Assessment of Radioactivity and the Exposure Doses from Local Cement Types in Saudi Arabia

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Abstract: Activity concentration of ^{232}Th, ^{226}Ra and ^{40}K in local cement factories (Aljouf, Yanbo, Alqassim, Tabuk, Jeddah and Alarabia) in Saudi Arabia has been measured using Na (Tl) detector. The mean values of radium equivalent fluctuated from 83 Bq/Kg in Yanbo to117 Bq/Kg in Aljouf. The average absorbed dose rate changes from 56.8 nGyh^{-1} and 38.6 nGyh^{-1} for Aljouf and Yanbo factories, respectively. These average values give rise to a mean effective dose 189 µSvy^{-1} and 279 µSvy^{-1} which are just about 2 % and 3 % of the 1.0 mSvy^{-1} recommended by the International Commission on Radiological Protection (ICRP, 1990) as the maximum annual dose to members of the public. The results indicate no radiological anomaly. The data presented here will serve as a baseline survey for primordial radionuclide concentrations in cement of the areas.

[J.H. AlZahrani **Assessment of Radioactivity and the Exposure Doses from Local Cement Types in Saudi Arabia** J Am Sci 2012;8(6):215-220]. (ISSN: 1545-1003). http://www.jofamericanscience.org. 27

Keywords: Radioactivity, radium equivalent, cement, Dose assessment.

1. Introduction

Human exposure to ionizing radiation is one of the scientific subjects that attract most public attention. As natural radiation is the largest contributor to the external dose of the population (UNSCEAR, 2000), it is important to assess the gamma radiation dose from natural sources. The main source of indoor gamma radiation is building materials besides terrestrial and cosmic radiation . Buildings are very important in human life as 80% of the life time is spent at home and /or office. Indoor elevated external dose rates may arise from high activities of radionuclides in building materials (UNSCEAR,1993).It will be possible to assess any possible radiological hazard by measuring radioactivity of building materials (Malance.A. et al ; 1993.

As is known, cement is the most important construction material of today civilization. The radioactivity content of cement varies considerably depending upon the geological characteristics of the initial raw materials from which the cement is processed. The knowledge of radioactivity in building materials, cement in particular, and the associated radiation doses due to inhalation are of paramount importance for assessment of radiological hazards to human health. Radioactivity levels in cements were reported by many authors from different geographical locations (Mollah, et al., 1986 , Croft, et al., 1999, Kumar. et al;1999, Kan and Kan. 2001, Sam.& Abbas. 2001, Xinwei *et al.,* 2007, Hizem *et al.,* 2005 & Brahmanandhan *et al.,* 2007).

Presently, there are no standards or guidelines prescribing the acceptable levels of radioactivity in building materials in the country as in some industrialised countries (Steger *et al.,* 1992). This study is aimed at establishing broad base-line data on activity concentration of natural series nuclides in local cement types as well as assessing the potential radiological hazards to man.

2. Sample Collection and Measurement

A total of 30 cement samples representing 6 different types used in Saudi Arabia were collected from the local market ,all of them manufactured locally, Aljouf, Yanbo alqassim, Tabuk, Jeddah and Alarabia. The samples are dried at 110^{o} for 48 hours .The dried samples were crushed and sealed in a cylindrical polyethylene containers. The applied spectrometer consists of Na (Tl) detector connected with 1024 microcomputer multichannel analyzer. The detector has the following characteristics: peak efficiency: 1.2 X10^{-5} at 1332kev, crystal dimensions 3 X 3 inch and resolution: 7.5 for 662 kev.

The ^{226}Ra activities (or ^{238}U activities for samples assumed to be in radioactive equilibrium) were estimated from ^{214}Pb (242.2, 295.2, 351.9 keV) and ^{214}Bi (609.3, 1120.3 keV). The Gamma-ray energies of ^{212}Pb (238.6 keV),^{228}Ac (338.4, 911 keV) and ^{208}Tl (583.2 keV) were used to measure the concentration of ^{232}Th, while the ^{40}K activity was determined from the 1460.7 keV emission. The sample was sealed and the measurements were made one month later to assure secular equilibrium between the ^{226}Ra and its daughters (Manazul *et al.,* 1999 & Abbady, 2010). The activity concentrations of the natural radionuclides in the measured samples were computed using the following relation (Noorddin, 1999):

$$A_S\ (\mathrm{Bq\ kg^{-1}}) = C_a / \varepsilon\ P_r M_s \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

Where C_a is the net gamma counting rate (counts per second), ε the detector efficiency of the specific γ–ray,

P_r the absolute transition probability of Gamma–decay and M_s the mass of the sample (kg).

3. Results and Discussion

3.1. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The variation of the mean activity concentration (Bq/kg) of ^{226}Ra ,^{232}Th and ^{40}K radionuclides in the cement samples under investigation from the different factories , Kingdom of Saudi Arabia were represented in Table (1) . From all samples (30 samples) , the ^{226}Ra activity (or ^{238}U activities for samples assumed to be in radioactive equilibrium) ranges from 23 Bq/kg in (Aljouf) to 47 Bq/kg in (Alarabia) with an average of 33±1.6 Bq/kg . The activity concentration of ^{232}Th ranges from 22 Bq/kg in (Tabuk) to 39 Bq/kg in (Alqassim) with an average of 30±1.5Bq/kg. Finally, the activity concentration of ^{40}K ranges from 131 Bq/kg in (Tabuk) to 731 Bq/kg in (Tabuk) with an average of 313 ±4.4 Bq/kg .The results show that the average concentration of ^{226}Ra,^{232}Th and ^{40}K in the cement samples are lower than the world average value of 500 $Bqkg^{-1}$ (UNSCEAR, 1993).The mean activity concentration (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K are plotted as histograms (1) for different locations from the cement factories.

3.2. Ra equivalent activity (Ra_{eq})

The radionuclide content of building materials has been reported in several publications (Sciocchetti *et al.,* 1983, El-Tahawy & Higgy, 1995,Yasir *et al.,* 2007, Xinwei *et al.,* 2007and Kilic & Aykamis, 2009).In comparing the radioactivity of materials that contain ^{226}Ra,^{232}Th and^{40}K a common index termed radium-equivalent activity is required to obtain the total activity and is also used to assess the gamma radiation hazards. Since 98 % of the radiological effects of the uranium series are produced by radium and its daughter products, the contribution from the ^{238}U and the other ^{226}Ra precursors is usually ignored, so that the Ra_{eq} of a sample can be calculated using the formula proposed by (Beretka & Mathew, 1985) :

$$Ra_{eq} = C_{Ra} + (10/7)\, C_{Th} + (10/130)\, C_k \ldots\ldots\ldots\ldots..(2)$$

where C_{Ra} ,C_{Th} and C_k are the activity concentrations of ^{226}Ra ,^{232}Th and ^{40}K in $Bq.kg^{-1}$.The mean and the range of radium equivalent of the total activity of the samples calculated on the basis of the aforementioned relationship are shown in Table(1).On average, Ra_{eq} concentration was found to be 117 $Bq.kg^{-1}$ for Aljouf, 104 $Bqkg^{-1}$ for Alarabia ,100 $Bqkg^{-1}$for Tabuk,94 $Bqkg^{-1}$ for Alqassim, 92 for Jeddah and 83 $Bqkg^{-1}$ for Yanbo cement. Upon comparing the results, it is seen that there are no significant variations in Ra_{eq} values of different cement samples analyzed. The highest value (117 $Bqkg^{-1}$) of Ra_{eq} was observed in Aljouf cement, while the lowest value (83$Bqkg^{-1}$) was found in Yanbo cement. However, all the values obtained here for radium equivalent activity below the internationally accepted value of 370 $Bqkg^{-1}$(Beretka & Mathew, 1985). Consequently, all of these types of local cement do not pose a significant radiological hazard when used for construction of buildings.

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).

3.3. Representative level index ($I_{\gamma r}$).

Representative level index ($I_{\gamma r}$) is used to estimate the level of $^{\gamma}$-radiation hazard associated with the natural radionuclides in specific building materials, is defined as (NAE-OECD, 1979, El-arabi, 2005) :

$$I_{\gamma r} = C_{Ra}/150 + C_{Th}/100 + C_k/1500 \ldots\ldots\ldots\ldots\ldots\ldots\ldots(3)$$

Where C_{Ra},C_{Th} and C_k are the activity concentrations of^{226}Ra,^{232}Th and^{40}Kin $Bqkg^{-1}$, respectively.

The calculated average values of I□r for the samples of cement ranged from 0.59 to 0.88 $Bqkg^{-1}$ Table (2). All the I□r values are below the internationally accepted value of 1 $Bqkg^{-1}$ (NAE-OECD 1979), these means that the external radiation dose within the building is less than the maximum suggested dose.

3.4. Absorbed dose and annual effective dose rate.

For materials containing naturally occurring radioactive materials such as ^{238}U, ^{232}Th and ^{40}K, the absorbed dose rate $\dot{D}$ can be defined if the radionuclide concentrations are known. It can be obtained in units of nGy h^{-1} using the formula proposed by UNSCEAR (1988):

$$\dot{D} = \sum_x A_x C_x \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (4)$$

where A_x (Bq kg^{-1}) are the mean activity of ^{226}Ra,^{232}Th and ^{40}K, and C_x (nGy h^{-1} per Bq kg^{-1}) their corresponding dose conversion factors. In the present work, we took the dose conversion factors reported by (UNSCEAR, 1988), namely 0.427, 0.662, 0.043 nGy/ h per Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. Table (1)gives the results for absorbed dose rate in air for cement samples from the different factories. We notice that cement from Aljouf shows the highest average value (56.8 $nGyh^{-1}$), while the lowest average value is found in Yanbo (38.6 $nGyh^{-1}$). Generally , all the calculated dose rate are within the estimate of average global primordial radiation of 55 nGy h^{-1} and comparable with the world range (28 -120 $nGyh^{-1}$) (UNSCEAR, 1988, 1999).

To estimate the annual effective dose rate , account must be taken of (a) the conversion coefficient from absorbed dose in air to effective dose and (b) the indoor occupancy factor. The average numerical values of those parameters vary with the age of the population and the climate at the location considered. In the(UNSCEAR 2000)Report, the Committee used 0.7 $SvGy^{-1}$ for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.8 for the indoor occupancy factor . Taking These values into account ,the annual effective dose rate was calculated according to ((UNSCEAR 1988, 1999) :

Effective dose rate ($mSvy^{-1}$) = D (nGy h^{-1}) ×8760 (h/y)×0.8×0.7($SvGy^{-1}$)×10^{-6} ....(5)

The world wide average annual effective dose in air is approximately 0.5 $mSvy^{-1}$ and the results for individual countries being generally within the 0.3 – 0.6 $mSvy^{-1}$ range (UNSCEAR, 1988, 2000). Table (2) represent the average annual effective dose rates which they varied from 189 μSvy^{-1} (Yanbo) to 279 μSvy^{-1} (Aljouf). All values are lower than the global primordial radiation for the effective dose rates in the air. The mean absorbed dose rate,the effective dose rate, the radium equivalent activity concentration(Ra_{eq}) and the level index ($I_{\gamma r}$) are plotted as histograms (2) for different cement factories.

3.5. External Hazard Index and Internal Hazard Index.

In the literature a number of criterion formulae have been derived over the years to assess the indoor radiation dose rate due to exposure to gamma radiation from the natural radionuclides contained in building materials. The merits of these have been reviewed by the OECD's Nuclear Energy Agency(NEA-OCED, 1979). (Karpov and Krisiuk, 1980) have proposed a model for the activity concentrations that limits the annual radiation exposer due to radioactivity in building materials to about 1 mSv , based on infinitely thick walls without windows and doors. The criterion of this model is called an external hazard index (H_{ex}) :

$H_{ex} = (C_{Ra}/370 + C_{Th}/259 + C_K/4810) < 1$ …………(6)

where C_{Ra} ,C_{Th} and C_K are the activity concentrations in $Bqkg^{-1}$ of ^{226}Ra ,^{232}Th and ^{40}K respectively . (Hewamanna *et al.*,2001) corrected this model by considering a finite thickness of walls and the existence of windows and considerations into account doors .Taking these considerations into account to obtain the criterion for building materials that are acceptable for construction of inhabited buildings ,the formula for the external index used as :

$H_{ex} = (C_{Ra}/740 + C_{Th}/520 + C_K/9620)$ ……………..(7)

The value of this index must be less than unity for the radiation hazard to be negligible .

In addition ,the radioactivity inert gas radon ^{222}Ra,a decay product of ^{226}Ra and its short lived decay products are also hazardous to respiratory organs .The internal exposure to radon and its decay products is quantified by the internal hazard index (H_{in}) and defined as(Krieger, 1981 , Beretka &Mathew, 1985) :

$H_{in} = (C_{Ra}/185 + C_{Th}/259 + C_K/4810)$ ……………..(8)

where C denotes the respective specific activity in $Bqkg^{-1}$,for the safe use of amaterial in the construction of dwelling ,H_{in} should be less than unity .Table (1) showes the calculated values of H_{ex} and H_{in} for the local cement samples .The average values of H_{ex} ranged from 0.12 to 0.16 and 0.30 to 0.41 for H_{in} ,all values less than unity .

Comparative study of average activity concentrations radiation hazard indices values for cement with the previous measurements from deferent countries of the world including Saudi Arabia presented in Table (2) . The differences of ^{226}Ra, ^{232}Th and ^{40}K concentration and their radiation hazard in cement samples of different areas are probably caused by raw materials and processing techniques (Alm *et al.;* 1999).

Table 1. Mean (and range) of specific activities of ^{232}Th, ^{226}Ra and ^{40}K ($Bqkg^{-1}$), radium equivalent activity (Ra_{eq}) , Dose rate values and H_{ex} ,H_{in} for local cement types used in Saudi Arabia.

Sample	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}($Bqkg^{-1}$)	Dose rate($nGyh^{-1}$)	Hex	Hin
Aljouf	35±1.2 (23–39)	31±1.4 (32 –36)	505±1.7 (329–704)	117±3.3 (80–144)	56.8±1.5 (39–70)	0.16	0.41
Yanbo	30±1.3 (24–38)	28±1.1 (25 –33)	120±2.1 (147– 185)	83±2.9 (78–99)	38.6±1.4 (26 –40)	0.12	0.30
Alqassim	33±1.1 (27–38)	34±1.6 (29– 39)	170±1.3 (150–189)	94±3.5 (97 – 108)	44.7±1.7 (37 –50)	0.13	0.34
Tabuk	29±1.2 (24–31)	26±1.1 (22– 29)	434±2.1 (131–731)	100±3.0 (70 – 129)	48.4±1.4 (30–65)	0.14	0.35
Jeddah	37±1.5 (30–47)	29±1.3 (22–36)	168±2.1 (144– 183)	92 ±3.6 (72 –113)	42.3±1.7 (33–52)	0.13	0.33
Alarabia	38±1.4 (32–47)	30±1.2 (25–35)	313±2.2 (159 –532)	104±3.3 (79 –138)	49.1±1.5 (37– 66)	0.14	0.38

Table 2. Comparison of radiation hazard indices obtained in this study with measurements from different countries.

Country	Absorbe dose (nGy/h)	Ra_{eq} (Bq/kg)	$I_{\gamma r}$ (Bq/kg)	Effective dose rate (µSv/y)	References
Malaysia	82.7	181	1.27	406	Chong & Ahmed(1982)*
China	54.8	110	0.8	269	Xinwei *et al*.(2007)
Angladesh	71.1	148	1.09	349	Alm *et al.* (1999)
U.S.A.	32.6	72	0.5	159	Ingersoll(1983)*
India	49.8	105	0.78	224	Kumar *et al.* (1999)**
Algeria	42.5	112	0.82	208	Amrani &Tahtat(2002)**
Pakestan	41.8	88	0.64	205	Kan &Kan (2001)**
Australian	58.9	129	0.9	289	Beretka&Methew(1985)**
Sweden	65.0	141	1.0	319	CLiff *et al.* (1984)*
Saudi Arabia					Present work
Aljouf	56.8	117	0.88	279	
Yanbo	38.6	83	0.59	189	
Alkassim	44.7	94	0.67	219	
Tabuk	48.4	100	0.74	237	
Jeddah	42.3	92	0.65	208	
Alarabia	49.1	104	0.75	240	

*References from Alm *et al.* (1999) ,**references from Xinwei Lu.*et al.* (2007)

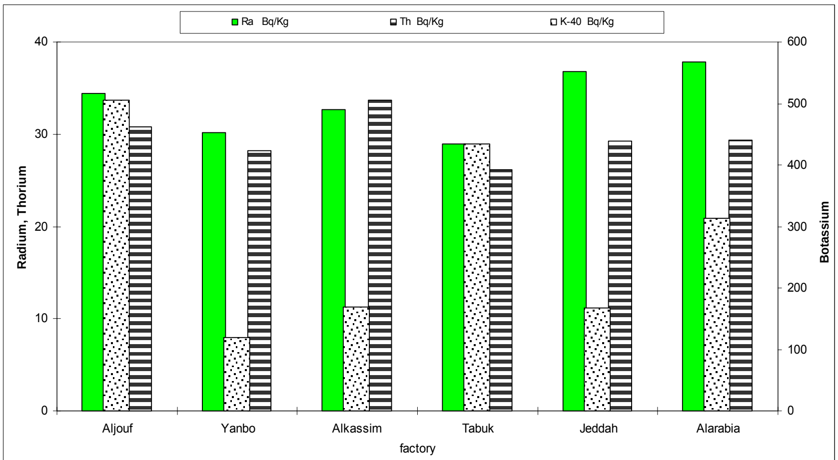

Fig.(1) Mean values of Radium , Thorium and Botassium (Bq/Kg) for cement from different Factories in Saudi Arabia.

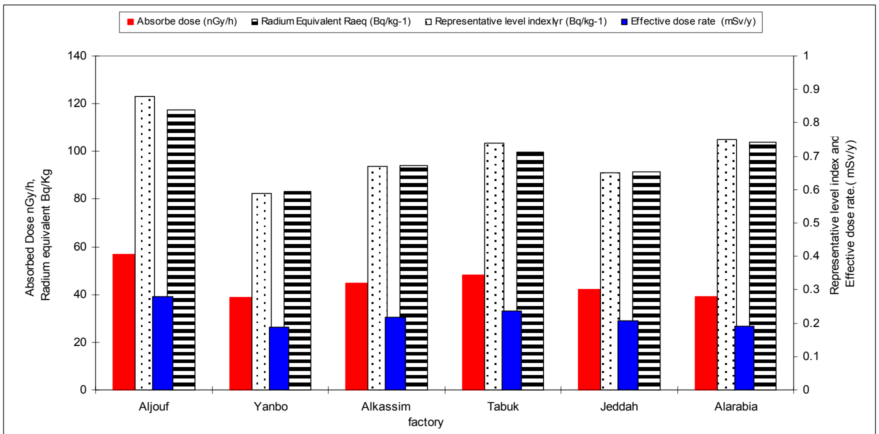

Fig.(2) Mean values of Absorbed dose rate (nGy/h),Radium equivalent (Bq/Kg),Representative level indexIyr (Bq/kg-1) and Effective dose rate (mSv/y) for cement from different Factories in Saudi Arabia.

Conclusions

The observed concentrations of ^{226}Ra ,^{232}Th and ^{40}K in most of the local cement smples are close or slightly above the corresponding world typical value of 500 $Bqkg^{-1}$ .The Ra_{eq} and $I_{\gamma r}$ values of all studied samples are below the internationally accepted values .The calculated mean Ra_{eq} values are also lower than the internationally recommended maximum level of equivalent of 370$Bqkg^{-1}$ for building materials .The annual effective dose is well below the recommended value (1mSv y^{-1}) . The values of both internal and external hazard indices are less than unity. The study has shown that all the types of cement in Saudi Arabia can safely be used as a construction material and poses no any radiological complication.

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