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Natural Radioactivity and its Associated Radiological Hazards in Ghanaian Cement

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Abstract: Radiation hazard due to presence of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in five Ghanaian cement types from different factories were assessed using gamma-ray spectrometry with HPGe detector. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the studied cement samples were 35.94±0.78, 25.44±0.80, and 233.0±3.95 Bq/kg, respectively. The radium equivalent activity (Ra_{eq}), the external and internal hazard indices, ^{222}Rn mass exhalation rate, the emanation coefficient, the indoor absorbed dose rate and the corresponding annual effective dose were estimated for potential exposure risks of the cement samples. The results obtained were compared with data from other countries. The mean indoor absorbed dose rate was 50.5% less than the population-weighted average of 84 nGy/h whiles the corresponding effective dose was 79% less than the dose criterion of 1 mSv/y. The results obtained in this study show no significant radiological hazards arising from using Ghanaian cement for construction of houses.

Key words: Annual effective dose, gamma spectroscopy, indoor absorbed dose rate, radium equivalent activity, radon emanation coefficient, radon mass exhalation rate

INTRODUCTION

Historical antecedents of studies conducted on natural radioactivity have established that the presence of the uranium-thorium series and potassium-40 in various materials constitute potential exposure to the global population. Empirical evidence suggest that these radionuclides are predominant in raw and produced materials used in the building industry, such as cement, bricks, sand, tile, limestone, gypsum and others derived from rock and soil. Radiation exposure due to the building materials can be divided into external and internal exposures. The external exposure is caused by direct gamma radiation whereas the internal exposure is caused by the inhalation of radioactive inert gas radon (^{222}Rn, a daughter product of ^{226}Ra) and its short-lived secondary decay products. The specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the building raw materials and products mainly depend on geological and geographical conditions as well as geochemical characteristics of these materials. The worldwide average specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the earth's crust are estimated at 32, 45 and 412 Bq/kg, respectively (UNSCEAR, 2000, 2008). In order to assess the possible radiological hazards to human health, it is important to study the radioactivity levels emitted by the building materials. The data obtained from such a study is essential for the development of standards and guidelines for use and the management of these materials (Turhan *et al*., 2008a, b; Mujahid *et al*., 2008)

Cement is one of the important and expensive materials used by the building industry in Ghana. Most buildings in Ghana are constructed from cement blocks and cement concrete. Cement is used for making blocks, concrete, and for plastering the buildings made of bricks, blocks or concrete. The development of a strong physical infrastructural base is cardinal in achieving the millennium development goals of the country to attain a middle income status by 2015, as this will aid industrialization and consequently promote a sound economic growth. Ghana's housing deficit currently stands at more than 1 million with an annual delivery of only 40,000 being provided. The country's cement industry is estimated to grow by over 8% every year for the next 20 years. This is because demand for cement in the country is expected to increase as a result of increasing population and expansion of infrastructure (Ministry of water resources, works and housing report, 2010).

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Portland cement results from the grinding of a clinker. The clinker is produced by burning a mixture of limestone, clay, and gypsum at high temperatures (1450-1600ºC for the materials, and approximately 2000ºC for the combustion fumes).

Portland cement dust is a gray powder with an aerodynamic diameter ranging from 0.05 to 5.0 μm (Kalacic, 1973a). This size is within the range of sizes of respirable particles, and, therefore, exposure to Portland cement dust has long been associated with respiratory symptoms and varying degrees of airway obstruction in people who work with Portland cement (Bazas, 1980; El-Sewefy *et al*., 1970; Kalacic, 1973a, b; Oleru, 1984; Shamssain *et al*., 1988; Saric *et al*., 1976; Noor *et al*., 2000; Yang *et al*., 1996).

To date, data on levels of natural radioactivity in Ghanaian cement are not available. Currently, the standards and guidelines with regard to acceptable levels of natural radioactivity in building materials are not defined by the regulatory authority in Ghana.

The objective of the present study was to determine the activity concentration due to ^{226}Ra, ^{232}Th and ^{40}K in five different brands of Ghanaian cement (total of 50 samples) and to assess the associated radiological risks from the use of these cements. The most important radiological parameters, namely; radon emanation coefficient, radon mass exhalation rate, effective dose, absorbed dose rate, radium equivalent activity, the external and internal hazard indices were also determined and evaluated in the studied samples.

MATERIALS AND METHODS

The study was carried out at the Radiation and waste safety Department of the Radiation Protection institute, Ghana Atomic Energy Commission from October, 2009 to September, 2010.

Sampling and sample preparation: A total of 50 cement samples representing the five main types of cement manufactured and used in building construction in Ghana were collected from different cement suppliers for the measurement of radionuclide activity concentrations. The cement samples were used without any processing since they are already in a powdered form. The samples were dried in a temperature-controlled furnace at 110ºC for 24 h to remove moisture. After moisture removal, these samples were cooled in moisture-free atmosphere and pulverized into powdered form. The samples were packed into a plastic 1litre Marinelli beaker, weighed and hermetically sealed. These sample containers were same in size and geometry of the reference materials (RG-set) used for calibrations. The sealed samples and the reference materials were stored for 6-8 weeks before counting so as to allow ^{224}Ra and ^{226}Ra to reach the secular equilibrium with their short-lived decay products.

Measurements of specific radioactivity: The activity concentrations of natural radionuclides were determined by high-resolution gamma-ray spectrometry using n-type HPGe detector Model GR 2518-7500SL (Canberra Industries Inc.) coupled to a computer based PCA-MR 8192 Multi-Channel Analyser (MCA) mounted in a cylindrical lead shield (100 mm thick) and cooled in liquid nitrogen. The detector has a relative efficiency of 25% to NaI detector, 1.8 keV energy resolutions at the energy peak of 1333 keV of ^{60}Co isotope, and a peak-to-Compton ratio of 55:1. The radionuclides were identified using gamma ray spectrum analysis software, ORTEC MAESTRO-32.

The absolute efficiency calibration of the gamma spectrometry system was carried out using the radionuclide specific efficiency method in order to reduce the uncertainty in gamma-ray intensities, as well as the influence of coincidence summation and self absorption effects of the emitting gamma rays (Nir-El, 2000; Stoulos *et al*., 2003; Turhan *et al*., 2008a, b; Mujahid *et al*., 2008). The IAEA reference materials RGU-1(U-ore), RGTh-1 (Th-ore) and RGK-1 (K_2SO_4), with densities similar to the cement samples to be measured after pulverization, were employed for the efficiency calibration of the system. The sample containers were placed on top of the detector for counting. The same geometry was used to determine peak area of samples and references. Each sample was measured during an accumulating time between 36,000 and 80,000 s. Background measurements were taken and subtracted in order to get net counts for the sample. The specific radioactivity of ^{40}K was measured directly by its own gamma ray at 1460.8 keV (10.7), while activities of ^{226}Ra and ^{232}Th were calculated based on the weighted mean value of their respective decay products in equilibrium. The specific radioactivity of ^{226}Ra was measured using the 295.2 (18.2), 351.9 (35.1) keV gamma rays from ^{214}Pb and the 609.3 (44.6), 1764.5 (15.1) keV from ^{214}Bi. The specific radioactivity of ^{232}Th was measured using the 911.2 (26.6) keV from ^{228}Ac, and the 583.2 (30.6) keV from ^{208}Tl. The values inside the parentheses following gamma-ray energy indicate the absolute emission probability of the gamma decay.

The specific activities (A_{SP}) in Bq/kg of the radionuclides in the samples were calculated after decay correction using the expression:

$$A_{SP} = \frac{N_{sam}}{P_E\,, \eta(E), T_c . M_{sam}}$$

Where M_{sam} is the dry-weight of sample (kg), N_{sam} is the net counts for the sample in the peak range, P_E is the gamma emission probability, T_c is the counting time, $\eta(E)$ is the photopeak efficiency .

The minimum detectable activity (MDA) of the γ-ray measurement system was calculated according to the equation (Currie, 1968):

$$MDA = \frac{k_{\alpha}\sqrt{N_B}}{P_E.\eta(E).T_c.M_{sam}}$$

where, (MDA) is in Bq/kg, k_{α} is the statistical coverage factor equal to 1.645 at 95% confidence level, and N_B is the background counts at the region of interest of a certain radionuclide. The MDA for each radionuclide were calculated as 0.12 Bq/kg for ^{226}Ra, 0.11 Bq/kg for ^{232}Th, and 0.9 Bq/kg for ^{40}K.

Assessment of radiation hazard associated with NORM in Ghanaian cement: In this study, radiological parameters such as the radon emanation coefficient, the radon mass exhalation rate, the radium equivalent activity, the external and internal hazard indices, the indoor absorbed dose rate, and the annual effective dose were determined to assess the potential radiation hazards associated with Ghanaian cement samples.

Measurement of radon emanation coefficient and mass exhalation rate: Cement as a building material may be a source of large amounts of dust polluting the air with ^{226}Ra. The ^{222}Rn gas emanated from the ^{226}Ra may constitute a radiation hazard to both people working directly with the cement material and inhabitants of dwellings constructed with cement. The fraction of ^{222}Rn that can diffuse through the building materials is known as the emanation coefficient or fraction of the material. The radon emanation coefficient (Rn_{EC}) is a very important radiological index used to evaluate the amount of the ^{222}Rn emanated fraction released from the building raw materials and products containing naturally occurring radionuclides such as ^{226}Ra in radioactive equilibrium with its parents. The radon emanation coefficient of samples was calculated based on two γ- measurements. The first measurement was carried out directly after sealing of samples, while the second measurement was carried out after attainment of secular equilibrium between radon and its short-lived decay daughters. Based on these measurements, the radon emanation coefficient was calculated according to the following expression (White and Rood, 2001; Turhan, 2008b):

$$Rn_{EC} = \frac{N}{\left(A_o + N\right)}$$

where, Rn_{EC} is the radon emanation coefficient, A_0 is the net count rate of ^{222}Rn at the time of sealing the sample container, N is the net count rate of ^{222}Rn emanated at the radioactive equilibrium with ^{226}Ra and its progeny.

The mass exhalation rate or radon mass exhalation rate is the product of the emanation coefficient and ^{222}Rn production rate (Chowdhury *et al*., 1998; Turhan, 2008b). The mass exhalation rate (E_{Rn}, in Bq/kgs) was determined through the following equation:

$$E_{Rn} = C_{Rn} \times Rn_{EC} \times \lambda_{Rn}$$

where, C_{Ra} is the specific activity of ^{226}Ra (in Bq/kg) and λ_{Rn} is the decay constant of ^{222}Rn (2.1× 10^{-6} per s).

Radium equivalent activity: To represent the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them, a common radiological index has been introduced. The index called radium equivalent activity ($^{226}Ra_{eq}$) is used to ensure the uniformity in the distribution of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K and is given by the expression (Beretka and Mathew, 1985; Mujahid *et al*., 2008):

$$Ra_{cq} = C_{Ra} + 1.43C_{Tn} + 0.077C_K$$

where, C_{Ra}, C_{Th} and C_K are the activities (Bq/kg) of ^{226}Ra (^{238}U-series), ^{232}Th and ^{40}K, respectively. It was assumed that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate. The maximum dose Ra_{eq} in building materials must be less than 370 Bq/kg for safe use, i.e., to keep the external dose below 1.5 mSv/y

External and internal hazard indices: To limit the external γ-radiation dose from building materials to 1.5 mSv/y per practice, the external hazard index (H_{EX}) was calculated as (Beretka *et al*., 1985; Mujahid *et al*., 2008):

$$H_{EX} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1$$

In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. To account for this threat the maximum permissible concentration for ^{226}Ra must be reduced to half of the normal limit (185 Bq/kg). The internal exposure to carcinogenic radon and its short lived progeny is quantified by the internal hazard index (H_{IN}) given by the expression (Xinwei *et al*., 2004; Mujahid *et al*., 2008).

Determination of absorbed gamma dose rate: The absorbed dose rates due to γ-radiation in air at 1m above the ground surface for a uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR (2000, 2008) reports. We assumed that the contributions from other naturally occurring radionuclides, such as ^{235}U, ^{87}Rb, ^{138}La, ^{147}Sm and ^{178}Lu,

Table 1: Specific activity of ^{226}Ra, ^{232}Th and ^{40}K and Radium equivalent activity from samples of different brand of Ghanaian cement

	Mean specific activity (Bq/kg)			
Sample ID*	^{226}Ra	^{232}Th	^{40}K	Ra_{eq} (Bq/kg)
CM I	34.56±0.80	38.18±0.90	655.13±3.50	139.60
CM II	51.04±1.24	26.90±0.77	124.22±2.87	99.07
CM III	32.13±0.68	22.40±1.10	208.76±6.40	80.24
CM IV	28.16±0.63	18.13±0.85	99.14±3.19	61.72
CM V	33.80±0.54	21.60±0.40	67.50±1.80	69.90
AVE±SD	35.94±0.78	25.44±0.80	233.0±3.95	90.12±13.52

*: CM I: Portland limestone cement; CM II: Portland limestone cement; CM III: Ordinary portland cement; CM IV: White cement (Tile adhesive cement cole); CM V: White cement (Tile adhesive cement)

to actual dose rates were insignificant. The D_γ (nGy/h) in air was calculated using the expression below:

$$D_\gamma = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K$$

Estimation of annual effective dose: To estimate the effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7 SvG/y) and indoor occupancy factor (0.8) proposed by UNSCEAR 2000 and 2008 reports are used. The annual effective dose, E, in units of mSv was calculated from the expression

$$E = D_\gamma \text{ (n Gy/h)} \times 10^{-6} \times 8760 \text{ h/y} \times 0.8 \times 0.7 \text{ SvG/y} C_K$$

where, D_γ (nGy/h) is the total absorbed dose rate due to gamma radiations from materials containing radionuclides of ^{226}Ra, ^{232}Th, and 0.7 SvG/y is the conversion coefficient from absorbed dose in air to effective dose.

RESULTS AND DISCUSSION

The activity concentrations due to ^{226}Ra, ^{232}Th and ^{40}K and radium equivalent activity in different brands of Ghanaian cement has been calculated and presented in Table 1. The specific activity of ^{226}Ra was found in the range 28.16±0.63 to 51.04±1.24 Bq/kg with an average of 35.94±0.78 Bq/kg, ^{232}Th from 18.13±0.85 to 38.18±0.90 Bq/kg with an average of 25.44±0.80 Bq/kg and ^{40}K varied from 67.50±1.80 to 655.13±3.50 Bq/kg with an average of 233.0±3.95 Bq/kg.There is a variation in the concentration of radionuclides in the samples of different brand of cement. For example, the lowest value for ^{226}Ra was observed in CM IV (Tile adhesive cement cole), and the highest in CM II (Portland limestone cement). For ^{232}Th, the minimum value was in CM IV (Tile adhesive cement cole) and the maximum was in CM I (Portland limestone cement). In the case of ^{40}K, the CM V (Tile adhesive cement) samples showed the lowest concentrations, whereas CM I (Portland limestone cement) samples showed the highest concentrations.

These variations in the activity concentration are due to the varying amounts of uranium, thorium and ^{40}K contents under the earth crust from where the raw material for particular brand of cement was obtained. Limestone is a type of sedimentary rock which contain, 1 ppm of uranium, but relatively lower level of radiation as compared with igneous rock which contain on an average 15 ppm of thorium and 5 ppm of uranium (Me`nager *et al*., 1993). The radionuclide concentration in a particular cement sample also depends on the origin of limestone used in the production. The cement samples may also contain naturally occurring radionuclide due to the presence of these radionuclides in large amounts of waste products used in the cement manufacturing, suh as phosphogypsum.

The mean specific activities of ^{232}Th and ^{40}K obtained for the different brands of Ghanaian cement were less than the average worldwide values of 45 and 412 Bq/kg for ^{232}Th and ^{40}K respectively. On the other hand, the average activity concentration value of ^{226}Ra was higher than its corresponding mean worldwide value of 32 Bq/kg.

The calculated Ra_{eq} values for the cement samples are given in Table 1. In all the cement samples, the Ra_{eq} values vary from 61.72 to 139.60 Bq/kg with an average of 90.12±13.52 Bq/kg. It is observed that the Ra_{eq} values for all the studied samples are lower than the recommended maximum value of 370 Bq/kg. Thus, these samples are within the recommended safety limit when used as building materials and products. In Table 2, the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K and the Ra_{eq} determined for cement are compared with the corresponding values determined in other different countries.

It is observed that the Ra_{eq} values for cement samples originating from different countries show considerable variations, which are likely related to the type of raw materials used in cement manufacture. The comparison also shows that the calculated mean Ra_{eq} in this study is lower than that calculated in Australia, Algeria,Bangladesh, Brazil, China, Egypt, Greece, Italy, India and Turkey and higher than that in Austria, Finland, Japan, Netherlands, Pakistan, and Tunisia.

The ^{222}Rn emanation coefficient and mass exhalation rate of cement samples are presented in Table 4. The calculated mean values of ^{222}Rn emanation coefficient and

Table 2: Comparison of the mean values of ^{226}Ra, ^{232}Th, ^{40}K and radium equivalent(Ra_{eq}) activities in Ghanaian cement samples with data from other countries

Country	No.	Specific Activity (Bq/kg) ^{226}Ra	^{232}Th	^{40}K	Ra_{eq}(Bq/kg)	References
Australia	7	51.50	48.10	114.7	129.4	Beretka *et al*. (1985)
Austria	18	26.70	14.20	210.0	63.1	Sorantin and Steger (1984)
Algeria	12	41.00	27.00	422.0	112.0	Amrani and Tahtat (2001)
Bangladesh	18	62.30	59.40	328.9	172.8	Chowdhury *et al*. (1998)
Brazil	1	61.70	58.50	564.0	188.8	Malanca *et al*. (1993)
China	46	56.50	36.50	173.2	122.0	Xinwei (2004)
Egypt	85	78.00	33 30	37.0	151.0	El Afifi *et al*. (2006)
Finland	11	40.00	20.00	251.0	88.0	Mustonen (1984)
Ghana	50	35.94	25.44	233.0	90.12	Present Study
Greece	20	62.80	23.80	284.1	117.0	Papastefanou *et al*. (2005)
India	1	37.00	24.10	432.2	104.7	Kumar *et al*. (1999)
Italy	7	38.00	22.00	218.0	92.0	Rizzo *et al*. (2001)
Japan	16	35.80	20.70	139.4	77.0	Suzuki *et al*. (2000)
Netherlands	6	27.00	19.00	230.0	71.9	Ackers *et al*. (1985)
Parkistan	25	26.70	28.60	272.9	87.9	Khan and Khan (2001)
Tunisia	2	21.50	10.10	175.5	49.7	Hizem *et al*. (2005)
Turkey	145	40.00	28.00	248.3	99.1	Turhan *et al*. (2008a)

Table 3: The absorbed dose rate in air and the annual effective dose from samples of different brand of Ghanaian cement, and the external and internal hazard indices

Sample ID*	Hazard Index H_{EX}	H_{IN}	$D_{\gamma r}$ (nGy/h)	Annual effective dose (mSv/y)
CM I	0.38	0.47	66.35	0.33
CM II	0.27	0.41	45.01	0.22
CM III	0.22	0.30	37.08	0.18
CM IV	0.17	0.24	28.10	0.14
CM V	0.19	0.28	31.48	0.15
AVE±SD	0.25±0.03	0.34±0.05	41.60±6.28	0.21±0.03

*: CM I: Portland limestone cement; CM II: Portland limestone cement; CM III: Ordinary portland cement; CM IV: White cement (Tile adhesive cement cole); CM V: White cement (Tile adhesive cement)

Table 4: Radon mass exhalation rates and radon emanation coefficient from the samples of different brand of Ghanaian cement

Sample ID*	Radon emanation coefficient	Radon mass exhalation rate (μBq/kgs)
CM I	0.62±0.04	44.99±4.54
CM II	0.50±0.02	53.60±3.44
CM III	0.65±0.03	43.86±2.81
CM IV	0.55±0.02	32.53±3.32
CM V	0.53±0.02	37.62±2.40
AVE±SD	0.57±0.03	42.52±3.30

*: CM I: Portland limestone cement; CM II: Portland limestone cement; CM III: Ordinary portland cement; CM IV: White cement (Tile adhesive cement cole); CM V: White cement (Tile adhesive cement)

mass exhalation rate were 0.57±0.03 (range from 0.53 to 0.65) and 42.52±3.30 μBq/kgs (range from 53.60 to 32.53 μBq/kgs), respectively.

The calculated internal (H_{IN}) and external (H_{EX}) hazard indices for cement samples as shown in Table 3 varied from 0.28 to 0.47 with an average of 0.34±0.05 and from 0.17 to 0.38 with an average of 0.25±0.03, respectively. These indices are, less than unity as required by the above-mentioned index formulae, therefore these cements are regarded as safe for construction purposes.

The estimated indoor gamma dose rate values for cement samples are also shown in Table 3. The D_γ values for cement samples range from 31.48 to 66.35 nGy/h with a mean of 41.60±6.28 nGy/h. This mean D_γ value from all cement samples is lower than the world average (populated-weighted) indoor absorbed gamma dose rate of 84nGy/h (UNSCEAR, 2000, 2008).

Table 3 shows calculated Effective dose rate. These values vary from 0.14 to 0.33 mSv/y with an average of 0.21±0.03 mSv/y. The NORM concentration, together with the radon exhalation rate, determines the total chronic (prolonged) radiation dose from building materials to the general public. It is noted that the average effective dose is about 79% less than the dose criterion of 1 mSv/y (IAEA, 1996).

CONCLUSION

Radiological significance of various Ghanaian cement samples has been investigated using gamma spectroscopy technique and mathematical models. The specific activity of ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent activity, the external and internal hazard indices, radon emanation coefficient, radon mass exhalation rate, the absorbed gamma dose rate in indoor air and the corresponding effective dose rate were determined to assess the radiological hazards from Ghanaian cement. The calculated average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the cement samples were 35.94±0.78, 25.44±0.80 and 233.0±3.95 Bq/kg, respectively. The results obtained in this study compares well with data from most countries but also showed some variations with values from other countries. These indicated considerable variations in the activity concentration are due to the varying amounts of uranium, thorium and ^{40}K contents as a result of different geological formations under the earth crust from where the raw material for particular brand of cement was obtained. The mean Ra_{eq} value (90.12±13.52) was determined to be lower than the recommended maximum level of radium equivalent of 370 Bq/kg for building raw materials and products. The estimated mean indoor absorbed dose rate was 50.5% less than the population-weighted average of 84 nGy/h. The corresponding annual effective dose rate was calculated to be 0.21±0.03 mSv/y. The calculated internal and external hazard indices in all the cement samples were less than unity. The average radon emanation coefficient and the radon mass exhalation rate were 0.57±0.03 and 42.52±3.30, respectively. Based on the values of the radiological parameters (radon emanation coefficient, radon mass exhalation rate, effective dose equivalent, absorbed dose rate, radium equivalent activity, the external and internal hazard indices) obtained in this study, all the cement samples considered do not pose any significant radiation hazard and the use of these cement samples in construction of dwellings is considered to be safe for inhabitants.

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