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Determination of natural radioactivity in building raw materials from the quarries of Assiut cement company, Assiut, Egypt.

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Abstract: By using gamma spectrometry $(NaI\ (Tl)\ 3" \times 3")$, the concentrations of (^{226}Ra, ^{232}Th and ^{40}K) were measured and its radiological hazards were presented in this paper for some building raw materials (sand, clay and limestone) collected from the quarries of Assiut cement company, Assiut, Egypt. The concentration values of (^{226}Ra, ^{232}Th and ^{40}K) were in between (3.6 $\pm$ 0.4 and 16.8 $\pm$ 1.4), (1.1 $\pm$ 0.2 and 11.9 $\pm$ 1.6) and (4.3 $\pm$ 0.3) and (155.2 $\pm$ 9) respectively. The radiation hazard indices like: Radium equivalent (Ra_{eq}), external hazard index (H_{ex}), activity concentration index (I_{ex}), the specific dose rates in door (D), the annual effective dose (DE) due to gamma radiation and the annual gonadal dose equivalent $(AGDE)$ were calculated , it was below the world average value 300 $Sv\ y^{-1}$. The excess lifetime cancer risk $(ELCR)$ have been calculated, its values were lower than the world's average value of (0.29 x 10^{-3}) comparing with internationally recommended values.

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

1 Introduction

We live with radiation every day and everywhere. Wherever we are, whenever we are, we will be in natural radioactive zone. The air we breathe, the food we eat, the drinks we drink even we ourselves are containing natural radioactive materials.Building materials can cause significant gamma dose indoors, due to their natural radionuclide content. Moreover, they can also be a source of indoor radon. A large database of activity concentration measurements of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in building material has been set up in the last years [1]. Knowledge of basic radiological parameters, such as radioactive contents in building materials, is important in the assessment of possible radiation exposure of the population.Because most people spend 80% of their time indoors,this knowledge is essential for the development of standards and guidelines for the use of these materials [2]. Normally, the two main routes of indoor exposure are terrestrial gamma-ray irradiation and radon isotope inhalation. It has been demonstrated in various studies that, if building materials with high natural radioactivity concentration are employed, dose rates indoors will be elevated accordingly [3]. Natural radioactive material in rocks and soilaccount for about 28 millirem or 8% of the radiation dose a person typically receives in a year from all sources including medical exposures. Since radiation of natural origin is responsible for most of the total radiation exposure,knowledge of the dose received from natural sources is very important in the discussion not only of its effects on health but also of the incidence of other radiation from man-made sources [4] . This study aims to determine the natural radioactive levels (^{226}Ra, ^{232}Th and ^{40}K) and evaluate its radiological hazards from natural building raw material samples extracted from the quarries of Assiut cement company, Assiut, Egypt.

2 Materials and methods

2.1 Sample collection and preparation

Sixteen samples were collected from five building material quarries in Assiut Cement Company between

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February 2011 and February 2013 these quarries and samples are:
1) Sands (3 samples: 1, 2 and 3).
2) (valley road 2091), Limestone, (3 samples: 4, 5 and 6).
3) (valley road 1252), Limestone ,(3 samples: 7, 8 and 9).
4) (valley road 1251), Clay (4 samples :10, 11, 12 and 13).
5) (Jahdam 1254), Clay (3 samples: 14, 15 and 16). Each sample was dried in an oven at about 110^oC to ensure that moisture was completely removed. The samples were crushed, homogenized and sieved through a 200 μm mesh, which is the optimum size enriched in heavy minerals. Weighed samples were placed in polyethylene beaker. 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]. This step is necessary to ensure that radon gas confined 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 8 hours at least. 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 energy calibration was made using ^{137}Cs (661.9 *KeV*) and ^{60}Co (1173.2 and 1332.5 *KeV*), however, the efficiency calibration was made by calibration cylindrical beaker standard source *IAEA-314* , where the specific activity was known [8], which containing three radionuclides: ^{226}Ra, ^{232}Th and ^{238}U. The same cylindrical beakers were used in measurements of samples and correction on geometry was not necessary. The absolute efficiency was calculated by using the equation [9]:

$$eff = \frac{N_p \times 100}{I_\gamma \times TOC \times A_{BOC}} \quad (1)$$

Where N_p the net peak area, I_γ the intensity of emitted gamma ray, TOC the time of counting and $ABOC$ the activity of the standard source at beginning of counting (BOC). $ABOC$ was calculated by equation [9]:

$$A_{BOC} = A_{DOR} exp(-\lambda(BOC - DOR)) \quad (2)$$

Where A_{DOR} is the activity of the standard source at date of reference (DOR) and λ is the decay constant. Some fitting function is needed to calculate the absolute efficiency for any considered gamma energy. A function 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 [10]:

$$\eta = a - bexp([-cE^d]) \quad (3)$$

Where E, represents energy in *MeV*, a, b, c and d are coefficient data. By equation (3), the absolute efficiency,η , can be determined at any specific energy E, 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 professional 1.5.0.

2.3 Uncertainty of efficiency

[9] The combined standard uncertainty of absolute efficiency $u(eff)$ consists of $u(N_p)$, $u(I_\gamma)$, $u(TOC)$ and $u(A_{BOC})$. So

$$[\frac{u(eff)}{eff}]^2 = [\frac{u(N_p)}{N_p}]^2 + [\frac{u(I_\gamma)}{I_\gamma}]^2 + [\frac{u(TOC)}{TOC}]^2 + [\frac{u(ABOC)}{ABOC}]^2 \quad (4)$$

Because $u(TOC) \ll TOC, u(TOC)$ was neglected. The value of $u(A_{BOC})$ was calculated by equation [9]:

$$[\frac{u(ABOC)}{ABOC}]^2 = [\frac{u(ADOR)}{ADOR}]^2 + (BOC - DOR)u^2(\lambda) \quad (5)$$

$u(N_p)$ was obtained from the code Genie 2000 [11] whereas $u(\lambda)$ and $u(I_\gamma)$ were taken from the compilation of Reus and Westmeier [12] The total uncertainty of the full-energy-peak efficiency of 5.28%.

3 Results and discussion

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. The 1460.6 *KeV* gamma ray was used to determine ^{40}K. The 186 *KeV* photon peak of ^{226}Ra was not used because of the interfering peak of ^{235}U, with an energy of 185.7 *KeV* [13]. The activity concentration in $B_q/k_g(A)$ in the environmental samples was obtained as follows [9]:

$$A = \frac{N_p \times 100}{\eta \times m \times I_\gamma} \quad (6)$$

Where N_p is the net peak area, I_γ the intensity of emitted gamma ray , η the measured efficiency for each gamma line and m the

Table 1: Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/kg) of the studied samples

Sample type	*Sample name*	^{226}Ra	^{232}Th	^{40}k
	1	3.6 ± 0.4	2.2 ± 0.4	40.3 ± 2.5
sand	2	4.2 ± 0.4	4.7 ± 0.8	48.6 ± 2.9
	3	11.3 ± 0.8	1.7 ± 0.2	43.7 ± 2.5
mean		6.4 ± 0.3	2.9 ± 0.3	44.2 ± 1.5
	4	5.5 ± 0.6	1.3 ± 0.3	4.3 ± 0.3
Limestone (valley road) (2091)	5	6 ± 0.5	1.4 ± 0.3	5.2 ± 0.3
	6	4.9 ± 0.4	1.1± 0.2	7.4 ± 0.5
mean		5.5 ± 0.3	1.3 ± 0.2	5.6 ± 0.2
	7	6.6 ± 0.7	1.5 ± 0.3	17.6 ± 1.2
Limestone (valley road) (1252)	8	5.3 ± 0.5	1.2 ± 0.3	10.4 ± 0.7
	9	5.7 ± 1.3	2.5 ± 0.5	16.5 ± 1
mean		5.9 ± 0.5	1.7 ± 0.2	14.8 ± 0.6
	10	9.6 ± 0.7	10.4 ± ±1.4	155.2 ± 9
Clay (valley road)	11	11.9 ± 2	12.5 ± 1.7	86.1 ± 5
	12	9.5 ± 0.7	11.6 ± 1.7	151.5 ± 8.8
	13	11.7 ± 1	10.9 ± 1.6	121.5 ± 7.5
mean		10.7 ± 0.6	11.4 ± 0.8	128.6 ± 3.9
	14	16.8 ± 1.4	8.8 ± 1.3	146.8 ± 9.2
clay (Jahdam)	15	15.1 ± 1.4	11.9 ± 1.6	147.9 ± 8.5
	16	15.3 ± 1.5	6.5 ± 0.9	112.6 ± 6.8
mean		15.7 ± 0.8	9± 0.8	135.8 ± 4.8

mass of the sample in kilograms. The activities of ^{226}Ra, ^{232}Th series and ^{40}K in Bq/kg determined for each of the measured samples together with their total uncertainties are presented in table (1).

The obtained results, in table (1), show that the values of the measured specific gamma ray activities ($Bq\ kg^{-1}$) in different samples as follows: for ^{226}Ra the activity concentrations are ranged from (3.6 ± 0.4) to (16.8 ± 1.4) $Bq\ kg^{-1}$ for sand (1) and Clay (Jahdam) sample (14), respectively, whereas the activity concentration values of ^{232}Th are between (1.1 ± 0.2) and (11.9 ± 1.6) $Bq\ kg^{-1}$ for Limestone (valley road)2091(6) and Clay (Jahdam) samples (15), respectively. The ^{40}K activity concentrations ranged between (4.3 ± 0.3) and (151.5 ± 8.8) $Bq\ kg^{-1}$ for Limestone (valley road) 2091 (4) and Clay (Jahdam) samples (12), respectively. The comparison between the specific activity of ^{226}Ra, ^{232}Th and ^{40}K for building material samples with other regions of the world was listed in table (2).

The obtained results indicate that the concentrations of natural radionuclides are different in different types of samples. This is due to the different compositions of these materials and the random distribution of the radionuclide within the samples. The variation observed in similar materials is also a function of the local geology as building materials are extracted from different regions of earth crust [14] . In figure (1), we can see the obtained results of Table 1 in graphical form; it is clearly indicating the high- and low activity samples. When we test the coloration between and ^{40}k as it appears in figure 2 and 3, we found that, the correlation between ^{232}Th and ^{226}Ra in samples under investigation is low ,with correlation coefficient (R^2 = 0.4758). In the other side, there is a good correlation between the concentrations of ^{40}K, and ^{232}Th with correlation coefficient (R2 = 0.8266) can be seen in figure (3).

3.1 Evaluation of radiological hazard effects

3.1.1 Radium equivalent activity:

In order to evaluate the radiation hazards associated with ^{226}Ra, ^{232}Th and ^{40}K, an index known as radium equivalent activity Ra_{eq} has been introduced. This concept allows a single index to describe the radiation hazard from different radionuclide mixtures in a material. Assuming 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

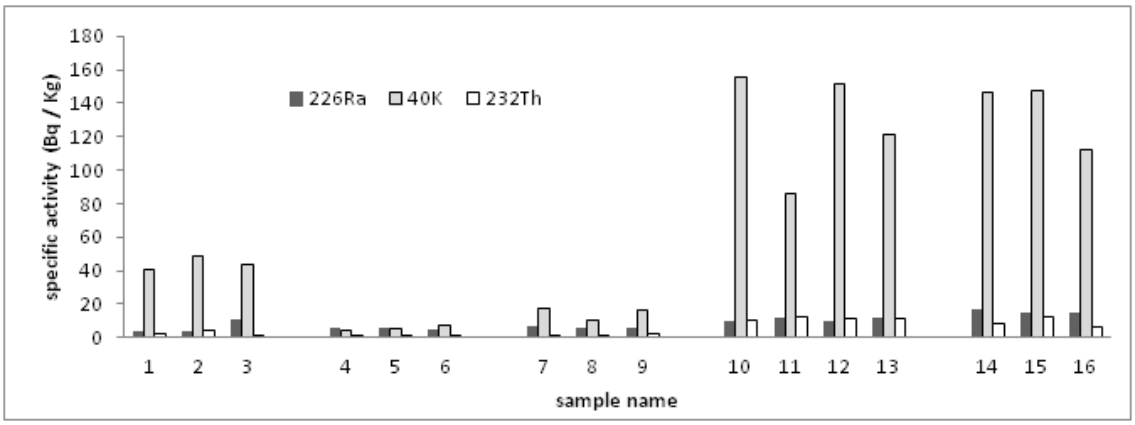

Fig. 1: Specific Activity of the radioelements (in Bq/kg) founded in studied samples.

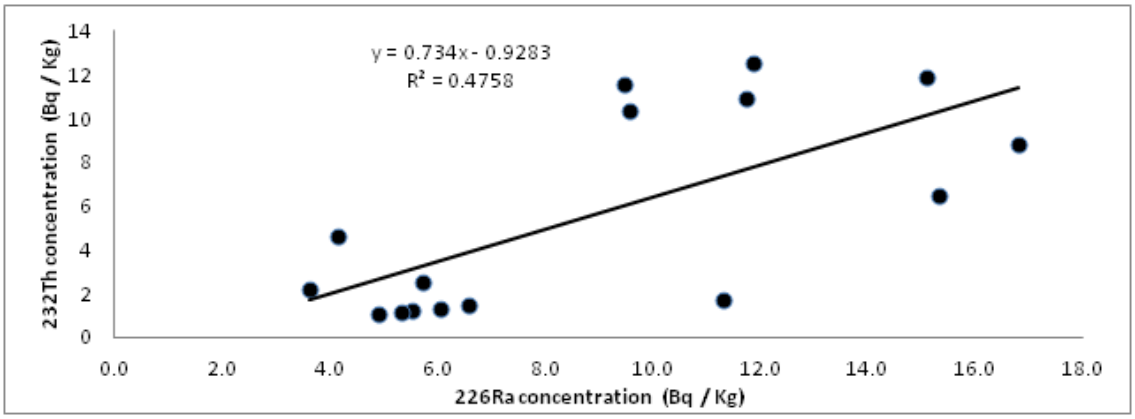

Fig. 2: The correlation between ^{226}Ra and ^{232}Th concentration in the samples.

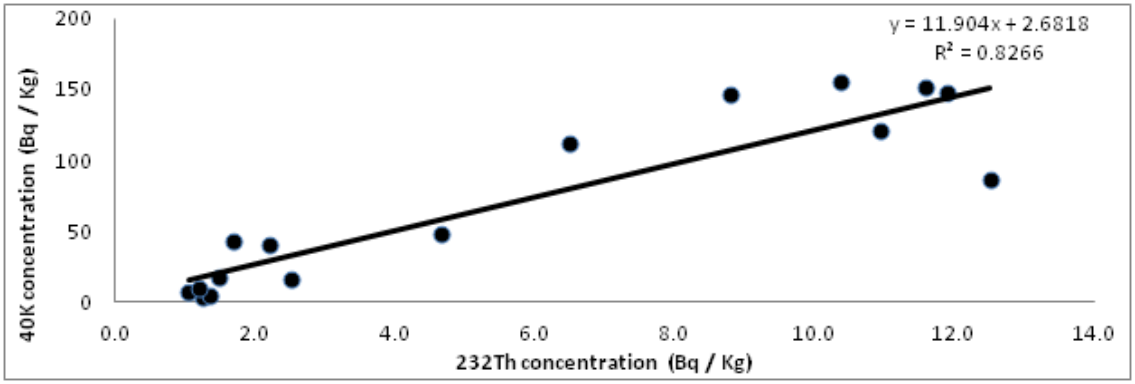

Fig. 3: The correlation between ^{40}K and ^{232}Th concentration in the samples.

Table 2: Comparison between average specific activity of ^{226}Ra ,^{232}Th and ^{40}K for studied samples with those of other countries

Region	Type	^{226}Ra	^{232}Th	^{40}K	Reference
Jordan	*limestone*	10.82 ± 0.22	0.7 ± 0.26	4 ± 0.18	[15]
Algeria	*limestone*	16	13	36	[18]
China	*limestone*	19.5	13.4	63.2	[19]
Turkey	*limestone*	11.9	5.4	52.7	[20]
Egypt, Assiut	*Limestone (valley road)*(2091)	5.5 ± 0.3	1.3 ± 0.2	5.6 ± 0.2	*Present work*
Egypt, Assiut	*Limestone (valley road)* (1252)	5.9 ± 0.5	1.7 ± 0.2	14.8 ± 0.6	*present work*
Greece	*sand*	12 ± 3	2.6 ± 3.6	–	[17]
Egypt	*sandstone*	7.5 ± 1.5	12.5 ± 3	263.911	[21]
Yemen Juban	*sandstone*	32.1	22.3	190.9	[22]
Pakistan	*Sand*	20	29	383	[23]
Egypt, Assiut	*Sand*	6.4 ± 0.3	2.9 ± 0.3	44.2 ± 1.5	*present work*

same gamma-ray dose rate (*UNSCEAR*, 1988), Ra_{eq} is calculated using the following equation :

$$Ra_{eq} = A_{Ra} + 1.43A_{th} + 0.077A_k \quad (7)$$

Where A_{Ra}, A_{Th} and $A_K$are the specific activity of ^{226}Ra, ^{232}Th and ^{40}K in $Bq\ kg^{-1}$, respectively. From table 3 we can see that the values of Ra_{eq} is ranged from 7 ($Bq\ kg^{-1}$) in (6) Limestone (valley road) 2091 to 43.5(Bq/Kg) in (15) Clay (Jahdam). These values are clearly smaller than the recommended maximum value for the safe use of materials in the construction of buildings 370 $Bq\ kg^{-1}$ [14].

3.1.2 Absorbed dose rate (D):

The measured activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K are converted into doses by applying the conversion factors 0.462, 0.604, and 0.0417 for uranium, thorium, and potassium, respectively (*UNSCEAR*, 2000). These factors are used to calculate the total dose rate *(D)(nGy* h^{-1}*)* using the following equation [13]:

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_k \quad (8)$$

From table (3) we can see that the values of (D) is ranged from 3.2 (*(D)(nGy* h^{-1}*)*) in Limestone (valley road) 2091 (6) to 20.3 (*(D)(nGy* h^{-1}*)*) in Clay (Jahdam) (15). These values are clearly smaller than the world average value of D (57 *(D)(nGy* h^{-1}*)*) (*UNSCEAR*, 2000).

3.1.3 Annual effective dose equivalent$(AEDE)$:

Annual estimated average effective dose equivalent $(AEDE)$ received by an individual was calculated using a conversion factor of 0.7 $Sv\ Gy^{-1}$, which was used to convert the absorbed rate to the human effective dose equivalent with an outdoor occupancy of 20%and 80% for indoors (*UNSCEAR*, 1993). The annual effective dose is determined using the following equations:

$$AEDE(outdoor)(\mu Sv\ y^{-1}) = absorbeddose\ (nGy\ h^{-1}) \times 8760\ h \times 0.7\ Sv\ Gy^{-1} \times 0.2 \times 10^{-3} \quad (9)$$

$$AEDE\ (indoor)\ (\mu Sv\ y^{-1}) = absorbeddose\ (nGy\ h^{-1}) \times 8760\ h \times 0.7\ Sv\ Gy^{-1} \times 0.8 \times 10^{-3} \quad (10)$$

From table (3) we can see that the values of *AEDE* (outdoor) is ranged from 3.9 ($\mu Sv\ y^{-1}$) in Limestone (valley road) 2091 (6) to 24.9 ($\mu Sv\ y^{-1}$) in Clay (Jahdam)(15). Also the values of AEDE (indoor) is ranged from 15.7($\mu Sv\ y^{-1}$) in (6) Limestone (valley road) 2091 to 99.7($\mu Sv\ y^{-1}$) in (15) Clay (Jahdam). These values are clearly smaller than the world average value of indoor *AEDE* (450 $\mu Sv\ y^{-1}$) and for outdoor *AEDE* (70 $\mu Sv\ y^{-1}$). Figure (4). Show Values of radium equivalent *Raeq* ($Bq\ .kg^{-1}$), the dose rate ($nGy\ h^{-1}$) and annual effective dose equivalent $(AEDE)$ ($\mu Sv\ y^{-1}$) for samples in graphical form.

From this figure we can see that sample (15) has the greatest values of Ra_{eq} (Bq/kg), the dose rate (nGy/h) and annual effective dose equivalent $(AEDE)$ ($\mu Sv\ y^{-1}$) and sample (6) has the smallest values but all values are in control with no significant radiation hazards. The greatest value of *AEDE* is 124.6 ($\mu Sv\ y^{-1}$) for sample (15) and it is very small compared with the allowed dose of 1.0 ($\mu Sv\ y^{-1}$) (*ICRP*-60 1990) as the maximum annual dose to members of the public.

3.2 Hazard indices:

Beretka and Mathew (1985) defined 2 indices that represent external and internal radiation hazards. The prime objective of these indices is to limit the radiation dose to a dose equivalent

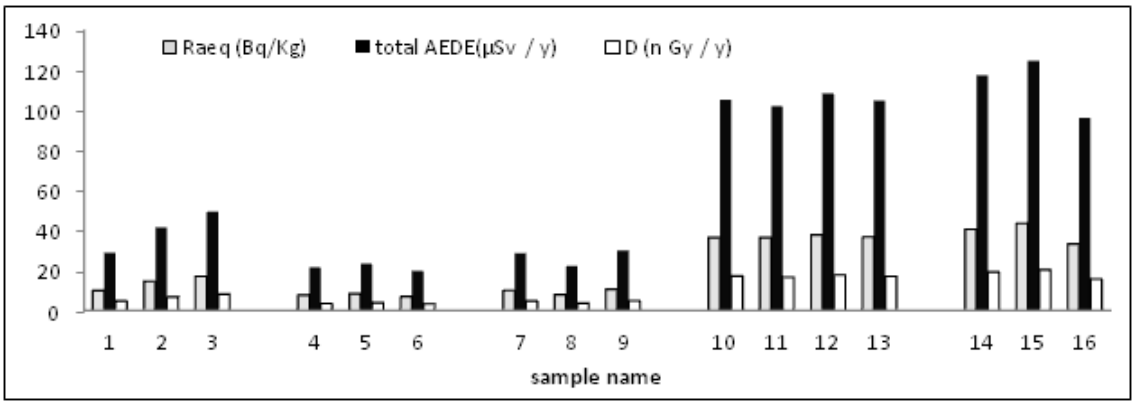

Fig. 4: Values of radium equivalent Ra_{eq} (Bq/kg)),the dose rate (nGy/h) and annual effective dose equivalent $(AEDE)$ $(\mu Sv\, y^{-1})$ for samples.

limit of 1 $mSv\ y^{-1}$. The external hazard index (H_{ex}) is calculated using the given equation [13]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \tag{11}$$

The H_{ex}must not exceed the limit of unity for the radiation hazard to be negligible.

From table (3) we can see that the values of H_{ex} is ranged from 0.0189 in (6) Limestone (valley road) 2091 to 0.1175 in (15) Clay (Jahdam). These values are clearly smaller than unity.

3.3 Gamma index (I_γ):

Another radiation hazard, called the gamma activity concentration index (I) , has been defined by the European Commission $(EC, 1999)$, and it is given below [13]:

$$I_\gamma = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_k}{4000} \tag{12}$$

The I_γ is correlated with the annual dose rate due to the excess external gamma radiation caused by superficial material. Values of I_γ of ≤ 2 correspond to a dose rate criterion of 0.3 $mSv\ year^{-1}$, whereas $I_\gamma \leq 6$ corresponds to a criterion of 1 $mSv\ y^{-1}$. Thus, I_γ should be used only as a screening tool for identifying materials that might be of concern to be used as construction materials, though materials with $I_\gamma > 6$ should be avoided since these values correspond to dose rates higher than 1 $mSv\ y^{-1}$, which is the highest value of the dose rates recommended for humans. From table 3 we can see that the values of I_γ is ranged from 0.02 to 0.15 . Figure (5) show values of external hazard index (H_{ex}) and gamma activity concentration index I_γ for samples in graphical form. From this figure we can see that sample (15) has the greatest values and sample (6) has the smallest values but all values are in control with no significant radiation hazards.

3.4 Annual gonadal dose equivalent:

The annual gonadal dose equivalent $(AGDE)$ due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using the following formula [23] :

$$AGDE\,(\mu Sv\ year^{-1}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \tag{13}$$

From table (3) we can see that the values of $AGDE$ is ranged from 21. to 142.8 $\mu Sv\, y^{-1}$, these values is lower than the world average values for soil 300 $\mu Sv\, y^{-1}$ [23].

3.5 Excess lifetime cancer risk $(ELCR)$

Excess lifetime cancer risk $(ELCR)$ was calculated by using the following equation:

$$ELCR = AEDE \times DL \times RF \tag{14}$$

Where DL is duration of life (70 year) and RF is risk factor (Sv^{-1}) fatal cancer risk per Sievert. For stochastic effects, $[ICRP\ 60]$ uses values of 0.05 for the public. From table (3) we can see that the values of $ELCR$ is ranged from 68.9 x 14^{-4} in (6) Limestone (valley road) 2091 to 436.2 x 14^{-4} in (15) Clay (Jahdam) all are lower than the world's average value of $(0.29x10^{-3})$. Figure (6) show values of annual gonadal dose equivalent $(AGDE)(Sv\ y^{-1})$ and Excess lifetime cancer risk $(ELCR)$ for samples in graphical form. From it, we can see that clay sample (14) has the greatest values while limestone sample (8) has the smallest one, but all values haven't significant radiation hazards.

4 Conclusions

The specific activity of natural radionuclides ^{238}U (^{226}Ra), ^{232}Th and ^{40}K in the row building materials samples ,which extracted from Assiut Cement company's quarries were found to be within the average worldwide ranges. Radium equivalent

Table 3: radiological hazard for the samples

Sample type	Sample name	Ra_{eq} (Bq / Kg)	H_{ex}	D nGy/h	AEDE(outdoor) (μ Sv y^{-1})	AEDE (indoor) (μ Sv y^{-1})	I_γ	AGDE (μ Sv y^{-1}	ELCR 10^{-4})
	1	9.9	0.03	4.7	5.7	23	0.03	33.1	100.6
sand	2	14.6	0.04	6.8	8.3	33.2	0.05	47.6	145.1
	3	17.1	0.05	8.1	9.9	39.6	0.06	55.7	173.2
Limestone (valley road) (2091)	4	7.7	0.02	3.5	4.3	17.2	0.03	23.8	75.2
	5	8.4	0.02	3.8	4.7	18.8	0.03	26	82.2
	6	7	0.02	3.2	3.9	15.7	0.02	21.9	68.9
Limestone (valley road) (1252)	7	10.1	0.03	4.7	5.7	22.9	0.03	32.1	100.4
	8	7.8	0.02	3.6	4.4	17.8	0.03	24.8	77.7
	9	10.6	0.03	4.9	6	23.9	0.03	33.5	104.4
Clay (vally road)	10	36.3	0.0982	17.2	21	84.1	0.12	121.6	368.2
	11	36.4	0.1	16.6	20.4	81.6	0.12	116	356.9
	12	37.7	0.1	17.7	21.7	86.8	0.1274	125.3	379.6
	13	36.7	0.1	17.1	21	83.9	0.12	120.2	367
Clay(Jahdam)	14	40.6	0.1	19.2	23.5	94	0.14	134.5	411.3
	15	43.5	0.1	20.3	24.9	99.7	0.15	142.8	436.2
	16	33.3	0.1	15.7	19.2	76.9	0.11	109.8	336.6

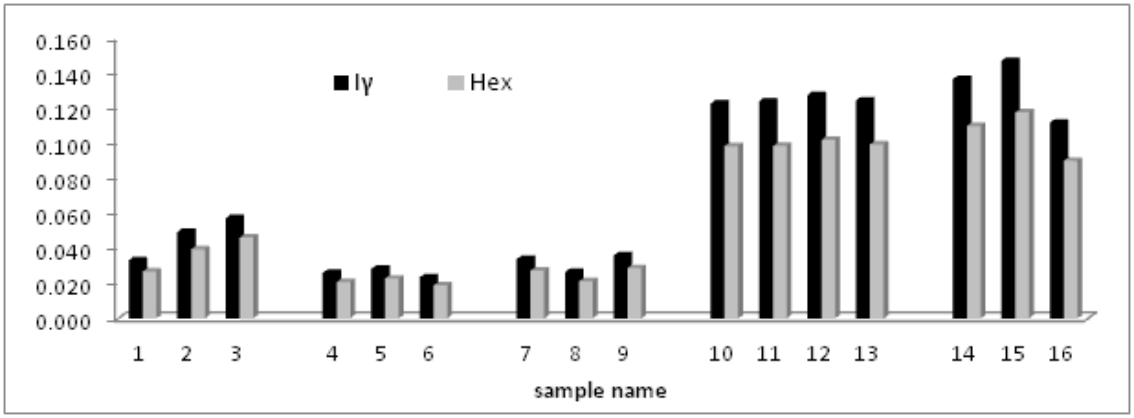

Fig. 5: Values of external hazard index (H_{ex}) and gamma activity concentration index $(I_\gamma)$for samples.

activity Ra_{eq} ,External hazard index (H_{ex}), Internal hazard index (H_{in}), Absorbed dose rate (D), Annual effective dose equivalent $(AEDE)$, Gamma index (I_γ) and Annual gonadal dose equivalent $(AGDE)$ were calculated and found to be within common values , with no significant radiation hazards arising from using such materials for construction of buildings.

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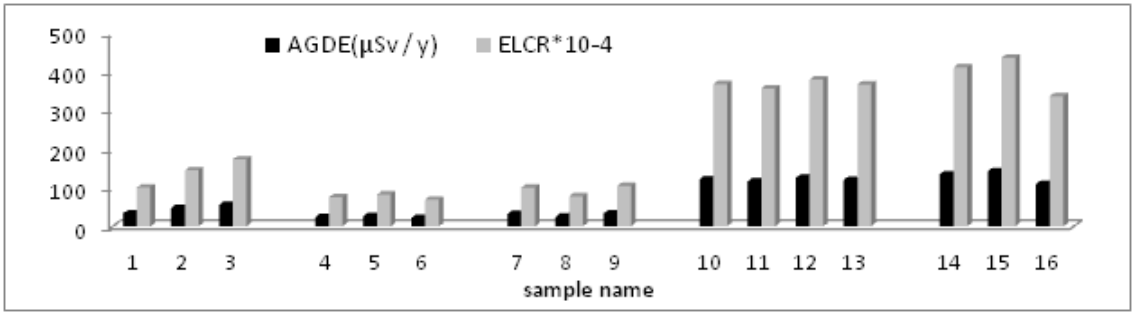

Fig. 6: Values of annual gonadal dose equivalent $(AGDE)(\mu Sv\ year^{-1})$ and Excess lifetime cancer risk $(ELCR)$ for samples

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