RAP Conference Proceedings, vol. 10, pp. 87–91, 2025
ISSN 2737-9973 (online) | doi: 10.37392/RapProc.2025.17
RAP-PROCEEDINGS.ORG

DETERMINATION OF NATURAL RADIOACTIVITY OF WHEAT FLOUR SAMPLES FROM SELECTED ALBANIAN MARKETS

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Abstract. *In this paper, the natural radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K were determined in 16 brands of wheat flour samples using HPGe gamma-ray spectrometry. Measurements of natural radioactivity in foodstuffs are essential for monitoring radiation levels to which the population is directly or indirectly exposed. Since wheat flour is one of the staple food products, establishing a national baseline of radioactivity exposure from different brands available in markets in Tirana, Albania, is significant. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in wheat flour are found to be (6.3 ± 0.6), (0.25 ± 0.08), and (45.9 ± 2.7) Bq kg^{-1}, respectively. The results demonstrate that activity concentrations in wheat flour samples are much below the world average values of 33 Bq kg^{-1} for ^{226}Ra, 42 Bq kg^{-1} for ^{232}Th, and 512 Bq kg^{-1} for ^{40}K. All evaluated radiological parameters, including radium equivalent activity, internal hazard index, and annual effective dose, were found to be well below the recommended international safety limits. These results indicate that the wheat flour samples analyzed pose a low and acceptable radiological risk. Therefore, from a radiological protection standpoint, their consumption does not represent a significant health concern for the public. The present study contributes to the continuation of similar investigations aimed at providing a comprehensive overview of radioactivity levels and radiological risk in foodstuffs in Albania.*

Keywords: *natural radionuclides, wheat flour, gamma spectrometry*

1. Introduction

Exposure to radiation in everyday life originates from several sources, including natural background radiation, medical applications, nuclear weapons testing, nuclear power generation, nuclear power, nuclear accidents, and certain occupations that involve increased exposure to artificial or natural radiation [1].

All materials on and within the Earth contain radionuclides. Among the most significant naturally occurring radionuclides are those belonging to the uranium-238 and thorium-232 decay series, as well as potassium-40. Together with their decay products, these radionuclides emit ionizing radiation [2]. Due to their very long half-lives, on the order of hundreds of millions to billions of years, they represent the main contributors to natural background radiation and are ubiquitously present in the environment, including soil, air, water, and food [1].

Some artificial radionuclides released into the environment from uranium-fueled reactors emit gamma radiation, while others are beta or alpha emitters. Among artificial radionuclides, cesium-137 (^{137}Cs) is one of the most significant gamma emitters due to its relatively long half-life compared to many other gamma-emitting radionuclides [3].

The primary sources of ^{137}Cs in the environment include atmospheric nuclear weapons testing, releases from the Chernobyl Nuclear Power Plant accident in 1986, as well as local discharges from nuclear reactors and nuclear waste reprocessing facilities [4].

Contaminated soil, air, and water act as direct sources of radionuclides, leading to the contamination of agricultural products, including wheat and, consequently, wheat flour [3].

The main contributors to radiation exposure are the naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K [1]. However, among radionuclides present in the environment, ^{137}Cs and ^{40}K exhibit relatively high soil-to-plant transfer factors. Nevertheless, the environmental abundance of ^{40}K is significantly higher than that of ^{137}Cs [5]. Radionuclides present in soil are absorbed by plants and can subsequently be transferred to humans through the consumption of contaminated food, resulting in internal exposure. Radionuclides ingested through various foodstuffs represent a major source of internal radiation exposure and may contribute significantly to radiation doses delivered to different organs and tissues over both short- and long-term periods.

Several radionuclides, such as ^{226}Ra and ^{232}Th, are considered radiotoxic. Among them, radium is of particular concern due to its radiotoxicity and its classification as a carcinogenic element.

These radionuclides may accumulate in specific organs and tissues, where their effects can manifest over both short- and long-term periods. Prolonged exposure

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may increase the risk of adverse health effects, including various radiation-induced disorders [6].

In Albania, information on radioactivity levels in foodstuffs is very limited [7, 8].

Therefore, this study aims to determine the radioactivity levels and assess the associated radiological risk from the consumption of selected brands of wheat flour by the population in Albania. Such an assessment is important for evaluating potential health impacts and ensuring food safety.

The results obtained in this work were compared with findings from similar studies, as well as with international reference values and recommendations provided by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the International Commission on Radiological Protection (ICRP), and the World Health Organization (WHO), which establish guidelines and dose limits related to radioactivity in foodstuffs.

2. Materials and methods

2.1. Sampling and sample preparation

Sixteen brands of wheat flour were selected from various markets in Tirana based on their sales volume. The samples were prepared following the recommendations of the International Atomic Energy Agency [3]. They were dried at 80 °C for approximately 10 hours, pulverized into a fine powder, sieved through a mesh of ≤2 mm, and thoroughly homogenized.

The prepared samples were then packed into plastic containers and tightly sealed for 30 days to allow secular equilibrium to be established between ^{226}Ra, ^{232}Th, and their respective daughter products. An empty Marinelli beaker was also measured under identical conditions to determine the background radiation.

2.2. Sample measurement

The activity concentrations of radionuclides in the samples were determined using a gamma-ray spectrometry system equipped with a high-purity germanium (HPGe) detector from Canberra. Spectrum analysis was performed with the Genie 2000 software. The HPGe detector was connected to an 8k-channel Multichannel Analyzer (MCA) and featured a carbon-epoxy window on the upper part of the detector during measurements.

The energy resolution (FWHM) of the detector was 1.8 keV at the 1.33 MeV γ-ray of 60Co, with a relative efficiency of 40% at the peak. To minimize interference from external radiation and other fluctuations, the detector was shielded with three layers consisting of lead, copper, and cadmium. Each sample was counted for 86,400 seconds, and background counts measured over the same duration were subtracted from the sample counts to obtain the net activity.

Energy calibration was performed over the range of 50 keV to 2000 keV using several reference sources. The detector efficiencies were determined using a power-fitting function provided by the LabSOCS software (Laboratory Sourceless Calibration Software), and the fitted efficiencies were subsequently applied in the calculation of the sample activities [9].

The efficiency calibration was validated using standard reference radioactive material supplied by the International Atomic Energy Agency (IAEA). The absolute efficiency uncertainties were evaluated for each peak, ranging from approximately 10% at low energies to 4% at high energies [10].

2.3. Activity concentration calculation

The activity concentrations of each radionuclide in the sample were calculated using the following equation (1)

$$A = \frac{N_{net}}{\varepsilon(E_\gamma) \cdot P_\gamma \cdot t \cdot m} \qquad (1)$$

where A is the activity concentration, expressed in $Bq\ kg^{-1}$, N_{net} is the net counts of peak after background subtracted, $\varepsilon(E_\gamma)$ is the absolute efficiency in the corresponding peak to energy Eγ, P_γ is the gamma-emission yield, t is the counting time and m is sample mass in kilogram [11]. The natural radionuclides of ^{226}Ra, ^{232}Th, ^{40}K were detected and the activity concentration was determined for every sample, respectively from the equation (1).

Assuming secular equilibrium was sufficient in the uranium and thorium decay series, the activity concentration of ^{226}Ra and ^{232}Th activities were calculated indirectly via the activities of their daughter products. Concretely, the activity of ^{226}Ra was calculated, by the averages of activities of decay daughters ^{214}Pb and ^{214}Bi, in energy peaks 295.2 and 351.9 keV for ^{214}Pb and for ^{214}Bi energy peaks are taken 609.3 keV and 1120.29 keV. The activity concentration of ^{232}Th was calculated by the activity of ^{228}Ac in energy peaks 338.4 and 911.2 keV. The true coincidence summing (TCS) effect was considered in the activity determination. The LabSOCS efficiency calibration applies to TCS corrections for cascade gamma emitters ^{214}Pb, ^{214}Bi and ^{228}Ac. The activity concentration of ^{40}K was calculated in the key line 1460.8 keV. All gamma-emission intensity, and gamma lines energy were taken from the Nuclide-LARA library [12]. Minimum detectable activity (MDA) was performed by using the Currie method [13].

2.4. Assessment of radiological parameters

In this section, the focus is the evaluation of radiological parameters such as radium equivalent activity Ra_{eq}, internal hazard index H_{in} and the annual effective dose (AED), to assess the radiological implications of wheat flour consumption. Ra_{eq} is used as a weighted sum of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, providing a convenient measure of the overall gamma radiation potential. The hazard index was applied to estimate the internal radiation risks, while AED represents the annual effective dose received by individuals due to ingestion. Together, these parameters allow a quantitative assessment of the radiological safety associated with wheat flour in daily human consumption.

The radium equivalent activity Ra_{eq} is a widely used radiological hazard index that represents a single quantity describing the combined gamma radiation output from ^{226}Ra, Th and ^{40}K. It allows the comparison in terms of their potential radiological risk. The radium equivalent activity is calculated using the activity concentrations of these radionuclides as follows [14]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \qquad (2)$$

where: A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The recommended safety limit for radium equivalent activity is 370 Bq/kg, corresponding to an annual effective dose of approximately 1 mSv for the public [14].

The internal hazard index H_{in} is a radiological risk indicator used to assess the potential internal exposure to radiation due to the ingestion of radionuclides. It is calculated using the activity concentrations of radionuclides as follows by equation (3) and the permissible values should be less than unity [14]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{Ra}}{4810} \qquad (3)$$

The denominators 185, 259, and 4810 are the maximum activity concentration limits in Bq kg^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, corresponding to a safe internal exposure level.

The annual effective dose (AED) is used to estimate the radiation dose received by an individual due to the ingestion of natural radionuclides present in food. In this study, the annual effective dose was calculated for each of the 16 wheat flour samples using the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, to assess the potential radiological risk associated with their consumption. It is calculated using the following formula, given in equation (4) [2].

$$(AED)_j = \sum_i I_j \times A_{ij} \times C_i \qquad (4)$$

$(AED)_j$ is dose of sample flour j, expressed in $\mu Sv\ y^{-1}$, A_{ij} is the activity of radionuclide i (^{226}Ra, ^{232}Th and ^{40}K) in $Bq\ kg^{-1}$, I_j is the annual intake consumption rate of flour, in kg/year and C_i is dose conversion factors for every radionuclide expressed in nSv/Bq. The conversion factor varies depending on radionuclides and the age of the individual. The conversion factors for adults are: 0.28 μSv Bq^{-1} for ^{226}Ra, 0.23 μSv Bq^{-1} for ^{232}Th and 0.0062 μSv Bq^{-1} for ^{40}K [15]. The international reference values for the annual dose from basic foods are in the range 200-1000 μSv y^{-1} by the report UNSCEAR (2024) and the allowable limits of 1000 μSv y^{-1} by the report ICRP (2007) and WHO (2021) for all age groups [16, 17].

3. Results and discussion

3.1. Activity concentrations in wheat flour samples

Table 1 presents the activity concentrations of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K, along with their associated uncertainties (±1σ). No artificial radionuclides were detected in any of the measured wheat flour samples. The Minimum Detectable Activities (MDA) for ^{226}Ra, ^{232}Th, and ^{40}K were determined to be 0.02, 0.01, and 4.20 Bq kg^{-1}, respectively.

The activity concentration of 226Ra ranged from 3.8 ± 0.2 Bq kg^{-1} (S2) to 10.6 ± 0.9 Bq kg^{-1} (S10), with an average value of 6.3 Bq kg^{-1}. For 232Th, the activity concentrations varied from below the Minimum Detectable Activity (MDA) to 0.30 ± 0.09 Bq kg^{-1} (S8), with an average of 0.25 Bq kg^{-1}. As shown in Table 1, ^{232}Th was detected in only three samples, and its activity was relatively low compared to the other radionuclides; in the remaining samples, it was below the MDA. The activity concentration of ^{40}K ranged from 38.3 ± 2.7 Bq kg^{-1} (S10) to 61.5 ± 2.3 Bq kg^{-1} (S8), with an average value of 45.9 Bq kg^{-1}.

Table 1. Activity concentration of radionuclides of ^{40}K, ^{226}Ra and ^{232}Th in sixteen samples of flour.

No.	Sample	Activity Concentration (Bq kg^{-1})		
		^{40}K	^{226}Ra	^{232}Th
1	S1	57.8±3.1	5.4±0.4	< MDA
2	S2	39.4±2.1	3.8±0.2	0.25±0.07
3	S3	48.1±2.6	4.5±0.3	< MDA
4	S4	48.1±2.6	4.5±0.3	< MDA
5	S5	40.8±3.1	3.8±0.4	< MDA
6	S6	41.9±2.3	6.1±0.5	< MDA
7	S7	61.0±2.2	4.4±0.7	< MDA
8	S8	61.5±2.3	6.4±0.5	0.3±0.09
9	S9	40.4±3.0	7.2±0.7	< MDA
10	S10	38.3±2.7	10.6±0.9	< MDA
11	S11	39.0±3.1	5.0±0.8	< MDA
12	S12	41.5±2.4	7.6±0.7	< MDA
13	S13	40.4±3.3	6.2±0.8	< MDA
14	S14	41.6±2.3	9.5±0.9	< MDA
15	S15	47.4±3.5	7.3±0.7	< MDA
16	S16	46.5±2.5	8.5±0.7	0.20±0.07

Among the radionuclides analyzed, ^{40}K exhibited the highest activity concentrations. This observation is expected, reflecting both the natural abundance of ^{40}K and the potential contribution from the use of phosphate fertilizers in the fields.

The levels of activity concentration of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in this study were comparable to those in the literature values studied elsewhere except for the values of Saudi Arabia, Turkey, Iraq, Spain and India [5, 6]. The lower levels of activity of ^{226}Ra, ^{232}Th and ^{40}K compared to some literature values obtained in different countries may be due to the dissimilarities of geological formation and condition, fertilizers methods, and the process of physical and chemical in soil in different countries.

The results of this study indicate that the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in all wheat flour samples were below the internationally recommended limits for food ingestion. The average activity concentrations were 6.3, 0.25, and 45.9 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, compared to the corresponding worldwide average values of 33, 42, and 512 Bq kg^{-1} [2].

Although the measured activity concentrations are lower than the global averages, indicating that the wheat flour is generally safe for consumption, periodic monitoring is recommended to ensure continued safety. Such measures also contribute to establishing a reference database for future assessments of radioactivity levels in foodstuffs.

3.2. Radiological risk assessment of wheat flour samples

The radiological parameters, including radium equivalent activity Ra_{eq}, the internal hazard index (H_{in}, and the annual effective dose AED, were evaluated and are presented in Table 2. The calculations were performed in accordance with Equations (2), (3), and (4). These parameters were estimated based on the measured activity concentrations of the investigated radionuclides. The obtained results provide an assessment of the potential radiological impact associated with the consumption of wheat flour.

Table 2. Radium equivalent, internal hazard index and annual effective dose in sixteen samples of flour.

Sample	Ra_{eq} (Bq kg^{-1})	H_{In}	AED (μS y^{-1})
S1	9.88	0.041	244.16
S2	7.60	0.032	192.46
S3	8.60	0.036	216.82
S4	8.60	0.036	216.82
S5	7.27	0.031	183.22
S6	9.99	0.045	279.94
S7	9.21	0.037	213.41
S8	12.33	0.053	319.38
S9	11.53	0.054	338.85
S10	14.00	0.068	433.24
S11	8.56	0.038	233.84
S12	10.88	0.050	313.11
S13	10.21	0.047	290.95
S14	13.24	0.063	398.74
S15	11.16	0.050	311.81
S16	13.08	0.060	378.99

The radium equivalent activity Ra_{eq} was calculated for the sixteen wheat flour samples to evaluate their overall gamma radiation hazard. The obtained Ra_{eq} values ranged from 7.27 to 14.00 Bq kg^{-1}, with a mean value of 10.38 Bq kg^{-1}. These values are substantially lower than the widely accepted reference limit of 370 Bq kg^{-1}, which corresponds to an annual effective dose of approximately 1 mSv y^{-1} for the public. The measured Ra_{eq} values in all analyzed flour samples represent less than 4% of this reference limit.

The internal hazard index H_{in} was calculated for the wheat flour samples to assess the potential internal radiological risk associated with the presence of natural radionuclides detected. The obtained H_{in} values ranged from 0.031 to 0.068, with a mean value of 0.046. All calculated values are significantly lower than the recommended safety criterion of $H_{in} \leq 1$, which represents the threshold below which internal radiation exposure is considered radiologically acceptable for the public. The maximum recorded value 0.068 corresponds to less than 7% of the reference limit.

These results clearly indicate that the investigated wheat flour samples do not pose any appreciable internal radiological hazard. The very low H_{in} values reflect the low activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the analyzed samples.

The mean annual effective dose AED due to the ingestion of natural radionuclides in the analyzed wheat flour samples was calculated to be 285.36 μSv y^{-1}. Although wheat flour consumption varies among countries and individuals, a value of 130 kg per year was adopted in this study based on the dietary profile reported by UNSCEAR (2024). This value is within the worldwide reference range of 200–1000 μSv y^{-1} reported by UNSCEAR (2024) for ingestion exposure from natural sources. Furthermore, the obtained mean dose is well below the recommended public dose limit of 1 mSv y^{-1} established by ICRP (2007) and adopted by WHO (2021). The calculated value corresponds to approximately 28.5% of the recommended annual dose limit for members of the public.

The results obtained for activity concentrations, radium equivalent activity, internal hazard index, and annual effective dose are comparable to those reported in similar studies [5, 6, 14, 18, 19, 20]. The observed variations in the evaluated parameters are expected, as activity concentrations are influenced by several factors. These include the geochemical characteristics of the soils in which the crops were cultivated, climatic conditions, physicochemical processes, local geology, and the use of chemical fertilizers. In addition, differences in the assumed consumption rates adopted for the calculation of the annual effective dose may further contribute to the variability among studies conducted in different countries.

However, all calculated values are lower than the international reference limits previously mentioned. Therefore, the ingestion of the investigated wheat flour does not represent a significant radiological health concern.

4. Conclusion

The levels of natural radioactivity in sixteen brands of wheat flour samples commonly consumed in Albania were investigated. The radionuclides of ^{226}Ra, ^{232}Th and ^{40}K were detected and the activity concentration of them was calculated. The results of activity concentration are lower than the worldwide average values, defined by UNSCEAR 2024 report.

The calculated radiological parameters, including radium equivalent activity, internal hazard index, and annual effective dose, were all found to be well below the internationally accepted reference and safety limits. The obtained Ra_{eq} values were significantly lower than the recommended limit, while all H_{in} values were far below the safety criterion of ≤ 1. In addition, the mean annual effective dose remained within the worldwide reference range and below the public dose limit of 1 mSv y^{-1}.

Based on the evaluation of these radiological indices, it can be concluded that the wheat flour samples analyzed present a low and acceptable level of radiological risk. Therefore, from a radiological protection perspective, the consumption of the investigated wheat flour does not pose any significant health concern to the public.

The results show that the daily consumption of these brands of wheat flours is safe and did not show any significant radiological risk for the public living in Albania.

Acknowledgements: *The authors would like to thank the Institute of Applied Nuclear Physics for support of this research.*

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