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Evaluation of radionuclides contamination in wheat flour and bread using gamma-ray spectrometry

Fatimh Alshahri

Department of Physics, University of Dammam, Dammam 1982-31441, Saudi Arabia

Abstract: Because of the increasing of cancer incidence rates, this study was carried out to evaluate the activity concentration of radionuclides in the most common food consumed in Saudi Arabia (wheat flour and bread) and to estimate their radiological impact in long-term. For this purpose, the activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in wheat flour and bread samples were measured using gamma-ray spectrometry. The result showed that the mean values of the activity concentrations in brown bread were higher than those in wheat flour and white bread. A decreasing trend of the mean values of their specific activities has been observed in the order: brown bread white bread wheat flour. The highest concentrations of ^{232}Th were found in brown bread that contained more bran and other grains. However, these values were lower than the acceptable limit. Furthermore, the radium equivalent activity (Ra_{eq}), absorbed dose rate in air (D), annual effective dose rate (E) and the internal hazard indices (H_{in}) were calculated. The radiation hazard indexes for all samples were lower than the acceptable values. The data were compared with those given in the literature.
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Keywords: ^{226}Ra, ^{232}Th, ^{40}K, wheat flour, bread, gamma-ray spectrometry

1. Introduction

Natural radioactivity is widespread throughout the earth's environment and it exists in various geological formations in soil, rocks, plants, water and air (**Yang *et al.,* 2005, Al-Hamidawi, 2015, Alsaffar *et al.,* 2015, Ahmad *et al.*, 2015**) The radionuclides concentrations in soil and plants are higher in cultivated lands due to the effect of phosphate fertilizers which contain high concentration of radionuclides (**Amaral *et al*,. 2005, Ahmed and El-Arabi, 2005, Fawzia, 2007, Lambert *et al.*, 2007, Alshahri and Alqahtani, 2015**). Radionuclides transfer through the environment by various pathways, for example, through the atmosphere, aquatic systems and soil sub-compartments; each of these pathways contribute to human exposure. A large fraction of radiation exposure occurs as a result of ingestion of foodstuffs produced in the natural environment due to the emission of gamma rays and the inhalation of radon and its daughters which can pose serious health hazards (**Hosseini *et al.*, 2006, Asaduzzaman *et al.*, 2014, Kant *et al.*, 2015**).

The most important terrestrial sources of natural radiation are the long-lived ^{40}K and the ^{238}U and ^{232}Th decay series. These radionuclides are mainly responsible for internal exposure, through ingestion of food and water and through inhalation of air particulates (**Amaral *et al.*, 2005**). The ^{238}U series decays via a chain containing eight alpha decays and six beta decays to ^{206}Pb. This chain includes radon gas, which is produced from the decay series of ^{238}U by the alpha decay of ^{226}Ra. Radon is an inert, colorless, odorless and tasteless gas with a half- life of 3.825 d. Radon is a cause of lung cancer when inhaled (**Akbari *et al.*, 2013, Alshahri and Alqahtani, 2015**). Thorium accumulates in human lungs, liver and skeleton tissues, uranium accumulates in lungs and kidneys and potassium accumulates in muscles. Depositions of large quantities of these radionuclides in particular organs produce radiation damage and biochemical and morphological changes (**Akhter *et al.*, 2007, Adeniji *et al.*, 2013**). Moreover, cesium-137, which can pass to humans through the food chain, is one of the most important radionuclides among man-made radionuclides due to the fact that it has similar chemical properties of potassium, has a long physical half-life (30.2 years) and emits beta particles and gamma rays (**Kilic *et al.*, 2009**). A radioactive isotope of cesium, ^{137}Cs, is found in the fallout from the detonation of nuclear weapons and the waste from nuclear power plants. ^{137}Cs is one of the most common radioisotopes used in industry. It is used in various measuring devices, such as moisture-density gauges (**Abd El Wahab and Morsy, 2006**). Contamination with cesium-137 can cause serious illness or death, depending upon the dose, and has been associated with the development of cancer long after exposure (**Strandberg, 2004, Abd El Wahab and Morsy, 2006, Lavi *et al.*, 2006**).

Cancer is a leading cause of disease worldwide. An estimated 14.1 million new cancer cases occurred in 2012. Lung, female breast, colorectal and stomach cancers accounted for more than 40% of all cases diagnosed worldwide (**WHO, 2014**). In Saudi Arabia, incidence rates of cancer are steadily increasing. There are more than 12,000 new cancer cases per year in the Kingdom of Saudi Arabia (KSA), with an incidence rate of 52.3 per 100,000 and the overall age-standardized incidence is 82.1 per 100,000 populations. Most of these cases present with advanced stages of the disease (**Ministry of Health, 2012**).

Wheat flour is an essential commodity to human existence through the centuries and is currently the most widely consumed staple food and cultivated in different regions of the world. Most breads are made with wheat flour; some breads contain other grains or more bran. The intake of radionuclides, due to breads consumption, is the largest contributor of radiation doses received by the human body. Therefore, it is important to establish databases of the concentration of long-lived radionuclides in wheat and its products which are the most popular food, to ensure that the radiation levels are within the specified safety limits. These databases can be useful as baseline values to estimate the radiation hazard indices from wheat flour and bread among various brand names in Saudi Arabia markets.

2. Material and Methods

2.1 Sample preparation

Thirty samples of the most available types of flour and wheat breads among various brand names were collected from the local markets in Saudi Arabia. To remove moisture, the bread samples were dried in an electric oven at 100^{o} C for 24 hours. After drying, the bread samples were crushed into a fine powder to pass through a 2 mm mesh sieve. For radiation measurements, each sample was packed into 152 ml standard size beakers and tightly sealed and stored for 28 days to reach equilibrium. Two reference materials were packed into the same standard size beakers for efficiency calibration.

2.2 Experimental setup

A Hyper pure germanium detector (HPGe), coaxial type, P-type with a relative efficiency of 20% was used. The detector was shielded with a low-level background lead shield. The HPGe was calibrated for efficiency using the reference material RGU-1 from IAEA. The certified activity of uranium is 400 ppm which refers to 4960 Bq kg^{-1}. The energy transitions of the ^{226}Ra daughters (^{214}Pb and ^{214}Bi) were used to develop the efficiency calibration curve. A fourth-degree polynomial fitting was performed to achieve the best R^2 value (≈ 0.97).

After subtracting the background, the radionuclides were measured at the gamma lines as given in Table (**1**). ^{226}Ra was measured using its progenies ^{214}Pb with energies of 295.2 keV (19.3%) and 351.93 keV (37.6%), and ^{214}Bi with energies of 609.31 keV (46.1%), 1120.29 keV (15.1%) and 1764.49 keV (15.4%). Radium was determined based on the above mentioned energy transitions after achieving secular equilibrium for 28 days after sample packing. For ^{232}Th, the specific activity concentration was determined using the gamma lines of 338.40 keV (12.4%) and 911.07 keV (25.8%) for ^{228}Ac and the gamma lines of 583.14 keV (84.5%) for ^{208}Tl. In the case of ^{40}K and ^{137}Cs, the specific activity concentrations were estimated directly by their gamma lines of 1460.75 keV (10.7%) and 661.7 keV (85.12%), respectively.

The software used for analysis and reduction of the gamma-ray spectra was **Quantum Gold**, Version 4.04.00.

The Minimum detectable activity (MDA) for each radionuclide (Ra, Th and K) in the background was calculated separately based on the sample's weight using the detection limit according to the formula (**Currie, 1986**):

$$MDA\ (counts) = \frac{2.7+4.65\sqrt{BG}}{\varepsilon\, I_{\gamma}\, t} \qquad (1)$$

where BG is the background count below the peak of interest, is the absolute efficiency, I is the gamma line intensity and t is the counting time in second. The MDAs for ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were 8.6, 5.6, 52 and 0.22 Bq kg^{-1}, respectively.

To assess the radiological hazard, it is useful to calculate an index called the radium equivalent activity, Ra_{eq}, which can be calculated from the following relation (**Boukhenfouf and Boucenna, 2011, Alshahri and Alqahtani, 2015**):

$$Ra_{eq}\ (Bq\ kg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \qquad (2)$$

where A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively, expressed in Bq kg^{-1}.

The absorbed dose rate in air 1 m above the ground surface for the radionuclides (^{232}Th, ^{226}Ra, and ^{40}K) was computed on the basis of guidelines provided by Ahmed and El-Arabi, **2015**. The conversion factors used to compute the absorbed dose rates (D) in air per unit activity concentration in 1 Bq kg^{-1} sand correspond to 0.606 nGy h^{-1} for ^{232}Th, 0.429 nGy h^{-1} for ^{226}Ra, and 0.0417 nGy h^{-1} for ^{40}K. Therefore, D could be obtained from the following relation:

$$D\ (nGy\ h^{-1}) = 0.429\ A_{Ra} + 0.606\ A_{Th} + 0.0417\ A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively.

The annual effective dose rate E (mSv y^{-1}) received by the population is calculated using the following equation (**UNSCEAR, 2000**):

$$E\ (mSv\ y^{-1}) = D\ (nGy\ h^{-1}) \times 8760\ (h\ y^{-1}) \times 0.2 \times 0.7\ (Sv\ Gy^{-1}) \times 10^{-6} \qquad (4)$$

where D (nGy h^{-1}) is the absorbed dose rate in air, 8760 h is the time for one year, 0.7 (Sv Gy^{-1}) is the conversion factor, which converts the absorbed dose rate in air to human effective dose and 0.2 is the outdoor occupancy factor (**UNSCEAR, 2000**).

Another radiation hazard index is called Internal Hazard Index (H_{in}) (**Nasim *et al.*, 2012, Ahmad *et al.*, 2015**). This index value must be less than unity and is defined as follow:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (5)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively.

Table 1. Gamma rays and their related isotopes used to calculate the activity concentrations of the nuclides in the first column (Mansour *et al.*, 2012,. Boukhenfouf and Boucenna, 2011)

Nuclide	Half life (yr)	Gamma ray energy (keV)	Isotope	Intensity (%)
^{226}Ra	1650	295.2	^{214}Pb	19.3
		351.93	^{214}Pb	37.6
		609.31	^{214}Bi	46.1
		1120.29	^{214}Bi	15.1
		1764.49	^{214}Bi	15.4
^{232}Th	1.405×10^{10}	338.40	^{228}Ac	12.4
		911.07	^{228}Ac	29.0
		583	^{208}Tl	84.5
^{40}K	1.277×10^{9}	1460.83	---	10.7
^{137}Cs	30.1	661.7	---	85.12

3. Results and discussion

3.1. Radionuclides in wheat flour and bread

The specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in all wheat flour and bread samples were measured. The results are given in Table **(2)**. The specific activities of ^{226}Ra ranged from 11.5 3.7 to 34.6 4.1 Bq kg^{-1} with a mean value of 22.7 3.2 Bq kg^{-1} for wheat flour samples, from 14.9 3.5 to 31.3 3.7 Bq kg^{-1} with a mean value of 23.3 3.1 Bq kg^{-1} for white bread samples and from 8.67 1.0 to 37.3 4.4 Bq kg^{-1} with a mean value of 23.8 2.7 Bq kg^{-1} for brown bread samples. The specific activities of ^{232}Th ranged from 8.56 1.7 to 28.3 3.3 Bq kg^{-1} with a mean value of 16.6 2.5 Bq kg^{-1}, from MDA value to 26 3.1 Bq kg^{-1} with a mean value of 16.4 2.3 Bq kg^{-1} and from 6.9 0.8 to 43.1 5.1 Bq kg^{-1} with a mean value of 19.9 2.3 Bq kg^{-1} for wheat flour, white bread and brown bread, respectively. The highest activity concentration of radium was 37.3 4.4 Bq kg^{-1} in brown bread for sample B2 and the highest activity concentration of thorium was 43.1 5.1 Bq kg^{-1} in brown bread for sample B7 which were within the range of the acceptable values (**UNSCEAR, 2000**). From Table **(2),** the activity concentrations of radium and thorium in all samples were within or lower than the range of the acceptable values (**UNSCEAR, 2000**)˙ The data show that the mean values of radium and thorium in brown bread were higher than the mean values in wheat flour and white bread whereas the radioisotopes of ^{40}K and ^{137}Cs were present in low concentrations in all samples.

Comparison of radium, thorium and potassium activities in all samples are given in Figures **(1), (2)** and **(3).** These figures compare between UNSCEAR values and the values of this study.

The variations of activity concentrations in all samples may be due to the different amount of radionuclides found in the soil which can be absorbed by wheat plants (**Amaral *et al.*, 2005, El- Taher and Makhluf, 2010**). The radionuclides in soil can be transferred from soil to plants via the root system and no differentiation was observed between the absorption of chemically analogous isotopes via the root system (**Vandenhove *et al.*, 2009, Asaduzzaman *et al.*, 2014**).

3.2. Activity concentration of ^{232}Th in white and brown bread

The activity concentration of thorium in most of the brown bread samples were higher than the values in white bread samples which may due to the contents of the brown bread as shown in Figure **(4).** Brown bread is made from whole grain wheat which contains bran. Bran is an integral part of whole grains and represent the hard outer layer of cereal grains. Bran is particularly rich in dietary fiber and contains significant quantities of minerals (**Nike *et al.*, 2005**). This result can be observed in samples B4 and B7, which contain more bran and other grains (e.g., linseed and millet).

3.3. Radiation hazard indexes

The radium equivalent activity (Ra_{eq}), total absorbed dose rate in air 1 m above the ground (D), annual effective dose (E) and internal hazard index (H_{in}) were calculated for all samples under investigation. The calculated values are presented in Table **(3).** The radium equivalent activity varied from 49.2 6.8 to 89.5 9.8 Bq kg^{-1} with a mean value of 66.1 6.7 Bq kg^{-1} for wheat flour samples, from 51.3 5.5 to 85.3 9.2 Bq kg^{-1} with a mean value of 65.2 8.1 Bq kg^{-1} for white bread samples and from 59.3 6.2 to 103 11 Bq kg^{-1} with a mean value of 77.7 7.7 Bq kg^{-1} for brown bread. The radium equivalent activities for all samples were lower than the acceptable value of 370 Bq kg^{-1} (**UNSCEAR 2000**) as shown in Figures **(1), (2)** and **(3).** Figure **(5)** shows a good correlation between Ra_{eq} and ^{232}Th in wheat flour, white bread and brown bread samples. The values of correlation coefficient are R^2 = 0.702, R^2 = 0.725 and R^2 = 0.770 for wheat flour, white bread and brown bread samples, respectively.

Table 2. Activity concentration in Bq kg^{-1} of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs for wheat flour and bread samples.

Type of sample	Sample code	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Wheat flour	F1	17.8 2.1	12.7 1.5	200 15	< MDA
	F2	12.7 1.5	14.6 2.9	203 15	3.13 0.5
	F3	23.4 2.8	14.4 2.8	312 16	< MDA
	F4	11.5 3.7	14.8 1.7	279 21	< MDA
	F5	29.2 3.4	17.4 2.0	379 19	< MDA
	F6	34.6 4.1	8.56 1.7	200 16	< MDA
	F7	21.4 2.5	13.3 1.5	256 17	2.73 0.5
	F8	30.7 3.6	28.3 3.3	238 19	2.33 0.4
	F9	23.8 4.7	19.2 5.1	267 21	2.73 0.5
	F10	21.5 2.5	22.4 2.6	242 19	< MDA
Mean		22.7 3.2	16.6 2.5	258 18	2.73 0.5
White bread	W1	17.4 2.1	18.9 2.2	203 26	< MDA
	W2	24.6 2.9	13.2 1.5	232 18	0.97 0.2
	W3	24.7 2.9	18.9 2.2	203 26	0.24 0.05
	W4	23.5 2.8	22.8 4.6	217 23	2.26 0.6
	W5	14.9 3.5	21.3 2.5	251 21	2.74 0.8
	W6	20.0 3.6	10.6 3.2	242 24	0.49 0.1
	W7	25.5 3.2	19.6 2.2	266 21	2.11 0.6
	W8	31.3 3.7	< MDA	260 23	6.15 1.8
	W9	25.3 3.0	26.0 3.1	297 23	< MDA
	W10	25.7 3.1	12.5 1.5	231 23	0.57 0.1
Mean		23.3 3.1	16.4 2.3	240 22	1.93 0.5
Brown bread	B1	32.8 3.9	13.2 1.5	349 24	1.42 0.4
	B2	37.3 4.4	13.8 1.6	347 26	1.86 0.5
	B3	30.3 3.6	6.9 0.8	248 19	< MDA
	B4	33.1 3.9	24.8 2.9	359 28	2.91 0.6
	B5	18.3 1.6	17.7 2.1	342 27	0.49 0.1
	B6	17.5 1.5	16.1 2.1	432 20	< MDA
	B7	22.6 2.7	43.1 5.1	241 19	3.39 0.6
	B8	16.8 2.0	23.3 2.7	310 17	< MDA
	B9	8.67 1.0	18.7 2.2	359 28	< MDA
	B10	20.9 2.5	21.7 2.3	302 16	< MDA
Mean		23.8 2.7	19.9 2.3	328 22	2.01 0.2

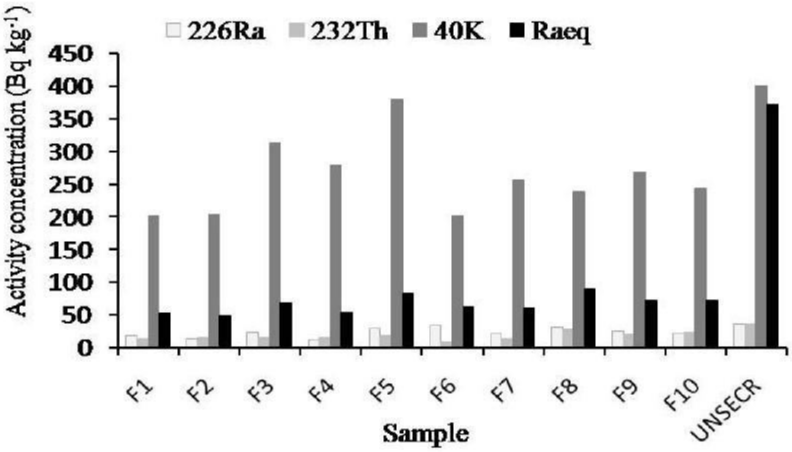

Figure 1. Comparison of ^{226}Ra, ^{232}Th, ^{40}K and Ra$_{eq}$ activities in Wheat flour samples with the allowed values by UNSCER, 2000.

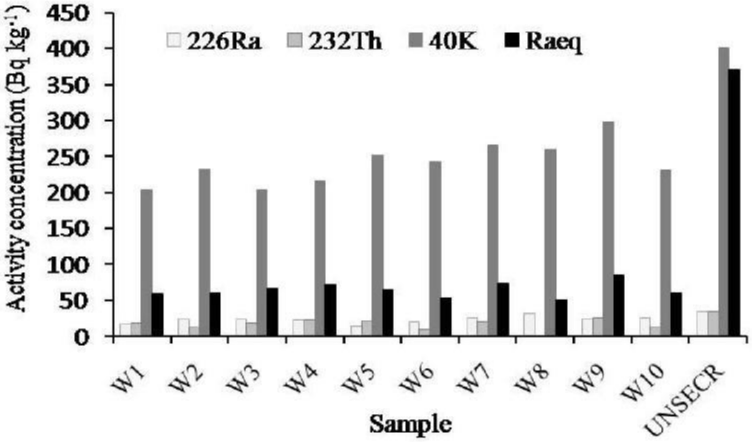

Figure 2. Comparison of ^{226}Ra, ^{232}Th, ^{40}K and Ra$_{eq}$ activities in white bread samples with the allowed values by UNSCER, 2000.

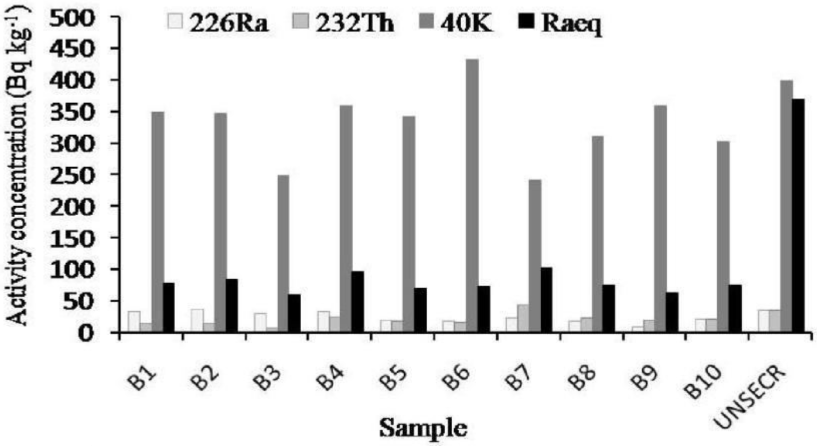

Figure 3. Comparison of ^{226}Ra, ^{232}Th, ^{40}K and Ra$_{eq}$ activities in brown bread samples with the allowed values by UNSCER, 2000.

Table 3. Radium equivalent activity (Bq kg^{-1}), absorbed gamma radiation dose rate in air (nGy h^{-1}), annual effective dose (mSv y^{-1}) and internal radiation hazard index (H_{in}) for sand samples and sediment.

Type of sample	Sample code	Ra_{eq}	D (nGy h^{-1})	E (mSv y^{-1})	H_{in}
Wheat flour	F1	51.4 5.4	16.1	0.02	0.19
	F2	49.2 6.8	22.8	0.03	0.17
	F3	68 8.0	31.8	0.04	0.25
	F4	53.8 8.5	25.5	0.03	0.18
	F5	83.3 7.7	23.1	0.03	0.30
	F6	62.2 7.7	28.4	0.03	0.26
	F7	60.1 5.9	27.9	0.03	0.22
	F8	89.5 9.8	40.3	0.05	0.33
	F9	71.8 13	33.1	0.04	0.27
	F10	71.7 7.6	32.9	0.04	0.25
Mean		66.1 6.7	30.6	0.04	0.24
White bread	W1	60.1 7.2	27.4	0.03	0.21
	W2	61.3 6.4	28.3	0.03	0.23
	W3	67.4 8.0	30.6	0.04	0.25
	W4	72.8 11	33.0	0.04	0.26
	W5	64.7 8.8	29.8	0.04	0.21
	W6	53.8 10	25.2	0.03	0.20
	W7	74 7.9	33.9	0.04	0.27
	W8	51.3 5.5	24.3	0.03	0.22
	W9	85.3 9.2	39.1	0.05	0.30
	W10	61.4 7.0	28.3	0.03	0.24
Mean		65.2 8.1	30.0	0.04	0.23
Brown bread	B1	78.5 7.9	36.7	0.05	0.30
	B2	83.8 8.7	38.9	0.05	0.33
	B3	59.3 6.2	27.6	0.03	0.24
	B4	96.2 10	44.3	0.05	0.35
	B5	69.9 6.7	32.9	0.04	0.24
	B6	73.7 6.0	35.4	0.04	0.25
	B7	103 11	45.9	0.06	0.34
	B8	74 7.2	34.3	0.04	0.25
	B9	63 6.3	30.1	0.04	0.19
	B10	75.2 7.0	34.8	0.04	0.26
Mean		77.7 7.7	36.0	0.04	0.28

The gamma adsorbed dose rate (D) in air and annual effective dose (E) for all samples ranged between 16.1-44.3 nGy h^{-1} and between 0.02-0.06, respectively. These results were within the estimated average global terrestrial radiation of 55 nGy h^{-1} and the acceptable value of annual effective dose (1 mSv y^{-1}) for the public (**UNSCEAR 2000**).

The internal hazard index ranged between 0.17 and 0.32 with a mean value of 0.24 for wheat flour, between 0.20 and 0.30 with a mean value of 0.23 for white bread and between 0.19 and 0.35 with a mean value of 0.28 for brown bread. From Figure **(6),** the data show a good correlation between radium equivalent activities and the internal hazard indices in all samples. However, the values of H_{in} in all samples are lower than unity.

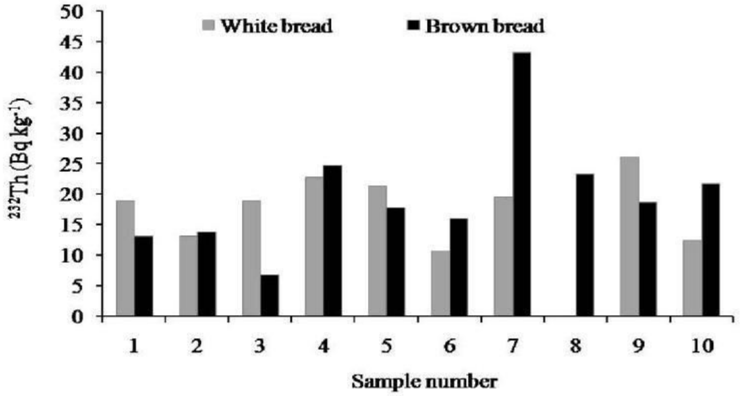

Figure 4. Activity concentrations of ^{232}Th in white and brown bread.

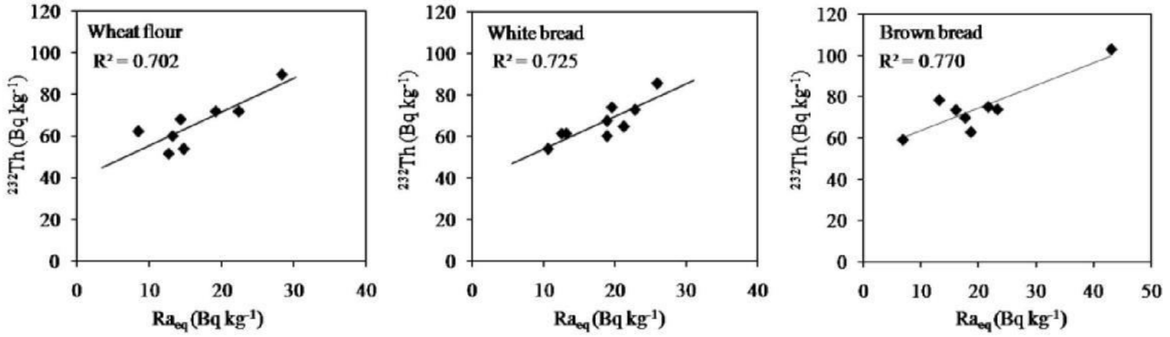

Figure 5. Relationship between Ra_{eq} and ^{232}Th in wheat flour, white bread and brown bread.

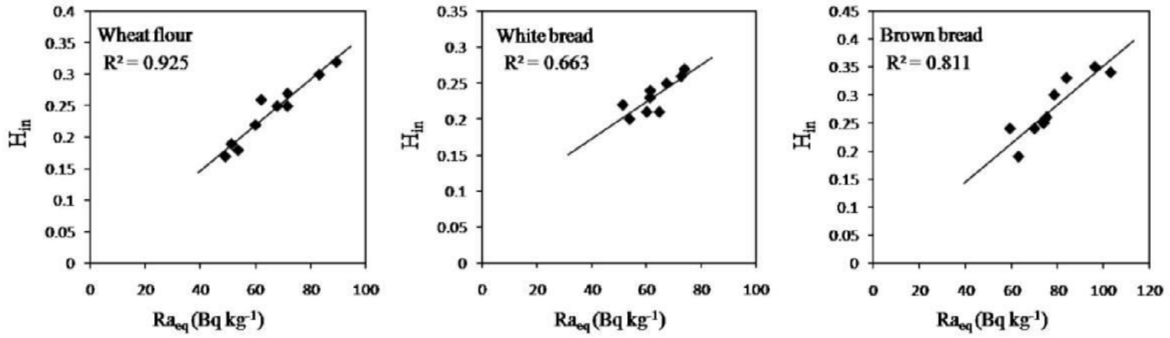

Figure 6. Relationship between Ra_{eq} and H_{in}.

3.4. Comparison of Activity Concentrations with Similar Studies

The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs for the present study were compared with the similar investigations of available studies (**Hoshi *et al.*, 1994, Santos *et al.*, 2002, Jeambrun *et al.*, 2012, Kimura *et al.*, 2012, Alharbi and El-Taher, 2013, Abid Abojassim *et al.*, 2014**). The obtained values for ^{226}Ra, ^{232}Th and ^{40}K concentrations were higher than the reported values from other studies whereas the activity concentrations of ^{137}Cs were within the range of values of the reported data in the literature. The variations in the activity concentrations of radionuclides in wheat flour and bread for this study and other studies may be due to the local geology of the different countries and the effect of phosphate fertilizers on cultivated land. Moreover, the food Additives may contribute to increase the concentration of radionuclides in bread.

Conclusion

The activity concentrations of ^{222}Ra, ^{232}Th, ^{40}K and ^{137}Cs in wheat flour and bread were evaluated using gamma-ray spectrometry. These data show that the mean values of ^{222}Ra, ^{232}Th, ^{40}K and ^{137}Cs were lower than the allowed limits for all samples. Thorium concentrations in most of the brown bread samples were higher than the values in white bread samples. This result can be observed clearly in sample B7 which contain more bran and other grains. The radium equivalent activities and the internal hazard indices were calculated to assess the radiological hazards from the consumption of wheat flour and bread. All of the calculated values were lower than the recommended level. Thus, the accumulation of radionuclides in wheat flour and bread samples under investigation do not pose any health risks. However, the obtained data emphasize the need for more studies on radionuclides in other foodstuffs to establish a baseline for radiation exposure and its impact on human health.

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Corresponding Author:
Dr. Fatimh Alshahri
Department of Physics, University of
Dammam Dammam 1982-31441, Saudi Arabia
E-mail: faalshehri@uod.edu.sa

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