaymak taşı ve mermer gibi bazı yapı malzemelerinde bulunan doğal radyoçekerdeklerin (^{226}Ra , ^{232}Th ve ^{40}K) özgül aktiviteleri bu makalede sunulmaktadır. Ölçümler 3"x 3" NaI (TI) gama spektrometresi kullanılarak yapılmıştır. Yapı malzemelerinden gelen gama radyasyonu ölçülerek, doğal radyoçekerdek yoğunlukları (C_{Ra} , C_{Th} ve C_K), radyum eşdeğer dozu (Ra_{eq}), harici risk indisi (H_{ex}), bina içi özgül doz oranları (D) ve yıllık etkin doz (DE) değerleri hesaplanmıştır. Doğal radyoçekerdek (^{226}Ra ve ^{232}Th) yoğunlukları olağan sınırlar içerisinde ve izin verilen en büyük değerlerin altındadır. En küçük H_{ex} değeri 0.15 (kum için) iken, en büyüğü ise 0,5'tir (kıvıllı tuğla için). DE parametresi 0,9 $\mu\text{Sv/y}$ – 3,5 $\mu\text{Sv/y}$ aralığında ve izin verilen en büyük değerinin altında olduğundan, incelenen materyallerin, taban, tavan, vb. yerlerin kaplanmasında kullanılmasının yanı sıra yeni yapıların iç ve dış kısımlarında da kullanılması uygundur.

Anahtar kelimeler: Yapı malzemeleri, aktivite yoğunluk indisi, doz oranı, yıllık etkin doz

1. Introduction

The exposure of human beings to ionizing radiation from natural sources is a continuing and inescapable feature of life on earth. There are two main contributors to natural radiation exposures: high-energy cosmic ray particles incident on the earth's atmosphere and radioactive nuclides that originated in the earth's crust and are present everywhere in the environment, including the human body itself.

It is well known that radioactive nuclides in the uranium and Thorium decay chains do occur with varying degrees of concentration in the earth's crust. While radioactive nuclides such as radium (^{226}Ra), radon (^{222}Rn) and bismuth (^{214}Bi) are the product in the decay chain of uranium (^{238}U), other radioactive nuclides, such as actinium (^{228}Ac), bismuth (^{212}Bi) and lead (^{212}Pb) do occur in the decay chain of the thorium element (^{232}Th). In addition, the radionuclide (^{40}K) does also occur in construction materials.

These radioactive elements can be found almost in all types of building materials containing naturally occurring radionuclides are the main source of exposure. The knowledge of natural radioactivity in these materials is then important for determining the amount of public exposure because people spend most of their time (about 80%) indoors [1]. Furthermore, knowledge of this radioactivity is useful in setting the standards and national guidelines in regard to the international recommendations and in assessing the associated radiation hazard.

The population-weighted average of indoor absorbed dose rate in air from terrestrial sources of radioactivity is estimated to be 84 nGy h^{-1} [2]. Elevated indoor external dose rates may arise from high activities of radionuclides in building materials. Large-scale surveys of concentrations of radioisotopes in construction materials were summarized by the United Nations Scientific Committee on the Effects of Atomic Radiation [3]. Consequently, this study was undertaken with the purpose of determining radioactivity in some Egyptian building materials and to assess the annual effective dose to the Egyptian population due to external gamma ray exposure in dwellings typical of Upper Egypt.

2. Materials and Methods

2.1. Sample Description and Preparation

A total of 43 samples of 7 different buildings materials six samples from each type (sand, redbrick, clay brick, limestone, alabaster and marble) while seven samples from soil were collected for the measurements of activity concentrations from Qena and Assiut governments in Upper Egypt. The materials were obtained from suppliers or gathered directly in demolished houses or buildings under construction. A brief description of these building materials follows.

Sand is a granular material made up of fine rock particles. The most common constituent of sand is silica (silicon dioxide, or SiO_2), usually in the form of quartz. The composition of sand varies according to local rock sources and conditions. Bricks may be made from clay, shale, soft slate, and calcium silicate. Limestone rocks are sedimentary rocks that are made from the mineral calcite, which comes from the beds of evaporated seas and lakes and from seashells.

Each sample was dried in an oven at about 110°C to ensure that moisture was completely removed. The samples were crushed, homogenized and sieved through a $200 \mu\text{m}$, which is the optimum size enriched in heavy minerals. Weighed samples were placed in polyethylene beaker, of 350-cm^3 volumes each. The beakers were completely

sealed for 4 weeks to reach secular equilibrium where the rate of decay of the progeny becomes equal to that of the parent (radium and thorium) within the volume and the progeny will also remain in the sample [4, 5].

2.2. Instrumentation and Calibration

Activity measurements were performed by gamma ray spectrometer, employing a scintillation detector (3" x 3"). It is hermetically sealed assembly, which includes a NaI (Tl) crystal, coupled to PC-MCA Canberra Accuspecs. To reduce gamma ray background, a cylindrical lead shield (100 mm thick) with a fixed bottom and movable cover shielded the detector. The lead shield contains an inner concentric cylinder of copper (0.3 mm thick) in order to absorb X-rays generated in the lead. In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The measurement time of activity or background was 43200s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program [6], from Canberra has carried out the online analysis of each measured gamma ray spectrum. The ^{232}Th concentration was determined from the average concentrations of ^{212}Pb (238.6 keV,) and ^{228}Ac (911.1 keV) in the samples, and that of ^{226}Ra was determined from the average concentrations of the ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 keV and 1764.5 keV) decay products. While the gamma line for ^{40}K is (1460.6 keV). The minimum detectable activity (MDA) was 25.2 Bqkg⁻¹ for ^{40}K , 6.5 Bqkg⁻¹ for ^{226}Ra and 5.7 Bqkg⁻¹ for ^{232}Th . All procedures and efficiency calibration are described in previous publications [7].

3. Results and Discussion

Activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in controlled samples of building materials are shown in Table 1.

Table 1. Activity concentrations range of ^{226}Ra , ^{232}Th and ^{40}K in different samples

Sample Name	Number Of samples	^{226}Ra A in (Bqkg ⁻¹)	^{232}Th A in (Bqkg ⁻¹)	^{40}K A in(Bqkg ⁻¹)
Soil	7	31±2 to 40±2	52±3 to 61± 3	149±7 to 210±11
Sand	6	11±1 to 29± 1	17±1 to 70 ±4	144±7 to155 ±8
Redbrick	6	54±3 to 65±3	65±3 to 81±4	285±14 to 376±19
Clay brick	6	16±1 to 52±3	30±2 to 89±5	167±8 to228±14
Limestone	6	23±1 to 74±3	17±2 to 51±3	115±10 to 135±13
Alabaster	6	29± 2 to 38 ±2	8.4±1 to13±1	121±13 to 156±14
Marble	6	32± 2 to51± 4	10±1 to14±1	124±12 to 178±17

From Table 1, it can be seen that ^{40}K always contributes to the most specific activity compared to ^{226}Ra and ^{232}Th . For different samples, the largest activity of ^{226}Ra is 74± 3 Bqkg⁻¹ for limestone, it is five times greater than that of the lowest value 11±1 Bq kg⁻¹ found in sand. ^{232}Th is in the wide range from 8.4±1 Bqkg⁻¹ in alabaster up to 89±5

Bqkg⁻¹ in clay bricks. Concentration of ⁴⁰K values ranges from 115±10 Bqkg⁻¹ in limestone to 376±19 Bqkg⁻¹ in redbrick.

Thus activities in redbrick were higher than in other building materials. This may suggest that it is advisable to monitor the radioactivity levels of materials from a new source before adopting it for use as a building material. We can say that, all materials under investigation would not present a significant radiological hazard when it is used in building constructions.

3.1. Radium Equivalent Activity and External Hazard Index

²²⁶Ra, ²³²Th and ⁴⁰K are non-uniformly distributed in building materials. In order to compare the specific activity of materials containing different amounts of ²²⁶Ra, ²³²Th and ⁴⁰K, the radium equivalent activity Ra_{eq} is used as defined by the following expression [8]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

Where A_{Ra} , A_{Th} and A_K are the mean activity in Bqkg⁻¹ of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. Eq. (1) is based on the fact that 370 Bqkg⁻¹ of ²²⁶Ra, 259 Bqkg⁻¹ of ²³²Th and 4810 Bqkg⁻¹ of ⁴⁰K, produce the same γ -ray dose equivalent. Column 2 of Table 2 summarizes the Ra_{eq} results for all the samples studied. These values range from 65 Bq kg⁻¹ in Alabaster to 187 Bq kg⁻¹ in redbrick. Thus, all materials will not present a significant radiological hazard when they are used in building constructions. However, from the results (Ra_{eq}), there is no considerably in the different materials and in the same type of material collected from different areas.

While the external hazard index (H_{ex}): is given by the following equation [9]:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (2)$$

Where C_{Ra} , C_{Th} and C_K are the concentration in BqKg⁻¹ of radium, thorium and potassium, respectively. This index must be less than unity so that the annual effective dose due to radioactivity in the material will be less or equal to 1.5 mSv. As indicated in Table 2, it appears that investigated materials meet this criterion.

3.2. Absorbed Dose, Activity Concentration Index and Annual Effective Dose Rate

For materials containing naturally occurring radioactive materials such as ²²⁶Ra, ²³²Th and ⁴⁰K, the absorbed dose rate D_o can be defined if the radionuclide concentrations are known. It can be obtained in units of nGyh⁻¹ using the formula:

$$D_o = 0.00333C_{Ra/200} + 0.005 C_{Th} + 0.000333C_K \quad (3)$$

Where C_{Ra} , C_{Th} and C_K are the concentration in (BqKg⁻¹) of radium, thorium and potassium respectively. Column 4 of Table 2 gives the results for absorbed dose rate in

air for building materials under investigation. We notice that redbricks show the highest value (0.7 nGy h^{-1}), where as the lowest value is found in sand (0.2 nGy h^{-1}).

Table 2: Range of Radium equivalent activity Ra_{eq} , external hazard index H_{ex} , absorbed dose rate D_o , and annual effective dose DE in building materials under investigation.

Sample name	Ra_{eq} (Bq kg ⁻¹)	H_{ex}	Dose rate D_o (nGyh ⁻¹)	Annual Effective Dose DE (μSv)
Soil	116.6 to 143.7	0.32 to 0.40	0.41 to 0.48	2.0 to 2.5
Sand	52.5 to 140.8	0.15 to 0.37	0.2 to 0.5	0.9 to 2.4
Redbrick	169.7 to 203.9	0.32 to 0.38	0.6 to 0.7	2.4 to 3.5
Clay brick	77.8 to 201.6	0.2 to 0.5	0.28 to 0.7	1.3 to 3.5
Limestone	66.8 to 118.1	0.2 to 0.3	0.3 to 0.4	1.0 to 2.0
Alabaster	59.3 to 70.3	0.1 to 0.2	0.18 to 0.25	0.9 to 1.2
Marble	59.8 to 79.8	0.16 to 0.22	0.21 to 0.28	1.0 to 1.4

The results are shown in Table 2 and these values were used to calculate annual effective doses, DE , due to gamma radiation from building materials was calculated as [10]:

$$DE = 0.7 \text{ SvGy}^{-1} \cdot 7000 \text{ h} \cdot D_o \tag{4}$$

Where D_o must be taken in μGyh^{-1} and 0.7 SvGy^{-1} is effective absorbed dose conversion factor and 7000 h is annual exposure time. Results are presented in Table 2 and the DE values are between 0.6 and $2.8 \mu\text{Svy}^{-1}$. According to Ref. [2], the annual effective dose of these samples doesn't exceed the average worldwide exposure of 2.4 mSv due to natural sources. Table 3 shows the average radionuclide concentrations and radium equivalent activities Ra_{eq} in building materials from other investigations for comparison.

Table 3: Comparison of activity and radium equivalent activities Ra_{eq} in building materials used in Upper Egypt with those of other countries.

Material	Country	Activity (Bq kg ⁻¹)			Ra_{eq}	Reference
		²³⁸ U	²³² Th	⁴⁰ K		
Sand	Algeria	12±1	7±1	74±7	28±2.1	Amrani and Tahtat (2001)[11]
	Bangladesh	14.53±1.8	34.78±2.4	303.11±14.9	87.52±3,8	Mantazul et al. (1998) [12]
	Brazil	10.2	12.6	51	34	Malanca et al. (1995) [13]
	Egypt	9.2	3.3	47.3	16.6	Sharaf et al. (1999) [14]
	Greece	18±7	17±10	367±20.4	—	Stoulos et al. (2003) [1]
	India	9.4	52.05	65.5	84.15	Kumar et al. (2003) [15]
	Kuwait	7.9±0.7	7.2±0.3	360±14	45.4	Bou-Rabee and Bem (1996)[16]
	Malaysia	60±3	13±2	750±53	136±33	Ibrahim (1999) [17]
	Pakistan	25.1	14.6	188.1	60.5	Ahmad and Hussein (1997) [18]

	Zambia	24±1	26±2	714±17	117±12	Hayambu et al. (1995) [8]
	Camiron	14±1	31±1	586±13	104.06	Ngachina et.al.(2006) [19]
	Upper Egypt	-----	44±2	150±8	96.6	This work
Red clay brick	Algeria	65±7	51±5	675±4	190±9.5	Amrani and Tahtat (2001) [11]
	Bangladesh	29.47±6.3	52.5±12.2	292.25±43.6	127.14±9.9	Mantazul et al. (1998) [12]
	Brazil	46.8±1.94	119.9±11.6	322±15.2	247.7±17.3	Malanca et al. (1995) [13]
	Egypt	24.5	24.4	227	77	Sharaf et al. (1999) [14]
	Egypt	24	24.1	258	78	El-Tahawy, Higgy (1995)[20]
	Greece	35.6	51.5	732	—	Savidou et al. (1995) [21]
	Hong Kong	82±3.6	—	—	—	Man and Yeung (1998) [22]
	India	18.03	33.33	44.8	69.15	Kumar et al. (2003) [15]
	Kuwait	11.9±0.7	6.6±0.2	332±4	41.6	Bou-Rabee and Bem (1996)[16]
	Malaysia	241±3	51±4	7541±272	895±107	Ibrahim (1999) [17]
	Qena(Egypt)	33±2.0	37±1.7	511±15.8	—	Ahmed (2005) [23]
	Zambia	32±2	81±7	412±19	180±22	Hayambu et al. (1995) [8]
	Camiron	49.6±0.3	91±2	172±4	193.34	Ngachin et.al.(2006) [19]
		Upper Egypt	-----	63±3	276±14	156

From the comparison of activity and radium equivalent activities Ra_{eq} in building materials used in Upper Egypt with those of other countries in Table 3, it is clear that our results are in average among other results.

4. Conclusions

The average value of the concentrations for ^{226}Ra , ^{232}Th and ^{40}K in investigated samples have been found to lie within the range 17 ± 1 – 79 ± 4 , 17 ± 1 – 115 ± 6 , 95 ± 5 – 376 ± 19 Bq kg^{-1} , respectively. The lowest ^{226}Ra and ^{232}Th activities were found in Sand and the highest in Redbrick. The lowest ^{40}K activity was found in limestone and the highest in redbricks. The absorbed dose rate in door was found to vary from 0.19 to 0.57 nGyh^{-1} , and the corresponding annual effective dose ranging from 0.9 to 3.5 μSvy^{-1} is lower than the value 1.5 mSv yr^{-1} set in the OECD report [24].

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