ANALYSIS OF NATURAL RADIOACTIVITY AND DETERMINATION OF POTASSIUM CONTENT IN FOODS

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ABSTRACT

Natural radionuclides, such as potassium-40 (^{40}K), radium-226 (^{226}Ra), and thorium-232 (^{232}Th) originating from the earth's crust can enter the human body through foods grown in the soil, which can cause internal irradiation. Information on radioactivity concentrations in foods allows us to assess the amount of exposure to radiation through the diet. K, a vitally important mineral for human health, is found in nature as a mixture of three isotopes ^{39}K, ^{40}K, and ^{41}K and of these, only ^{40}K is radioactive and constitutes 0.0117% of natural K. Because natural K is abundant in the soil, the biggest contribution among natural radionuclides that are taken into the body through food comes from ^{40}K, and beta and gamma radiation emitted from ^{40}K can increase the level of radiation exposure through internal irradiation. In the present study, the concentrations of radionuclides found naturally in some grains and legumes (e.g., pumpkin seeds, cowpeas, bulgur, sunflower seeds, rice, broad beans, chickpeas, beans, lentils, wheat, rye, corn, barley, and oats) were measured using a spectrometer, and the annual effective radiation dose resulting from these radionuclides was evaluated. Accordingly, we found that the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th radionuclides contained in food samples had values varying between 33.27 ± 5.17 and 370.4 ± 16.73, 1.19 ± 0.18 and 3.55 ± 0.34, and 0.98 ± 0.06 and 3.65 ± 0.37 Bq kg^{-1}, respectively. We determined that the annual effective dose resulting from these radionuclides varied between 0.09 and 49.98 µSvy^{-1}. The results obtained were compared with those from other studies. The present study also aimed to develop an alternate method, which is much easier and more sensitive, using ^{40}K concentrations to determine the K content in foods. Using this method, we observed that the amount of K in the food samples varied between 1.2 and 11.93 gkg^{-1}

KEYWORDS:
Cereals, legumes, potassium, natural radioactivity, annual effective dose, gamma spectrometer

INTRODUCTION

All living things on earth can be exposed to terrestrial and cosmic radiation, which create natural radiation sources, through internal and external factors that depend on the region in which they live and their living conditions. External exposure to radiation is caused by the ground crust–based radioactive potassium isotope (^{40}K) and radionuclides in the uranium-238 (^{238}U) and thorium-232 (^{232}Th) series, while gamma and alpha–beta radiation emitted from those radionuclides taken into the body through food cause internal irradiation. According to the report published by the United Nations Scientific Committee on the Effects of Atomic Radiation, the total exposure per person from ingestion of radionuclides worldwide is 0.29 mSv, of which approximately 60% comes from ^{40}K and 40% from radionuclides in the ^{238}U and ^{232}Th series [1]. Exposure to radiation and the risk of developing cancer are directly proportional. Studies have been conducted to determine amounts of radiation dose exposure and create a database that would help develop relevant standards.

In a study conducted in Brazil, the activity concentration of radium-226 (^{226}Ra) was determined to be 1.43 Bq kg^{-1} and that of ^{40}K was 434.0 Bq kg^{-1} [2]. In another study conducted in Rio de Janeiro, Brazil, ^{40}K and ^{226}Ra were found in different food samples in concentrations of 75.58 and 6.72 Bq kg^{-1}, respectively, and the annual effective dose value was 155.73 µSvy^{-1} [3]. In a study conducted in Egypt, it was determined that the annual effective dose from food was 1.54 µSvy^{-1} [4]. A study conducted in Egypt on imported foodstuffs found that the average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K were 1.87, 2.73, and 168.7 Bq kg^{-1}, respectively [5]. In a study conducted in Accra, Ghana, the researchers found that the total effective dose exposed through food was 4.46 mSvy^{-1} [6]. In a study conducted on various foods in Italy, it was determined that ^{40}K activity concentrations varied between 70.5 and 181.1 Bq kg^{-1}, and 94% of the total dose was from ^{40}K [7]. In a study conducted in Bangladesh, the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in food samples were determined to be 0.97, 0.13, and 3.56 Bq kg^{-1}, respectively [8]. In a study in Rize, Turkey, the average ^{40}K activity concentrations in various foods were reported to be 465.8 Bq kg^{-1} [9,10]. Another study was conducted in Rize on ^{40}K, ^{238}U, and ^{232}Th to determine the activity concentrations and annual effective doses from some vegetables and fruits [11]. In a study conducted using peanuts in Turkey on ^{226}Ra, ^{232}Th, and ^{40}K, the average activity concentrations were reported to be 5.4, 6.9, and 427.1 Bq kg^{-1}, respectively, and the effective dose of exposure varied from 6.5 to 10.1 µSvy^{-1} [12]. The aim of the present study was to determine the activity concentrations of natural radionuclides found in different food samples produced in Muğla soils with high agricultural potential and to evaluate annual effective dose resulting from consuming that food. Optical spectroscopy methods are commonly used to determine the minerals contained in foods [13]. These methods require special equipment and complex operations. In the present study, we used the 1460 keV gamma advantage in which ^{40}K activity concentration was determined and the K contents of the foods were determined.

The importance of (potassium) K. K, a vitally important mineral for health, regulates functions such as digestion, blood pumping, muscle contraction, nerve flow, and water retention in the cells. It helps to maintain the water balance in the body and is the main positive cation inside the cells; therefore, it helps balance fluids and electrolytes. The risk of hypertension decreases in those who eat foods rich in K. K helps to remove excess water, sugar, and salt from the body and is also very important for plant growth. K deficiency can be caused by the body's mineral loss through malnutrition and diarrhea, diuretic drugs, vomiting, or sweating. This deficiency can cause diseases and is among the most important causes of injuries in athletes. Excessive K intake can adversely affect the kidneys and heart.

MATERIALS AND METHODS

Sampling. Located in the Aegean region of Turkey (between 37° and 28° 12 north latitude and 21 east longitude), Muğla is a residential center and an important agricultural town dominated by a Mediterranean climate. The measured food samples were provided from the public markets in Muğla Province (Fig. 1). The food samples provided were dried in an oven in the laboratory and then ground in stone mills and turned into powder. These powdered food samples were then passed through 0.5-µm sieves and put into 6- by 5-cm standard sample containers. The mouth of each container was sealed with parafilm and left for ~1 month to allow for a permanent equilibrium to be reached between ^{226}Ra and radon-222 (^{222}Rn). Following this equilibrium, approximately 80.000 counts were taken for each food sample using the NaI (TI) detector gamma spectrometer at the Nuclear Physics Laboratory of Ondukuzmayıs University, which provided the gamma spectrum on each sample.

Experiment. We used the ORTEC 905-4 (3" x 3") model NaI (TI) Gamma Spectrometer with a sodium iodide scintillation detector to measure the gamma spectrum. The resolution of the system was 2% at 0.5 MeV and 1.3% at 2 MeV (1 µCi for 137 Cs at a 10-cm distance). Energy and efficiency calibration, mixed point (cobalt-60–cesium-137 (^{60}Co–^{137}Cs) sources with known gamma energies, and the International Atomic Energy Agency (IAEA) referenced RGU-1, RGTh-1, and RGK-1 in standard vessels with the same geometry were used as the sample resources. ScintiVision was used for analyses. This program has four different fit types—interpolative, linear, quadratic, and polynomial. The calibration

FIGURE 1
Simplified map showing study area.

curve was created by fitting the data obtained for the yield calibration to the polynomial curve, and the energy efficiency equation for the graph was created by drawing the efficiency calibration graph. The activity was calculated by using the obtained yield values in Eq 1 [14].

$$A = \frac{N_{net}}{\mathcal{E}_{\gamma}.I_{\gamma}t\ \ m}, \qquad (1)$$

where, A (Bq kg^{-1}) is the activity concentration of the relevant radionuclide, Nnet is the net area of the corresponding photopeak, and Ɛγ is the relative efficiency of the radionuclide Iγ, the gamma emission probability of the radionuclide m (kg), and the mass of the sample (kg).

Activity concentration measurement. Because ^{226}Ra, ^{232}Th, and their radioisotopes have very long half-lives and their concentrations in nature are very low, it is difficult to determine the radioactivity of these isotopes using the current gamma spectrometric method; therefore, while calculating the activity of these isotopes, the photopics formed by the degradation products in the ^{226}Ra and ^{232}Th series are used. In the present study, the photopic of lead-214 (^{214}Pb) at 351.9 keV and bismuth-214 (^{214}Bi) at 609 keV were used for the ^{226}Ra activity concentration, the photopic of actinium-228 (^{228}Ac) at 338.4 keV for the ^{232}Th activity concentration, and the photopic of thallium-208 (^{208}TI) at 583.2 keV. The activity concentration of the ^{40}K radioisotope was obtained using 1460 keV photopic [15].

Radiological evaluations. According to the type and energy of the radiation taken into the body, the biological damage can be expressed by equivalent dose, with light particles doing less damage than heavy particles. Determining the equivalent dose takes into account the radiation type and energy. The annual effective equivalent does is obtained by multiplying the radiation conversion factors per unit activity determined for each radionuclide and the activity concentration determined per unit volume or weight by the dose conversion factors based on the radiation type and energy emitted by the radionuclides (Eq 2).

$$\mathrm{H}\ \mathcal{E}(\mathrm{Sv}) = \sum(\ \ \mathrm{C}_{\mathrm{i}}\ \left(\frac{\mathrm{Bq}}{\mathrm{kg}}\right)\mathrm{V}_{\mathrm{i}}\ \ (\mathrm{kg})\mathrm{G}(\mathcal{E}), \quad (2)$$

where, i is the different substance, C$_i$ (Bq kg^{-1}) is the activity concentration of that substance, V$_i$ (kg) is the annual consumption amount of that substance, and G (Ɛ) is the dose conversion factor for the radionuclide in question [16,17] .

Determining K (potassium) in foods. The ^{40}K activity concentrations in foods and the annual effective dose values within the body after consuming these foods can be determined using the gamma spectrometric method developed for the present study. With this method, the measured ^{40}K activity concentrations were used in to calculate the amount of K in light of radioactive decay rules (Eq 3) [18].

$$A(\mathrm{Bq\ kg^{-1}}) = \quad \lambda N, \qquad (3)$$

where, N is the number of radioactive ^{40}K per kg, and λ is the decay constant of the ^{40}K. Because the activity concentration can be measured, the value of λ can also be found to calculate N (Eq 4).

$$\mathrm{t}_{1/2} = \frac{0{,}693}{\lambda}, \qquad (4)$$

where, t$_{1/2}$ = ^{40}K half-life of ^{40}K.

$$N = \frac{m(40_K)N_A}{\mathrm{A\ (g)}} \qquad (5)$$

When A (g) mass number and N$_A$, the Avogadro number, are written in Eq 5, the value of m (^{40}K) is found in grams. Because there is 0.0117 g ^{40}K in 100 g K, the value of m (^{40}K) g in a specific amount of K can be calculated. Thus, the amount of K in a kilogram of food sample can be determined.

RESULTS AND DISCUSSION

^{40}K, ^{226}Ra, and ^{232}Th activity concentrations in different grain and legume samples and the annual effective doses calculated based on these results are shown in Tables 1 and 2, respectively. Table 3 shows the results compared with those from similar studies. As seen in Table 1, the lowest value of the ^{40}K activity concentration was 33.27± 5.17 Bq kg^{-1} in the rice sample, and the highest value as 370.4± 16.73 Bq kg^{-1} in the bean sample. The ^{40}K activity concentration had relatively high values in the broad bean, pumpkin seed, cowpea, and lentil food samples. The lowest value of ^{226}Ra activity concentration was 1.19±0.18 Bq kg^{-1} in the wheat and corn samples, and the highest value was 3.55±0.34 Bq kg^{-1} found in the broad bean samples. The lowest value of ^{232}Th activity concentration was 0.98±0.06 Bq kg^{-1} in the bean samples and the highest was 3.65±0.37 Bq kg^{-1} found in the broad bean samples. The chickpea sample had the highest activity concentrations of both ^{226}Ra and ^{232}Th. In general, the highest value of activity concentration of all three radionuclides was found in the broad bean sample. The Turkey Atomic Energy Agency (TAEK) has reported that the ^{40}K activity concentrations in lentils, rice, corn, and wheat concentrations are approximately 300, 170, 90, and 130 Bq kg^{-1}, respectively. In the present study, the ^{40}K activity concentration was determined to be 272.6±13.60 Bq kg^{-1} in lentils, 33.27±5.17 Bq kg^{-1} in rice, 38.12±5.90 Bq kg^{-1} in corn, and 39.88±6.16 Bq kg^{-1} in wheat.

These determined values were lower than those explained in the TAEK report. In another TAEK report from 2013, the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th in Muğla soil samples were 259.90, 17, and 19 Bq kg^{-1}, respectively, which were well below the accepted reference values of 400 Bq kg^{-1} for ^{40}K, 35 Bq kg^{-1} for ^{238}U, and 30 Bq kg^{-1} for ^{232}Th [19-20]; therefore, the low activity concentration detected for food samples is predicted to result from the relatively low activity concentration in the soil. The annual effective doses that were calculated based on activity concentrations are shown in Table 2. The maximum total contribution to the annual effective dose exposed from ^{40}K, ^{226}Ra, and ^{232}Th was 136.64 µSvy^{-1} in the wheat sample, which was associated with the high levels of wheat consumption and/or wheat-containing foods. The lowest total contribution to the annual effective dose was 1.49± 0.17 µSvy^{-1} in the barley sample. In the report prepared by TAEK, the annual effective doses from exposure to ^{40}K from consumption of lentils, rice, corn, and wheat were 14.79–16.35, 1.19–18, 6.5–9.72, and 113.89–175.04 µSvy-1, respectively. In the present study, the annual effective doses calculated for the same food samples was considerably lower than that presented in the TAEK report, which listed 300 µSv as the accepted reference dose worldwide [21]; This situation was associated with the radiological structure of the soil in Muğla Province.

Determining the K (potassium) value. The best way to get adequate levels of K, an important mineral for many vital activities in the human body, is to consume adequate amounts of different foods. Within the scope of the International Atomic Energy Agency (IAEA) Basic Safety Standards, the recommended levels for K are 600 mg/d for infants up to 1 year, 1000–2000 mg/d for children up to 10 years old, and 2000–3000 mg/d for adults [30]. The K and ^{40}K values calculated using the ^{40}K radioactivity

TABLE 1
Natural radioactivity of ^{40}K, ^{226}Ra and ^{232}Th isotopes in food samples.

Beans	370.4 ± 16.73	1.47 ± 0.13	0.98 ± 0.06
Lentil red	272.6 ± 13.60	2.86 ± 0.24	1.56 ± 0.14
Wheat	39.88 ± 6.16	1.19 ± 0.18	1.44 ± 0.12
Rye	44.4 ± 6.17	2.16 ± 0.22	2.52 ± 0.27
Corn	38.12 ± 5.9	1.19 ± 0.18	1.98 ± 0.20
Barley	81.6 ± 9.85	1.81 ± 0.19	2.15 ± 0.23
Oat	66.2 ± 8.21	3.41 ± 0.31	1.85 ± 0.19
Mean	141.34 ± 10.45	2.33 ± 0.20	1.69 ± 0.18

TABLE 2
Annual effective doses resulting from ^{40}K, ^{226}Ra, and ^{232}Th isotopes.

Sample	**Annual Effective Dose (µSvy^{-1})**			
	^{40}K	**^{226}Ra**	**^{232} Th**	**Total Contribution**
Pumpkin seed	1.34 ± 0.18	0.35 ± 0.02	0.35 ± 0.02	2.04 ± 0.22
Kidney bean	1.96 ± 0.28	0.60 ± 0.06	0.33 ± 0.02	2.89 ± 0.36
Bulgur	12.50 ± 1.69	7.56 ± 1.08	4.41 ± 0.63	24.47 ± 3.40
Sunflower seed	0.73 ± 0.09	1.20 ± 0.17	0.48 ± 0.04	2.41 ± 0.30
Rice	1.13 ± 0.14	4.20 ± 0.60	4.12 ± 0.57	9.45 ± 1.31
Broad beans	0.09 ± 0.01	0.39 ± 0.04	0.33 ± 0.03	0.81 ± 0.08
Chickpea	7.81 ± 1.1	2.76 ± 0.38	2.33 ± 0.32	12.90 ± 1.80
Beans	7.50 ± 0.96	1.35 ± 0.19	0.74 ± 0.07	9.59 ± 1.22
Red lentil	8.45 ± 1.2	4.00 ± 0.57	1.79 ± 0.24	14.24 ± 2.01
Wheat	37.08 ± 5.29	49.88 ± 7.01	49.68 ± 6.59	136.64 ± 18.89
Rye	0.41 ± 0.05	0.90 ± 0.11	0.86 ± 0.08	2.17 ± 0.24
Corn	3.42 ± 0.45	4.83 ± 0.65	6.60 ± 0.88	14.85 ± 1.98
Barley	0.50 ± 0.07	0.50 ± 0.05	0.49 ± 0.05	1.49 ± 0.17
Oat	0.57 ± 0.06	1.33 ± 0.19	0.59 ± 0.05	2.49 ± 0.30
Mean	5.97 ± 0.83	5.71 ± 0.79	5.22 ± 0.69	16.89 ± 2.15
Standard deviation	9.77	12.91	12.94	35.16
Min	0.09	0.39	0.33	0.81
Max	37.08	49.98	49.68	136.64

TABLE 3
Comparsion of the data obtained from similar studies.

Sample	Radioactivity Concentrations (Bq kg^{-1})			References
	^{40}K	^{226}Ra	^{232}Th	
Rice	-	2.6 x 10^{-3}	3.2 x 10^{-3}	Santos et al., 2002 [22]
Rice	14.7	-	-	Venturini &Sordi,1999 [2]
Rice	70.3	-	-	Ramachandran & Mishra,1989 [23]
Rice	150	-	-	Donatello et al., 2014 [7]
Rice	3.56	0.97	0.13	Nahar et al.,2019 [8]
Beans	434	-	-	Venturini & Sordi,1999 [2
Beans	-	9.00	-	Asefi et al., 2005 [24]
Beans	453.6	-	18.9	Jibiri et al.,2007 [25]
Beans	1664.00	34.32	-	Bolca et al., 2007 [26]
Beans	739.79	0.28	0.78	Görür et al., 2012 [17]
Chickpea	150.8	-	-	Ramachandran &Mishra,1989 [23]
Wheat	91.73	1.67	0.5	Changızı et al., 2013 [27]
Wheat	102.9 ± 9.8	0.7 ± 0.2	1.1 ± 0.02	Pulhani et al., 2005 [28]
Wheat flour	132.4 ± 0.82	5.7 ± 0.41	1.9 ± 0.2	Khan et al.,2020 [29]
Wheat flour	155	-	-	Donatello et al., 2014 [7]
Corn	101.52	0.81	0.85	Changızı et al., 2013 [27]
Corn flour	150	-	-	Donatello et al., 2014 [7]
Corn	185.8	3.37	-	Awudu et al., 2012 [6]
Rice	33.27	2.73	3.26	Present study
Beans	370.4	1.47	0.98	Present study
Chickpea	217.4	1.70	1.75	Present study
Wheat	39.88	1.19	1.44	Present study
Corn	38.12	1.19	1.98	Present study

TABLE 4
Potassium values in food samples.

Sample	Potassium (K) Value g kg^{-1}	Potassium (^{40}K) Value g kg^{-1}
Pumpkin seed	6.96	8.15 x 10^{-4}
Kidney bean	8.74	9.91 x 10^{-4}
Bulgur	5.46	6.39 x 10^{-4}
Sunflower seed	2.50	2.96 x 10^{-4}
Rice	0.93	1.25 x 10^{-4}
Broad beans	11.70	13.87 x 10^{-4}
Chickpea	7.00	8.9 x 10^{-4}
Beans	11.93	13.96 x 10^{-4}
Red lentil	8.70	10.27 x 10^{-4}
Wheat	1.28	1.50 x 10^{-4}
Rye	1.42	1.67 x 10^{-4}
Corn	1.22	1.43 x 10^{-4}
Barley	2.62	3.07 x 10^{-4}
Oat	1.12	2.49 x 10^{-4}

concentration are shown in Table 4. After sorting the measured foods by K content, we observed the K levels to be from, most to least, bean > broad bean > lentil > cowpea > broad bean > pumpkin seed > bulgur > barley > sunflower > rye > wheat > corn > oat > rice, which indicates that beans, broad beans, and lentils are rich in K. The average consumption of foods per person in a year was taken from ITS data [31]. From that data, the K intake from each food was determined.

CONCLUSIONS

Being an important tourism and cultural center, Muğla is also known for its Mediterranean cuisine. There are no similar studies on selected foods within this region using the gamma spectroscopic method. In the present study, natural radioactivity was measured in food samples using a gamma spectrometer with a NaI (TI) detector. As seen in Table 1, the average activity concentrations of ^{40}K ^{226}Ra, and ^{232}Th in foods were 141.34 ±.

10.45, 2.33 ± 0.20, and 1.69 ± 0.18 Bq kg^{-1}, respectively. The total effective dose from ^{40}K, ^{226}Ra, and ^{232}Th varied between 0.81 ± 0.08 and 136.64 ± 18.89 µSvy^{-1}, with an average of 16.89 ± 2.15 µSvy^{-1}. The determined average dose was considerably lower than the annual permitted value of 300 µSvy^{-1} listed by the International Commission on Radiation Protection. We believe this level reflects the relatively low activity concentration in the soil of the region chosen as the study area. In addition, in this study, an alternative method has been try to developed in which the measured ^{40}K activity concentration can be used to determine the amounts of K and ^{40}K.

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