Evaluation of natural radioactivity and the associated radiation hazards in local cement used in Saudi Arabia

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Abstract: The natural radionuclide (^{226}Ra, ^{232}Th and^{40}K) concentrations for 14 brands of gray Ordinary Portland Cement and 9 brands of white cement used as building and construction material in Saudi Arabia were measured by using gamma spectrometry method. This subject is important in environmental radiological protection, since cement are widely used as building materials.The average value of ^{226}Ra, ^{232}Th, and ^{40}K for gray cement is 32.21 Bq/kg, 23.30 Bq/kg and 177.32 Bq/kg respectively. The average values for the white cement are 23.90 Bq kg^{-1}, 25.60 Bq kg^{-1} and 125.64 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The total average content of ^{226}Ra, ^{232}Th, and ^{40}K for all the cement brand samples are 28.59 Bq kg^{-1}, 24.30 Bq kg^{-1}, and 154.85 Bq kg^{-1} respectively. These values obtained are lower than the world average values for building materials. The estimated radium equivalent activities (Ra_{eq}), representative index (Iγ), absorbed γ-dose rate (D), the annual effective dose rate (AEDE) and external and internal hazard indices are lower than the recommended safe limit and are comparable with results from similar studies conducted in other countries. The measurements help in the development of standards and guidelines for the use and management of building materials.

[Shadiah S. Baz. **Evaluation of natural radioactivity and the associated radiation hazards in local cement used in Saudi Arabia.** *Life Sci J* 2015;12(2):189-195]. (ISSN:1097-8135). http://www.lifesciencesite.com. 30

Key words: Radioactivity, Radium equivalent, Cement, Dose assessment

1. Introduction

The concentration of primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K in building materials depends upon the origin of crustal material of which they are composed of The highest concentrations of these radionuclides are found in mineral-based building materials such as rocks, sand, bricks, gypsum and cement (Turtiainen *et al.,* 2008). Portland cement is a major building material used worldwide. Most construction cements today are hydraulic, and most of these are based upon Portland cement, which it is derived from mixing natural clay, limestone and gypsum at high temperature (Kpeglo *et al.,* 2011). White cement is made from specially selected raw materials, usually pure chalk and white clay (kaolin) containing very small quantities of iron oxides and manganese oxides. The specific levels of gamma radiation in cement depend upon the content of thorium, uranium and potassium in the waste products, used in cement industry.

In Saudi Arabia the use of cement as a basic building and construction material. It is used (when mixed with other materials such as fine sand) as binding material for concrete blocks and for floors, walls and even roof finishing (Sam and Abbas, 2001). The grain size of cement is such that it is aerodynamic (Esubiyi, 1995), which could easily pass through respiratory track, or get blown by air into food and water. Consequently, the presence of radionuclide in cement does not only pose potential external radiation hazard but could also cause internal radiological contamination as well. So, it is important to have knowledge of the amount of natural radioactivity present in the materials which are used in the construction of dwellings. The amount of activity present in building materials will decide its use in the construction of dwellings which are very important in human life as most of the lifetime is spent (about 80%) at home and/or office. (Stoulos *et al.,* 2003). Therefore, measuring the radioactivity in building materials will enable us to assess any possible radiological hazard to human health (El-taher 2011). As natural radiation is the largest contributor to the external dose of the human body, it is important to assess the gamma radiation dose from natural sources (UNSCEAR, 1988). The population-weighted average of indoor absorbed dose rate in air from terrestrial source of radioactivity was estimated to be 84 nGyh^{-1} (UNSCEAR, 2000). The worldwide average indoor effective dose due to gamma ray from building materials is estimated to be about 0.4 mSvy^{-1} (UNSCEAR, 1993). The radioactivities of natural radionuclides in cement have also been reported in many countries (Fathivand 2007, Mujahid *et al.,* 2008, Eltaher *et al.,* 2010, Gharbi 2012, Rahman1 2012, Amran *et al.,* 2013, XinweiL *et al.,* 2014), where they have shown that the radioactivity concentration present in cement materials varied from one country to another,according to geological and geographical conditions of the region. Information about radioactivity of building materials in Saudi Arabia is limited and there are neither standards nor guidelines prescribing the acceptable levels of radioactivity of these materials. So, the objective of the present work was to measure the activity concentration due to ^{226}Ra, ^{232}Th and ^{40}K in different brands of local gray Portland cement and white cement that are used commonly in Saudi Arabia. Knowledge of this radioactivity is useful in setting the standards and national guidelines for the use and management of these materials and in assessing the radiation hazard associated with the usage of building materials by computing the radium equivalent activity, external and internal hazard indices, representative index, absorbed dose rate and the annual effective dose. The results obtained in the present study are compared with those results available in some other countries of the world.

2. Material and methods

2.1 Materials

Fourteen gray (Portland) cements and Nine white cements are generally used in Saudi Arabia as building materials, were collected randomly at 2014 from factories, construction sites as well as from various agencies supplying raw materials for building construction. All the gray cement and white cement brands considered in this work are local materials. Each sample was air dried and pulverized into powdered form. 400 g of each powdered sample was put into Standard Marinelli beakers and sealed for about 4weeks in order to establish secular equilibrium between ^{232}Th and ^{226}Ra with their progeny. All samples were analyzed using Hyper Pure Germanium (HPGe) gamma-ray detector coupled to a PC with a special electronic card to make it equivalent to a 4096 CANBERRA multi-channel analyzer. The energy resolution was 1.95 keV for the 1332 keV gamma-ray of ^{60}Co with relative efficiency of 25%. A lead cylinder with a Copper cover shielded the detector to reduce gamma-ray background. An empty container was measured for subtraction of background. The energy calibration was performed using a mixed multi-nuclide calibration source and the absolute efficiency calibration of the spectrometer was measured by using a standard point sources of ^{152}Eu and ^{226}Ra (IAEA 1989). The samples were counted for a period of 82800 s and the spectra are analyzed for the photo peak of uranium, thorium daughter products and K-40. The activity of the radionuclides is calculated from the background subtracted area of prominent gamma ray peaks. The gamma-ray transitions energies of 352 keV (^{214}Pb), and 609 keV (^{214}Bi) were used to determine the concentration of ^{226}Ra. The gamma-ray transitions energies of 583 keV (^{208}Tl) and 911 keV (^{228}Ac) were used to determine the concentration of the ^{232}Th series. The ^{40}K activities were determined from the line at 1460 keV.

2.2 Natural specific activity

The activity concentrations of the natural radionuclides in the measured samples were computed using the following relation (El-Taher, 2011):

$$A\ (\mathrm{Bq\ kg^{-1}}) = N / \varepsilon\ \beta\ M \quad \ldots\ldots\ldots\ldots\ldots\ldots (1)$$

where N is the net gamma counting rate (counts per second), ε the detector efficiency of the specific γ–ray, β the absolute transition probability of Gamma–decay and M the mass of the sample (kg).

2.3 Calculation of the radiological parameters

2.3.1 Radium equivalent activity

The distribution of ^{226}Ra, ^{232}Th and ^{40}K in building materials is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity in Bq/kg unit, defined according to the estimation that 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 (Berekta & Mathew, 1985). This index Ra_{eq} is given as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \text{ ----------- } (2)$$

Where, A_{Ra}, A_{Th} and A_K are specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in Bqkg^{-1}. The maximum value of Ra_{eq} in building materials must be less than 370 Bqkg^{-1} for safe use i.e., to keep the external dose below1.5mSvy^{-1} (UNSCEAR, 2000).

2.3.2 Representative level index (I_γ)

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 European Commission (EC), NAE-OECD (1979):

$$I_\gamma = C_{Ra}/300 + C_{Th}/200 + C_K/3000 \leq 1 \text{----- } (3)$$

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

2.3.3 external and Internal gamma indices

For limiting the radiation dose from building materials to 1.5mGr y^{-1} (Krieger, 1981) proposed the following conservative model based on infinitely thick walls without windows and doors to serve as a criterion for calculating the external hazard index (H_{ex}). The external hazard index for samples under investigation is given by the following equation (Beretka and Mathew, 1985):-

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \ \ldots (4)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of this index must be less than unity for the radiation hazard to be negligible.

In addition to the external irradiation radon and its short-lived products are also hazardous to the respiratory organs. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}) which is given by the following equation (Berekta & Mathew,1985):-

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \leq 1 \ \ldots\ldots (5)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations (Bq kg^{-1}) of the specific radiation. The maximum value of H_{ex} to be less than unity corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}). If the maximum concentration of radium is half that of the normal acceptable limit then, H_{in} will be less than 1. The prime objective of this index is to limit the radiation dose to dose equivalent limit of 1mSv/y, UNSCEAR, 2000.

2.3.4 Absorbed gamma dose rate

The absorbed dose rate in air is due to gamma ray emission from the isotopes ^{226}Ra ^{232}Th and ^{40}K inside the building materials. It can be defined in units of nGyh^{-1} using the following formula (UNSCEAR,2000):

D_{indoor} (nGyh^{-1}) = $\sum_i Ri \times Ai$ ……. (6)

where Ai (Bq kg^{-1}) are the mean activities of ^{226}Ra,^{232}Th and ^{40}K, Ri (nGyh^{-1} per Bqkg^{-1}) their corresponding dose factor. According to an EC(1999) report, the dose conversion coefficients (Ri) were calculated for cement materials as 0.92, 1.1 and 0.08 nGyh^{-1}per Bqkg^{-1} for ^{226}Ra,^{232}Th and ^{40}k respectively, using the standard room model with Dimensions of 4m ×5m× 2.8m, wall thickness 20 cm and density 2350 kg m^{-3}. The world average value of the indoor absorbed gamma dose rate is 84 nGyh^{-1} UNSCEAR, 2000.

2.3.5 Annual effective dose rate

To estimate the annual effective dose rates indoor, one has to take into account the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv/Gy) and indoor occupancy factor 0.8 proposed by UNSCEAR (1993). Therefore, the annual effective dose rate (mSv y^{-1}) was calculated by the formula (UNSCEAR, 2000):

$(AEDE)_{indoor}$ = D (nGyh^{-1}) ×8760 (hy^{-1})× 0.7(× (10^3 mSv / nGy 10^9))× 0.8 … (7)

The worldwide annual effective dose from the natural sources of radiation in areas of normal background is estimated to be 1 mSvy^{-1} by UNSCEAR (2000).

4. Results and Discussion

4.1 Activity concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the various brands of cement considered in this work is given in Table (1). In the gray cement brands,the activity concentration for ^{226}Ra range from 21.32 Bq kg^{-1} to 52.54 Bq kg^{-1} with an average value 32.21 Bq/kg. ^{232}Th activity concentration range from 9.77 Bq kg^{-1} to 46.56 Bq kg^{-1} with an average 23.30 Bq/kg. The activity concentration for ^{40}K range from 97.54 Bq kg^{-1}to 295.75 Bq kg^{-1} with an average of 177.32 Bq/kg. While the activity concentrations of the white cement for ^{226}Ra range from 13.46 Bqkg^{-1} to 39.23 Bqkg^{-1}. For ^{232}Th, the concentration range from 13.52 Bq kg^{-1} to 51.84 Bq kg^{-1}, and finally, ^{40}K concentration range from 68.43 Bq kg^{-1} to 134.14 Bq kg^{-1}. The corresponding average values are 23.90 Bq kg^{-1}, 25.60 Bq kg^{-1} and 125.64 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively. The variation of the average activity concentrations of the cement brands could be attributed to the variations in the geological origin of the raw material used in their production. It is clear that the average activity concentration of ^{40}K was the highest in all the samples when compared with the other two radionuclides. This is typical and expected from any geologically derived material due to the relative abundance of ^{40}K in the natural environment (IAEA, 2003). The average concentrations are ^{40}K>^{226}Ra>^{232}Th and all values are less than the world average, which are 50, 50 and 500 Bqkg^{-1}, respectively (UNSCEAR, 2000). The average percentage contribution of ^{226}Ra,^{232}Th and ^{40}K in gray cement samples are 14%, 10% and 76% respectively, while in the white cement, the contribution of the three radionuclides are 14%,15 % and 71%, which shows that ^{40}K contributes the highest radioactivity to the cement activity concentration. Fig (1), shows the comparison of the average concentration of the three naturally occurring radionuclides examined in the two cement brands in Saudi Arabia.

4.2 Radiation hazards results

Radiological hazards for various cement brand samples under investigation were calculated. Summary of the results are presented in Table (2) with the world standard values. As shown from this Table, the results of the hazard parameters indicates that:-

1- Radium equivalent values for all cement samples range between 37.24 Bq/kg in sample (C21) and 166.45 Bq/kg in sample (C15). As can be seen, all cement brand samples studied in this work, the Ra_{eq} values are well below the upper limit values (370 Bq/kg) suggested for building material (UNSCEAR, 2000). Table (3) lists the calculated average radium equivalent concentrations of the results on Gray and White cement in Saudi Arabia in comparison with similar studies performed in other countries.

2- The external gamma indices for all the cements considered in this work are less than unity. For all cement brands, It varies from 0.07 in sample (C3) to 0.45 in sample (C15). Also, the internal hazard index (H_{in}) quantifies the internal exposure to carcinogenic radon and its short lived progeny. The values of the calculated H_{in} for the various brand of cement are also given in Table (2), and are all less than 1. The evaluation of the internal and the external hazard indices reveal that the two brands of cement are most safe cement for building construction.

3- The representative level index (Iγ) value obtained for all the cement brand samples range from 0.1 Bq kg^{-1}in sample (C3) to 0.83 Bq kg^{-1} in sample (C18) with an average value 0.37 Bq kg^{-1}. This shows that the (Iγ) values of the cements are within the world standard range.

4- The absorbed dose rate varies from 42.20 nGy h^{-1} in sample (C15) to 110.98 nGy h^{-1}in sample (C21) with an average value of 65.42nGy h^{-1}. This indicates that the obtained data for absorbed dose rate is still within the standard limit of 84 nGyh^{-1} as reported in UNSCEAR, 2000. This variation in absorbed dose is also an indication of the different geological sites where these cement raw materials are gotten from, which is a factor of the geological formation of the source rock.

5- The estimated indoor annual effective dose rate$(AEDE)_{indoor}$ obtained in the general brands of cements range between 0.21mSv y^{-1} in sample (C21)to 0.54mSv y^{-1} in sample (C7)with an average alue of 0.32mSv y^{-1}. effective dose of these samples does not exceed the recommended limit of 1mSv/y, UNSCEAR, 2000. This indicates that the cements used in Saudi Arabia are radiologically safe. The exposure assessment studies of this type are highly needed as they provide necessary baseline information on radiation level in any environment that may be prone to radioactive contamination.

Figure (2) shows comparison between the average values of radium equivalent activity (Bq/Kg) and absorbed dose rate (nGy/h) for the two cement brand samples under investigation.

***5. Comparison between the results of present study with* Similar Studies in other countries.**

The calculated average values of activity concentrations and the radium equivalent (Raeq) in this work and those obtained from recent published work from other countries is given in Table (3). It can be concluded that the results obtained in this study fall within the range of values reported in similar studies and are below the values of Turkey and China, which their activity concentrations above the world average.The comparison of the average value of radium equivalent activities obtained in different countries of the world indicate that the values of radium equivalent of gray cement in China, Egypt, and Turkey are well above the average value obtained in Saudi gray cement. The Ra_{eq} for Saudi white cement is lower than the values of Ra_{eq} in white cement of Egypt, Nigeria and Turkey and twice the value of Iraq. The variation in the Ra_{eq} for the different countries can be attributed to the difference in the Geology and consequent geochemical constituent of the rock from which the cements were derived.

Table 1. Activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K ($Bqkg^{-1}$) in gray cement and white cement samples used in Saudi Arabia.

Cement brand	Sample code	Radioactivity concentration($Bqkg^{-1}$)		
		^{226}Ra	^{232}Th	^{40}K
Gray Cement	C1	16.64 ±0.00083	17.47±0.0016	136.12±0.0005
	C2	21.93±0.0008	9.77±0.0018	123.01±0.0005
	C3	8.42±0.0009	8.67±0.0011	79.3±0.0005
	C4	36.42±0.0008	23.30±0.0019	97.54±0.0005
	C5	21.32±0.0008	11.20±0.0018	178.31±0.0005
	C6	52.54 ±0.0008	34.61±0.0019	132.45 ±0.0005
	C7	39.24 ±0.0008	46.56±0.0018	295.75 ±0.0005
	C8	45.24±0.0009	41.08±0.0019	254.56 ±0.0005
	C9	26.72±0.0007	36.53±0.0017	162.28 ±0.0005
	C10	31.19±0.0008	24.6±0.0019	133.55 ±0.0005
	C11	35.83±0.0009	21.46±0.0014	333.90±0.0005
	C12	38.98±0.0009	17.32±0.0018	248.51 ±0.0005
	C13	44.3±0.0009	10.32±0.0015	129.94±0.0005
	C14	25.95±0.0008	23.65±0.0018	226.45±0.0005
	Range	21.32-52.54	9.77-46.56	97.54-295.75
	Average	32.21	23.30	177.32
White Cement	C15	19.38±0.0008	15.98±0.0019	122.87±0.0005
	C16	16.97±0.0007	23.96 ±0.0017	134.14±0.0005
	C17	20.97±0.0008	25.24±0.0019	68. 43±0.0005
	C18	39.23±0.0009	51.84±0.0019	75.90±0.0005
	C19	13.64±0.0008	25.32±0.0020	129.79±0.0005
	C20	23.65±0.0005	36.575±0.0016	104.37±0.0005
	C21	17.91±0.0008	13.52±0.0018	135.6±0.0005
	C22	26.67±0.0009	15.94±0.0013	132.23±0.0005
	C23	34.59±0.0013	23.98±0.0033	126.64±0.0005
	Range.	13.64- 39.23	13.52- 51.84	68. 43 – 134.14
	Average	23.90	25.60	125.64
Average over all		28.59	24.30	154.85
World average		50	50	500

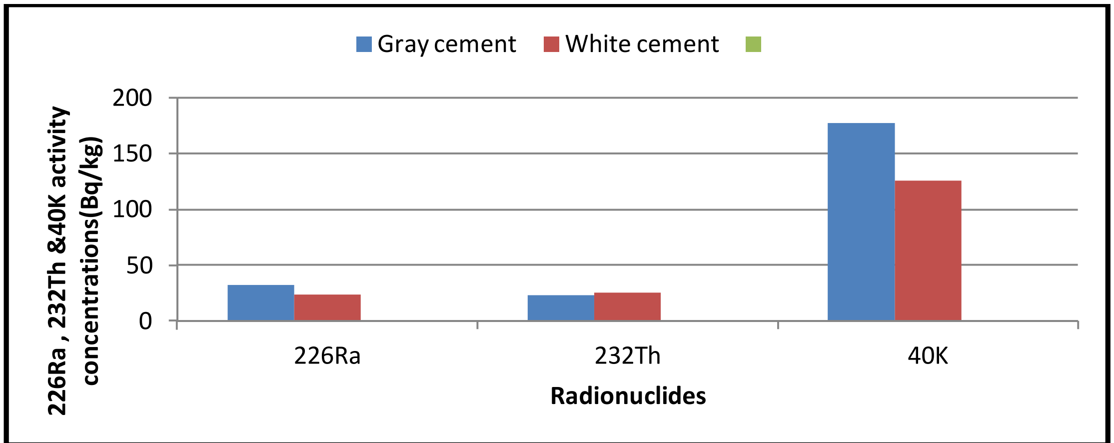
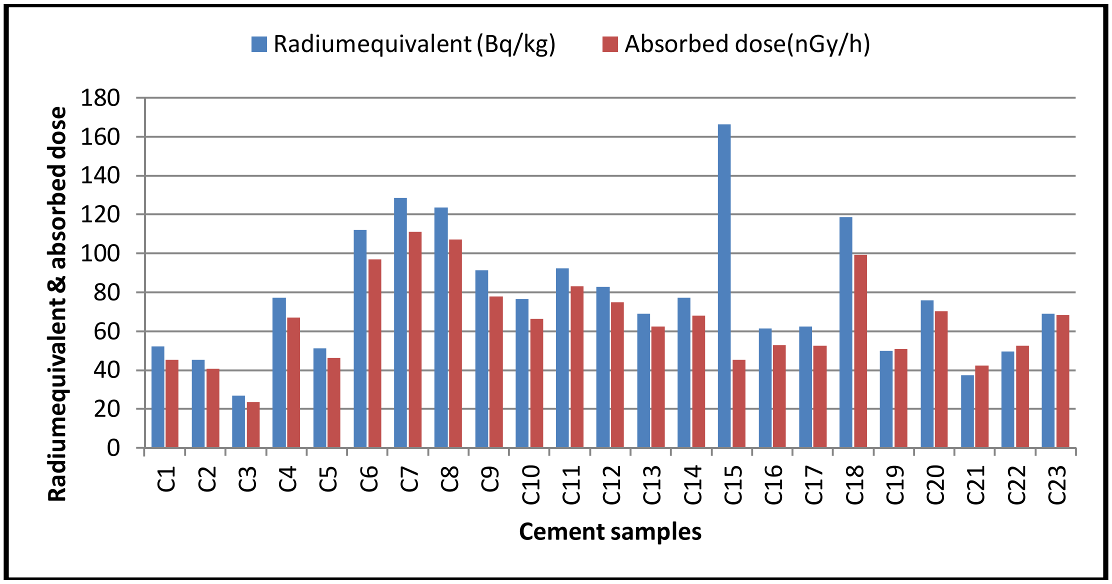


Fig (1). Shows the comparison of the average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in gray and white cement are used in Saudi Arabia.

Table 2: Radiological hazards calculated for gray and white cement samples used in Saudi Arabia

Material	Sample code	*Radium equivalent Raeq (Bq/kg)*	*External index Hex*	*Internal index Hin*	*Gamma index Iγr*	*Dose rate D indoor (nGy/h)*	*Annual effective Dose (mSv/y)*
Gray cement	C1	52.10	0.14	0.19	0.19	45.42	0.22
	C2	45.37	0.12	0.18	0.16	40.76	0.20
	C3	26.92	0.07	0.09	0.10	23.63	0.12
	C4	77.25	0.21	0.31	0.27	66.94	0.33
	C5	52.10	0.14	0.20	0.19	46.20	0.23
	C6	112.23	0.30	0.45	0.39	97.01	0.48
	C7	128.59	0.35	0.45	0.46	110.98	0.54
	C8	123.59	0.33	0.46	0.44	107.17	0.53
	C9	91.45	0.25	0.32	0.33	77.75	0.38
	C10	76.65	0.21	0.29	0.27	66.44	0.33
	C11	92.23	0.25	0.35	0.34	83.28	0.41
	C12	82.88	0.22	0.33	0.30	74.79	0.37
	C13	69.06	0.19	0.31	0.24	62.50	0.31
	C14	77.21	0.21	0.28	0.28	68.01	0.33
White cement	C15	166.45	0.45	0.81	**0.57**	45.24	0.22
	C16	61.56	0.17	0.21	0.33	52.70	0.26
	C17	62.33	0.17	0.23	0.44	52.53	0.26
	C18	119. 21	0.32	0.43	0.83	99.19	0.49
	C19	49.85	0.16	0.20	0.43	50.78	0.25
	C20	75.95	0.23	0.29	0.59	70.34	0.35
	C21	37.24	0.13	0.18	0.35	42.20	0.21
	C22	49.46	0.13	0.18	0.35	52.65	0.26
	C23	68.88	0.14	0.19	0.19	68.33	0.34
Range		37.24- 166.45	0.07-0.45	0.18-0.81	0.1-0.83	42.20-110.98	0.21- 0.54
Average		78.13	0.22	0.31	0.37	65.42	0.32
World standard		370	≤ 1	≤ 1	≤ 1	84	1



Fig(2).Radium equivalent and Absorbed dose rate in cement samples

Table (3). Comparison of Activity concentrations and Radium Equivalent Activities (Raeq) in gray and white cement used in Saudi Arabia with those of other Countries

Material	Country	Activity concentration			Ra_{eq}	Références
		^{226}Ra	^{23}2Th	^{40}K		
Gray cement	China	118.7	36.1	444.5	154.4	XinweiLu *et al.* (2014)
	Egypt	33.8	61.8	89.0	129	Uosif (2014)
	Malaysia	34.7	32.9	190.6	96.42	Amran (2013)
	Nigeria	30.2	24.6	251.3	84.7	Agbalagba *et al.* (2014)
	Pakistan	37	28	200	92.0	Rahman1 *et al.* (2012)
	Tunis	45.5	15.2	118.6	76.37	Gharbi *et al.* (2012)
	Austria	27	14	210	63.19	Trevisi *et al.* (2012)
	Turkey	24.7	20.7	2493.1	246.1	Baykara *et al.* (2011)
	Saudi Arabia	32.21	23.30	177.32	79. 26	Present work
White cement	Egypt	40.7	65.5	77.9	140.3	Uosif (2014)
	Nigeria	41.9	30.1	340.2	111.1	Agbalagba *et al.* (2014)
	Iraq	31.0	3.8	10.2	37. 2	Kamal K. Ali (2012)
	Turkey	12.5	2.7	1141.9	104.2	Oktay baykara *et al.* (2011)
	Saudi Arabia	23.90	25.60	125.64	76.75	Present work

Conclusion

The natural radioactivity of ^{40}K, ^{232}Th, ^{238}U and ^{226}Ra and their consequent radiation hazard indices were evaluated in gray and white cement used in Saudi Arabia. The data show that the activity concentration of the three natural radionuclides in the two cement brand samples are lower than the world average. Calculations of hazard indices show that none of the samples exceeded their recommended permissible level. A comparison of the measured specific activity values with the corresponding worldwide average values shows that the present activities were comparable to the results of similar studies undertaken in other countries. The results of this study shows that the cement used in Saudi Arabia for construction of dwellings and workplaces is radiologically safe and may not cause any significant health hazard.

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2/17/2015