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Distribution of Natural Radionuclide and Radiation Hazards of Building Materials Used in Assiut, Egypt

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Abstract

The concentration of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K have been measured in Clay (C), gypsum (G), limestone (L), sand (S), brick (B), soil (So), cement (Ce), which are used as building materials in Assiut, Egypt, using gamma spectrometry employing a 3×3 inch scintillation NaI (Tl) detector. The radium equivalent activity (Ra_{eq}), indoor gamma absorbed dose rate (D), annual effective dose (An), (AUI), alpha index (Iα), gamma index (Iγ), external radiation hazard index (Hex), internal radiation hazard index (Hin), representative level index (RLI), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) associated with the natural radionuclides are calculated to assess the radiation hazard of the natural radioactivity in the building materials. Basic statistics (skewness and Kurtosis) and frequency distributions for all radionuclides were used to describe the statistical characteristics of the radionuclide activities.

Keywords: *Natural radionuclids, Building materials, Annual effective dose, Excess lifetime cancer risk, skewness and Kurtosis statistics*

1. Introduction

Humans are always exposed to environmental radiation of terrestrial and cosmic origin. The biggest contribution to environmental radiation comes from radon gases and its decay products. Terrestrial radiation mainly originates from radioactive nuclides existing in the first phase of the formation of the solar system. These radioactive nuclides exist in air, water, soil, rocks and building construction materials, depending on the geological and geographical features of the region. Cosmic radiation originates from outer space and contributes to background radiation depending on variations with elevation and latitude [1].

The assessment of the population's exposure to indoor radiation is very important; therefore, knowledge regarding the concentration of natural radionuclides in construction materials is required. Construction materials are derived from both natural sources (*e.g.,* rock and soil) and waste products (*e.g.,* phospho-gypsum, alum shale, coal, fly ash, oil-shale ash, some rare minerals and certain slugs) as well as from industry products (*e.g.,* power plants, phosphate fertilizer and the oil industry). Although building materials act as sources of radiation to the inhabitants in dwellings, they also shield against outdoor radiation. Knowing the level of the natural radioactivity in building materials is important to assess the associated radiological hazards to human health and to develop standards and guidelines for the use and management of these materials [2].

In the present work, the concentration of natural radionuclides was measured in twenty-one samples of building materials that were commonly used in Assiut, Egypt, using gamma spectrometry employing 3×3 inch scintillation NaI (Tl) detector. The radiological hazards associated with the studied materials were assessed by calculating the radium equivalent activity (Ra_{eq}), indoor gamma absorbed dose rate (D), annual effective

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dose (A_n), alpha index (I_α), gamma index (I_γ), radiation hazards, representative level index (RLI), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE). The results were compared to the recommended values to assess the radiation hazards to humans resulting from the building materials, and with corresponding values of building materials from different countries.

2. Materials and Methods

2.1. Sampling and Sample Preparation

A total of 21 samples of natural and manufactured building materials commonly used in Assiut city, Egypt were collected randomly from sites where housing and other building were constructed and from the building material suppliers for the measurement of the specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K. Deposit samples were oven dried at a temperature of 105° C for 12 h and sieved through a 200 mesh. The dried samples were transferred to polyethylene Marinelli beakers. Each deposit sample was left for at least 4 weeks to reach secular equilibrium between radium and thorium, and their progenies [3].

2.2. Gamma-Ray Spectroscopic Technique

Activity measurements have been performed by a gamma ray spectrometer, employing a scintillation detector (3×3 inch). It is hermetically sealed assembly, which includes a NaI (Tl) crystal, coupled to PC-MCA Canberra Accuspec. 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 43200 s. The background spectra were used to correct the net peak area of gamma rays of measured isotopes. A dedicated software program, Genie 2000 from Canberra, was used to carry out the on-line analysis of each measured gamma-ray spectrum.The ^{226}Ra radionuclide was estimated from the 351.9 keV (36.7%) γ-peak of ^{214}Pb and 609.3 keV (46.1%), 1120.3 keV (15%), 1728.6 keV (3.05%) and 1764 keV (15.9%) γ-peaks of ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U with energy of 185.7 keV. ^{232}Th radionuclide was estimated from the 911.2 keV (29%) γ-peak of ^{228}Ac, 238.6 keV (43.6%) γ-peak of ^{212}Pb and 583.1 kev (84.5) γ-peak of ^{208}Tl. ^{40}K radionuclide was estimated using 1,461 keV (10.7%) γ -peak from ^{40}K itself. All procedures were described in previous publications [4].

3. Results and Discussion

3.1. Radionuclide Activity Concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}Kin building materials are presented in Table 1. Table 1 shows that, the highest values observed for the specific activities of ^{226}Ra, ^{232}Th and ^{40}K are 48±2 (G1), 26±2 (B3) and 195±10 Bq kg^{-1} (B3), respectively, while the lowest observed values of the specific activities of the same radionuclides are 14±1 (C1), 9.7±1 (L2) and 90±4 Bq kg^{-1} (L2), respectively. As shown in Table 1, the activity of ^{226}Ra varies from 14 to 48 Bq kg^{-1} and the arithmetic mean is 30 Bq kg^{-1}.

Table 1. Activity Concentration (Bq kg^{-1}), in Different Types of Building Materials: Clay (C), Gypsum (G), Limestone (L), Sand (S), Brick (B), Soil (So), Cement (Ce)

Material	Activity concentrations Bq kg^{-1}		
	^{226}Ra	^{232}Th	^{40}K
C1	14±1	24±1	151±8
C2	40±2	11±04	149±7
C3	15±1	10.4±04	172±9
G1	48±2	10±04	154±8
G2	34±2	10.5±1	133±6
G3	31±2	12±1	126±5
L1	23±1	12±0.3	108±5
L2	26±2	9.7±1	90±4
L3	29±2	13±1	94±4
S1	16±0.3	13±0.3	147±7
S2	20±1	12±0.3	133±7
S3	22±1	14±1	172±9
B1	41±2	20±1	190±9
B2	42±2	22±1	189±9
B3	36±1	26±2	195±10
So1	30±2	12±0.3	123±6
So2	46±2	11±1	180±9
So3	17±1	10.3±1	172±9
Ce1	37±2	11±1	194±10
Ce2	31±1	12±1	99±6
Ce3	38±2	13±0.4	106±7
Min.	**14**	**9.6**	**90**
Max.	**48**	**26**	**195**
Mean	**30**	**14**	**147**
SD	**10**	**5**	**35**
SE	**2**	**1**	**8**
Variance	**110**	**24**	**1211**
Skewness	**-0.03**	**1.7**	**-0.2**
Kurtosis	**-1.09**	**1.5**	**-1.3**

The activity concentration of ^{232}Th varies from 9.7 to 26 Bq kg^{-1}, and the arithmetic mean is 14 Bq kg^{-1}. The activity concentration of ^{40}K varies from 90 to195 Bq kg^{-1}, and the arithmetic mean is 147 Bq kg^{-1}.

Activities of ^{226}Ra, ^{232}Th and ^{40}Kare lower by factors of 0.85, 0.46 and 0.36 than the world average values, respectively, which are 35, 30 and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively [5]. Figure 1 shows the Distribution of ^{226}Ra, ^{232}Th and ^{40}K for the different building materials. Figure 2 shows that, the concentrations of ^{226}Ra, ^{232}Th and ^{40}K occupy the total activity of 7.5% in clay (C3) to 24.3% in cement (C3), 4.7% in gypsum (G1) to 12.1% in clay (C1) and 69.2% in cement (Ce3) to 87.2% in clay (C3), respectively, which indicates that the specific activity due to ^{40}K is the largest contributor to the total activity for all samples.

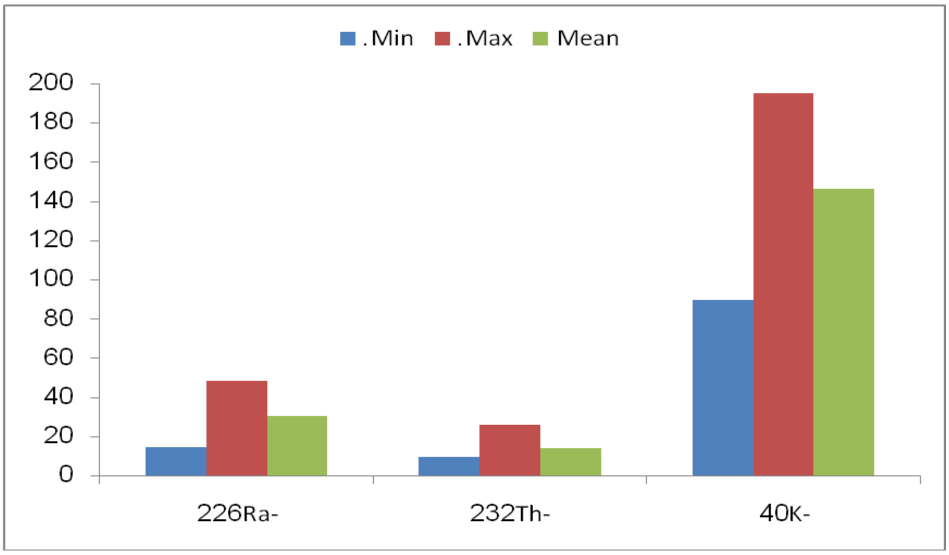

Figure 1. Distribution of ^{226}Ra, ^{232}Th and ^{40}Kin Building Materials

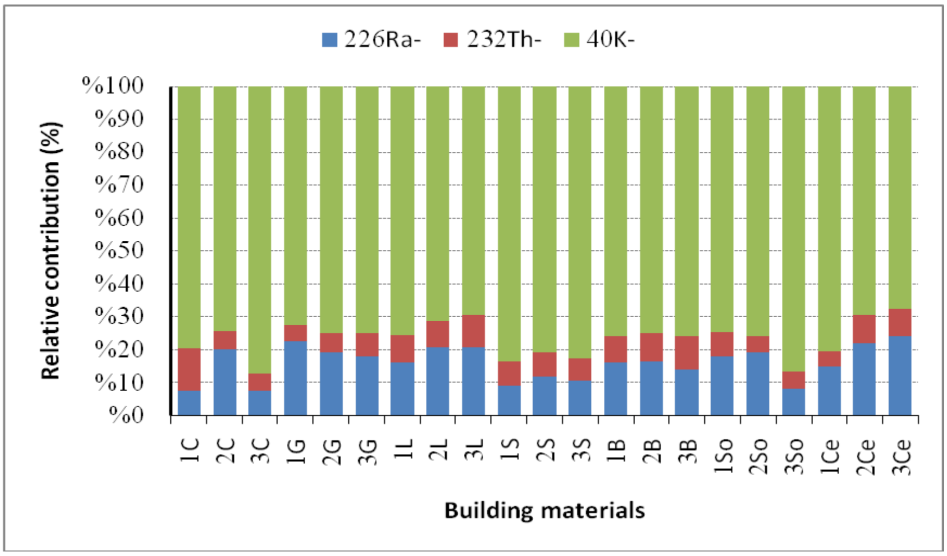

Figure 2. The Relative Concentration of ^{226}ra, ^{232}th and ^{40}k to the Total Activity in Building Materials

The frequency distributions of all the radionuclides were analyzed, and the histograms are given in Figures. 3–5. Graphs for ^{226}Ra, and ^{40}K show that these radionuclides demonstrate a normal (bell-shaped) distribution. However ^{232}Th exhibit some degree of multi-modality. This multi-modal feature of the radioactive elements demonstrates the complexity of minerals in building materials.

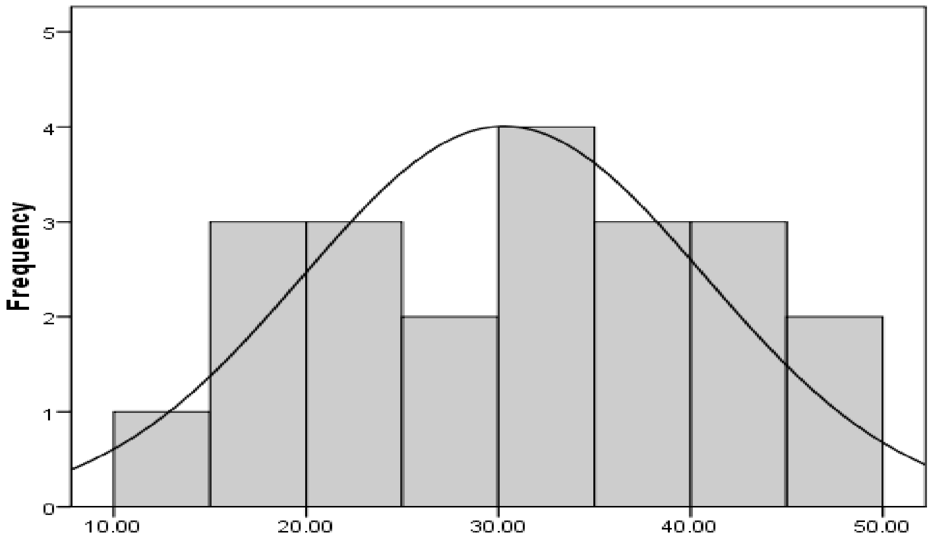

Figure 3. The Frequency Distribution of the Activity of ^{226}Ra

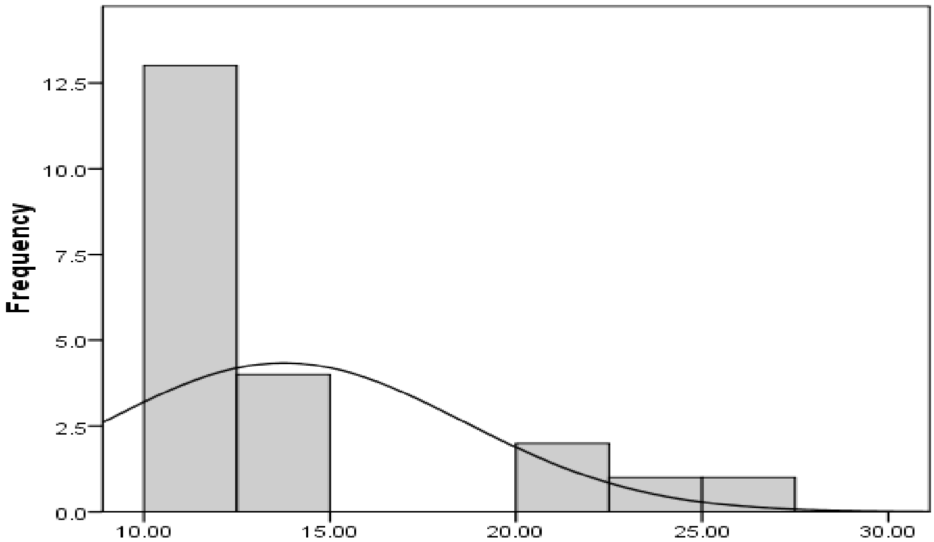

Figure 4. The Frequency Distribution of the Activity of ^{232}Th

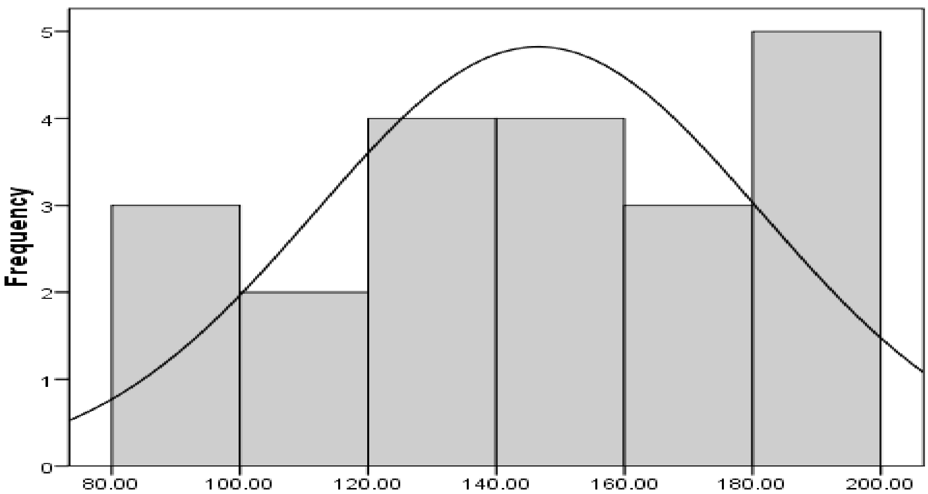

Figure 5. The Frequency Distribution of the Activity of ^{40}K

Table 1 presents the basic statistics were used to describe the statistical characteristics of the radionuclide activities. skewness is a measure of the asymmetry of the probability distribution of a real-valued random variable. The normal distribution has a skewness of zero. However, in reality, data points may not be perfectly symmetric. Therefore, an understanding of the skewness of the data set indicates whether deviations from the mean are likely to be positive or negative. Skewness characterizes the degree of asymmetry of a distribution around its mean [6]. Positive skewness indicates a distribution with an asymmetric tail extending towards values that is more positive. Negative skewness indicates a distribution with an asymmetric tail extending towards values that are more negative. Lower skewness values indicate generally normal distributions. The skewness values of ^{226}Ra, ^{232}Th and ^{40}K in this study are -0.03, 1.7 and -0.2, respectively, small skewness values (Table 1), which indicate that the distributions are asymmetric in nature.

Kurtosis is a measure of the peakedness of the probability distribution of a real-valued random variable. It characterizes the relative peakedness or flatness of a distribution compared with the normal distribution. Positive kurtosis indicates a relatively peaked distribution. Negative kurtosis indicates a relatively flat distribution. Higher kurtosis means that more of the variance is the result of infrequent extreme deviations, as opposed to frequent modestly sized deviations [2]. In our study, the ^{226}Ra and ^{40}Kdistributions have negative kurtosis values (Table 1), indicating flat distributions, while the ^{232}Th has positive kurtosis value indicating peaked distribution. The main statistical software that was used was "Microsoft Office Excel 2007".

3.2. Radium Equivalent Activities (Ra_{eq})

The radium equivalent concept allows a single index or number which is a widely used hazard index to describe the gamma output from different mixtures of uranium, thorium and potassium in samples from different locations [7]. The radium equivalent activities (Ra_{eq}) were calculated based on the estimation that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq $kg^{-1}$of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K each produce the same gamma-ray dose rate [8, 9]. Therefore, the Ra_{eq} of a sample is given by

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

Table 2. Radium Equivalent (Ra_{eq}), Representative Level Index (RLI), Absorbed Dose Rate (D), Annual Effective Dose Rate (E), Alpha index (I_α), gamma index (I_γ), Radiation Hazards, Excess Lifetime Cancer (ELCR), Annual Gonadal Dose Equivalent (AGDE), in Different Types of Building Materials of Assiut

Material	Ra_{eq} Bq kg^{-1}	RLI Bq kg^{-1}	Absorbed dose (D) (nGy h^{-1})	Annual effective dose (E)(mSv y^{-1})	I_α	I_γ	Radiation hazards		ELCR×10^{-4}	AGDE (μSv y^{-1})
							H_{ex}	H_{in}		
C1	61	0.44	52	0.256	0.07	0.22	0.17	0.20	2.2	194
C2	68	0.48	61	0.300	0.20	0.24	0.18	0.29	2.6	218
C3	43	0.32	39	0.190	0.07	0.16	0.12	0.16	1.7	143
G1	75	0.53	68	0.333	0.24	0.26	0.20	0.33	2.9	240
G2	60	0.42	54	0.264	0.17	0.21	0.16	0.25	2.3	192
G3	57	0.41	51	0.253	0.15	0.20	0.16	0.24	2.2	184
L1	49	0.35	43	0.212	0.12	0.17	0.13	0.19	1.9	156
L2	47	0.33	42	0.207	0.13	0.17	0.13	0.20	1.8	150
L3	55	0.39	49	0.239	0.14	0.19	0.15	0.23	2.1	174
S1	46	0.34	41	0.202	0.08	0.17	0.13	0.17	1.8	151
S2	47	0.34	42	0.205	0.10	0.17	0.13	0.18	1.8	152
S3	56	0.40	50	0.244	0.11	0.20	0.15	0.21	2.1	181
B1	84	0.60	75	0.367	0.20	0.30	0.23	0.34	3.2	269
B2	88	0.63	78	0.382	0.21	0.31	0.24	0.35	3.3	281
B3	88	0.63	77	0.380	0.18	0.32	0.24	0.34	3.3	281
So1	57	0.40	51	0.249	0.15	0.20	0.15	0.23	2.2	182
So2	76	0.54	69	0.341	0.23	0.27	0.21	0.33	3.0	247
So3	45	0.33	40	0.198	0.08	0.16	0.12	0.17	1.7	148
Ce1	67	0.48	61	0.300	0.18	0.24	0.18	0.28	2.6	220
Ce2	56	0.40	50	0.246	0.16	0.20	0.15	0.24	2.2	179
Ce3	65	0.45	58	0.284	0.19	0.23	0.18	0.28	2.5	205
Min.	**43**	**0.32**	**39**	**0.190**	**0.07**	**0.16**	**0.12**	**0.16**	**1.7**	**143**
Max.	**88**	**0.63**	**78**	**0.382**	**0.24**	**0.32**	**0.24**	**0.35**	**3.3**	**281**
Mean	**61**	**0.44**	**55**	**0.269**	**0.15**	**0.22**	**0.17**	**0.25**	**2.4**	**197**
SD	**14**	**0.10**	**13**	**0.062**	**0.1**	**0.05**	**0.1**	**0.22**	**0.5**	**44**

Where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, $^{232}Th$and ^{40}K respectively, in units of Bq kg^{-1}. The calculated values vary from 43 (C3) to 88 (B1 and B2) Bqkg^{-1} with an average of 61Bq kg^{-1} (Table 2). The estimated average value of Ra_{eq} in this study is lower (6 times) than the recommended maximum value of 370 Bq kg^{-1} [5] and thus does not pose any radiological hazard when used for the construction of buildings. All values of Ra_{eq} in the studied samples are found to blower than the criterion limit of 370 Bq kg^{-1} [10].

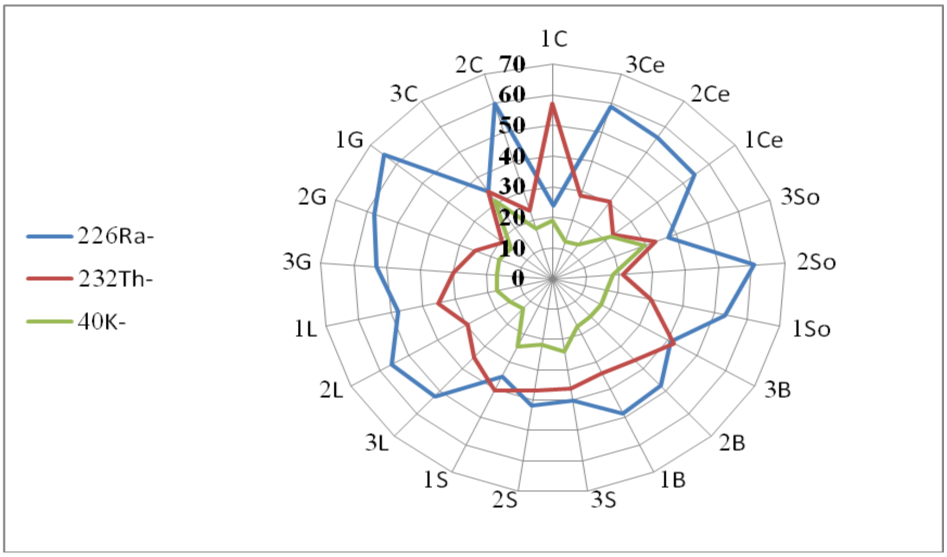

Figure 6. The Relative Concentration (%) of ^{226}Ra, ^{232}Th and ^{40}K to the Ra_{eq} in Building Materials

As shown in Figure 6, ^{226}Ra is the main contributor to Ra_{eq} in all samples, except in clay (C1), sand (S1) and brick (B3) the main contributor to Ra_{eq} is ^{232}Th. The sum of the relative contribution of ^{226}Ra and ^{232}Th to Ra_{eq} is 69.2–86.8% in all analyzed building materials. Tables 3, 4 and 5 presented a comparison of activity concentrations and radium equivalent activities (Bq kg^{-1}) in cement, sand and clay bricks in different areas of the world, respectively.

3.3. Representative Level Index (RLI)

To estimate the level of gamma radioactivity associated with different concentrations of certain specific radionuclides, known as the representative level index [2], the formula is given as

$$RLI = (A_{Ra}/150) + (A_{Th}/100) + (A_K/1500) \quad (2)$$

where A_{Ra}, A_{Th}, and A_K are the average activity concentrations of, ^{226}Ra ^{232}Th and ^{40}K, respectively. The representative level index for building material samples are presented in Table 2. The calculated RLI varies from 0.32 to 0.63 with an average of 0.44. It is clear that this average value does not exceed the upper limit for the RLI, which is unity [29]. Therefore, building materials present no radiation hazard and are not harmful to human beings.

3.4. Absorbed Gamma Dose Rate (D)

The absorbed dose rates in indoor air (D) attributed to gamma-ray emission from the radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building materials were evaluated using data and formulas provided by [5]. The dose conversion coefficients were calculated for the center of a standard room. The dimensions of this room are4 m × 5 m × 2.8 m. The thickness of the walls, floors and ceiling and the density of the structure are 20 cm and 2350 kg m^{-3} (concrete), respectively. The absorbed dose rate in indoor air (D) was determined through the following equation

Table 3. Comparison of Activity Concentrations and Radium Equivalents ($Bq\ kg^{-1}$) in Cement in Different Areas of the World

Country	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	Reference
Present work	35	12	133	63	-
Australia	51.8	48.1	115	129	[9]
Austria	26.1	14.2	210	63.1	[11]
China	69.3	62	169	189	[12]
Brazil	61.7	58.5	564	189	[13]
Germany	<26	<18	241	70.3	[10]
United kingdom	22	7	141	42.8	[10]
Sweden	55	47	241	141	[10]
Norway	30	18	241	74.3	[10]
Finland	44	26	241	99.7	[10]
Pakistan	31.3	26.8	212	85.9	[14]
Egypt	31.3	11.1	40.6	50.9	[15]
Cuba	23	11	467	74	[16]
Sicily	38	22	218	92	[17]
India	37	34	188	102	[2]
World	35	30	400	-	[5]

Table 4. Comparison of Activity Concentrations and Radium Equivalents ($Bq\ kg^{-1}$) in Sand in Different Areas of the world

Country	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	Reference
Present work	19	13	151	50	-
Australia	3.7	40	44.4	65.3	[9]
China	39.4	47.2	573	151	[18]
Brazil	14.3	18	807	102	[13]
Netherland	8.1	10.6	200	38.6	[19]
USA	37	33.3	18.5	86	[20]
Hong Kong	24.3	27.1	841	128	[18]
India	43.7	64.4	455.8	170.8	[21]
Pakistan	21.5	31.9	520	107	[14]
Egypt	9.2	3.3	47.3	16.6	[15]
Cuba	17	16	208	55	[16]
World	35	30	400	-	[5]

$$D\ (nGy\ h^{-1}) = 0.92A_{Ra}+1.1A_{Th}+0.08A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $Bq\ kg^{-1}$, respectively. The absorbed dose rate in indoor air for building materials has been listed in Table 2, fourth column. The absorbed dose rate in indoor air ranged from 39 in clay (C3) to 78 $nGy\ h^{-1}$ in brick (B3), with average value is 55 $nGy\ h^{-1}$. Figure 7 shows that the ^{226}Ra is the main contributor to the absorbed dose rate in indoor air in all samples, except in clay (C1) the main contributor to the absorbed dose rate in indoor air is ^{232}Th. The sum of the relative contribution of ^{226}Ra and ^{232}Th to the absorbed dose rate in indoor air is 64.4–85.4% in all analyzed building materials.

Table 5. Comparison of Activity Concentrations and Radium Equivalents ($Bq\ kg^{-1}$) in Clay Bricks in Different Areas of the World

Country	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	Reference
Present work	23	15	157	57	-
Australia	41	89	681	220	[9]
China	41	52	717	171	[12]
Egypt	24	24.1	258	78	[22]
Finland	78	62	962	241	[10]
Germany	59	67	673	207	[10]
Greece	49	24	670	135	[23]
Netherlands	39	41	560	141	[19]
Norway	104	62	1058	276	[24]
Sweden	96	127	962	352	[10]
Sri lanka	35	72	585	183	[25]
Kuwait	6.6	6.6	332	41.6	[26]
Malaysia	233	229	685	612	[27]
Bangladesh	29	52	292	127	[28]
Pakistan	45	61	692	187	[14]
India	5	23	374	61	[2]
World	35	30	400	-	[5]

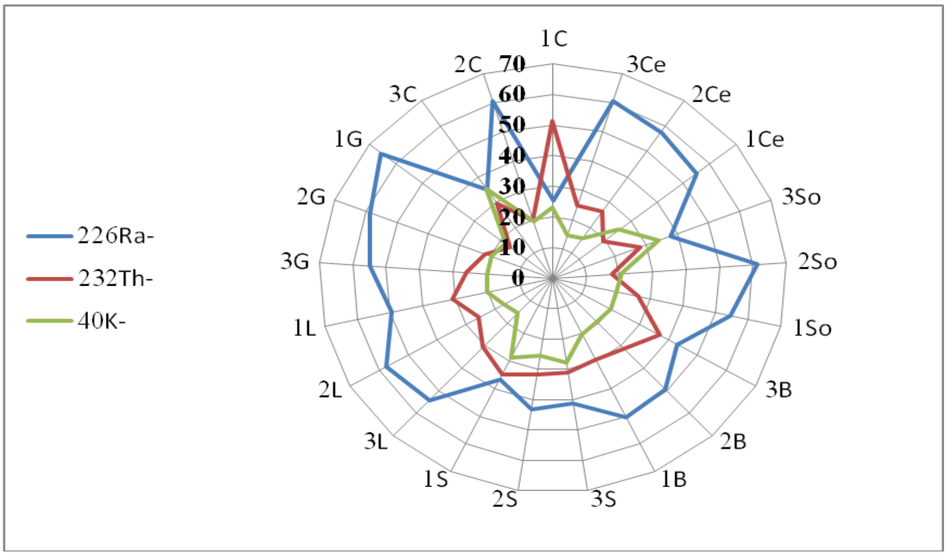

Figure 7. The Relative Concentration (%) of ^{226}Ra, ^{232}Th and ^{40}K to Absorbed Dose Rate in Building Materials

The average values of D for all studied building materials in Assiut are lower than the world population-weighted average indoor absorbed gamma dose rate of 84 $nGy\ h^{-1}$ [5].

3.5. Annual Effective Dose (E)

To estimate the annual effective dose rates, it is necessary to use the conversion coefficient from the absorbed dose in air to the effective dose (0.7 $Sv\ Gy^{-1}$) and the outdoor occupancy factor (0.2) proposed by [5]. Therefore, the effective dose rate is determined as follows:

$$\text{Annual effective dose } (mSv\ y^{-1}) = D\times8766\times0.8\times0.7\times10^{-6} \quad (4)$$

where D is the absorbed dose rate in indoor air. Annual effective dose rates (E) are given in the fifth column of Table 2. As shown in table 2, the annual effective dose rates ranged from 0.19 in clay (C3) to 0.382 mSv y^{-1} in brick (B2), with mean value of the annual effective dose rate of 0.269 mSv y^{-1}. The relative contributions of ^{226}Ra, ^{232}Th and ^{40}K to Annual effective dose are the same at absorbed dose rate in indoor air.

3.6. Alpha Index (I_α) and Gamma Index (I_γ)

Also, several indexes dealing with the assessment of the excess alpha radiation due to the radon inhalation originating from building materials called ‘‘alpha-indexes’’ or ‘‘internal-indexes’’ (I_α) have been developed [8]. In the present work, the alpha-indexes were determined through the following formula:

$$I_\alpha = (A_{Ra}/200) \quad (5)$$

When the ^{226}Ra activity concentration (A_{Ra}) of building material exceeds the value of 200 Bq kg^{-1}, it is possible that the radon exhalation from this material could cause indoor radon concentration exceeding 200 Bqm^{-3}. The calculated values of (I_α) are listed in Table1. The recommended exemption level and recommended upper level for the ^{226}Ra activity concentrations in building materials are 100 Bq kg^{-1} and 200 Bq kg^{-1}, respectively, in building materials as suggested by the Radiation Protection Authorities in Denmark, Finland, Iceland, Norway and Sweden [30]. Table 1 presented that, the values of the alpha index in the studied samples are below the recommended limit, i.e., $I_\alpha < 1$, Therefore, radon inhalation from the building material samples under investigation is not so large as to restrict the use of these materials in construction.

In order to assess whether the safety requirements for building materials are being fulfilled, a gamma index I_γ is calculated as proposed by the European Commission [31]:

$$I_\gamma = (A_{Ra}/300) + (A_{Th}/200) + (A_K/3000) \quad (6)$$

$I_\gamma \le 2$ correspond to an absorbed gamma dose rate of 0.3 mSv y^{-1}, whereas $2 < \gamma \le 6$ corresponds to an absorbed gamma dose rate of 1 mSv y^{-1} [31, 32]. Thus, the activity concentration index should be used only as a screening tool for identifying materials that might be of concern when used as construction materials; although materials with $I_\gamma > 6$ should be avoided, these values correspond to dose rates higher than 1 mSv y^{-1}, which is the highest dose rate value recommended for the population [2]. The gamma index I_γ for the building materials varies between 0.16in clay (C3) and 0.32in brick (B3) with an average of 0.22, as presented in Table 1. Therefore, the annual effective dose delivered by the building materials is smaller than the annual effective dose constraint of 1 mSv y^{-1}. Therefore, these building materials can be exempted from all restrictions concerning radioactivity.

3.7. External and Internal Hazard Indices

The external hazard index Hex can be calculated by the following equation [9]:

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/258) + (A_K/4810) \quad (7)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. The value of this index must be less than the unity in order to keep the radiation hazard to be insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}). Table 2 shows that, the H_{ex} values ranged from 0.12 to 0.24 with average value 0.17 is below the criterion value < 1. As shown in Figure 8, the main contributor to external radiation hazard is the ^{226}Ra for all building materials, except for clay (C1), sand (S1) and brick (B3). The sum of the relative contribution of ^{226}Ra and ^{232}Th to external radiation hazard is 69.2–87.5% in all analyzed building materials

In addition to the external hazard index, radon and its short-lived progeny are also hazardous to the respiratory organs. The internal exposure to radon and its daughter progenies is quantified by the internal hazard index H_{in} [5], which is given by the equation:

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_K/4810) \quad (8)$$

The internal hazard index is defined to reduce the acceptable maximum concentration of ^{226}Ra to half the value appropriate to external exposure alone. For the safe use of materials in the construction of dwellings, the H_{in} must be less than unity [8]. As presented in table 1, the internal radiation hazard is less than one, indicating that the internal hazard is below the critical value. ^{226}Ra is the main contributor to internal hazard index in all building materials except for clay (C1), Figure 9. The sum of the relative contribution of ^{226}Ra and ^{232}Th to internal hazard index is 77.1–92.1% in all analyzed building materials.

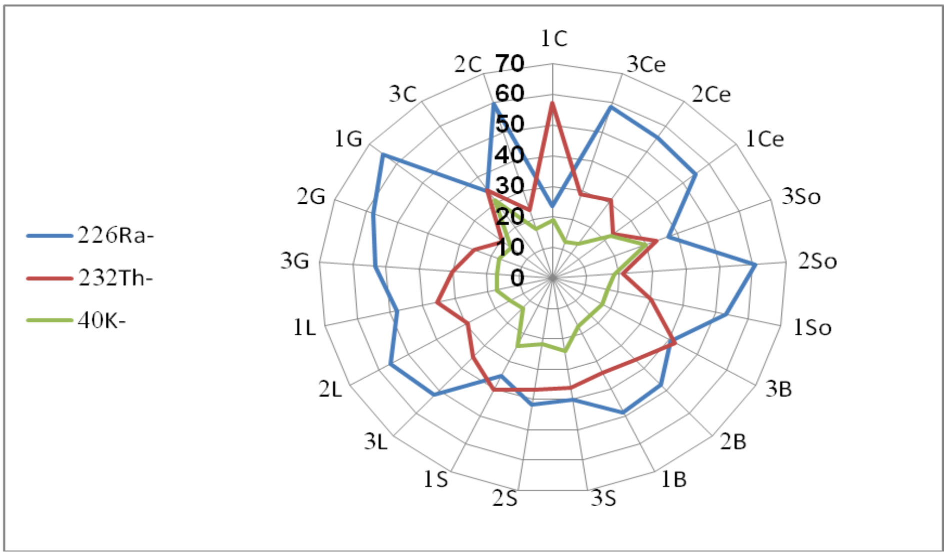

Figure 8. The Relative Concentration (%) of ^{226}Ra, ^{232}Th and ^{40}K to External Radiation Hazard (H_{ex}) in Building Materials

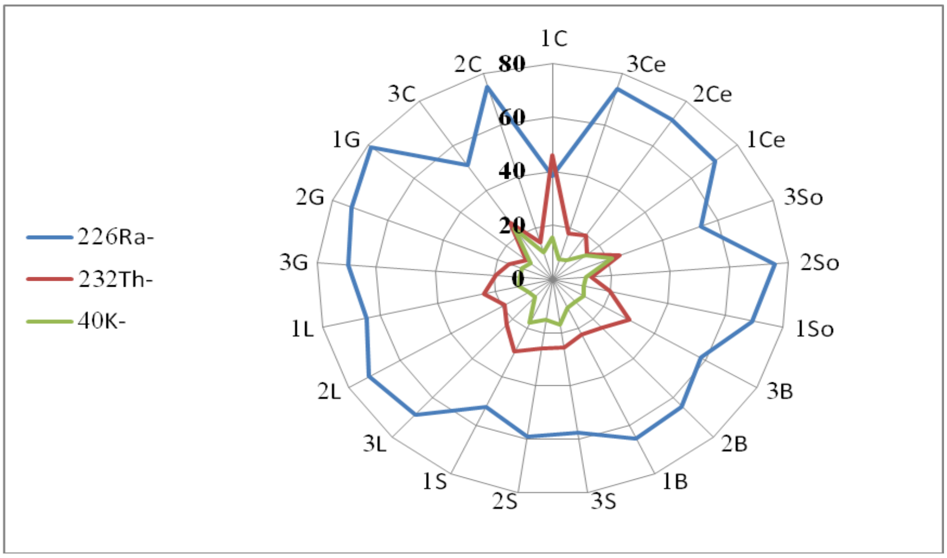

Figure 9. The Relative Concentration (%) of ^{226}Ra, ^{232}Th and ^{40}K to Internal Hazard Index (H_{in}) in Building Materials

3.8. Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) was calculated using the following equation and presented in Table 2.

$$ELCR = AEDE \times DL \times RF \quad (9)$$

where AEDE, DL and RF are the annual effective dose equivalent, duration of life (70 y) and risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public [33]. The calculated value of ELCR showed that the highest value was in brick (B2 and B3), while the lowest value was in clay (C3), with an average of 2.4×10^{-4}. The average ELCR value is lower than average (0.29×10^{-3}) [5].

3.9. Annual Gonadal Dose Equivalent (AGDE)

In the same context, the activity of bone marrow and bone surface cells are considered to be organs of interest by [34]. Therefore, the annual gonadal dose equivalent (AGDE) arising from the specific activities of ^{226}Ra, ^{232}Th and ^{40}K was calculated using the following formula [35]:

$$AGDE\ (\mu Sv\ y^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (10)$$

The AGDE values are presented in Table 2. The highest value is found to be 281 $\mu Sv\ y^{-1}$. In the literature, the average AGDE value for the Eastern Desert of Egypt was found to be 2398 $mSvy^{-1}$ [36]. This value is higher than our results. The annual gonadal dose equivalent results do not exceed the permissible recommended limits, indicating that the hazardous effects of the radiation are negligible.

4. Statistical Analysis

4.1 Principal Component Analysis (PCA)

Principal component analysis (PCA) was applied between the studied variables using varimax rotation with Kaiser normalization method (Table 6). Factor analysis yielded two factors with eigen value <1, explaining 95.215% of the total variance.

Table 6. Rotated Factor Loading of the Variables

Variables	Component	
	1	2
^{226}Ra	0.999	0.009
^{232}Th	0.064	0.910
^{40}K	0.183	0.736
Ra_{eq}	0.804	0.592
RLI	0.777	0.629
D	0.832	0.554
E	0.831	0.556
Iα	0.999	0.009
Iγ	0.773	0.632
H_{ex}	0.783	0.616
H_{in}	0.929	0.367
ELCR	0.849	0.523
AGDE	0.799	0.601
Variance explained in %	**83.972**	**11.243**

Normally, an ordination result was good if the value was 75% or better [37]. As seen from Table 6, the first component (PC1) explained 83.972% of the total variance and loaded heavily on ^{226}Ra series associated with all radiological parameters. The second component (PC2) was correlated very strongly with ^{232}Th with a high loading value (0.910), accounting for 11.243% of the total variance. Figure 10 shows the rotated factor loadings of radiological parameters.

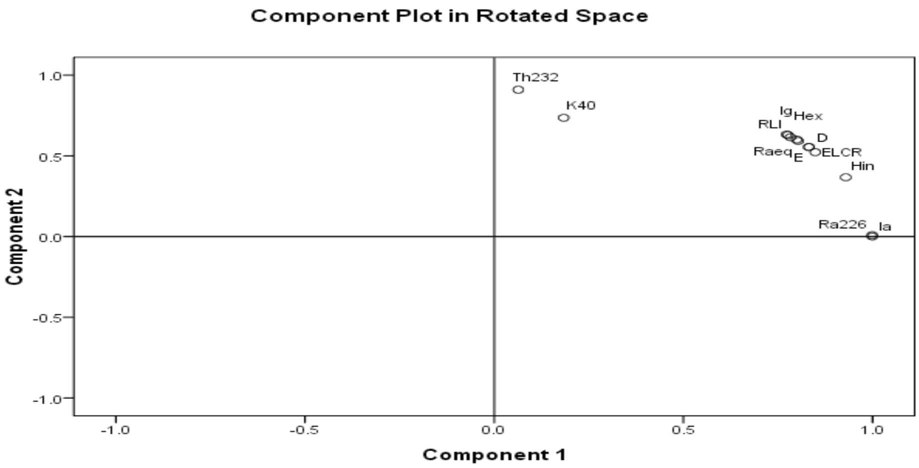

Figure 10. Graphical Representation of Factors 1 and 2

4.2 Cluster Analysis (CA)

To confirm the existing correlation between the variables, cluster analysis (CA) is carried out. It is a multivariate technique, whose primary purpose is to classify the objects of the system into categories or clusters based on their similarities, and the objective is to find an optimal grouping for which the observations or objects within each cluster are similar, but the clusters are dissimilar from each other. The dendrogram visually displays the order in which parameters or variables combine to form clusters with similar properties. The 100% similarity means that the clusters were zero distance apart in their sample measurements, whereas similarity of 0% means the cluster areas are as disparate as the least similar region. In this study cluster analysis is performed using the average linkage method, to calculate the Euclidean distance between the variables. The derived dendrogram is shown in Figure11.

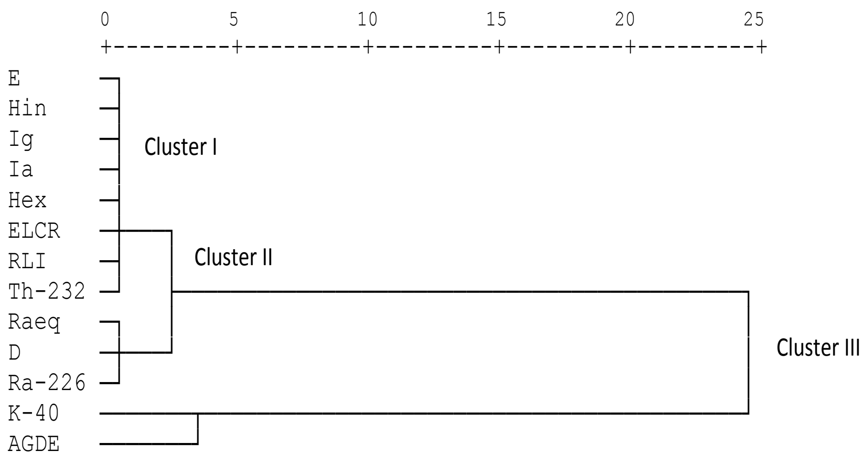

Figure 11. Dendrogram Shows Cluster Formation Between Radiological Parameters of Building Material Samples

In this dendrogram, all 13 parameters are grouped into three statistically significant clusters. Cluster-I consists of annual effective dose rates, external and internal indices, alpha and gamma indices, excess lifetime cancer risk, representative level index and ^{232}Th. Cluster-II consists of ^{226}Ra and mainradiological parameters distribution, such as D and Ra_{eq}, which means that, the radium equivalent and the absorbed gamma dose rate in the building materials are due to the concentration of ^{226}Ra. Cluster-III consists of annual gonadal dose equivalent and ^{40}K. It means that, the AGDE in the building materials is due to the content of potassium.

4.3 Pearson's Correlation Coefficient Analysis

Correlation analysis was carried out in terms of bivariate statistics to determine the mutual relations and strengths of association between pairs of variables through the calculation of the linear Pearson correlation coefficients. The results for the Pearson correlation coefficients between all the studied radioactive variables for the building materials are shown in Table 7. All radioactive variables have strong positive correlation coefficients with ^{226}Ra, ^{232}Th and ^{40}K.

4. Conclusion

Natural radioactivity levels in building materials (clay, gypsum, limestone, sand, brick, soil and cement) in the environments of Assiut, Egypt have been measured using gamma-spectrometry system. The radium equivalent activity (Ra_{eq}), indoor gamma absorbed dose rate (D), annual effective dose, alpha index (I_{α}), gamma index (I_{γ}), external radiation hazard index (H_{ex}), internal radiation hazard index (H_{in}), representative level index (RLI), excess lifetime cancer risk (ELCR) and annual gonadal dose equivalent (AGDE) were determined. The obtained results in our study are within the recommended safety limits, demonstrating that these building materials do not pose any significant radiation hazard; thus, the use of these materials in the construction of dwellings can be considered safe for the inhabitants.

Acknowledgments

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Table 7. Pearson Correlation Matrix Among the Variables

variables	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	RLI	D	E	I_{α}	I_{γ}	H_{ex}	H_{in}	ELCR	AGDE
^{226}Ra	1												
^{232}Th	0.057	1											
^{40}K	0.213	0.415	1										
Ra_{eq}	0.806	0.611	0.551	1									
RLI	0.780	0.632	0.589	0.998	1								
D.	0.836	0.561	0.555	0.998	0.995	1							
An.	0.835	0.562	0.556	0.998	0.995	1	1						

I_{α}	0.998	0.060	0.191	0.803	0.776	0.831	0.831	1					
I_{γ}	0.776	0.639	0.583	0.997	0.998	0.993	0.994	0.772	1				
H_{ex}	0.785	0.636	0.554	0.996	0.996	0.993	0.993	0.781	0.997	1			
H_{in}	0.931	0.400	0.429	0.965	0.953	0.976	0.976	0.928	0.951	0.955	1		
ELCR	0.853	0.514	0.564	0.990	0.986	0.995	0.995	0.848	0.985	0.982	0.978	1	
AGDE	0.804	0.599	0.586	0.999	0.999	0.998	0.998	0.799	0.997	0.995	0.963	0.992	1

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