A STUDY OF NATURAL RADIONUCLIDE ACTIVITIES AND RADIATION HAZARD INDEX IN SOME GRAINS CONSUMED IN JORDAN

(Satu Kajian Mengenai Aktiviti Radionuklida Semulajadi dan Indeks Hazard Sinaran Di Dalam Beberapa Bijirin Yang Di makan di Jordan)

A.A. Tawalbeh1*, K.M. Abumurad2, S.B. Samat1, M.S. Yasir1

1 School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
2 Department of Physics, Faculty of Science, Yarmouk University, Irbid, Jordan

*Corresponding author: flower77alaa@yahoo.com

Abstract

Forty samples of different types of imported and locally produced grains consumed in Jordan were analyzed using gamma-ray spectroscopy system with a high Purity germanium (HPGe) detector. The concentrations of the natural radionuclides $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ present in the studied samples were measured, and the radium equivalent activities $\text{Ra}_{\text{eq}}$ were calculated. In addition to that, the hazard index HI, was calculated. The average concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ were in the range of $(2.0 \pm 0.5) \times 10^{-3}$, $(7.0 \pm 2) \times 10^{-3}$ and 49.93 ± 1.69 ppm, respectively. The values of $\text{Ra}_{\text{eq}}$ ranged between 17.70 – 245.64 Bq/kg. The HI were ranged between 0.05 – 0.66, which is less than one (the higher limit of HI). The obtained results were compared with the standard accepted international values, and found to be within the acceptable limits.

Keywords: $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$, NORM, Grains, Activity concentration, Radium equivalent, Hazard index

Introduction

Ionizing Radiation is dangerous to the health, especially the charged particles and the high energy photon [1]. The main natural radioactive sources of ionizing radiation are the long lived $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$. The radiological hazard can be the consequence of external or internal exposure. Radionuclides can enter human body through inhalation and ingestion. The ingested radionuclides could be concentrated in certain parts of the body. For examples, $^{238}\text{U}$ accumulated in human lungs and kidney, $^{232}\text{Th}$ in lungs, liver and skeleton tissues and $^{40}\text{K}$ in muscles. Depositions of large quantities of these radionuclides in particular organs will affect the health condition of the human such as weakening the immune system, induce various types of diseases, and finally increase in mortality rate. Grain and its products, which is the main component of daily serving such as breads, rice, and pasta, are considered as staple food. Generally, Jordan consumes a large amount of grains (789 × 10^3 ton in one year) [2]. Since naturally occurring radioactive materials (NORM) are present in all food commodities, the levels in some grains consumed in Jordan need to be established in order to forecast any possible radiological risk associated...
Tawalbeh et al: A STUDY OF NATURAL RADIONUCLIDE ACTIVITIES AND RADIATION HAZARD INDEX IN SOME GRAINS CONSUMED IN JORDAN

with the consumption of the grains. Thus, the main objectives of this research are to quantify the presence of natural radionuclides in some important grains consumed in Jordan, and to estimate the radium equivalent index ($\text{Ra}_{eq}$) and hazard index (HI).

**Experimental**

**Materials**

Thirteen types of grains were taken as the sample. All samples used were both local and imported grains and were obtained from local markets. The name of the samples and their number are as follows: rice (9), starch (5), bulgur (4), beans (4), peas (1), lentil (4), chickpea (3), corn (3), lupine (2), fava beans (2), wheat (1), sesame seeds (1) and black seeds (1). This makes the total number of samples collected as forty as shown in Table 1.

**Sample Preparation**

The samples were prepared for the natural radioactivity measurement. Each dried sample weighing between (300-500 g) was sealed in 500 ml marlinelli bottles and kept at room temperature (25°C) for at least 30 days before counting in to allow reaching the secular equilibrium of $^{232}$Th and $^{238}$U with their respective decay products, in which the activities of all radionuclide within each series are nearly equal. Each sample was given a code as shown in Table 1.

**Natural radioactivity measurement**

For each grain type, a total of 40 samples were prepared for the natural radioactivity measurement. The measurement was conducted for 24 hours using a Ray HPGe – APTEK counting system. The system was calibrated using $^{57}$Co, $^{212}$Pb, $^{199}$Ir, $^{238}$U, $^{137}$Cs, $^{60}$Co and $^{40}$K, for their known energy (which covers the energy range from 122 keV to 1461 keV) and peak width of gamma-ray emission. The counting efficiency was determined previously for all of its counting geometry. The radionuclides were identified according to their individual photopeak, which are 609 keV ($^{214}$Bi) and 351.9 keV ($^{214}$Pb) for $^{238}$U, 238 keV ($^{212}$Pb), 583.191 and 510.80 keV, ($^{208}$Tl) for $^{232}$Th. And 1460 keV for $^{40}$K. The activity of $^{226}$Ra during the equilibrium was assumed to be the same as its parent, $^{238}$U. The specific activity for each radionuclide was calculated using equation proposed by Jibiri and Ajao, 2005[3]:

$$A_2 = \frac{C_n}{\eta} Y_2 M_2$$  \hspace{1cm} (1)

where $A_2$ is the specific activity of each radionuclide in the grains in Bq/kg, $C_n$ the count rate in cps, $\eta$ is the detection efficiency, $Y_2$ is the number of gammas per disintegration of nuclide for a transition and $M_2$ is the mass of the sample in kg.

Using the equation of $A_{2n} = A_{2a}$ at secular equilibrium, the daughter activity is equal to that of the parent. In terms of the numbers of atoms of the parent ($N_P$) and daughter ($N_D$), secular equilibrium can be also expressed as [1]:

$$N_P \lambda_P = N_D \lambda_D$$  \hspace{1cm} (2)

where $\lambda_P$ are the decay constant of the parent and daughter respectively.

Thus, the concentration of the parent $m$ can be calculated in the sample from the relation [4].:

$$m = \frac{N_P}{N_{Av}} \times M$$  \hspace{1cm} (3)

where $N_{Av}$ is the Avogadro’s number ($6.022 \times 10^{23}$ atom/ mol), $M$ is the mass number of the parent nucleous.

From above two equations 2 and 3, and substitution the values of half- lives we get the concentrations of the radioisotopes in the unit of mg/kg = 1 part per million (ppm), as follows:
\[ m^{(238}\text{U}) = 0.08 \times A^{(238}\text{U}} \]  
\[ m^{(232}\text{Th}) = 0.246 \times A^{(232}\text{Th}} \]  
\[ m^{(40}\text{K}) = 0.038 \times A^{(40}\text{K}} \]

where \( m \) is the concentration of the parent nuclides in the unit of ppm, \( A \) is the specific activity in the unit of Bq/kg. The specific activity of \(^{40}\text{K}\) for example is well documented \[5\].

**Radium equivalent index \((\text{Ra}_{eq})\)**

The radium equivalent activity \((\text{Ra}_{eq})\) concept allows a single index or number to describe the gamma output from different mixtures of \(^{238}\text{U}\) (i.e., \(^{226}\text{Ra}\)), \(^{232}\text{Th}\), and \(^{40}\text{K}\) in a Material \[6\]. \(\text{Ra}_{eq}\) for each sample in Bq/kg, is calculated using the following formula proposed by UNSCEAR \[7\] in Bq/kg.

\[ \text{Ra}_{eq} = A^{(226}\text{Ra}} + 1.43 A^{(232}\text{Th}} + 0.077 A^{(40}\text{K}} \]  

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

**Table 1: List of the collected samples and their codes**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Name</th>
<th>Symbol</th>
<th>Sample</th>
<th>Name</th>
<th>Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>U.S.A Rice(_1)</td>
<td>UsRi(_1)</td>
<td>21</td>
<td>Lebanese Beans</td>
<td>LeBe</td>
</tr>
<tr>
<td>2</td>
<td>U.S.A Rice(_2)</td>
<td>UsRi(_2)</td>
<td>22</td>
<td>Argentina Beans</td>
<td>ArBe</td>
</tr>
<tr>
<td>3</td>
<td>U.S.A Rice(_3)</td>
<td>UsRi(_3)</td>
<td>23</td>
<td>Brazilian Peas</td>
<td>BrPe</td>
</tr>
<tr>
<td>4</td>
<td>U.S.A Rice(_4)</td>
<td>UsRi(_4)</td>
<td>24</td>
<td>Jordanian Lentil</td>
<td>JoLe</td>
</tr>
<tr>
<td>5</td>
<td>Italian Rice(_1)</td>
<td>It Ri</td>
<td>25</td>
<td>Turkish Lentil</td>
<td>TuLe</td>
</tr>
<tr>
<td>6</td>
<td>Australian Rice</td>
<td>AuRi</td>
<td>26</td>
<td>Lebanese Lentil</td>
<td>LeLe</td>
</tr>
<tr>
<td>7</td>
<td>Pakistani Rice</td>
<td>PaRi</td>
<td>27</td>
<td>Canadian Lentil</td>
<td>CaLe</td>
</tr>
<tr>
<td>8</td>
<td>Indian Rice</td>
<td>InRi</td>
<td>28</td>
<td>Jordanian Chickpea</td>
<td>JoCh</td>
</tr>
<tr>
<td>9</td>
<td>Egyptian Rice</td>
<td>EgRi</td>
<td>29</td>
<td>Turkish Chickpea</td>
<td>TuCh</td>
</tr>
<tr>
<td>10</td>
<td>Jordanian Starch</td>
<td>JoSt</td>
<td>30</td>
<td>Lebanese Chickpea</td>
<td>LeCh</td>
</tr>
<tr>
<td>11</td>
<td>Qatari Starch</td>
<td>QaSt</td>
<td>31</td>
<td>Jordanian Corn</td>
<td>JoCo</td>
</tr>
<tr>
<td>12</td>
<td>Lebanese Starch</td>
<td>LeSt</td>
<td>32</td>
<td>Lebanese Corn</td>
<td>LeCo</td>
</tr>
<tr>
<td>13</td>
<td>Turkish Starch</td>
<td>TuSt</td>
<td>33</td>
<td>Argentina Corn</td>
<td>ArCo</td>
</tr>
<tr>
<td>14</td>
<td>Italian Starch</td>
<td>ItSt</td>
<td>34</td>
<td>Jordanian Lupine</td>
<td>JoLu</td>
</tr>
<tr>
<td>15</td>
<td>Jor/ Bulgur/ White</td>
<td>JoBu(_1)</td>
<td>35</td>
<td>Turkish Lupine</td>
<td>TuLu</td>
</tr>
<tr>
<td>16</td>
<td>Jor/ Bulgur/ Red</td>
<td>JoBu(_2)</td>
<td>36</td>
<td>Jor/Fava beans</td>
<td>JoFa</td>
</tr>
<tr>
<td>17</td>
<td>Leb/ Bulgur/ White</td>
<td>LeBu(_1)</td>
<td>37</td>
<td>Syrian Fava beans</td>
<td>SyFa</td>
</tr>
<tr>
<td>18</td>
<td>Leb/ Bulgur/ Red</td>
<td>LeBu(_2)</td>
<td>38</td>
<td>Jordanian Wheat</td>
<td>JoWh</td>
</tr>
<tr>
<td>19</td>
<td>Jordanian Beans</td>
<td>JoBe</td>
<td>39</td>
<td>Jor/Sesame Seeds</td>
<td>JoSe</td>
</tr>
<tr>
<td>20</td>
<td>Brazilian Beans</td>
<td>BrBe</td>
<td>40</td>
<td>Jor/ Black Seeds</td>
<td>JoBl</td>
</tr>
</tbody>
</table>
Hazard index
This index is used to estimate the level of γ-radiation hazard associated with the natural radionuclides in grains samples. The hazard index (HI) is calculated from the equation [8]:

\[ HI = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810} \leq 1. \]  

Results and Discussion
The activity concentration of \(^{238}\text{U}\), \(^{226}\text{Ra}\), \(^{232}\text{Th}\) and \(^{40}\text{K}\) in Bq/kg present in the samples are given in Fig. 1, Fig. 2 and Fig. 3 respectively. In Fig. 4, Fig. 5 and Fig. 6 the concentration of the parent nuclides of the natural radioactive series for each sample had been calculated in the unit of ppm. The average concentrations of \(^{238}\text{U}\), \(^{232}\text{Th}\) and \(^{40}\text{K}\) were in the range of \((2.0 \pm 0.5) \times 10^{-3}\), \((7.0 \pm 2) \times 10^{-3}\) and \(49.93 \pm 1.69\) ppm, respectively. The concentration of \(^{238}\text{U}\) ranged from 0.0033± 0.0006 to 0.0012± 0.0002 ppm. In general the measured concentration values of \(^{238}\text{U}\) lies within the acceptable values \(8 \times 10^{-5} - 3.3 \times 10^{-2}\) ppm as was reported in [9] except in Jordanian sesame seed sample in which the concentration of \(^{238}\text{U}\) was found to be 0.0033±0.0006 ppm, which is the highest permissible limit allowed. The highest concentration of \(^{232}\text{Th}\) (0.010±0.002) ppm was found in JoBe sample, while the lowest concentration (0.002±0.001) ppm was found in ArBe sample. A result of the \(^{232}\text{Th}\) concentration measurements in all samples lies within the acceptable value, which should be less than 0.02 ppm for safe use[9]. The maximum value of \(^{40}\text{K}\) concentration was found in ArBe with a value of 123.57±2.56 ppm. The lowest concentration was found in ItRi with a value of 8.90±0.89 ppm. \(^{40}\text{K}\) concentration is comparable with the results of other researchers and lies within the acceptable value, which is less than 133 ppm for safe consumption.[7].

Figure 1: \(^{238}\text{U}\) mean specific activity of the studied samples in the units of Bq/kg.
Figure 2: $^{238}$Th mean specific activity of the studied samples in the units of Bq/kg.

Figure 3: $^{40}$K mean specific activity of the studied samples in the units of Bq/kg.
Figure 4: $^{238}\text{U}$ concentration of the studied samples in the unit of ppm.

Figure 5: $^{238}\text{Th}$ concentration of the studied samples in the unit of ppm.
Figure 6: $^{40}$K concentration of the studied samples in the unit of ppm

Figure 7: The radium equivalent ($Ra_{eq}$) of the studied samples.
Radium equivalent index ($Ra_{eq}$)

Fig. 7 illustrates the calculated values of $Ra_{eq}$ corresponding to sample number, the values of $Ra_{eq}$ ranged between 17.70 and 245.64 Bq/kg. Of all the 40 samples measured in this study, Argentinian beans and all the other samples of beans, Jordanian, Lebanese and Brazilian kinds appeared to have the highest concentrations of $Ra_{eq}$ 245.64, 241.79, 235.90 and 234.75 Bq/kg respectively. Whereas Italian, Indian and Egyptian rice showed the lowest concentration 17.70, 19.64 and 22.17 Bq/kg respectively. The average radioactivity concentration of $Ra_{eq}$ was found to be 99.26 Bq/kg, it is clear that the values of $Ra_{eq}$ in all samples were much less than the safe value 370 Bq/kg [8].

Hazard index (HI)

As shown in Fig. 8, the HI ranged between 0.05 and 0.66. It is obviously clear that the values of HI were much less than the safe value, which should be less than one [8].

![Safe limit line HI ≤ 1](image)

Figure 8: The hazard index (HI) of the studied samples.

Conclusion

The results have shown that the concentration of natural radionuclides of $^{238}$U, $^{232}$Th, and $^{40}$K in grain samples studied are relatively lower than the world recommended limits. Except Jordanian sesame seed sample the concentration of $^{238}$U was found to be 0.0033±0.0006 ppm, which is the highest permissible limit allowed. The calculated Radium equivalent and hazard index (HI) are also not exceeding the maximum suggested values. Thus, the grains in Jordan are not supposed to acquire any radiological complication.

Acknowledgement

This project is funded by Yarmouk University. This project also used various facilities provided by Hashemite University, Jordan. The author would like thank Dr. Jamal Al-Jundi, Ms. Nadia AL-Ahmad and Ms Ghada Assayed for their help in preparation and analysis of the samples.
References