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 to 117 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 µSv y$^{-1}$ and 279 µSv y$^{-1}$ which are just about 2 % and 3 % of the 1.0 mSv y$^{-1}$ 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.

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 (Steiger 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° 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 X 10$^{-5}$ at 1332 keV, 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):

$$A_S \ (Bq \ kg^{-1}) = \frac{C_a}{\epsilon \ P \ M_a} \ ……………\ (1)$$

Where $C_a$ is the net gamma counting rate (counts per second), $\epsilon$ the detector efficiency of the specific γ-ray,
3. Results and Discussion

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

The variation of the mean activity concentration (Bq/kg) of ²²⁶Ra, ²³²Th and ⁴⁰K 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 ²²⁶Ra activity (or ²³⁸U activities for samples assumed to be in radioactive equilibrium) ranges from 23 Bq/kg in (Alarabia) with an average of 33±1.6 Bq/kg. The activity concentration of ²³²Th ranges from 22 Bq/kg (in (Alarabia)) with an average of 30±1.5 Bq/kg. Finally, the activity concentration of ⁴⁰K 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 ²²⁶Ra, ²³²Th and ⁴⁰K in the cement samples are lower than the world average value of 500 Bq kg⁻¹ (UNSCEAR, 1993). The mean activity concentration (Bq/kg) of ²²⁶Ra, ²³²Th and ⁴⁰K are plotted as histograms (1) for different locations from the cement factories.

3.2. Ra equivalent activity (Raₐₑｑ)

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., 2007 and Kilic & Aykamis, 2009). In comparing the radioactivity of materials that contain ²²⁶Ra, ²³²Th and ⁴⁰K a common index termed radium-equivalent activity is required to assess the gamma radiation hazards. Since 98% of the radiological effects of the uranium series are produced by radium radiation hazards. Since 98% of the radiological effects of ²³⁸U and the other ²²⁶Ra precursors is usually ignored, so that the Raₑ𝐪 of a sample can be calculated using the formula proposed by (Beretka & Mathew, 1985):

\[
Ra_{eq} = C_{Ra} + (10/7) C_{Th} + (10/130) C_{K} \quad \text{(2)}
\]

where \(C_{Ra}\), \(C_{Th}\) and \(C_{K}\) are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹. The mean and the range of radium equivalent activity of the total activity of the samples calculated on the basis of the aforementioned relationship are shown in Table (1). On average, Raₑ𝐪 concentration was found to be 117 Bq kg⁻¹ in Aljouf, 104 Bq kg⁻¹ for Alqassim, 92 for Jeddah and 83 Bq kg⁻¹ for Yanbo cement. Upon comparing the results, it is seen that there are no significant variations in Raₑ𝐪 values of different cement samples analyzed. The highest value (117 Bq kg⁻¹) of Raₑ𝐪 was observed in Aljouf cement, while the lowest value (83 Bq kg⁻¹) was found in Yanbo cement. However, all the values obtained here for radium equivalent activity below the internationally accepted value of 370 Bq kg⁻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.

3.3. Representative level index (Iᵣ). Functional level index (Iᵣ) is used to estimate the level of γ-radiation hazard associated with the natural radionuclides in specific building materials, is defined as (NAE-OECD, 1979, El-arabi, 2005):

\[
I_r = C_{Ra}/150 + C_{Th}/100 + C_{K}/1500 \quad \text{(3)}
\]

Where \(C_{Ra}\), \(C_{Th}\) and \(C_{K}\) are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹. The calculated average values of 1, for the samples of cement ranged from 0.59 to 0.88 Bq kg⁻¹ (Table 2). All the 1 values are below the internationally accepted value of 1 Bq kg⁻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 ²³⁵U, ²³²Th and ⁴⁰K, the absorbed dose rate \(D\) 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):

\[
D = \sum x A_x C_x \quad \text{(4)}
\]

where \(A_x\) (Bq kg⁻¹) are the mean activity of ²²⁶Ra, ²³²Th and ⁴⁰K, 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⁻¹ per Bq kg⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K, 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 nGy h⁻¹), while the lowest average value is found in Yanbo (38.6 nGy h⁻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 nGy h⁻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 Sv Gy⁻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)):

\[
\text{Effective dose rate (mSv⁻¹)} = \text{D (nGy h⁻¹)} \times 8760 (\text{h/y}) \times 0.8 \times 0.7 (\text{Sv Gy⁻¹}) \times 10^{-6} \text{..........................(5)}
\]

The world wide average annual effective dose in air is approximately 0.5 mSv⁻¹ and the results for individual countries being generally within the 0.3 – 0.6 mSv⁻¹ range (UNSCEAR, 1988, 2000). Table (2) represents the average annual effective dose rates which they varied from 189 µSv⁻¹ (Yanbo) to 279 µSv⁻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 (Raeq) and the level index (Iₚ) 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 radiation dose rate due to exposure to gamma radiation 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 (Hex) :

\[
\text{H}_{\text{ex}} = (\text{CRa}/740 + \text{CTh}/520 + \text{CK}/9620) \text{..........................(6)}
\]

where CRa, CTh and CK are the activity concentrations in Bq kg⁻¹ of ²²⁶Ra, ²³²Th and ⁴⁰K 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:

\[
\text{H}_{\text{ex}} = (\text{CRa}/740 + \text{CTh}/520 + \text{CK}/9620) \text{..........................(7)}
\]

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

In addition, the radioactivity inert gas radon ²²²Ra, a decay product of ²²⁶Ra 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 (Hin) and defined as(Krieger, 1981, Beretka & Mathew, 1985) :

\[
\text{H}_{\text{in}} = (\text{CRa}/185 + \text{CTh}/259 + \text{CK}/4810) \text{..........................(8)}
\]

where C denotes the respective specific activity in Bq kg⁻¹ for the safe use of a material in the construction of dwelling, Hin should be less than unity. Table (1) shows the calculated values of Hex and Hin for the local cement samples. The average values of Hex ranged from 0.12 to 0.16 and 0.30 to 0.41 for Hin, 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 ²²⁶Ra, ²³²-Th and ⁴⁰K 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>
<thead>
<tr>
<th>Sample</th>
<th>²²⁶Ra (Bq/kg)</th>
<th>²³²Th (Bq/kg)</th>
<th>⁴⁰K (Bq/kg)</th>
<th>Raeq (Bq/kg)</th>
<th>Dose rate (mGy/h)</th>
<th>Hex</th>
<th>Hin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aljouf</td>
<td>35±1.2</td>
<td>31±1.4</td>
<td>505±1.7</td>
<td>117±3.3</td>
<td>56.8±1.5</td>
<td>0.16</td>
<td>0.41</td>
</tr>
<tr>
<td>Yanbo</td>
<td>30±1.3</td>
<td>28±1.1</td>
<td>120±2.1</td>
<td>83±2.9</td>
<td>38.6±1.4</td>
<td>0.12</td>
<td>0.30</td>
</tr>
<tr>
<td>(24–38)</td>
<td>(25–33)</td>
<td>(147–185)</td>
<td>(78–99)</td>
<td>(26–40)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Alqassim</td>
<td>33±1.1</td>
<td>34±1.6</td>
<td>170±1.3</td>
<td>94±3.5</td>
<td>44.7±1.7</td>
<td>0.13</td>
<td>0.34</td>
</tr>
<tr>
<td>Tabuk</td>
<td>29±1.2</td>
<td>26±1.1</td>
<td>434±2.1</td>
<td>100±3.0</td>
<td>48.4±1.4</td>
<td>0.14</td>
<td>0.35</td>
</tr>
<tr>
<td>Jeddah</td>
<td>37±1.5</td>
<td>29±1.3</td>
<td>168±2.1</td>
<td>92±3.6</td>
<td>42.5±1.7</td>
<td>0.13</td>
<td>0.33</td>
</tr>
<tr>
<td>Alarabia</td>
<td>38±1.4</td>
<td>30±1.2</td>
<td>313±2.2</td>
<td>104±3.3</td>
<td>49.1±1.5</td>
<td>0.14</td>
<td>0.38</td>
</tr>
</tbody>
</table>
Table 2. Comparison of radiation hazard indices obtained in this study with measurements from different countries.

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

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

The observed concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K in most of the local cement samples are close or slightly above the corresponding world typical value of 500 Bq kg$^{-1}$. The Ra$_{eq}$ and $I_{r}$ 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 370Bq kg$^{-1}$ for building materials. The annual effective dose is well below the recommended value (1 mSv 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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4. References.


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