Assessment of Natural Radioactivity for Some Secondary Ceilings Samples in Iraq

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Abstract. This research focuses on measuring of specific activity of natural radioactive sources (238U, 232Th and 40K) in some types of secondary ceilings that used as a decorate material and available in Iraq markets. The measurements were done using gamma ray spectroscopy system that based on NaI(Tl) Detector. Also, the radiological hazard indices have been assessed for all samples in present study. The results show that the specific activity vary from 8.7±0.6 to 32.9±2.3 Bq/kg for 238U, 2.9±0.2 to 40.3±1.5 Bq/kg for 232Th, and 117.4±2.6 to 649.1±7.9 Bq/kg for 40K. The latter levels were compared with the world mean values that reported by the UNSCEAR 2008. It was found that all values of 238U and 232Th were below the world wide published values, while the values of only two samples of 40K were above the upper range of the world wide published values. Regarding the average values of radiological hazard risk were found to be within the permissible limit according to the OECD, UNSCEAR 2000, and ICRP. It can be concluded that natural radioactivity levels together with radiological hazard risk studied from the most of the secondary ceilings samples available in local markets of Iraq within natural rates of permissible limits and may not cause any danger to the human when being used.

1. Introduction

Radiological and toxicological contaminants are one the of environmental pollutions that can cause cancerous disease and other changes in the health of humans and the living world in general. This is, therefore, substantially influencing the changes in the environment [1]. The earth and its residents are exposed to radiation emitted from radioactive materials that originally found in the crust of the earth, the sun, and other sources. Radioactivity on the earth can be originated from two types of sources: natural and artificial ones. These sources are currently part of the ecosystem. The natural radioactivity major sources are the nuclides of long half-lives that existed since the formation of the earth and nuclides produced via cosmic rays [2]. The natural radionuclides of concern are mainly 238U, 235U, and 232Th, and their progenies and 40K. Radiation is existed in all parts of our life. It, perhaps, has been found on the earth since it was formed. As a result, life has been developed in an environment that may have a few amounts of ionized radiation [3]. Therefore, one can conclude that radiation is available in everywhere around us (in the atmosphere, the water sources, the buildings and even in the construction materials used in these buildings). Natural materials such as soil, sand, cement and, rock etc., which contain
amounts of natural radioactivity of $^{238}$U, $^{232}$Th and $^{40}$K were used as building materials for construction of buildings and houses [4]. Life development has brought the people to spend most of their time (almost ninety percent of life) in enclosed and confined spaces such as residential buildings (houses and offices) and transportation vehicles. Buildings that are used as shelter should protect us from the environmental conditions (Pollutants, heat, cold and noise). Unfortunately, these shelters do not appear to be safe enough due to the many types of pollutants within them. In this regards, the latter pollutants may have detrimental effects on our health, which in turn be increased dramatically with time. On a daily basis, our body is exposing to many natural sources of radiations that are presented in water, food, and building materials. The motivation of the present study is to assess the specific activity of natural radioactivity in some types of secondary ceilings samples that used in most Iraqi building materials as a decorate materials. Exposure to high levels of radiation could be a reason for the significant damage to humans bodies and can lead to death [5] (WHO, 2016). In this study building materials (secondary ceilings) samples were selected because it is in direct touch with human. There are many previous studies for measuring radioactivity levels in building materials using gamma ray spectroscopy [6-8]. The purpose of this study is to evaluate the natural radioactivity raised from ($^{238}$U, $^{232}$Th and $^{40}$K) in some types of secondary ceilings that available in Iraqi markets using NaI(Tl) detector with dimensions of "3x3". Moreover, ten radiological hazard parameters using different equations were calculated.

2. Materials and Method

2.1. A Collection of Samples
Twenty samples were collected in this study that reflects various types of secondary ceilings. They were taken from the local markets in Al-Najaf governorate, as shown in Tables (1).

Table 1. Information about the secondary ceilings

| No. | Sample name | Sample code | Country of Origin |
|-----|-------------|-------------|-------------------|
| 1   | Saten       | S1          | Germany           |
| 2   | Cardcles    | S2          |                   |
| 3   | Turbo       | S3          | Turkey            |
| 4   | Fuga        | S4          |                   |
| 5   | Akwsatak    | S5          |                   |
| 6   | Gypsum Board| S6          |                   |
| 7   | Rota trofor | S7          |                   |
| 8   | Flogan      | S8          |                   |
| 9   | Summit      | S9          |                   |
| 10  | ABS         | S10         | Iran              |
| 11  | Azran       | S11         |                   |
| 12  | Arcopa      | S12         |                   |
| 13  | Fs.AB       | S13         |                   |
| 14  | MDF- KS     | S14         | China             |
| 15  | CNC - KS    | S15         |                   |
| 16  | Peld        | S16         |                   |
| 17  | Against fire| S17         |                   |
| 18  | Anti-humidity| S18        | Saudi             |
| 19  | Cement board| S19         |                   |
| 20  | Techno      | S20         | Bulgaria          |

2.2. Preparations of Samples
The collected samples were crushed into small pieces first, then they were converted into a fine powder, using electric grinder. The fine powder, after that, was sieved to obtain grain size of about 300μm for
about 750gm in weight using special sieves. Then, the samples were dried at 100 °C for 2 hours using an oven (Model Memmert GmbH+ Co. KG, Germany). Next, the samples were packed in (1L) polyethylene Marinelli beaker of constant volume ((See Figure 1). Then, All samples were stored for about four weeks before the measurement, to allow secular equilibrium between $^{222}$Rn and $^{226}$Ra [9].

![FIGURE 1. Samples in Marinelli beaker](image)

2.3. Measurement of Samples
Gamma-ray spectrum from each samples was recorded using Sodium iodide doped with thallium NaI(Tl) with "3×3" crystal dimensions (See Figure 2). It was processed using the MAESTRO-32 software. NaI(Tl) detector was calibrated by radioactive standard sources (137Cs, 60Co, 22Na, 54Mn, and 152Eu) of known energies and activity. The specific activity for $^{238}$U and $^{232}$Th were determined using gamma-lines 1765 keV ($^{214}$Bi) and the gamma-ray lines 2614 keV ($^{208}$Tl), respectively [9,10]. The specific activity for $^{40}$K was determined directly by its gamma-line of 1460 keV [9,10]. The samples were placed on the detector and measured for a period of 18000 sec.

![FIGURE 2. Block diagram of NaI(Tl) spectrometer system.](image)

2.4. Theoretical calculations
2.4.1. Specific Activity (A)
The specific activities of $^{238}$U, $^{232}$Th, and $^{40}$K ($A_{U}$, $A_{Th}$ and $A_{k}$) radionuclides were calculated using following equation [11,12]:

$$A \left(\frac{Bq}{kg}\right) = \frac{N}{I_{\gamma} \varepsilon MT} \quad \ldots \ldots \quad (1)$$

where, N is net area under photo peak, $I_{\gamma}$ is the probability of gamma decay , $\varepsilon$ is the efficiency of detector, M is the mass of sample, and T is time measured.
2.4.2. External Hazard Index (H_{ex})

The external hazard index was calculated using the following equation [13]:

\[ H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ldots \ldots (2) \]

2.4.3. Internal Hazard Index (H_{in})

The internal hazard index was calculated using the following equation [14]:

\[ H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ldots \ldots (3) \]

2.4.4. Representative Level Index (I_r)

Representative level index was calculated using the following equation [15].

\[ I_r = \left( \frac{1}{150} \right) A_U + \left( \frac{1}{100} \right) A_{Th} + \left( \frac{1}{1500} \right) A_K \ldots \ldots (4) \]

2.4.5. Alpha Index (I_\alpha)

Alpha index was calculated using the following equation [13]:

\[ I_\alpha = \frac{A_U}{200 \left( \frac{Bq}{kg} \right)} \ldots \ldots (5) \]

2.4.6. Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity was calculated using the following equation [16]:

\[ Ra_{eq} \left( \frac{Bq}{kg} \right) = A_U + 1.43 A_{Th} + 0.077 A_K \ldots \ldots (6) \]

2.4.7. Exposure Rate (\dot{X})

The exposure rate was calculated as the following equation [14,17]:

\[ \dot{X} \left( \frac{\mu R}{h} \right) = 1.90 A_U + 2.82 A_{Th} + 0.197 A_K \ldots \ldots (7) \]

2.4.8. Absorbed Dose Rate in Air (D_r)

The absorbed dose rate in air 1 meter was calculated using the following equation [18]:

\[ D_r \left( \frac{nGy}{h} \right) = 0.462 A_U + 0.604 A_{Th} + 0.0417 A_K \ldots \ldots (8) \]

2.4.9. Annual Gonadal Equivalent Dose (AGED)

Annual gonadal equivalent dose was calculated using the following equation [19-21] as:

\[ AGED \left( \frac{mSv}{y} \right) = 3.09 A_U + 4.18 A_{Th} + 0.314 A_K \ldots \ldots (9) \]

2.4.10. Annual Effective Dose Equivalent (AEDE)
Annual effective dose equivalent indoor was calculated using the following equation [22].

\[
AEDE_{\text{indoor}} = \left[ D_r \left( \frac{mSv}{hr} \right) \times 8760 \text{ hr} \times 0.8 \times \frac{0.75\text{Gy}}{\text{Sv}} \right] \times 10^{-6} \ldots \ldots (10)
\]

2.4.11. Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk indoor according to Duration of Life (DL = 70 year) and Risk Factor (RF = 0.05 y/Sv) was calculated using the following equation [11, 16]:

\[
ELCR = AEDE \times DL \times RF \ldots \ldots (11)
\]

3. Results and Discussion

The results of specific activity for $^{238}$U, $^{232}$Th, and $^{40}$K in different types of secondary ceilings samples of the present study were shown in Table (2), all were measured in units of Bq/kg. Examining the results in Table 2, the range of specific activity of $^{238}$U with the standard error was 8.7±0.6 Bq/kg in the sample S16 to 32.9±2.3 Bq/kg in the sample S14, with an average of 22.4±1.6 Bq/kg. Also, the specific activity of $^{232}$Th was ranged between relatively high values in S14, where it reached 40.3±1.5 Bq/kg, and the lowest value in the S4 which was 2.9±0.2 Bq/kg, with an average of 12.2±1.8 Bq/kg. While, the results of specific activity for $^{40}$K were ranged from 117.4±2.6 Bq/kg in sample S4 to 649.1±7.9 Bq/kg in sample S6, with an average of 282.2±26.6 Bq/kg. According to the above results, it was found that the specific activity of $^{238}$U in all samples present study are within the permissible limit set by the UNSCEAR (i.e. 33 Bq/kg) [18], as shown in Figure 3. From figure 4, it was found that the specific activity of $^{232}$Th in all samples in present study were less than the permissible limit set by the UNSCEAR (i.e. 45 Bq/kg) [18]. The present results show that the values of the specific activity of $^{40}$K in all samples were less than the recommended value of (420) Bq/kg, that given by worldwide UNSCEAR 2008 [18], except samples S6 and S12 (see Figure 5). So, It recommend not to use this type of secondary ceiling (S6 and S12). The results of the natural radioactivity in secondary ceilings samples as building materials in the present study were varied, because of the different geological nature of original materials and basic components that made of the samples in the present study. The calculation of ten hazard indices such as (Ra<sub>eq</sub>, H<sub>ex</sub>, H<sub>in</sub>, I<sub>y</sub>, I<sub>r</sub>), and (Exposure, D<sub>r</sub>, AGED, AEDE<sub>indoor</sub>, ELCR) in secondary ceilings samples from Iraq markets are listed in tables 3 and 4, respectively. Form Table 3, it can be seen that the average value of R<sub>aq</sub>, H<sub>ex</sub>, H<sub>in</sub>, I<sub>y</sub>, I<sub>r</sub>) were 61.6±4.9 Bq/kg, 0.166±0.013, 0.227±0.016, 0.459±0.036, and 0.112±0.008, respectively. The highest value of R<sub>aq</sub>, and (H<sub>ex</sub>, H<sub>in</sub>, I<sub>y</sub>, I<sub>r</sub>) were 107.8 Bq/kg, (0.291, 0.380, 0.772, 0.164) in S14 which were less than the recommended value of 370 Bq/kg [23], and [24] (see Figure 6), respectively. Form Table 4, it is found that the average value of Exposure, D<sub>r</sub>, AGED, AEDE<sub>indoor</sub>, and ELCR in secondary ceilings samples were 132.6±10.5µR/h, 29.5±2.3 nGy/h, 208.9±16.3 mSv/y, 0.145±0.011 mSv/y, and (0.500±0.039)×10<sup>−3</sup>, respectively. The highest value of D<sub>r</sub>, and AEDE<sub>indoor</sub> were 48.9 nGy/h, and 0.240 mSv/y in S14 which were less than the recommended value of 55 nGy/h [25], and 0.42 [26], respectively. While, the highest value of AGED was 340.7 mSv/y in S14 which were larger than the average value of 300 Bq/kg [27]. Comparing the results of the specific activity for $^{238}$U, $^{232}$Th and $^{40}$K that shown in the present study sample that produced in different countries, it can be seen that the highest average value of the specific activities for $^{238}$U and $^{232}$Th was in Chinese sample, but for $^{40}$K was in Turkey sample as shown in Figure 7. While, the radiological hazard index (R<sub>aq</sub>, H<sub>ex</sub> and H<sub>in</sub>) were highest values in Chines, as shown in Figure 8. This increase in values may be due to geological nature for the origin of soil that made of secondary ceilings samples. But, the average values for all samples of secondary ceilings of specific activity for ($^{238}$U, $^{232}$Th, and $^{40}$K) and radiological hazard index were within the world's average according to UNSCEAR 2008, UNSCEAR2000, OECD and ICRP.

| No. | Specific activity in Bq/kg |
|-----|--------------------------|
| 238U | 40K | 232Th |
| 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 |

Table 2. Results of specific activity for $^{238}$U, $^{232}$Th, and $^{40}$K in present samples.
| Sample code | Average ± Error | Average ± Error | Average ± Error |
|-------------|-----------------|-----------------|-----------------|
| **U**       |                 |                 |                 |
| 1 S1        | 14.8 ± 0.9      | 6.7 ± 0.4       | 234.0 ± 3.8     |
| 2 S2        | 22.9 ± 1.2      | 9.9 ± 0.5       | 270.4 ± 4.1     |
| 3 S3        | 22.4 ± 1.4      | 15.8 ± 0.7      | 315.6 ± 5.5     |
| 4 S4        | 14.1 ± 0.9      | 2.9 ± 0.2       | 117.4 ± 2.6     |
| 5 S5        | 12.1 ± 0.9      | 11.0 ± 0.5      | 250.3 ± 4.3     |
| 6 S6        | 23.6 ± 1.4      | 22.6 ± 0.9      | 649.1 ± 7.9     |
| 7 S7        | 19.9 ± 1.2      | 18.8 ± 0.7      | 348.5 ± 5.4     |
| 8 S8        | 25.2 ± 1.4      | 7.6 ± 0.5       | 241.9 ± 4.6     |
| 9 S9        | 32.8 ± 1.6      | 8.1 ± 0.5       | 149.7 ± 3.5     |
| 10 S10      | 25.2 ± 1.7      | 7.0 ± 0.4       | 300.0 ± 5.0     |
| 11 S11      | 24.1 ± 1.5      | 9.0 ± 0.5       | 290.0 ± 4.5     |
| 12 S12      | 31.0 ± 2.0      | 11.6 ± 0.8      | 460.9 ± 8.2     |
| 13 S13      | 32.6 ± 2.1      | 13.0 ± 0.8      | 393.6 ± 7.6     |
| 14 S14      | 32.9 ± 2.3      | 40.3 ± 1.5      | 224.8 ± 6.2     |
| 15 S15      | 26.0 ± 1.8      | 12.5 ± 0.8      | 332.8 ± 6.8     |
| 16 S16      | 8.7 ± 0.6       | 3.9 ± 0.3       | 143.3 ± 2.7     |
| 17 S17      | 30.6 ± 1.4      | 13.0 ± 0.6      | 310.5 ± 4.8     |
| 18 S18      | 18.6 ± 1.4      | 16.6 ± 0.8      | 254.5 ± 5.5     |
| 19 S19      | 17.3 ± 1.0      | 5.8 ± 0.3       | 178.4 ± 3.3     |
| 20 S20      | 14.0 ± 0.8      | 7.4 ± 0.3       | 178.9 ± 2.9     |

Minimum: 8.7±0.6, 2.9±0.2, 117.4±2.6
Maximum: 32.9±2.3, 40.3±1.5, 649.1±7.9
Average±S.E: 22.4±1.6, 12.2±1.8, 282.2±26.6

Worldwide [18]: 33, 45, 420
**FIGURE 3.** The comparison of specific activity between present samples with UNSCEAR limit, for $^{238}$U.

**FIGURE 4.** The comparison of specific activity between present samples with UNSCERAR limit, for $^{232}$Th.
FIGURE 5. The comparison of specific activity between present samples with UNSCEAR limit, for \( {}^{40}K \).

Table 3. Results of Ra\(_{eq}\), H\(_{ex}\), H\(_{in}\), \( I_{\gamma} \), and \( I_{\alpha} \)

| No. | Sample code | Ra\(_{eq}\) (Bq/kg) | H\(_{ex}\) | H\(_{in}\) | \( I_{\gamma}\) | \( I_{\alpha}\) |
|-----|-------------|-----------------|-------|-------|--------|--------|
| 1   | S1          | 42.4            | 0.115 | 0.155 | 0.322  | 0.074  |
| 2   | S2          | 57.9            | 0.156 | 0.218 | 0.432  | 0.114  |
| 3   | S3          | 69.3            | 0.187 | 0.248 | 0.518  | 0.112  |
| 4   | S4          | 27.3            | 0.074 | 0.112 | 0.201  | 0.071  |
| 5   | S5          | 47.1            | 0.127 | 0.160 | 0.358  | 0.061  |
| 6   | S6          | 105.9           | 0.286 | 0.350 | 0.816  | 0.118  |
| 7   | S7          | 73.6            | 0.199 | 0.253 | 0.553  | 0.099  |
| 8   | S8          | 54.7            | 0.148 | 0.216 | 0.405  | 0.126  |
| 9   | S9          | 55.9            | 0.151 | 0.240 | 0.399  | 0.164  |
| 10  | S10         | 58.3            | 0.158 | 0.226 | 0.438  | 0.126  |
| 11  | S11         | 59.3            | 0.160 | 0.225 | 0.444  | 0.120  |
| 12  | S12         | 83.1            | 0.224 | 0.308 | 0.630  | 0.155  |
| 13  | S13         | 81.5            | 0.220 | 0.308 | 0.610  | 0.163  |
| 14  | S14         | 107.8           | 0.291 | 0.380 | 0.772  | 0.164  |
| 15  | S15         | 69.5            | 0.188 | 0.258 | 0.520  | 0.13   |
| 16  | S16         | 25.3            | 0.068 | 0.092 | 0.193  | 0.043  |
| 17  | S17         | 73.1            | 0.197 | 0.280 | 0.541  | 0.153  |
| 18  | S18         | 61.9            | 0.167 | 0.218 | 0.460  | 0.093  |
| 19  | S19         | 39.3            | 0.106 | 0.153 | 0.292  | 0.086  |
| 20  | S20         | 38.4            | 0.104 | 0.141 | 0.287  | 0.07   |
|     | Average± S.E. | 61.6±4.9       | 0.166±0.01 | 0.227±0.01 | 0.459±0.03 | 0.112±0.00 |
|     | Worldwide   | <370[23]        | <1[24]    | <1[24]   | <1[24]   | <1[24]   |
Figure 6. The comparison of $R_{\text{eq}}, H_{\text{ex}}, H_{\text{in}}, I_{\gamma}$ and $I_{\alpha}$ between present samples with world limit.

Table 4. Results of $X$, $D_{\alpha}$, AGED, AEDE$_{\text{indoor}}$, and ELCR

| No. | Sample code | Exposure ($\mu$R/h) | $D_{\alpha}$ (nGy/h) | AGED (mSv/y) | AEDE$_{\text{indoor}}$ (mSv/y) | ELCR x $10^{-3}$ |
|-----|-------------|---------------------|---------------------|--------------|--------------------------------|------------------|
| 1   | S1          | 93.1                | 20.6                | 147.2        | 0.101                          | 0.354            |
| 2   | S2          | 124.7               | 27.8                | 197.0        | 0.137                          | 0.478            |
| 3   | S3          | 149.3               | 33.1                | 234.4        | 0.162                          | 0.567            |
| 4   | S4          | 58.1                | 13.2                | 92.6         | 0.065                          | 0.226            |
| 5   | S5          | 103.3               | 22.7                | 162.0        | 0.111                          | 0.389            |
| 6   | S6          | 236.4               | 51.6                | 371.2        | 0.253                          | 0.886            |
| 7   | S7          | 159.5               | 35.1                | 249.5        | 0.172                          | 0.602            |
| 8   | S8          | 117.0               | 26.3                | 185.6        | 0.129                          | 0.452            |
| 9   | S9          | 114.7               | 26.3                | 182.2        | 0.129                          | 0.451            |
| 10  | S10         | 126.7               | 28.4                | 203.1        | 0.139                          | 0.487            |
| 11  | S11         | 128.2               | 28.6                | 203.1        | 0.141                          | 0.492            |
| 12  | S12         | 182.4               | 40.5                | 289.0        | 0.199                          | 0.696            |
| 13  | S13         | 176.1               | 39.3                | 278.7        | 0.193                          | 0.675            |
| 14  | S14         | 220.4               | 48.9                | 340.7        | 0.240                          | 0.840            |
| 15  | S15         | 150.2               | 33.4                | 237.1        | 0.164                          | 0.574            |
| 16  | S16         | 55.8                | 12.4                | 88.2         | 0.061                          | 0.212            |
| 17  | S17         | 156.0               | 34.9                | 246.4        | 0.171                          | 0.600            |
| 18  | S18         | 132.3               | 29.2                | 206.8        | 0.143                          | 0.502            |
| 19  | S19         | 84.4                | 18.9                | 133.7        | 0.093                          | 0.325            |
| 20  | S20         | 82.7                | 18.4                | 130.4        | 0.090                          | 0.316            |
Average ± S.E. | 132.6±10.5 | 29.5±2.3 | 208.9±16.3 | 0.145±0.011 | 0.500±0.03
Worldwide | ----- | 55[25] | ≤ 300 [27] | 0.42 [26] | ----- 

FIGURE 7. The comparison of specific activity in present samples for different countries.
FIGURE 8. The comparison of radiological hazard index in present samples for different countries.

4. Conclusions

Natural radioactivity and radiological hazard index in samples of present study were within the allowed limit according to world limit (UNSCEAR, UNSCEAR, OECD, and ICRP). Therefore, our gamma spectroscopic investigations allow us to confirm that samples of secondary ceilings were safe, except samples S6 and S12.

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