Natural radioactivity measurement and evaluation of radiological hazards in some environmental materials from Aswan area, Upper Egypt

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Abstract: Natural radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K in the common environmental materials collected from Aswan area in upper Egypt were performed using a NaI (Tl) gamma-ray spectrometer. The average activity concentrations of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K for all samples under investigation were found to be lower than the world average (50 Bq / kg for ^{226}Ra and ^{232}Th and 500 Bq / kg for ^{40}K) present in environmental materials (UNSCEAR, 2000). The Ra_{eq} values of the environmental materials are below the internationally accepted values (370 Bq/kg (21). The values of internal and external hazard index in the present environmental materials are less than unity. The annual effective dose of all measured Aswan environmental materials are at an acceptable level. The measurements enable one to assess any possible radiological risks to human health and help in the development of standards and guidelines for the use and management of environmental materials.

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1. Introduction

The presence of the naturally occurring radionuclides ^{40}K, ^{238}U, ^{232}Th and their daughter products in environmental materials is a source of radioactive exposure to population. The dweller of houses, offices and buildings, where they spend about 80% of their time are exposed to radiation emitted by these radionuclides) Mollah *et al.,* 1987). Continuous exposure to even low level radiation may adversely affect human health. To limit the exposure of the population to ionizing radiation, there is need to control and minimize the materials used in human needs with higher contents of radionuclides materials. Therefore, it is important to monitor the natural radioactivity level of environmental materials which can vary considerably according to the geological locations and geochemical characteristics of those materials. So, it is necessary to determine the natural radioactivity level of environmental materials from different areas. The natural radioactivity level of environmental building materials has been reported in many countries (El-Taher 2010, Rahman *et al.,* 2012, El-Taher *et al.,* 2013, Amran *et al.,* 2013, Sharaf., Hamideen, 2013, Senthilkumar *et al.,* 2014, XinweiLu *et al.,* 2014).

The aim of the present study is to determine the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in some environmental materials collected from Aswan, an important region in Egypt. The knowledge of these radionuclides present in the selected environmental materials helps to assess the possible radiological hazards as equivalent radium (Ra_{eq}), absorbed dose rate, annual effective dose and external and internal hazard indices and compare the results with the recommended limits from UNSCEAR 2000. The data obtained are essential for development of standards and national guidelines concerning the use and management of environmental materials in the light of international recommendations.

2. Materials and method

2.1 Sample treatment

The samples belonged to six types of environmental building materials: Black Clay, Shale, Sand, Concrete, Marble and Red Brick. For each environmental material, a total of 3 samples were prepared for the natural radioactivity measurement. The selected samples had been collected at 2014 from Aswan region in upper Egypt. All samples were ground into a fine powder with a particle size, 1 mm. They were then dried at 110°c for 20 h to eliminate moisture and filled into a cylindrical plastic containers and stored for a period of four weeks to allow ^{226}Ra and ^{232}Th decay series to reach radioactive equilibrium with the short-lived progenies.The activity concentrations of ^{226}Ra,^{232}Th and ^{40}K for all homogenized and equilibrium samples were measured by a gamma ray spectrometry using a NaI (TI) detector 3x3 inch with a 1024-chanel computer analyzer.The detector has a peak efficiency of 1.2x10^{-5} at 1332.5KeV Co-60 and an energy resolution (FWHM) of 7.5% for 662kev, detector employed with adequate lead shielding which reduces the background radiation. The specific activity of ^{226}Ra was evaluated from gamma-ray lines of ^{214}Bi at 609.3, 1120.3 keV and ^{214}Pb at 351 keV, while the specific activity of

^{232}Th was evaluated from gamma-ray lines of ^{228}Ac at 338.4, 911.1 and 968.9 keV. The specific activity of ^{40}K was determined directly from its 1460.8 keV line.

The activity concentration in $Bqkg^{-1}$ (A) in the environmental samples was obtained as follows:

$$A = N_{ap} / e\, \eta\, m \quad \ldots\ldots\ldots\ldots (1)$$

Where N_{ap} = net count rate (cps), measured count rate minus background count rate, *e* is the abundance of the γ-line in a radionuclide, η is the detection absolute efficiency at a specific energy and m the mass of the sample in kilograms.

2.2 Estimation of radiation hazards:

2.2.1 Radium equivalent activity Raeq

The main objective of calculating Radium equivalent activity Ra_{eq} is to make an estimate of radiation dose likely to be externally due to gamma radiation. Ra_{eq} provides a useful guidelines in regulating the safety standards on radiation protection for the general public. The Ra_{eq} was calculated using the following equation UNSCEAR(2000):

$$Ra_{eq}\,(Bqkg^{-1}) = A_R + 1.43\, A_{Th} + 0.077 A_K \qquad (2)$$

Where A_{Th}, A_{Ra}, A_K are the activities of ^{232}Th, ^{226}Ra and ^{40}K (Bq/Kg) in the sample respectively. The Ra_{eq} is the summation of above mentioned radio nuclides and based on the assumption that, 1 Bq/Kg of ^{226}Ra, 0.7 Bq/Kg of ^{232}Th and 14 Bq/Kg of ^{40}K produce the same gamma radiation dose rate (5). The maximum value of Ra_{eq} must be 370 Bq/Kg to keep the external dose to 1.5 mSv/y. The environmental materials whose Ra_{eq} exceeds 370 Bq/Kg is discarded to reduce radiation hazards associated with these materials.

2.2.2 External hazard index H_{ex}.

The external hazard index *H*ex and internal hazard index can be calculated by the following equations (UNSCEAR, 2000):

$$H_{ex} = A_{Ra}/370\ Bq\ kg^{-1} + A_{Th}/259\ Bq\ kg^{-1} + A_K/4810\ Bq\ kg^{-1} \quad \ldots\ldots .(3)$$

$$H_{in} = A_{Ra}/185\ Bq\ kg^{-1} + A_{Th}/259\ Bq\ kg^{-1} + A_K/4810\ Bq\ kg^{-1} \quad ---------- \ (4)$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations of uranium, thorium, and potassium in $Bq\ kg^{-1}$, respectively.

For the safe use of a material in the construction of dwellings and other human needs, the external hazard index(Hex), and the internal hazard index (Hin) should be less than unity.

2.2.3 Representative level index (I_r)

Representative level index ($I_γ$) is used to estimate the level of γ -radiation hazard associated with the natural radionuclides in specific building materials. The value is calculated using the formula derived by the Eropean Commission (EC, NAE-OECD,1979):

$$I_r = C_{Ra}/150 + C_{Th}/100 + C_K/1500 \leq 1 \quad --------- \quad (5)$$

where C_{Ra}, C_{Th} and C_K (in Bq/kg) are the concentration of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

2.2.4 Absorbed dose rates (D) and annual effective dose D_{eff}

The absorbed dose rates (D) due to gamma radiation in air at 1m above the ground surface, assuming uniform distribution of the naturally occurring radionuclides have been calculated according to UNSCEAR, 2000 as follows:

$$D\,(nGy\,h^{-1}) = 0.427 ARa + 0.623 A_{Th} + 0.043 A_K \ldots (6)$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq/Kg.

The annual effective dose equivalent was calculated from the absorbed dose by applying the dose conversion factor of 0.7 Sv Gy^{-1} with an indoor occupancy factor of 0.8 (UNSCEAR 1993, 2000):

$$D_{(eff)\ indoor} = D\,(nGyh^{-1}) \times 8{,}766\ h \times 0.7 (SvGy^{-1}) \times 0.8 \times 10^{-6} \quad \ldots\ldots\ldots (7)$$

where 8,766 h is the number of hours in 1 year. 10^{-6} is conversion factor of nano and milli.

3. Results and discussion

3.1 Activity concentration

The activity concentrations and an average values of radium-226, thorium-232 and potassium-40 for building materials (Black Clay, Shale, Sand, Concrete, Marble and Red Brick) collected from Aswan region, are listed in table (1). Each sample was counted four times before an average was calculated. From the obtained average values present in table (1), the following observations can be recorded: -

1- The average values of radium-226 activity concentrations are 10.9±3.5 ($Bq.kg^{-1}$) for Black clay, 14.9±3.7 ($Bq.kg^{-1}$) for Shale, 7.9±2.8 ($Bq.kg^{-1}$) for sand, 9.2±3.0 ($Bq.kg^{-1}$) for concrete, 21.6±4.4for marble and 12.8±3.6 ($Bq.kg^{-1}$) for red brick. The lowest average value of ^{226}Ra activity concentrations is found in Sand samples while the highest average value is found in Marble samples.

2- The a average values concentrations of Thorium-232 are 34.9±5.8 $Bq.kg^{-1}$ for Black clay, 29.8±5.5 $Bq.kg^{-1}$ for shale, 21.6±4.6 $Bq.kg^{-1}$ for Sand, 20.5±4.2 $Bq.kg^{-1}$ for Concrete, 37.6±6.0 $Bq.kg^{-1}$ for Marble and 24.0±4.9 $Bq.kg^{-1}$ for red brick. The lowest average value of ^{232}Th activity concentrations is found in Concrete samples while the highest average value is found in Marble samples.

3- The a average values concentrations of potassium-40 are 421.1±20.2 $Bq.kg^{-1}$, 377.5±18.3 $Bq.kg^{-1}$, 416.5±20.4 $Bq.kg^{-1}$, 454.5±19.2 $Bq.kg^{-1}$, 440.5±20.9 $Bq.kg^{-1}$ and 466.6±21.5 $Bq.kg^{-1}$ for Black clay, Shale, Sand, Concrete, Marble and red brick, respectively. The lower average value is found in Sand samples and the higher value in Red Brick samples.

Generally, the average concentration of natural radionuclides in the samples under studying are lower than the world average (50 Bq / kg for ^{226}Ra and ^{232}Th and 500 Bq / kg for ^{40}K) present in environmental materials (UNSCEAR, 2000).The results may be important from the point of view of selecting suitable materials for all purposes of human needs. Fig (1) Shows comparison between the average values of activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K, in building materials in the present study.

Table (1).The values ^{226}Ra,^{232}Th and ^{40}K activity in the environmental building samples from Aswan region, Egypt.

Sample Type	Sample Number	Activity concentrations Bq/Kg		
		^{226}Ra	^{232}Th	^{40}K
Black Clay	Cl-1	9.5±3.1	34.7±5.9	416.7±20.4
	Cl-2	10.9±3.3	30.7±5.5	421.4±19.6
	Cl3	12.3±3.7	39.4±6.3	425.1±20.8
Average		10.9±3.5	34.9±5.8	421.1±20.2
Shale	Sh-1	16.6±4.1	25.3±5.0	394.3±19.9
	Sh-2	15.4±3.9	43.5±6.4	316.0±17.8
	Sh-3	12.8±3.5	20.6±4.5	422.3±20.2
Average		14.9±3.7	29.8±5.5	377.5±18.3
Sand	Sa-1	4.4±2.1	20.7±3.9	379.7±19.5
	Sa-2	10.1±3.2	19.8±4.6	384.7±16.8
	Sa-3	9.1±3.0	23.3±4.2	485.2±22.1
Average		7.9±2.8	21.6±4.6	416.5±20.4
Concrete	Co-1	8.6±2.9	16.0±4.3	411.2±13.6
	Co-2	7.5±2.7	22.1±4.7	471.9±21.7
	Co-3	11.4±3.4	23.3±3.6	480.4±22.4
Average		9.2±3.0	20.5±4.2	454.5±19.2
Marble	Ma-1	23.1±4.8	32.6±5.2	418.1±20.4
	Ma-2	17.8±4.2	41.9±6.5	444.7±21.1
	Ma-3	24.0±4.6	37.2±6.1	458.6±21.3
Average		21.6±4.4	37.6±6.0	440.5±20.9
Red Brick	Br-1	15.3±3.9	25.3±5.0	411.7±20.3
	Br-2	11.5±3.4	24.0±4.9	406.8±18.2
	Br-3	11.6±3.2	22.6±4.1	581.3±24.3
Average		12.8±3.6	24.0±4.9	466.6±21.5

The average results for each radionuclide were compared with the average activity concentration for other countries as shown in Table (3) From this table, the average values of activity concentrations of ^{226}Ra for all samples are lower than all corresponding values in other countries. While the average values of ^{232} Th activity concentrations for all measured samples are comparable with the corresponding values of other countries. The average values of ^{40}K activity concentrations for Black clay is higher than Saudi Arabia (Alharbi &Alzahrani, 2013) and *lower than the most corresponding values* in other countries as shown in table (3). The average value in present study for Sand is lower than the values in Malaysia (Amran *et al.,*2013) and Pakstan (Rahman *et al.,*2012) and higher than the others. Concrete average values is lower than the value of China (Caifeng Zhao *et al.,*2012) and comparable with the other countries. Marble average value in present study is much higher than all countries listed in table (3). Finally, the average values of red brick is lower than the average value of Iraq (Hussain, 2010) and comparable with the *most corresponding values in other countries. As* shown in table (3), the radioactivity in building materials varies from one country to another of different materials and also within the same type of material. It is important to point out that these values are not the representative values for the countries mentioned but for the regions from where the samples were collected.

3.2 Radiation hazards

As shown in Table (3), the result of the hazard parameters indicates that:-

1- The radium equivalent activity Ra_{eq} for all selected materials, the highest value (109.29 Bq/kg) is observed in Marble, while the lowest value (70.86 Bq/kg) is found in Sand. However, the maximum Ra_{eq} values recorded are still lower than the maximum values (370 Bq/kg) suggested for building material (UNSCEAR, 2000).Therefore, we can recommend those materials for contractures. A comparative study of Ra_{eq} concentrations with similar data from some other countries calculated on the basis of the above formula are presented in Table(2).

Table (2) Comparison between the activity concentrations of various environmental materials in the present study with those of other countries.

Material	Country	Activity Concentration (Bq/Kg)			Raeq	Reference
		^{226}Ra	^{232}Th	^{40}K		
Clay	Iraq	59	12	934	146	Hussain 2010
	Malyzia	32	31	488	114	Yasir *et al.,* 2006
	Algeria	65	51	675	190	Amrani&Tahtat 2001
	Saudi Arabia	26	15	234	58	Alharbi & Alzahrani (2013)
	Egypt (Aswan)	10.9	34.9	421.1	93.23	Present study
Shale	Egypt, Nile valley	85.2	93.3	303.1	241.96	Abbady *et al*. (2005)
	Egypt, Eastern desert south Egypt	22.1	38.3	435.7	110.4	Shadiah S. Baz, A. Abbady(2014)
	Egypt (Aswan)	14.9	29.8	377.5	86.58	Present study
Sand	Malyziya	43.2	39.8	523.9		Amran *et al*. (2013)
	China	32.5	47.7	249.6	119.9	XinweiLu(2014)
	Pakistan	24	39	462		. Rahman *et al*. (2012)
	Saudi Arabia	23	21	172	65	Alharbi & Alzahrani (2013)
	Egypt (Aswan)	7.9	21.6	416.5	70.86	Present study
Concrate	Malyziya	25	22	324	84	Yasir *et al*., 2006
	China	22	41	725	140	Caifeng Zhao *et al.* (2012)
	Saudi Arabia	22	16	226	62	Alharbi & Alzahrani (2013)
	Egypt (Aswan)	9.2	20.5	454.5	73.51	Present study
Marble		28	6.2	42.3		Al-Jundi(2006)
	Egypt	15.9	12.3	60	37.76	El-Taher(2010)
	Saudi Arabia	27	28	54	66	El-Taher(2013)
	Egypt (Aswan)	21.6	37.6	440.5	109.29	Present study
Brik (red)	Malyziya	33	24	443	101	Yasir *et al.*, 2006
	Iraq (Najaf)	120	15	978	215	Hussain 2010
	Saudi Arabia	27	21	279	79	Alharbi & Alzahrani (2013)
	Egypt (Aswan)	12.8	24.0	466.6	83.05	Present study

2- For representative level index (Iγ), the maximum suggested value is 1. Any value less than 1 shows that the external radiation dose within the building is less than the maximum suggested dose. Gamma index for the investigated materials are ranged from 0.27 (Sand) to 0.41 (Marble).

3- The average values of outdoor radiation hazard index (H_{ex}) and indoor hazard index (Hin) for all samples, are in the range from 0.17 (Concrete) to 0.32 (Clay) and from 0.21(Sand) to 0.35 (Marble), respectively. Where, all average values are less than the critical value of unity.

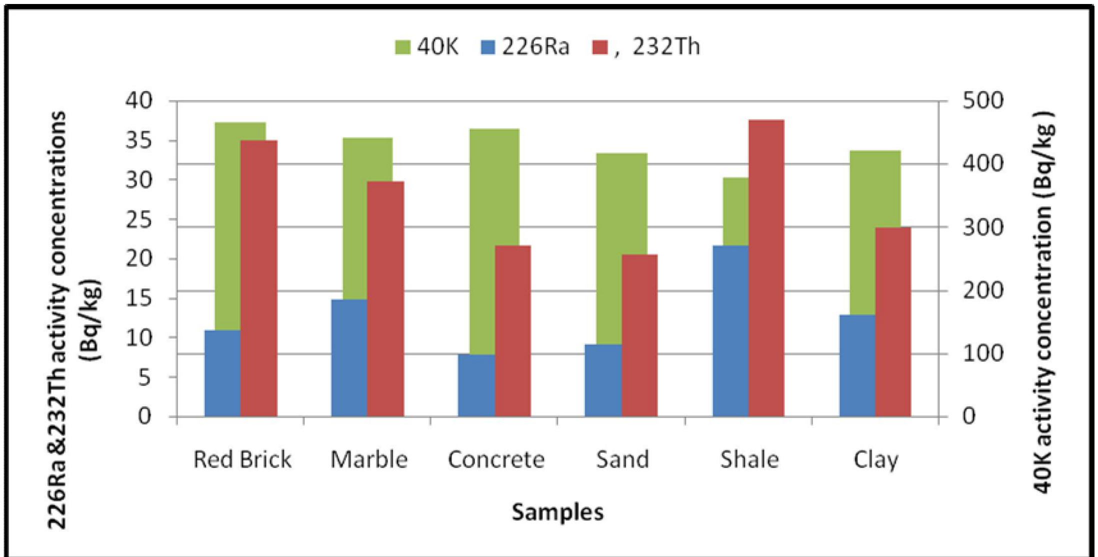
4- The estimated average absorbed dose rate (D), range from 34.74 $nGyh^{-1}$ (Sand)) to 51.59 $nGyh^{-1}$ (Marble). These values are lower than the recommended upper limit 55$nGyh^{-1}$

5- The values of the annual effective dose, varied from 0.170 (Sand) to 0.253 (Marble). The recommended upper limit of 1 $mSvy^{-1}$ is not exceeded. In general the dwellers inside the building are not supposed to acquire any radiological complication.

Figure (2). Shows comparison between Radium equivalent activity (Bq/Kg) and absorbed dose rate (nGy/h) for building materials under investigation.

Table (3): The average radiation hazard parameters for environmental building materials under investigation.

Sample Type	Radium equivalent Ra_{eq} (Bq/kg)	Level index $I_{\gamma r}$	External index H_{ex}	Internal index H_{in}	Absorbed dose rate*(D)* (nGy/h)	Annual effective dose rate (D_{eff}) (mSv/y)
Clay	93.23	0.35	0.32	0.28	44.50	0.218
Shale	86.58	0.33	0.25	0.27	41.16	0.202
Sand	70.86	0.27	0.23	0.21	34.74	0.170
Concrete	73.51	0.28	0.17	0.22	36.24	0.178
Marble	109.29	0.41	0.18	0.35	51.59	0.253
Red Brick	83.05	0.32	0.30	0.26	40.48	0.198



Fig(1): Comparison between the mean values of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in building materials of Aswan, Egypt

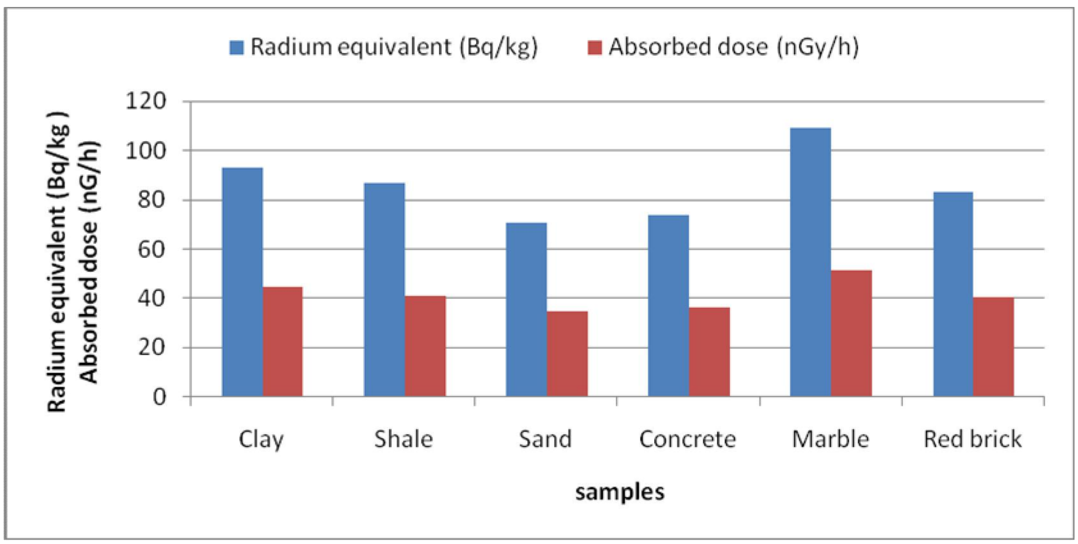


Figure (2): Shows comparison between the average values of radium equivalent Bq/kg) and average values of absorbed dose (nG/h) for the building materials used in Aswan region, Egypt

4. Conclusion

The average of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples of l building materials in the present study were below the acceptable limits. The building materials were found to have Ra_{eq} values less than the upper recommended value of 370 Bq/ kg and close to values in many other countries. The concentrations of the natural radionuclides and Ra_{eq} in the studied samples were compared with the corresponding results of other countries. The external and internal hazard indices(H_{ex}, H_{in}), representative level index ($I_{\gamma r}$) for all samples investigated are below unity. The annual effective dose(D_{eff}) is well below the recommended value (1 mSv/y). Therefore, the use of these materials for all purposes of human needs is considered to be safe.

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