Study of natural radionuclides and its radiation hazard index in building materials used in Saudi Arabia

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Abstract: Forty samples for thirteen different types of building materials (cement, gypsum, Insulators of water, Ceramic Ceiling, ceramic, granite, marbles, sand, wood, bricks, iron, aluminum and Paints are used in Jeddah were analyzed by a gamma spectrometer based on HPGe detector. The total average content of ^{226}Ra, ^{232}Th, and ^{40}K for all the building brand samples are 18.25 Bq kg^{-1}, 21.22 Bq kg^{-1}, and 260.11 Bq kg^{-1} respectively. The estimated radium equivalent activities (Ra_{eq}), representative index (Iγ), absorbed γ-dose rate (D_R), the annual effective dose rate (D_{eff}) and external hazard indices are lower than the recommended safe limit. The results indicated that all the samples collected from the market used as building materials in Jeddah are safe in general for the radioactivity levels.

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1. Introduction:

Millions of tons of industrial waste and byproducts are used in the reduction of building materials as phosphogypsum and iron in the manufacturing of cement and bricks. These waste products often contain naturally occurring radioactive materials (NORM) with different concentrations. Also; recycling of iron scrap which sometimes contains manmade radio nuclides as ^{137}Cs & ^{60}Co. It was found that burden from inhalation of radon is about ten thousand times than of its effect when it is soluble in water (Leker, 1996). Recently effects from NORM in building materials and its relation with the building construction were studied by Eltaher *et al.,* 2010, Kamal; 2012, Gharbi; 2012, Rahman1; 2012, Amran *et al.,* 2013, Sharaf & Hamideen; 2013, XinweiL *et al.,* 2014. World Health Organization (WHO) studied the effects of exposing from NORM in building materials from 1996 to 1998. In Austria some factories producing building materials send their products to be analyzed for the contents and concentrations of radioactivity before their use in local markets (Bossew, 2003). International Commission for Radiation Protection (ICRP) & United Nation Scientific Committee of the Effects of Atomic Radiation (UNSCEAR), estimated the average annual absorbed dose for inhabitants and updating these data (UNSCEAR 2000).

Thomson *et al.,* 2003 calculated the absorbed dose from some building materials produced by burning as ceramic and bricks which contains low concentrations from NORM. The existence of radioactivity in building materials increases the internal and external absorbed dose to the residence so the analysis of building materials for the concentrations of either natural or manmade radioactivity give good information about the internal as well as the external dose to the inhabitance. The source of the increase of the absorbed dose was found to be mostly of Rn-222 or Rn-220 which are members of the radioactive U-238, Ra-226 and Th-232 series respectively, about 45% of the inhabitant areas of the world are recorded by UNSCEAR, 2000. The average dose per person range from (84 n Gy /h to 200 nGy /h), the lowest value in Newzeland, Iceland & USA (40 nGy/h because the construction of buildings mostly of wood. The highest value (95-115 nGy/h) in Hungary, Malezia and some other countries which use stones in construction (UNSCEAR, 2000).

Hayumbu *et al*., 1995 studied the natural radioactivity in Zambia by gamma spectroscopy they compared their results by some other published results also they calculated the radium equivalent which was found to be less than the recommended value 370 Bq/kg (Amrani & Tahtat, 2001).

Malanca et *al*., 1995 studied 48 samples of building and raw materials used in Italy, also; they calculated the radium equivalent (range from 14.6 to 1930 Bq/kg) which was converted to absorbed dose. Also; samples from the building materials used in Bengaledis were analyzed by gamma spectroscopy (Chowdhury *et al*., 1998) the highest value was found to gypsum. Croft & Hutchinson, 1999 measured uranium, thorium and potassium concentrations by Harwell laboratories UK in some sand and cement samples to be used in buildings to check their suitability. All over the world building materials were checked for radioactivity concentrations and calculating the expected internal and external burden before being in markets for quality assurance.

Risica *et al.*, 2001 put room model analysis to calculate 1st the absorbed dose rate inside a room, 2nd, to calculate the dose from the concentrations of the natural radioactivity, 3rd the dose rate calculated from the concentrations of the natural radioactivity referring to the room geometry by the mathematical model. Markkanen, 2001 and group of EU put limits for concentrations for natural radioactivity in building materials to be used in the EU (Article 31 Expert Group and the European Commission). Jawad & Kearfott, 2005; studied the environmental factors affecting the radon concentrations inside buildings and estimate relation between radon concentrations indoor and outdoor. They took into consideration the difference in temperature, the speed and direction of wind in addition to the environmental statistical factors which can affect the internal dose.

The aim of this study is to measure concentrations of radioactive 226Ra, 232Th series and40K as well as the man made Cs-137, to estimate the radiological hazards associated with the natural radioactivity levels in the materials (cement, gypsum Insulators of water, Ceramic Ceiling, ceramic, granite, marbles, sand, wood, bricks, iron, aluminum and Paints), which are used as building materials in Saudi Arabia.

2. Materials and method

2.1 Sampling, and sample preparation:

Forty samples of thirteen different brands of building materials (cement, gypsum, Insulators of water, Ceramic Ceiling, ceramic, granite, marbles, sand, wood, bricks, iron, aluminum and Paints) were collected from the local market in Jeddah. The samples were sieved with 1m x 1m mesh sieve, and then dried at 80 C to keep the volatile polonium and cesium. The dried sample were put in a polyethylene Marinelli beaker and sealed for about 4weeks in order to establish secular equilibrium between ^{232}Th and ^{226}Ra with their progeny obeying the method given by RADREM, 1980.

2.2 Gamma spectrometry:

The gamma-ray spectra of the samples were measured using Gamma spectrometer based on HPGe crystal, Tennelec model number CPVD 530 – 15200 of coaxial vertical configuration, with 20% efficiency and 2 keV resolution at 1332 keV, The detector is housed inside a thick lead shield to reduce the background of the system. For energy calibration Cs-137 (661.66 keV) and Co-60 (1.173 & 1.3325 MeV) were used. The efficiency calibration was performed by using point source of 152Eu and ^{226}Ra (IAEA; 1989). The samples were counted for a period of 82800 s and the spectra were analyzed for the photo peak of uranium, thorium daughter products and K-40. The specific activity of ^{226}Ra was evaluated from gamma-ray lines of (1001 keV Pa-234m, 295.09, 351.87 keV Pb-214 & 609.31, 1120.27 & 1764.49 keV of Bi-214). For Th-232 series energies of (338.42, 911.16, 964.6 & 968.97 keV of Ac-228 & 727.25 of Bi-212 and 583.02 & 2614.48 keV of Tl-208) were used,(Saito & Moriuchi, 1985 & Holden Norman, 2003). 40K activities were estimated using the 1460 keV gamma-ray peak and for the man made Cs-137 the 661.66 keV of Ba-137.

2. 3. Calculations

2.3.1 Activity concentrations

The activities of the radionuclides were calculated using the following equation:

$$A_c\,(^{A}x) = C/(m\,T\,P_c\,\xi) \quad \text{---------- (1)}$$

where $A_c(^{A}X)$ is the activity concentration of the radionuclide ^{A}X (Bq kg^{-1}) in the sample, C the count rate obtained under the corresponding peak, m the sample mass (kg), T is the counting time(s), P_c the emission probability, and ξ is the detection absolute efficiency at a specific energy.

2.3.2 *Radiation hazard index*

Radium- equivalent activity index

The radium equivalent activity (Raeq),defined according to the estimation that 1 Bq.kg-1 of 226Ra, 0.7 Bq.kg-1 of 232Th and 13 Bq.kg-1 of 40K produce the same -ray dose (Berekta & athew1985). This index Raeq is given as (UNSCEAR, 1982):

$$Raeq = ARa + 1.43ATh + 0.077AK \quad \text{----------- (2)}$$

Where, ARa, ATh and AK are specific activities of 226Ra, 232Th and 40K, respectively, in Bq.kg-1. The maximum value of Raeq in building materials must be less than 370 Bq.kg-1for safe use i.e., to keep the external dose below1.5mSv.y-1 (UNSCEAR, 2000).

external and Internal gamma indices

The external hazard index for samples under investigation is given by the following equation (Beretka and Mathew, 1985):-

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \ \ldots\ (3)$$

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.

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):

$$I_\gamma = C_{Ra}/300 + C_{Th}/200 + C_K/3000 \leq 1 \quad \text{----- (4)}$$

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

Absorbed gamma dose rate and annual effective dose

The absorbed dose rates (DR) 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_R (nG h^{-1}) = $0.427C_{Ra}$+ $0.623C_{Th}$+ $0.043C_K$ ……(5)

where C_K, C_{Ra} and C_{Th} are the activity concentrations (Bq kg^{-1}) of ^{40}K,^{226}Ra and ^{232}Th, respectively, in the samples.

The annual effective dose rate is calculated by applying the dose conversion factor of 0.7SvGy^{-1} with an indoor occupancy factor of 0.8 (UNSCEAR., 2000):

D_{eff} (mSv/y) = D(nGyh^{-1}) ×(0.7Sv/Gy ×8760h/year ×0.8) ×10^{-6} ------(6)

where 8,766 h is the number of hours in 1 year. Taking into account the indoor exposure, D(nGyh^{-1}) ~ 80 nGyh^{-1} to terrestrial gamma-rays for the case of a typical masonry, Eq. (5) gives D_{eff} (mSv/y) = 0.39 mSvy^{-1} as the annual effective dose for individuals indoors, the recommended upper limit is 1 mSvy^{-1}, UNSCEAR.2000.

3. Results and Discussion

3.1 Activity concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the various brands of building materials considered in this work is given in Table (1). The range and mean values of activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K in various building materials used in Saudi Arabia are presented in Table 1. From these results, it can be seen that the lowest mean value of the ^{226}Ra concentration is 1.3 Bq/ kg^{-1} measured in Insulators of water, whereas the highest mean value for the same radionuclide is 64.1 Bq kg^{-1} measured in granite. The lowest mean value of ^{232}Th is 3.2 Bq kg^{-1} recorded in Paints and the highest mean value is 86.5Bq kg^{-1} measured in Ceramic. The lowest mean value for ^{40}K is 4.4 Bq/kg in Insulators of Water and the highest mean value is 1082.3 Bqkg^{-1} measured in granite, 40K is lower than the detected limit in iron and Paints. Generally, the mean value 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 building materials except for ^{232}Th and ^{40}K for ceramic and granite. The artificial nuclide 137Cs is not detected in the present samples. Figure 1 shows comparison between the mean activity concentration of ^{226}Ra, 232Th and ^{40}K in building under investigation.

3.2 Radiation hazard index results

Radiological hazards for different brand samples under investigation 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 building materials samples range from 7.07 Bq/kg in Insulators of water to 267.41 Bq/kg in Granite sample with an average value 54.55 Bq/kg. As can be seen, all building 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)..

2- The external gamma indices for all brand samples considered in this work are less than unity. It varies from 0.02 in Insulators of water to 0.72 in granite with an average value 0.19. All values are less than 1.

3- The representative level index (Iγ) value obtained for all the brand samples range from 0.03 Bq kg^{-1}in two samples (Insulators of water and Paints) to 0.99 Bq kg^{-1} in Granite with an average value 0.25. This shows that the (Iγ) values of all samples are within the world standard value.

4- The absorbed dose rate varies from 5.73 nGy h^{-1} in Insulators of water to 237.85 nGy h^{-1}in granite, with an average value of 60.94 nGy h^{-1}. This indicates that the obtained data for absorbed dose rate is still below the standard limit of 84 nGyh^{-1} as reported in UNSCEAR, 2000.

5- The estimated indoor annual effective dose rate $(AEDE)_{indoor}$ obtained in different brands of building materials, range from 0.03mSv y^{-1} in pains to 1.17mSv y^{-1} in granite with an average value of 0.29 mSv y^{-1}. Only granite sample effective higher than the the recommended limit of 1mSv/y, UNSCEAR, 2000, all the other samples does not exceed this limit. This indicates that the building materials under study which are 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 all brand samples under investigation.

Table (1): Range and average values of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K ($Bqkg^{-1}$) in different building materials used in Saudi Arabia.

Sample Type	Radioactivity concentration($Bqkg^{-1}$)		
No. of sample	**226Ra** **Range (average)**	**232Th** **Range (average)**	40K **Range (average)**
Cement (3)	3.89 – 66.02 (28.5)	2.28 – 12.95 (7.6)	LDL – 458.39 (167.4)
Gypsum (3)	3.54 – 5.34 (4.5)	6.78 – 13.87 (9.2)	LDL – 26.84 (12.0)
Insulators of water	0.64 – 2.32 (1.3)	LDL – 8.52 (3.8)	LDL – 4.06 (4.4)
Ceramic Ceiling (3)	7.84 – 9.11 (8.64)	6.40 – 7.97 (7.15)	26.07 – 438.43 (165.33)
Ceramic (3)	38.5 –129.22 (71.3)	57.30 – 140.48 (86.5)	370.32 – 637.30 (522.0)
Granite (3)	53.76 – 78.11 (64.1)	72.46 – 98.99 (83.9)	1053.89 – 1108.31 (1082.3)
Marble (3)	1.05 – 7.38 (3.2)	2.86 – 8.52 (5.2)	9.00 – 33.84 (22.8)
Sand (3)	3.63 – 7.36 (5.3)	2.75 – 13.51 (7.7)	LDL – 326.78 (112.0)
Bricks (3)	5.28 – 0.93 (8.7)	7.52 – 9.73 (8.6)	142.06 – 454.47 (271.2)
Wood (3)	3.35 –23.73 (13.1)	13.98 – 32.46 (19.7)	LDL – 2108.63 (1016.4)
Iron (4)	7.67 –46.92 (21.6)	8.52 – 52.79 (24.1)	LDL
Aluminum (3)	1.25 – 6.51 (4.3)	4.77 – 14.91 (9.2)	LDL – 7.60 (5.6)
Paints (3)	1.19 – 3.92 (2.7)	2.13 – 4.25 (3.2)	LDL
Total average	18.25	21.22	260.11

Table 2: Radiological hazards calculated for building materials samples used in Saudi Arabia.

Sample code	***Radium equivalent Raeq (Bq/kg)***	***External index Hex***	***Gamma index Iγr***	***Dose rate D indoor (nGy/h)***	***Annual effective Dose(mSv/y)***
Cement	52.26	0.14	0.19	47.97	0.24
Gypsum	18.58	0.05	0.07	15.22	0.07
Insulators of water	7.07	0.02	0.03	5.73	0.03
Ceramic Ceiling	31.59	0.09	0.12	29.04	0.14
Ceramic	52.26	0.64	0.84	202.51	0.99
Granite	267.41	0.72	0.99	237.85	1.17
Marble	12.39	0.03	0.04	10.49	0.051
Sand	24.94	0.07	0.09	22.31	0.11
Bricks	41.88	0.11	0.16	39.16	0.19
Wood	119.53	0.32	0.48	115.03	0.56
Iron	56.06	0.15	0.19	46.38	0.23
Aluminum	17.89	0.05	0.06	14.52	0.07
Paints	7.28	0.19	0.03	6.0	0.03
Range	7.07-267.41	0.02-0.72	0.03-0.99	5.73-237.85	0.03-1.17
Average	54.55	0.19	0.25	60.94	0.29
World standard	370	≤ 1	≤ 1	84	1

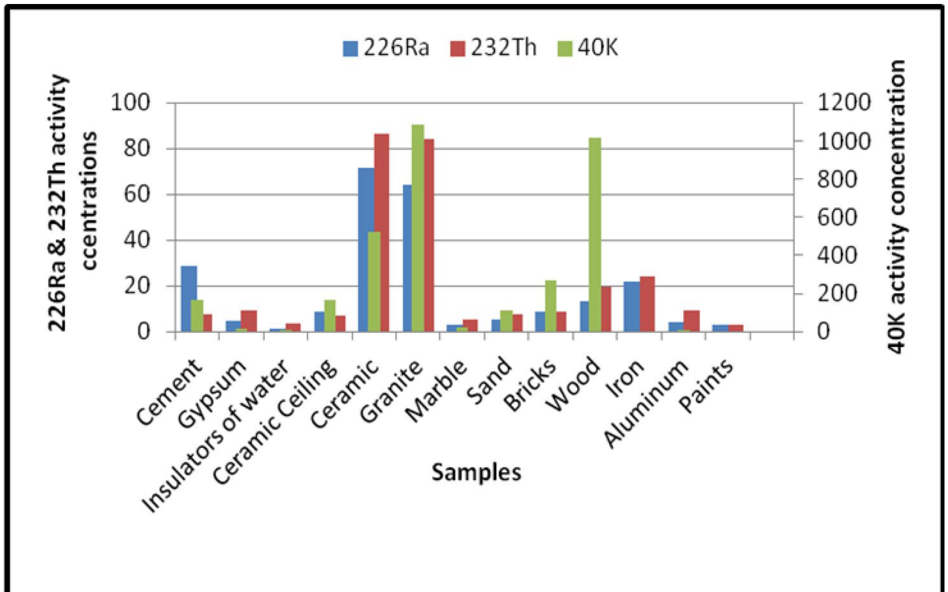

Fig (1). The comparison of the average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in building materials are used in Saudi Arabia.

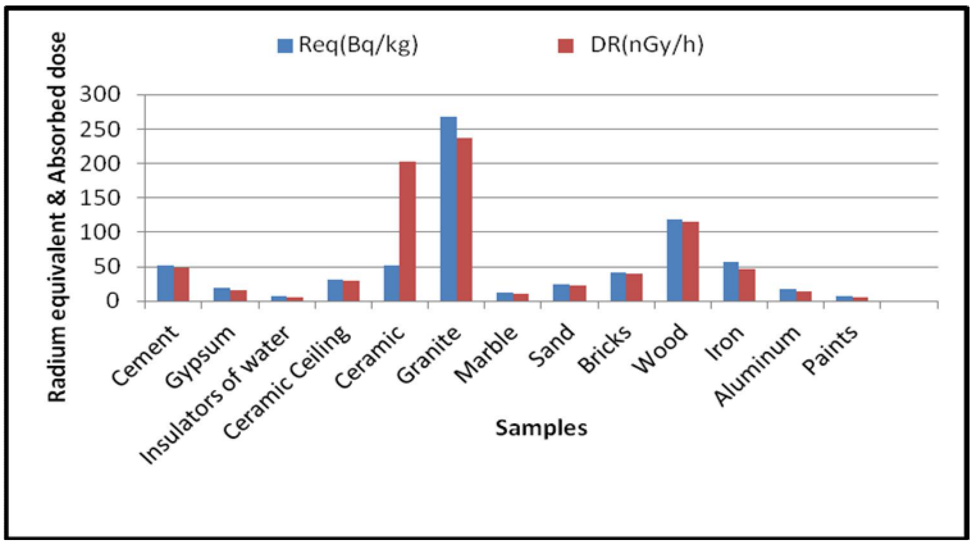

Fig (2). Radium equivalent and Absorbed dose rate in building materials under investigation.

Conclusion:

Most of the analyzed samples have radioactive concentrations within the accepted range which gives radium equivalent less than 370 Bq/kg dry weight except ceramic and granite samples have high concentrations. Wood samples have high concentrations of ^{40}K which contribute the absorbed dose. The present building materials as cement, gypsum Insulators of water, Ceramic Ceiling, marbles, sand, bricks, iron, aluminum and Paints have low concentrations for all the radioactive nuclides. The analyzed samples show the absence of manmade nuclides as Cs-137. We conclude to decrease the areas of the granite as well as ceramic, also some wood and advise not to use wood for walls.

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