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RESEARCH ARTICLE

NATURAL RADIOACTIVITY AND HEAVY METALS MEASUREMENT IN RICE AND FLOUR CONSUMED BY THE INHABITANTS IN K.S.A

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ABSTRACT

The natural radionuclide's ^{226}Ra, ^{232}Th and ^{40}K and some heavy metals (Fe, Cd, Zn, Cu, Mn, Ni, and Pb) were measured in the rice and the flour samples consumed in Saudi Arabia. Gamma ray spectrometry was used to determine the activity concentration of the three nuclides. Heavy metals were analyzed by an inductively coupled plasma optical emission spectrophotometer (ICP-OES). The results indicate that the average concentration of 226Ra, 228Th, and 40K in the rice samples were 1.08, 1.19 and 83.08 Bq kg^{-1}, respectively. While, in the flour samples, the average concentrations were 1.65, 1.61 and 171..31 Bqkg^{-1}, respectively. The ingestion doses were calculated to be 0.224μSvY^{-1}for the rice and 0.471 μSvY^{-1} for the flour samples which are below the recommended 1 mSv limit. The concentration of heavy elements (Fe. Cd, Mn, Ni, and Pb) in the rice and the flour samples were below the detection limits. Whereas, the mean contents of Cu were 3.75 mg/kg and 3.6 mg/kg of the rice and the flour samples, respectively. The mean values of Zn in the rice and the flour samples were 19.42 mg/kg and 17.3 mg/kg, respectively. The daily intake of Cu and Zn through the rice and the flour samples were lower than the tolerable daily intakes by FAO/WHO; this indicates there is no risk due to the intake of these foods to the people.

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INTRODUCTION

Naturally occurring radioactive elements 238U, 232The, 40K are the primary source of natural radiation exposure to the human. "The consumption of the foodstuff caused at least, one-eighth of the mean annual effective dose due to natural sources" (Hosseini, Fathivand *et al.*, 2006). "Foodstuffs are known to contain natural and man-made radionuclide's which after ingestion, contribute to an internal effective dose" (Venturini and Sordi 1999). Also, the heavy metals may enter the human being body through the intake of foodstuffs, gather in the main organs of the human body and are causing many health disorders (Duruibe, Ogwuegbu *et al*. 2007)."Heavy metal pollutants such as Cd, Zn, Cu and Pb are common and essential for human nutrition, but when they are consumed in high levels can cause health issues" (Kovalchuk, Titov *et al*., 2001). For contamination assessment of foodstuffs consumed by the population, it is important to know the baseline value of dose level of both natural and heavy metals.

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Therefore, natural radioactivity and heavy metals concentrations measurements in foodstuffs have been performed in several countries, e.g., e.g.(Singh, Sharma *et al.* 2010, Nadal, Casacuberta *et al.* 2011, Thomas B, Robert Martin *et al.* 2011, Awudu, Faanu *et al.* 2012, James, Dileep *et al.* 2013, Abojassim, Al-Gazaly *et al.* 2014, Desideri, Meli *et al.* 2014, Patra, Mohapatra *et al.* 2014). In Saudi Arabia, 80% of foods are imported from various countries, and very few researchers on exposure from radioactivity in foodstuffs have been conducted(Al-Ghamdi 2014).

It is important to carry out regular monitoring of foods like the rice and the flour, which are considered the main daily foodstuff consumed not only by people in Saudi Arabia but in all Arab countries. Thus, the objective of this work was to investigate the concentration of natural radioactivity (226Ra,232Th and 40K) and some heavy metals (Fe. Cd, Zn, Cu, Man, Ni, and Pb) in rice and flour samples. These concentrations can be useful as a guideline background to estimate the risk exposure of radionuclide's and heavy metals content through the individual intake foodstuff.

MATERIALS AND METHODS

Sample collection, preparations, and measurement

In this study, two types of essential foodstuff samples, including 12 samples of rice (imported) and 12 samples of flour (local and imported) were selected randomly from different markets in Saudi Arabia. The sample types and their origins were listed in Table 1. All samples were prepared according to the recommendations given by (IAEA, 1989). The samples were ground and sieved through a 2 mm mesh, homogenized and then stored in Marginally beakers. The beakers filled, weighed, sealed, and aged one month before measurement procedures, to ensure that radioactive equilibrium was reached between 226Ra,22Rn and its progeny (Abbady 2010). The concentration of the radionuclide's (^{226}Ra, ^{232}Th, and ^{40}K) in the foodstuff samples has been determined by using a high-resolution gamma-ray spectrometry system combined with a high-purity germanium detector (HPGe, Canberra). The counting time for the samples and background were 3600 Sec. ^{226}Ra activities were estimated from ^{214}Pb (295.2, 351.9 keV) and ^{214}Bi (609.3). ^{232}Th concentration was measured at the Gamma-ray energies of ^{212}Pb (238.6 keV), ^{228}Ac (911 keV) and^{208} To (583.2 keV), while the 40K activity was determined from the 1460.7 keV emission. Heavy elements (Fe. Cd, Zn, Cu, Man, Ni, and Pb) were measured by an inductively coupled plasma optical emission spectrometer (ICP-OES).

Calculations

Activity concentrations

The activity concentrations (A_c) of the natural radionuclides' in the measured samples were computed using the following relation (El-Taher 2015):

$$A_c(Bqkg^{-1}) = C\ /\varepsilon P_r M \quad (1)$$

Where C is the net gamma counting rate (counts per second), ε is the detector efficiency of the specific γ–ray, P_r is the absolute transition probability of Gamma–decay and M is the mass of the sample (kg).

Assessing the Annual Effective dose from ingested foods

The annual intake of radionuclide's with food is dependent on the concentration of radionuclide's in the various foodstuffs and on food consumption. It was calculated using the following formula (UNSCEAR, 2000):

$$D = AEI \quad (2)$$

Where D is the effective dose by ingestion of the radionuclide (mSy^{-1}), **A** is the activity concentration of the nuclide in the ingested food (By kg^{-1}); It is the annual intake of food (kg y^{-1}). For adults, the rice intake is 75 kg year^{-1} (Kuwait Government, 2009), and the average flour intake is 140 kg year^{-1} (UNSCEAR 2000). E is the radionuclide's dose conversion factor.For adults the values of (E) or 2.8 × 10^{-4}, 7.2 × 10^{-5} and 6.2 × 10^{-6} mSv Bq^{-1} of 226Ra, 228Th, and 40K, respectively, (ICRP 1996).

RESULTS AND DISCUSSION

Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

226Ra, 232Th and 40K activity concentration measurement in rice and flour samples are shown in Table 2. From this Table, ^{238}U content in the rice samples ranged from 0.38±0.09 (sample R9) to 2.67±0.29 Bqkg^{-1} (sample R2) with an average 1.08 Bqkg^{-1}, while that in the flour ranged from 0.89±0.25 (sample F5) to 2.67±0.64 Bqkg^{-1} (sample F11) with an average 1.65 Bq kg^{-1}. 232Th concentrations, in the rice samples, ranged from 0.18±0.02(sample R7) to 2.31±0.67 Bq kg^{-1} (sample R6) with an average 1.19 Bq kg^{-1} and for the flour samples 232Th content ranged from 0.64±0.14 (sample F10) to 2.62±0.64Bqkg^{-}1 (sample F3) with an average 1.61 Bq kg^{-1}. The highest concentrations of 238U and 232th in the rice samples are found in Indian rice and Egyptian rice, respectively, while, the American rice samples (F9 and F12) represent the lowest values in 238U and 232Th, respectively.The maximum and the minimum concentration values of 238U and 232Th in flour samples were found in Yemeni wheat flour (F11) and Saudi wheat flour samples (F5), correspondingly.

^{40}K content in the rice samples ranged from 56.24±1.71 (sample R5) to 110.33±2.12 Bq kg^{-1} (sample R9), while that in the flour samples ranged from 80.04±2.35 (sample F10) to 268.21±4.61 Bq kg^{-1} (sample F8) with average values 83.08Bq kg^{-1} and 171.31Bq kg^{-1}, respectively. These results for ^{40}K concentration be in- agreement with the world range from 40 to 240 Bq kg-1 reported by (Maul and O'hara 1989). From the results, it can be concluded that 40K is the most predominant radionuclide in the rice and flour samples, this is because, Potassium is an essential element and plant isotopic differentiation, thus, ^{40}K is preferred to the other two radionuclides (Musa Hamzah *et al*., 2011).

The results show; the variation in radionuclide concentrations was found even within the same kind of food samples, which were not collected from same farmlands in one region or different regions. This variation observed, can be probably caused by in the chemical and physical properties of the various farms of the producing areas, in which the plant grown may lead to variability in the concentration of the radionuclide in the food crops. Also, the variety may be caused by using of many phosphate fertilizers by the farmers to get the optimum product in a short term (Khater and Bakr 2011). The obtained results showed that for all the investigated samples, the specific activity concentrations of ^{226}Ra, ^{232}The and ^{40}K appeared lower than the standard recommended limit for foodstuffs (UNSCEAR 2008, 2000). The activity concentrations of 238U, 232Th and 40K in presently studied samples are given in Figure 1. Comparison between the present results with the reported results of the same foodstuffs in different countries was displayed in Table 3. It was observed that the average values of ^{226}Ra, 232Th and 40K activities for the rice samples are lower than the obtained values in Italy, India and Ghana. For the flour samples, the average concentration values of 226Ra and 232Th are higher than the values reported in Brazil and lower than Iraq values, but the average value of 40K concentration is greater than the reported values for the two countries. The concentrations of the radioactivity in the foodstuff may be varied from one country to another, depend on their climate and geological properties of the soil, and also, on the phosphate fertilizers were applied to the agricultural lands. (UNSCEAR 2000).

Annual effective dose

As shown in Table 4, for adults, the average annual effective doses of 226 Ra, 232Th, and 40k in the rice samples were estimated to be 0.062, 0.056, 0.106 μSv/year, respectively. While, for the flour samples the annual doses of 226Ra, 232Th and 40K were 0.178, 0.076 and 0.218 μSv/year, respectively.

Table 1. Origin of the samples and the brands of the flour samples

Rice		Flour		
Sample code	Sample origin	Sample code	Sample origin	Flour brand
R1	India	F1	Saudi Arabia	Wheat
R2	India	F2	Saudi Arabia	Wheat
R3	India	F3	Saudi Arabia	Wheat
R4	India	F4	Saudi Arabia	Co r n
R5	Thailand	F5	Saudi Arabia	White
R6	Egypt	F6	Omani	Wheat
R7	Egypt	F7	Kuwait	White
R8	Egypt	F8	Dubai	Wheat
R9	America	F9	Australia	Wheat
R10	America	F10	Yemen	Co r n
R11	America	F11	Yemen	Wheat
R12	America	F12	Yemen	White

Table 2. Activity concentration (By kg^{-1}) of 226Ra, 232Th and 40K in the rice and flour samples

Sample code no.	Activity concentration (Bq kg l^{-1})		
	226Ra	232Th	40K
R1	1.72±0.44	0.49±0.01	74.34±2.08
R2	2.67±0.29	0.88±0.11	84.37±2.32
R3	0.78±0.02	2.04±0.66	62.75±1.92
R4	0.81±0.04	0.24±0.07	96.23± 0.57
R5	0.25±0.05	0.83±0.21	56.24±1.71
R6	1.37±0.41	2.31±.0.67	64.90±1.71
R7	0.68±0.22	1.98±0.31	101.68±2.89
R8	1.07±0.08	1.94±0.41	71.39±1.41
R9	0.38±0.09	1.29±0.11	110.33±2.12
R10	1.32±0.36	1.76±0.43	90.86±2.13
R11	0.98±0.12	0.34±0.03	106.01±2.03
R12	0.91±0.18	0.18±0.02	77.87±2.54
Range	0.38±0.09 - 2.67±0.29	0.18±0.02 -2.31±.0.67	56.24±1.71 -110.33±2.12
Average	1.08	1.19	83.08
F1	2.14±0.43	1.28±0.37	218.50±5.15
F2	1.86±0.31	2.17±0,62	205.52±5.15
F3	1.03±0.08	1.37±0.54	99.52±2.77
F4	1.13±0.05	0.89±0.06	114.21±2.54
F5	0.89±0.25	0.64±014	128.19±3.23
F6	2.24±0.37	1.65±0.51	188.22±4.61
F7	1.97±0.36	2.11±0.39	229.32±5.28
F8	2.39±.0.09	1.05±0.07	268.21±4.61
F9	0.48±0.15	1.92±0.25	212.55±4.71
F10	1.78±0.43	2.13±0.16	199.03±4.86
F11	2.67±0.64	2.62±0.64	80.04±2.35
F12	1.26±0.48	1.45±0.29	112.45±4.13
Range	0.89±0.25 -2.67±0.64	0.64±014-2.62±0.64	80.04±2.35 - 268.21±4.61
Average	1.65	1.61	171.31

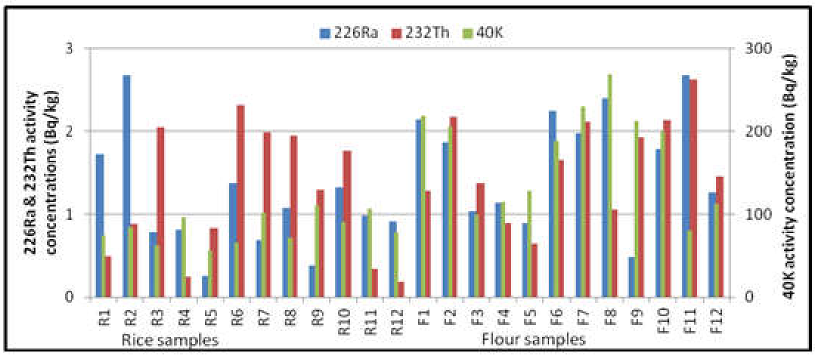

Fig.1 Activity concentrations of 226Ra, 232Th and 40K for the rice and the flour samples consumed in Saudi Arabia

The highest annual dose was for 40K; this radionuclide is usually of limited interest because it is an essential element, its concentration in the human body is under homeostatic control, and hence, an adequate dose 40Kwithin the body is constant (UNSCEAR1982). Table 4 shows that the average total annual doses due to ingestion the rice and the flour samples were 0.224 and 0.471 µSv/year, respectively. The annual dose of the flour samples is higher than the rice samples dose, may be referred to the high consumption rate. Fig. 2 shows the calculated contributions to a total effective dose of U, Th series and 40K for the rice as 37.40%, 16.08%, and 46.20%, respectively, and in the flour samples as 27.68%, 25.09%, and 47.23% respectively. In general, the current annual effective doses of the three terrestrial gamma radiations are lower than the recommended limit of 1 mSv/year (IAEA 2005). Therefore, consumption of the studied rice and flour samples in Saudi Arabia is still safe and pose no detrimental health effect.

Heavy metal concentrations in foodstuffs

In this study, heavy elements (Fe. Cd, Zn, Cu, Mn, Ni, and Pb) were measured in the rice and the flour samples. It was established that the concentrations of the measured elements (Fe, Cd, Mn, Ni and Pb) in all instant rice and flour samples were below the detection limits. The concentration of Cu ranged from 2 to 6 mg/kg with a mean value 3.75 and ranged from 2-7mg/kg with a mean value 19.42 mg/kg for the rice and the flour samples respectively. Zn concentration in the rice samples ranged from 2-27 mg/kg with a mean value 19.42 mg/kg, and in the flour ranged from 2-37 mg/kg with a mean value 17.3mg/kg. The current mean values for Cu and Zn concentrations in rice were below the reported values in South China (Cu:20.3 mg/kg and Zn:31.9 mg/kg) by (Jing Zhen *et al* 2013) and in India (Cu:36.4, and Zn: 9.5 mg/kg) by (Sharma *et al*., 2008). WHO/FAO 2007 established the permitted maximum concentrations (MPCs) of copper and zinc values as 30

Table 3. Comparison of activity concentrations results of 226Ra, 232Th and 40K from various countries

Country	Foodstuff	Activity concentration (Bq kg^{-1})			Reference
		226Ra	232Th	40K	
Saudi Arabia	Rice	1.08	1.19	83.08	Present work
Italy	Rice	2.9	2.8	119.3	Desideri, Meli *et al*. 2014
India	Rice	3.07	34.3	120.8	Shansi *et al* 2009
Ghana	Rice	4.72	4.33	104.36	Awudu, Fasano James, *et al*. 2012
Saudi Arabia	Flour	1.65	1.61	171.31	Present work
Brazil	Flour	0.18	0.12	36.2	V. Schreiber *et al*.,2006
Iraq	Flour	6.60	1.95	133. 10	Ali Abed Abu Jassim *et al*., 2014

Table 4. Annual effective dose (µSv y^{-1}) due to the intake of the natural radionuclide's of 226Ra, 232Th and 40K from the foodstuffs (rice and flour)

Sample code	Effective dose (µSv y^{-1})			Total effective dose
	226Ra	232Th	40K	
R1	0.123	0.060	0.278	0.461
R2	0.108	0.102	0.261	0.470
R3	0.059	0.065	0.126	0.250
R4	0.065	0.042	0.145	0.252
R5	0.051	0.030	0.163	0.244
R6	0.129	0.078	0.239	0.446
R7	0.113	0.099	0.291	0.504
R8	0.137	0.049	0.341	0.528
R9	0.028	0.091	0.270	0.388
R10	0.102	0.100	0.253	0.456
R11	0.153	0.124	0.102	0.379
R12	0.072	0.068	0.143	0.284
Range	0.028 - 0.153	0.030 - 0.124	0.102 -0.341	0.244 -0.528
Average	0.062	0.056	0.106	0.224
F1	0.230	0.113	0.520	0.863
F2	0.199	0.192	0.489	0.881
F3	0.111	0.121	0.237	0.469
F4	0.122	0.079	0.272	0.472
F5	0.096	0.057	0.305	0.457
F6	0.241	0.146	0.448	0.835
F7	0.212	0.186	0.546	0.944
F8	0.257	0.093	0.639	0.988
F9	0.052	0.169	0.506	0.727
F10	0.191	0.188	0.474	0.853
F11	0.287	0.231	0.191	0.709
F12	0.135	0.128	0.268	0.531
Range	0.096 -0.287	0.057 - 0.231	0.191 - 0.639	0.469 - 0.988
Average	0.178	0.076	0.218	0.471

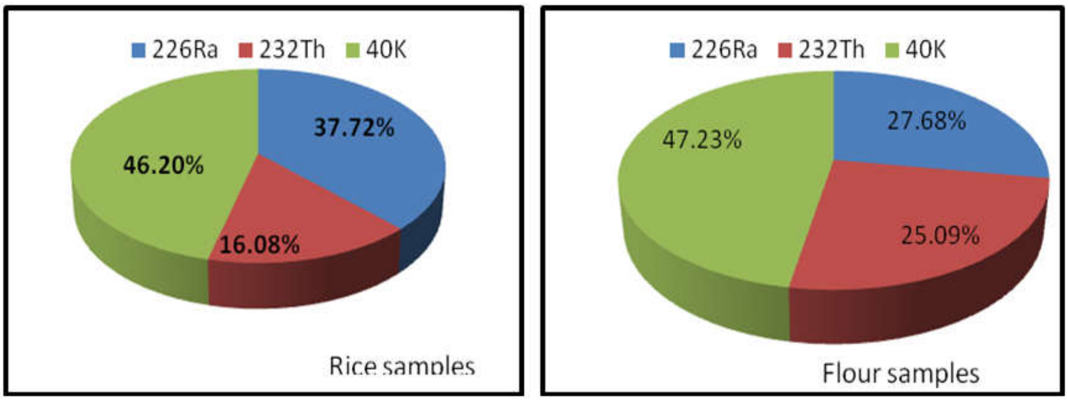

Fig 2. Percentage contribution to the total effective dose of ^{226}Ra, ^{232}Th and ^{40}K in the rice and flour samples

and 50 mg/kg, respectively, no values of Cu and Zn exceeding the MPCs were detected in the rice and the flour samples in the present study.

Estimated daily intake of heavy metals

The average daily dose (EDI) of metals was determined by dividing the daily intake by the human body weight as the following equation (Zhuang, McBride *et al*. 2009):

$$EDI = C_{metal} \times W/m \quad (3)$$

The estimated daily intake of heavy metals depends on the metal concentration level(C_{metal}) and the average daily consumption of a foodstuff (g/ person/day). In the present study, the calculations were made for adults with a body weight of 70 kg and average daily consumptions of 205g rice and 384g flour. The average daily intake of Cu in the analyzed rice and flour samples were 0.011 mg kg^{-1} day^{-1} bw and 0.020 mg kg^{-1} day^{-1} bw, respectively. The EDI of Zn was 0.057 mg kg^{-1} day^{-1} bw for the rice samples and 0.095 mg $kg^{-1}day^{-1}$ bw for the flour samples. The estimated daily intakes of copper and zinc in analyzing samples were found to be lower than the maximum intake 3mg day^{-1} kg^{-1} and 5mg day^{-1} kg^{-1} for Cu and Zn respectively, recommended by FAO/WHO. So, the concentrations of Cu and Zn elements for daily intake are below safety levels for human consumptions. As a result, Cu and Zn were not a cause of any risk to the local population.

Conclusion

In this project, the concentration levels of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K, and some heavy metals (Fe. Cd, Zn, Cu, Mn, Ni, and Pb) were found in an essential foodstuff (rice and flour) consumed in Saudi Arabia. The activity concentration of the three radionuclides in the present study was found to be within the values reported by UNSCEAR(2008). The calculated total annual effective dose is lower than the permitted limit 1mSv. The obtained concentrations of heavy elements (Fe. Cd, Mn, Ni, and Pb) were below the detection limit, whereas, Cu and Zn concentrations were below the recommended values of the WHO/ FAO.

Therefore, there is no harm effect due to the consumption of rice and flour samples presenting the concentration levels found in this work. The obtained data can provide a baseline of the natural radioactivity and the heavy metal's exposure to the population from the consuming of daily foodstuffs as rice and flour.

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