Estimation of Radiological Hazards of Some Egyptian Building Materials Due to Natural Radioactivity

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Abstract

Concentrations of natural radionuclides (226Ra, 232Th and 40K) content and its radiological hazards were presented in this paper for variety of commonly used building materials soil, sand, redbrick, clay brick, limestone and cement. Measurements were done by using gamma spectrometry (NaI (Tl) 3"x 3"). The radiation hazard indices like: Radium equivalent (Raeq), external hazard index (Hex), activity concentration index (Iex), the specific dose rates in door (D), and the annual effective dose (DE) due to gamma radiation were calculated and compared with internationally recommended values.

Keywords: *Building materials; Activity concentration index; Dose rate; Annual effective dose*

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.

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 (Stoulos *et al.*, 2003)[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 84nGy h^{-1} (UNSCEAR, 2000)[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 (UNSCEAR, 1977)[3]. Consequently, this study comes within the framework of the estimation of the possible radiological hazards resulting from the natural radiation, which can be exposed to the Egyptian population when they use of such building materials.

2. Materials and Methods

2.1. Sample Description and Preparation

A total of 39 samples of 6 different buildings materials (soil, sand, clay brick, limestone, gypsum and cement) were collected for the measurements of activity concentrations from Assiut, Qena and Souhag 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:

- **Soil** is a mixture of mineral and organic matter, it is mainly made up of oxygen (46.7%), silicon (27%), aluminum (8.1 %) and iron (5.0%). Plant nutrients like Ca, Mg, K, Na, P and S are present in the minerals and in the soil solution. O_2, Si, and Al occur as constituents of minerals and as oxides. Fe occurs mainly in the form of oxides and ferromagnesian minerals. Ca occurs mainly in calcite, gypsum, apatite and dolomite. Mg is present mainly in dolomite and hornblend. K occurs mainly in microcline and mica. P occurs as aluminum phosphate and calcium phosphate and in the organic form as phospholipids, inositol, choline, etc. N occurs mainly in the organic form as proteins, amino acids, etc. All micronutrients like Mo, Fe, Mn, Zn, Cu, B occur in the inorganic form that is capable of supporting plant growth

- **Sand** is a granular material made up of fine rock particles. The most common constituent of sand is silica (silicon dioxide, or SiO2), 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.

- **Brick clay** ,its chemical composition is as follows:
  -Silica(55%) ,it is present in brick clay as the chemical composition of alumina and forming silicate of alumina. Sometimes silica is present in pure form termed as flint. Silica prevents shrinking, cracking and warping of brick clay.
  -Alumina(30%): it imparts plasticity to brick clay which is very important in molding. It also imparts density.
  -Iron oxide(8%): The presence of iron oxide increases impermeability and durability of brick. The color of brick depend upon iron oxide and color changes from light yellow to orange and red as iron oxide goes up to 8%.
  -Magnesia(5%): Presence of small quantity of magnesia decreases shrinkage and gives yellowish tint.
  -Lime(1%): Lime melts silica in burning and binds the particles of brick together. It also reduces the shrinkage of brick during drying. Excess of lime cause the brick to fuse and shape will be lost.
  -Alkali and Organic Matter(1%): Small quantity assist burning of brick clay. It also reduces the fusion point. If excess amount is present and not burned properly the brick would be porous.

- **Limestone** is a sedimentary rock composed largely of the minerals calcite and aragonite, which are different crystal forms of calcium carbonate (CaCO3). Many limestones are composed from skeletal fragments of marine organisms such as coral or foraminifera. Limestone makes up about 10% of the total volume of all sedimentary rocks. It is made from the mineral calcite, which comes from the beds of evaporated seas and lakes and from seashells.

- **Gypsum** is a very soft sulfate mineral composed of calcium sulfate dihydrate, with the chemical formula $CaSO4 \cdot 2H2O$. It can be used as a fertilizer, is the main constituent in many forms of plaster and is widely mined. A much harder and very fine-grained white or lightly tinted variety of gypsum, called alabaster, has been used for sculpture by many cultures including Ancient Egypt and Ancient Rome. Gypsum is moderately water-soluble (~2.0–2.5 g/l at 25℃) and, in contrast to most other salts, it exhibits a retrograde solubility, becoming less soluble at higher temperatures. When gypsum is heated in air it loses water and converts first to calcium sulfate hemihydrate, (bassanite, often simply called "plaster") and, if heated further, to anhydrous calcium sulfate (anhydrite). As for anhydrite, its solubility in saline solutions and in brines is also strongly dependent on NaCl concentration (Smith Joshua, 2007) [4].
- **Cement** is a mixture of compounds made by burning limestone and clay together at very high temperatures ranging from 1400 to 1600. The chemical composition of cement is Tricalcium silicate $3CaO \bullet SiO_2$ (50 – 70%), Dicalcium silicate $2CaO \bullet SiO_2$ (15–30%), Tricalcium aluminate $3CaO \bullet Al2O3$ (5-10%) ,Tetra calcium aluminoferrite $4CaO \bullet Al_2O_3 \bullet Fe_2O_3$ (5 – 15%) and Calcium sulfate dehydrate $CaSO4 \bullet 2H2O$ ~ 5%.

Each sample under investigation was dried in an oven at about 110 ^{o}C to ensure that moisture was completely removed. The samples were crushed, homogenized and sieved through a 200 μm, 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) [5,6] within the volume and the progeny will also remain in the sample.

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 Accuspes. 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 contained 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 Genie 2000[7] from Canberra has carried out the online analysis of each measured gamma ray spectrum.

The efficiency calibration curve was made using different energy peaks covering the range up to ≈2000 keV. Measurements were performed with calibrated source samples, which contain a known activity of one or more gamma ray emitters of the radionuclides ^{60}Co (1173.2 and 1332.5 keV), ^{133}Ba (356.1 keV), ^{137}Cs (661.9 keV) and ^{226}Ra(1764.49 keV), with certified accuracies of ≈2%, Equation (1) is used for calculating the absolute efficiency:

$$Eff = \frac{100 \cdot N_p}{I_\gamma \cdot TOC \cdot A_{BOC}} \qquad (1)$$

Where, Np the net peak area at E_γ, I_γ the intensity of emitted gamma ray, TOC the time of counting and A_{BOC} the activity of the standard source at beginning of counting (BOC). A_{BOC} was calculated by equation (2)

$$A_{BOC} = A_{DOC} \cdot \exp(-\lambda \cdot (BOC-DOC)) \qquad (2)$$

Where A_{DOC} is the activity of the standard source at date of calibration DOC, and λ is the decay constant.

Daily efficiency and energy calibrations for each sample measurement were carried out to maintain the quality of the measurements. The absolute efficiency of the detector was calculated at the specific energy of the standard sources for the same geometry of the samples. But, the gamma spectra of the samples have different gamma energies. So, some fitting function is needed to calculate the absolute efficiency for any considered gamma energy. A function [8] is used, for this purpose, for calculating the absolute efficiency at any gamma energy of interest in the energy range below 2000 keV, which is in the following form:

$$\eta = a - b \times \exp(-c \times E_\gamma^d) \qquad (3)$$

Where, $E\gamma$ represents energy in MeV, where a, b, c and d are coefficient data. By equation (3), the absolute efficiency, η, can be determined at any specific energy $E\gamma$, if the energies and the coefficient data are known. From the experimental efficiency curves, the coefficient data were determined, by using the curve-fitting program Curve Expert 1.34[9].

2.2.1. Uncertainty of Efficiency

The combined standard uncertainty of absolute efficiency $u(EFF)$ consists of $u(N_p)$, $u(I_\gamma)$, $u(TOC)$ and $u(A_{BOC})$ so,

$$\left[\frac{u(EFF)}{EFF}\right]^2 = \left[\frac{u(N_p)}{N_p}\right]^2 + \left[\frac{u(I_\gamma)}{I_\gamma}\right]^2 + \left[\frac{u(TOC)}{TOC}\right]^2 + \left[\frac{u(A_{BOC})}{A_{BOC}}\right]^2 \qquad (4)$$

Because $u(TOC) << TOC$, we neglected $u(TOC)$. The value of $u(A_{BOC})$ was calculated by equ. (5)

$$\left[\frac{u(A_{BOC})}{A_{BOC}}\right]^2 = \left[\frac{u(A_{DOC})}{A_{DOC}}\right]^2 + (BOC - DOC)^2 \cdot u^2(\lambda) \qquad (5)$$

We got $u(N_p)$ from the code Genie 2000, while $u(\lambda)$ and $u(I_\gamma)$ were taken from the compilation of Reus and Westmeier [10]. The calibration standards used had a certified accuracy of $\leq 2\%$. By measurements for many times it could be verified with a total uncertainty of the full-energy-peak efficiency of 5 %.

2.2.2. Calculation of Activity

Calculations of count rates for each detected photo peak and radiological concentrations (activity per mass unit or specific activity) of detected radionuclides depend on the establishment of secular equilibrium in the samples. The ^{232}Th concentration was determined from the average concentrations of ^{212}Pb(238.6 keV) and ^{228}Ac(911.1keV) in the samples, and that of ^{226}Ra was determined from the average concentrations of the ^{214}Pb(351.9keV) and ^{214}Bi(609.3 and 1764.5 keV) decay products[11]. The activity concentration in Bqkg^{-1} (A) in the environmental samples was obtained as follows:

$$A = \frac{N_p}{e \times \eta \times m} \qquad (6)$$

Where N_p = the (cps) sample - (cps) B.G, *e is* the abundance of the γ-line in a radionuclide, η is the measured efficiency for each gamma-line observed for the same number of channels either for the sample or the calibration source, and *m* the mass of the sample in kilograms. The uncertainty of activity *u (A)* was calculated by the following equation:

$$u(A) = A\sqrt{[u(N_p)/(N_p)]^2 + [u(\eta)/(\eta)]^2 + [u(m)/(m)]^2} \qquad (7)$$

From the equation, we can find that, there are many sources of uncertainties of the Activity and some may result in considerable uncertainties. The following sources of uncertainties were considered:

2.2.2.1. Uncertainty of the Determination of Net Peak Areas

The uncertainty of each single net-peak area is determined by the spectrum-evaluation code. It takes into account the Poisson uncertainties of the counts in the individual channels as well as the uncertainty of the background determination. Sometimes a peak cannot be attributed unambiguously to a single nuclide. If it seems that the contributions of other nuclides to a peak are very small no correction was applied. Due to this procedure we think that we have to assume a maximum inaccuracy of 2 % due to contributions of other nuclides but it must be pointed out that in the average this uncertainty should be smaller. By repeated measurements it could be verified that the total uncertainty of the efficiency calibration was 5%.

3. Results and Discussion

3.1. Activity Cncentrations

This study is a continuation of our ongoing project in physics department (faculty of science, Al-Azher university, Assuit branch, Egypt) related to the measurement of specific activity of ^{226}Ra, ^{232}Th and ^{40}K in environmental samples from Upper Egypt using a gamma-ray spectrometric technique and estimation of the gamma dose rate from these radionuclides. The results of analysis of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in controlled samples of building materials are presented in Table 1. Activity is reported in Bqkg^{-1} on the basis of the sample's dry weight.

Table 1. Activity Concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Different Samples

Sample Name	^{226}Ra A in (Bq kg^{-1})	^{232}Th A in (Bq kg^{-1})	^{40}K A in(Bq kg^{-1})
Soil(Assiut)	39.7±2.0	56.9±2.8	196.4±9.8
Soil(Assiut)	31.2±1.6	51.7±2.6	149±7.4
Soil(Assiut)	35.3±1.8	53.3±2.7	210.3±10.5
Soil(Qena)	37.5±1.9	71.3±4.1	239.9±12
Soil(Qena)	32.5±1.4	59.3±2.1	233.2±11
Soil(Souhag)	29±1.1	67.3±3.9	219.6±9
Soil(Souhag)	37.5±1.9	61.3±3.1	237.9±12
Average	*34.6±1.7*	*60.2±3*	*212.3±10.3*
Range of values	*29-39.7*	*51.7-71.3*	*149-239.9*
Sand(Assiut)	16.5±0.8	17.4±0.9	144±7.2
Sand(Assiut)	17.7±0.9	18.4±0.9	124±7.2

Sand(Qena)	*28.7±1.4*	*29±1.6*	*155.4±8.8*
Sand(Qena)	*23.5±0.8*	*31.3±0.9*	*164±7.8*
Sand(Souhag)	*19.5±0.9*	*17.9±1*	*140±6.2*
Sand(Souhag)	*18.7±1.2*	*21±3.5*	*165.4±9*
Average	***20.8±1***	***22.5±1.2***	***148.8±13.3***
Range of values	***16.5-28.7***	***17.4-31.3***	***124-165.4***
Clay brick (Assiut)	*54.3±2.7*	*65.3±3.3*	*285.4±14.3*
Clay brick (Assiut)	*65±3.3*	*78.1±3.9*	*352.1±17.6*
Clay brick (Qena)	*59.6±3*	*80.6±4*	*375.5±18.8*
Clay brick (Qena)	*36±1.8*	*63.4±3.2*	*216.3±108*
Clay brick (Qena)	*29.3±1.5*	*35.3±1.8*	*166.8±8.3*
Clay brick (Souhag)	*35.5±1.8*	*30.3±1.5*	*245.5±12.3*
Clay brick (Souhag)	*51.7±2.6*	*89.3±4.5*	*288.2±14.4*
Average	***47.3±2.4***	***68.9±4.1***	***275.7±12.3***
Range of values	***29.3-65***	***50.3-89***	***166.8-375.5***
Limestone (Assiut)	*30±1.5*	*47.2±2.4*	*120.6±6*
Limestone (Assiut)	*35.3±1.8*	*51.1±2.6*	*125.8±6.3*
Limestone (Qena)	*27.1±1.4*	*35.5±1.8*	*114.8±5.7*
Limestone (Qena)	*23±2.1*	*52±3.6*	*74.5±5.2*
Limestone (Souhag)	*33.5±2*	*45.6±3.7*	*89.3±4.5*
Limestone (Souhag)	*31.6±3.2*	*79.2±5.1*	*81.3±3.3*
Average	***30.1±2***	***51.8±3.5***	***101.1±10.9***
Range of values	***23-35.3***	***35.5-79.2***	***74.5-125.8***
Gypsum(Assiut)	*40.1±2*	*56.1±2.8*	*66.8±2.9*
Gypsum(Assiut)	*28.5±2*	*69.2±3.5*	*72±2.1*
Gypsum(Qena)	*37.6±1.9*	*72±3.6*	*88.2±4.1*
Gypsum(Qena)	*57.5±2.9*	*69.4±3.3*	*65.2±3.3*
Gypsum(Souhag)	*46.4±4.1*	*71±5.7*	*78.5±1.4*
Gypsum(Souhag)	*34.1±1.7*	*55.1±2.8*	*96.6±4.8*
Average	***40.7±2.4***	***65.5±4.2***	***77.9±9.6***
Range of values	***28.5-57.5***	***55.1-72***	***65.2-96.6***
Cement (Assiut)	*30.8±3.6*	*66.5±3.6*	*91.3±7.1*
Cement (Assiut)	*29±2.5*	*55.2±4.8*	*97.3±5.6*
Cement (Qena)	*33.5±1.7*	*59.4±3*	*88.9±4.7*
Cement (Qena)	*29.8±3.3*	*42.6±3.6*	*85.5±3.3*
Cement (Souhag)	*31.8±3.4*	*75.2±3.2*	*76.6±5.4*
Cement (Souhag)	*44.2±2.2*	*67.7±2.4*	*94.8±2.7*
Cement (Souhag)	*37.2±3.9*	*66.1±2.7*	*88.5±3.9*
Average	***33.8±3.1***	***61.8±3.3***	***89.0±5***
Range of values	***29.8-44.2***	***42.6-75.2***	***76.6-97.3***

From the obtained results, which listed in Tble 1, large variation among the radioactivity concentration for different samples has been observed. It may be due to geological condition and the raw material that used in making the building materials under investigation. It can be seen that ^{40}K almost always contributes to the most specific activity compared to ^{226}Ra and ^{232}Th. The results tell us that for all samples under investigation , the largest average activity of ^{226}Ra is ($47.3\pm2.4Bq.kg^{-1}$) for clay bricks , it is more than two times greater than that of the lowest value ($20.8\pm1Bq.kg^{-1}$) ,which was found in *sand samples.* ^{232}Th is in the wide range from ($17.4\pm0.9Bq.kg^{-1}$) in *sand (Assiut)* up to ($80.6\pm4Bq\ kg^{-1}$) in *Clay brick (Qena)*. Concentration of ^{40}K values ranging from ($66.8\pm2.9Bq\ kg^{-1}$) in *Gypsum(Assiut)* to ($375.5\pm18.8\ Bq.kg^{-1}$) in redbrick (Qena), it is approximately the maximum admissible value of (370 *Bq kg*$^{-1}$) set in the UNSCEAR report (1988)[12]. Figure 1 shows the results of Table 1 in graphical form, clearly indicating the high- and low-activity samples.
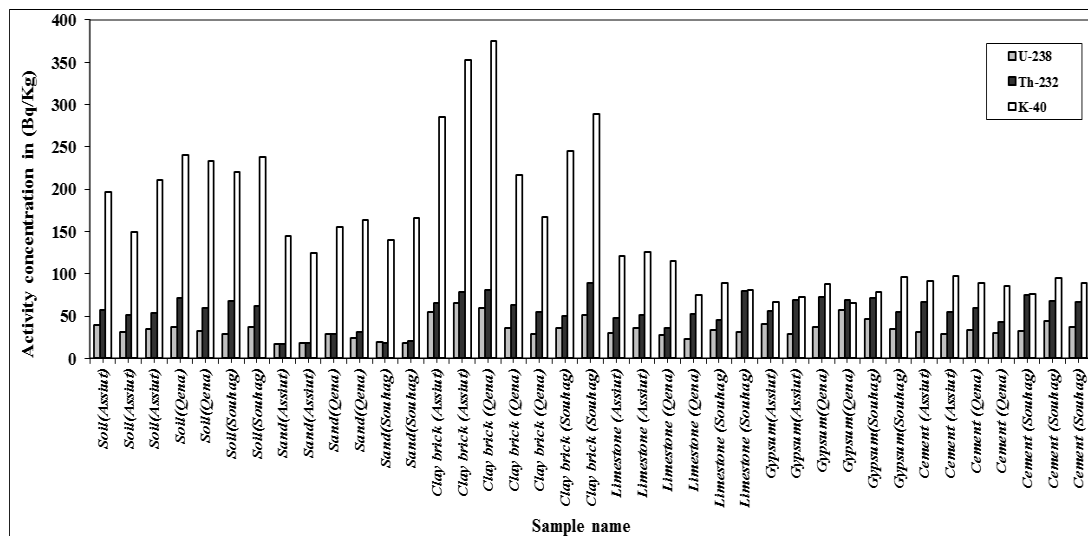

Figure 1. Activity Concentrations of the Radioelements (in Bq kg^{-1}) Found in Studied Samples

3.2. Radium Equivalent Activity

Because of ^{226}Ra, ^{232}Th and ^{40}K are non-uniformly distributed in building materials. In order to compare the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent activity Ra_{eq} is used as defined by the following expression (Hayambu *et al.*, 1995)[13]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad (8)$$

Where A_{Ra}, A_{Th} and A_K are the mean activity in (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. Eq. (8) is based on the fact that *370 (Bq kg^{-1}) of ^{226}Ra, 259 (Bq kg^{-1}) of ^{232}Th and 4810 (Bq kg^{-1}) of ^{40}K*, produce the same γ-ray dose equivalent. The values of Ra_{eq} are listed in column 2 of Table (2) for all the samples under investigation. These values ranged from *52.49 Bq kg^{-1}* in *sand (Assiut)* to 203.8 Bq kg^{-1} in *Clay brick (Qena)*. All results are less than the maximum admissible value of 370 Bq kg^{-1} set in the UNSCEAR report (1988), which is equivalent to an external dose of 1.5 mSvyr^{-1} (Organization for Economic Cooperation and Development, 1979)[14].

3.3. External Hazard Index (Hex)

The external hazard index is obtained from R_{aeq} expression through the supposition that its maximum allowed value (equal to unity) corresponds to the upper limit of R_{aeq} *(370 Bqkg^{-1})*. This index value must be less than unity in order to keep the radiation hazard insignificant; *i.e.*, the radiation exposure due to the radioactivity from construction materials is limited to 1.0 mSv y^{-1}, then the external hazard index (H_{ex}) is given by the following equation:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \qquad (9)$$

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 mSvyr^{-1}. As indicated in Table 2, it appears that investigated materials meet this criterion.

Table 2. The Values of Radiation Hazard Parameters for Building Materials under Investigation

Sample	Raeq	Hex	Gamma index	Dose rate ($\dot{D}$)	Annual Effective Dose	Excess lifetime cancer risk	Usability of a building material factors	
name	(Bq/kg)	1000	I_{ex}	(nGy/h)	DE(μSv)	ELCR	F1	F2
Soil(Assiut)	136.2	0.37	0.48	114.8	0.06	0.20	0.40	39.7
Soil(Assiut)	116.6	0.31	0.41	97.5	0.05	0.17	0.35	31.2
Soil(Assiut)	127.4	0.34	0.45	107.7	0.05	0.18	0.38	35.0
Soil(Qena)	157.9	0.43	0.56	132.1	0.06	0.23	0.47	37.5
Soil(Qena)	135.3	0.37	0.48	113.8	0.06	0.20	0.41	32.5
Soil(Souhag)	142.1	0.38	0.51	118.3	0.06	0.20	0.43	29.0
Soil(Souhag)	143.5	0.39	0.51	121.0	0.06	0.21	0.43	37.5
Averange	**137.0**	**0.37**	**0.49**	**115.0**	**0.06**	**0.20**	**0.41**	**34.6**
Range of values	**116.6-157.9**	**0.31-0.43**	**0.41-0.56**	**97.5-132.1**	**0.05-0.06**	**29-39.7**	**0.35-0.47**	**29-39.7**
Sand(Assiut)	52.5	0.14	0.19	45.8	0.02	0.08	0.16	16.5
Sand(Assiut)	53.6	0.14	0.19	46.4	0.02	0.08	0.16	17.7
Sand(Qena)	82.1	0.22	0.29	70.7	0.03	0.12	0.24	28.7
Sand(Qena)	80.9	0.22	0.29	69.2	0.03	0.12	0.24	23.5
Sand(Souhag)	55.9	0.15	0.20	48.8	0.02	0.08	0.17	19.5
Sand(Souhag)	61.5	0.17	0.22	53.5	0.03	0.09	0.19	18.7
Averange	**64.4**	**0.17**	**0.23**	**55.8**	**0.03**	**0.10**	**0.19**	**20.8**
Range of values	**52.5-82.1**	**0.14-0.22**	**0.19-0.29**	**45.8-70.7**	**0.02-0.03**	**16.5-28.7**	**0.16-0.47**	**16.5-28.7**
Clay brick (Assiut)	169.7	0.46	0.60	144.6	0.07	0.25	0.50	54.3
Clay brick (Assiut)	203.8	0.55	0.72	173.9	0.09	0.30	0.61	65.0
Clay brick (Qena)	203.8	0.55	0.73	173.5	0.09	0.30	0.61	59.6
Clay brick (Qena)	143.3	0.39	0.51	120.2	0.06	0.21	0.43	36.0
Clay brick (Qena)	121.2	0.33	0.43	101.1	0.05	0.17	0.36	29.3
Clay brick (Souhag)	126.3	0.34	0.45	107.6	0.05	0.18	0.38	35.5
Clay brick (Souhag)	201.2	0.54	0.71	168.5	0.08	0.29	0.60	51.7
Averange	**167.0**	**0.45**	**0.59**	**141.4**	**0.07**	**0.24**	**0.50**	**47.3**
Range of values	**121.2-203.8**	**0.33-0.55**	**0.51-0.73**	**101.1-173.9**	**0.05-0.09**	**29.3-65**	**0.36-0.61**	**29.3-65**
Limestone (Assiut)	106.8	0.29	0.38	89.2	0.04	0.15	0.32	30.0

Limestone (Assiut)	118.1	0.32	0.42	98.8	0.05	0.17	0.35	35.3
Limestone (Qena)	86.7	0.23	0.31	73.2	0.04	0.13	0.26	27.1
Limestone (Qena)	103.1	0.28	0.36	84.3	0.04	0.14	0.31	23.0
Limestone (Souhag)	105.6	0.29	0.37	88.1	0.04	0.15	0.31	33.5
Limestone (Souhag)	151.1	0.41	0.53	122.7	0.06	0.21	0.45	31.6
Averange	**111.9**	**0.30**	**0.39**	**92.7**	**0.05**	**0.16**	**0.33**	**30.1**
Range of values	**86.7-151.1**	**0.23-0.41**	**0.31-0.53**	**73.2-122.7**	**0.04-0.07**	**23-35.3**	**0.26-0.45**	**23-35.3**
Gypsum(Assiut)	125.5	0.34	0.44	103.9	0.05	0.18	0.37	40.1
Gypsum(Assiut)	133.0	0.36	0.47	108.1	0.05	0.19	0.39	28.5
Gypsum(Qena)	147.4	0.40	0.51	120.8	0.06	0.21	0.43	37.6
Gypsum(Qena)	161.8	0.44	0.56	134.5	0.07	0.23	0.47	57.5
Gypsum(Souhag)	154.0	0.42	0.54	127.1	0.06	0.22	0.45	46.4
Gypsum(Souhag)	120.3	0.32	0.42	99.7	0.05	0.17	0.36	34.1
Averange	**140.3**	**0.38**	**0.49**	**115.7**	**0.06**	**0.20**	**0.41**	**40.7**
Range of values	**120.3-161.8**	**0.32-0.44**	**0.42-0.56**	**99.7-134.5**	**0.05-0.07**	**28.5-57.5**	**0.36-0.47**	**28.5-57.5**
Cement (Assiut)	132.9	0.36	0.47	108.8	0.05	0.19	0.39	30.8
Cement (Assiut)	115.4	0.31	0.41	95.2	0.05	0.16	0.34	29.0
Cement (Qena)	125.3	0.34	0.44	103.3	0.05	0.18	0.37	33.5
Cement (Qena)	97.3	0.26	0.34	81.1	0.04	0.14	0.29	29.8
Cement (Souhag)	145.2	0.39	0.51	118.1	0.06	0.20	0.43	31.8
Cement (Souhag)	148.3	0.40	0.52	122.7	0.06	0.21	0.44	44.2
Cement (Souhag)	138.5	0.37	0.48	114.0	0.06	0.20	0.41	37.2
Averange	**129.0**	**0.35**	**0.45**	**106.2**	**0.05**	**0.18**	**0.38**	**33.76**
Range of values	**97.3-148.3**	**0.26-0.40**	**0.34-0.52**	**81.1-122.7**	**0.04-0.06**	**29-44.2**	**0.29-0.44**	**29.8-44.2**

3.4. Activity Concentration Index(I)

Because more than one radionuclide contribute to the dose, it is practical to present investigation levels in the form of an *activity concentration index, (I),* or shortly, gamma index *(I),* is defined in the following way:

$$I = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_{K}}{3000} \qquad (10)$$

Activity concentration index is derived to identify whether a dose criterion is met. The limit value that activity concentration index should not exceed depends on the dose criterion (EC 1999, 1999)[15]. The activity concentration index takes into account typical ways and quantities in which the material is used in a building D.Krsti ́c *et.al.*, 2007)[16]. The activity concentration index should be used only as a screening tool for identifying materials which might be of concern. In this paper, the *Activity concentration index (I)* was calculated and the results were listed in column 4, Table 2. Obtained values of *I* ranged from 0.19 to 0.73 in studied samples.

3.5. Dose Rate Indoor ($\dot{D}$)

Dose rate indoors are calculated according to (EC 1999, 1999)[15] for investigated samples as:

$$\dot{D} = 0.92C_{Ra} + 1.1C_{Th} + 0.08C_{k} \qquad (11)$$

The specific dose rates (in units nGy h^{-1}per Bq kg^{-1}) for ^{226}Ra, ^{232}Th and ^{40}K are given for different materials. The results were listed also in Tables 2 and these values were used to calculate annual effective doses. The values of the absorbed dose rate in indoor air are in between (45.8 to 173.9) nGy h^{-1}.

3.6. Annual Effective Dose Rate

The annual effective dose, DE, due to gamma radiation from building materials was calculated as:

$$DE = 0.7 \text{SvGy}^{-1} \cdot 7000 \text{ h} \cdot \dot{D} \qquad (12)$$

Where $\dot{D}$ must be taken in $\mu Gy\ h^{-1}$; $0.7SvGy^{-1}$ is effective absorbed dose conversion factor , $7000\ h$ is annual exposure time and $\dot{D}$ .The resulting values of the annual effective dose rate have been listed in Table 2. We can find that, the lowest value of DE is $0.02mSv$ while the highest is 0.09mSv. According to (UNSCEAR, 2000) [2] the annual effective dose of these samples does not exceed the average worldwide exposure of 2.4 mSvy^{-1} due to natural sources.

4. Comparison of Results with Similar Studies in Other Countries

Our results for average specific activities of ^{226}Ra, ^{232}Th and ^{40}K and radium equivalent activities Raeq in various types of building materials are compared in table 4 with the results for other countries.

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
		^{226}Ra	^{232}Th	^{40}K		
Soil	Turkay	37±17	40±18	667±282	—	H. Taskin et al.2009[16]
	Worldwide	35	30	400	—	UNSCEAR (2000)[2]
	Egypt	34.6±1.7	60.2±3	212.3±10.3	137	This work
Sand	Algeria	12±1	7±1	74±7	28±7.1	Amrani and Tahtat (2001)[17]
	Bangladesh	14.53±8.2	34.78±2.4	303.11±141.91	87.52±38.05	Mantazul et al. (1998)[18]
	Brazil	10.2	12.6	51	34	Malanca et al. (1995)[19]
	Egypt	9.2	3.3	47.3	16.6	Sharaf et al. (1999)[20]
	Greece	18±7	17±10	367±204	—	Stoulos et al. (2003)[21]
	India	9.4	52.05	65.5	84.15	Kumar et al. (2003)(23)
	Kuwait	7.9±0.7	7.2±0.3	360±14	45.4	Bou-Rabee and Bem (1996)[24]
	Malaysia	60±3	13±2	750±53	136±33	Ibrahim (1999)(25)
	Pakistan	25.1	14.6	188.1	60.5	Ahmad and Hussein (1997)(26)
	Zambia	24±1	26±2	714±17	117±12	Hayambu et al. (1995)[13]
	Camiron	14±1	31±1	586±13	104.06	Ngachina et.al.(2006)(28)
	Egypt	20.8±1	22.5±1.2	148.8±13.3	64.4	This work

clay brick	*Algeria*	*65±7*	*51±5*	*675±4*	*190±9.5*	*Amrani and Tahtat (2001)[17]*
	Bangladesh	*29.47±6.3*	*52.5±12.19*	*292.25±43.65*	*127.14±9.85*	*Mantazul et al. (1998)[18]*
	Brazil	*46.8±19.4*	*119.9±110.6*	*322±152*	*247.7±170.3*	*Malanca et al. (1995)[19]*
	Egypt	*24.5*	*24.4*	*227*	*77*	*Sharaf et al. (1999)[20]*
	Greece	*35±11*	*45±15*	*710±165*	*—*	*Stoulos et al. (2003)[1]*
	India	*18.03*	*33.33*	*44.8*	*69.15*	*Kumar et al. (2003)[21]*
	Malaysia	*241±3*	*51±4*	*7541±272*	*895±107*	*Ibrahim (1999)[22]*
	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)[13]*
	Camiron	*49.6±0.3*	*91±2*	*172±4*	*193.34*	*Ngachina et.al.(2007)[24]*
	Egypt	***47.3±2.4***	***68.9±4.1***	***275.7±12.3***	***167***	***This work***
	Brazil	24.3	7	205	—	*Malanca et al. (1995)[19]*
	Bangladesh	68	107	166	349	Alam, M.(1999)[25]
	Egypt	20.4±2.8	4.4±0.8	19.3±2	25.4	*Sharaf et al. (1999)[20]*
	Algeria	16±3	13±2	36±3	37±4.7	*Amrani and Tahtat (2001)[17]*
	Italy	11	2.0±2.0	22±3	14±11	Rizzo, S. et al.(2001)[26]
	Egypt	27.8±1.4	46.6±2.3	66±3.3	79.85	A. El-Taher (2010)[27]
	Egypt	***30.1±2***	***51.8±3.5***	***101.1±10.9***	***111.9***	**This work**
Limestone	Finland	7 2	25	—	—	NEA–OECD(1979)[14]
	UK	22	7	141	—	NEA–OECD(1979)[14]
	Brazil	6.3	N.D.	154	18.1	*Malanca et al. (1995)[19]*
	Italy	6.0	2.0	32	12	Rizzo, S. et al.(2001)[26]
	Egypt	31.7±4.6	55±2.7	88.7±4.4	116	A. El-Taher (2010)[27]
	Egypt	***40.7±2.4***	***65.5±4.2***	***77.9±9.6***	***140.3***	***This work***
Gypsum	Cameron	27±4	15±1	277±16	70	*Ngachina et.al.(2007)[24]*
	Finland	44	26	241	—	NEA–OECD(1979)[14]
	Norway	30	18	241	—	NEA–OECD(1979)[14]
	Sweden	55	47	241	—	NEA–OECD(1979)[14]
	UK	22	18	155	—	NEA–OECD(1979)[14]
	Brazil	61.7	58.5	564	188.8	*Malanca et al. (1995)[19]*
	Bangladesh	29.7	54.3	523	148	Alam, M.(1999)[25]
	Algeria	41±7	27±3	422±3	112±8.2	*Amrani and Tahtat (2001)[17]*
	Italy	38±14	22±14	218±248	92±60	Rizzo, S. et al.(2001)[26]
	Zambia	23±2	32±3	134±13	79±11	*Hayambu et al. (1995)[13]*
	Egypt	31.3±3.6	11.1±1.1	48.6±4	50.9	*Sharaf et al. (1999)[20]*
	Egypt	36.6±4.4	43.2±2.2	82±4.1	103	A. El-Taher (2010)[27]
	Egypt	***33.8±3.1***	***61.8±3.3***	***89.0±5***	***129***	***this work***

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 4, it is clear that our results are in average between other results.

5. Conclusions

This study determined that the average natural radioactivity (^{226}Ra, ^{232}Th and ^{40}K) concentrations and related radiation hazards in various building materials used in Egypt. All results were within the worldwide range. The radium equivalent activities obtained for the building materials under investigation were below the criterion limit of γ-radiation dose (370 Bq kg^{-1}) adopted by the OECD 1979[14] criterion. The adsorbed dose rate in door was found to vary from 45.8 to 173.9 nGyh^{-1} per BqKg^{-1}, and the corresponding Annual Effective dose ranging is lower than the value 1.5 mSv yr^{-1} set in the OECD report [14]. In regard to the above results, Major radiation risk from examined materials comes from clay bricks and all the samples investigated can be safely used as building materials for dwelling constructions. It should be mentioned that anther building materials that were not studied here, so further investigations are needed to estimate the average exposure of people living in Upper Egypt dwellings.

Acknowledgements

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