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ASSESSMENT OF RADIOLOGICAL HAZARD FOR VARIOUS FOOD COMMONLY USED IN REPUBLIC OF NORTH MACEDONIA

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Abstract

Consuming food containing radionuclides is particularly dangerous. If anyone ingests or inhales a radioactive particle, it continues to irradiate the body as long as it remains radioactive and stays in the body. However, studies on the radioactivity of consumable foods assume importance as it is necessary to estimate the ingestion dose to the public. Due to all this, the focus of this research was on determining the activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th. Forty-nine samples in three categories of vegetables, cereals (rice, wheat, corn), and milk, were collected from local markets (city of Skopje) in the Republic of North Macedonia and they were analysed by using high-purity germanium (HPGe) detector to assess natural and artificial radioactivity. The average activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th of the tested samples were 2.85±1.15, 2.48±0.85, and 80.64±5.45 Bq kg^{-1}, respectively. No artificial radionuclide was found in any of these samples. The average value of the radium equivalent activity in all samples was 12.56 Bq kg^{-1}, which was less than the maximum permitted value of 370 Bq kg^{-1}. The values of the external hazard indices for vegetables, cereals and milk samples vary with an average value of 0.11, which is less than one in all samples indicating the non-harmfulness of the samples. The mean activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th (Bq kg^{-1}) in the samples were used to calculate the absorbed dose rate whose mean value for all food samples was 6.16 Bq kg^{-1}. It was determined that the measured values are within the globally accepted values, i.e., they are quite lower than the data in literature. These data would be useful to establish a baseline for natural radioactivity concentrations in food products consumed in the Republic of North Macedonia.

Key words: *natural spectrometry, food, gamma spectrometry, radiation risk*

INTRODUCTION

Humans are constantly exposed to natural radioactivity at different levels depending on the natural radioactive elements present in each area; therefore, researchers are studying natural environmental radiation and radioactivity in order to conduct background checks and detect environmental radioactivity (Radhakrishna et al., 1993). Considering that radionuclides occur naturally in both rocks and soils, when these rocks decompose by natural processes, radionuclides are transported to the soil by rain and lowlands (Agbalagba & Onoja.,2011; Essiett et al., 2015) By means of migration, the present radionuclides can easily accumulate in the food chain through the soil - plants and the human (Skwarzec & Falkowski.,1988). Due to this migration, radioactivity has always been present in all food products to some extent. In general, anthropogenic radionuclides in food products originate from the effects introduced into the biosphere. In fact, radionuclides can cause a number of health conditions and diseases as a result of their exposure, hence resulting in bio-accumulation and bio-toxicity (Ferdous et al., 2015). Therefore, the IAEA and international experts (Natural and induced radioactivity in food IAEA-TECDOC-1287, 2002) are developing guidelines for measuring and determining acceptable levels of natural radioactivity in food, with the ultimate goal of improving food safety. Many studies have been conducted on the concentration of radioactivity in food products (Alsaffar et al., 2015) (Angeleska et al., 2021) where the natural radioactivity in food is usually within the range from 40 to 600 Bq kg^{-1} of food (IAEA, 2002). This work aims to identify radionuclides in certain food samples that are commonly used in households in the Republic of North Macedonia, by measuring the amount of specific concentrations of ^{226}Ra, ^{232}Th and ^{40}K. Based on these results, the potential radiological health risks associated with the consumption of these analysed types of food were assessed.

MATERIAL AND METHODS

- Sample collection and preparation

A total of 66 samples of locally widely consumed food products were collected from various markets in the Republic of North Macedonia. The main food selection was based on a survey questionnaire on the diet of 30 residents. The main food groups were selected, including wheat, rice, and milk. The samples were prepared fresh, i.e., they were homogenized and packaged in standard Marinelli cups (500 g) and sealed in order to achieve a radioactive balance between the parents and their daughter radionuclides. (IEEE,1997; Walley El-Dine et al., 2001)

- Sample analysis

The analytical techniques for gamma-ray spectrum were used for determination of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K. This measurement system includes a typical high-resolution gamma-spectrometer based on a high-purity germanium detector with energy resolution of 1.80 keV full width at half maximum for the gamma ray line of 1332 keV from ^{60}Co. According to the energy of the photopic line, the activities of ^{232}Th were measured by taking the mean activity of the photopeaks of the daughter nuclides ^{228}Ac (338.40, 911.07 and 968.90 keV) and 212 Pb (238.63 keV). The activities of ^{226}Ra were calculated with ^{226}Ra. keV, line energy of ^{214}Bi. The activities of ^{40}K were determined directly from its gamma emission at 1460.83 keV. (Righi et al.,2009) The counting time for each sample was 108 000 seconds.

- The activity concentration

For all samples, the corresponding specific activities according to the methods of Shanthi were determined (Shanthi et al., 2009) as shown in Equation 1.

$$A = \frac{\frac{N}{t} - \frac{N_0}{t_0}}{\varepsilon \cdot \gamma \cdot m} \quad (Bq \cdot kg^{-1}) \ (1)$$

Where, N is clean surface of peak accumulated from a specific radionuclide in analysis of a specific sample (number of readings), N_0 is clean surface of peak accumulated from the spot of a specific radionuclide without an analysis of sample (number of readings), t is live time of accumulation of the sample spectrum (s), t_0 is live time of accumulation of the phone spectrum (s), ? is detector efficiency for a given energy (for a specific peak), ? is intensity of gamma transition in radioactive decay for a respective radionuclide (%), and m is sample mass (kg).

- <u>Radiological hazard assessment</u>

- Assessment of Radium Equivalent

Gamma-ray radiation hazards caused by specific radionuclides of ^{226}Ra,^{232}Th, and ^{40}K were evaluated using different indices, among which, the radium equivalent activity is the most widely used radiation hazard index (Beretka et al.,1985) It is determined by the weighted sum of activities of the three radionuclides, based on the assumption that 370 Bq, 259 Bq, and 481 Bq produce the same gamma-ray dose rate as given by (Kessaratikoon & Awaekechi, 2008)

$$Ra_{eq}(Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.07A_k \ (2)$$

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

To keep the annual radiation dose below 1.5 m Gy y^{-1}, the maximum value must be less than 370 Bq kg^{-1}.

- Absorbed Dose Rate in Air

The absorbed dose rates (nGy h–1) in outdoor air from global gamma radiation at 1 m above the ground were analysed by the next equation (Özmen et al.,2014; Curtin et al., 2008)

D (nGy / h) = 0,462 A_{Ra} + 0,604 A_{Th} +0,0417 A_k (3)

- External hazard index

The internal and external hazard indexes are calculated by the following expressions (Rafique et al., 2013)

$H_{eks} = A_{Ra}/370 + A_{Th}/259 + A_k/4810 \leq 1$ (4)

A_{Ra}, A_{Th}, A_k - specific activities (Bq/kg), ^{226}Ra ^{232}Th and ^{40}K, respectively

- Ingestion effective dose

The Ingestion effective dose due to the intake of ^{238}U, ^{233}Th and ^{40}K in foods can be evaluated using the following expression ICRP (ICRP., 2006).

$HT,r = \sum_i U_i \times C_{i,r} \times gT,r$ (5)

where, *i* denotes a food group, the coefficients *Ui* and $C_{i,r}$ denote the consumption rate (kg/y) and activity concentration of the radionuclide *r* of interest (Bq kg^{-1}), respectively, and $g_{T,r}$ is the dose conversion coefficient for ingestion of radionuclide *r* (Sv Bq^{-1}) in tissue *T*. For adult members of the public, the recommended dose conversion coefficient $g_{T,r}$ for ^{40}K, ^{226}Ra (^{238}U), and ^{232}Th, are 6.2×10^{-9}, 2.8×10^{-7} and 2.2×10^{-7} Sv Bq^{-1} respectively IAEA (IAEA., 1996). In fact, the aggregate quantities for Average annual intake for adults in the Republic of North Macedonia by groups are: vegetables 44.5 kg, cereals 75.7 kg, legumes 8 kg and milk 35l kg.

- Annual effective dose equivalent (AEDE)

The annual effective dose equivalent received was computed from absorbed dose rate by applying a dose conversion factor of 0.7 Sv Gy^{-1} and the occupancy of 0.8 (19/24 h) recommended by UNSCEAR. Therefore, the annual effective dose equivalent (μSv y^{-1}) was calculated using the equation 6 (UNSCEAR., 2000) AEDE (μSv y^{-1}) = absorbed dose (nGy h^{-1}) $\times 8760$ h $\times$ 0.7Sv Gy$^{-1} \times 0.8 \times 10^{-3}$ (6)

RESULTS AND DISCUSSION

Table 1. Mean values of specific activities (A) of values of ^{226} Ra, ^{232}Th and ^{40}K in foods.

Sampling sites	A±SD (Bq kg^{-1}) ^{226}Ra	A±SD (Bq kg^{-1}) ^{232}Th	A±SD (Bq kg^{1}) ^{40}K
Potatoes (n=5)	0.41±0.15	1.96±0.05	92.15±5.50
Tomatoes (n=5)	0.71±0.05	1.26±0.02	102.50±6.50
Beans (n=7)	0.24±0.01	0.95±0.02	69.70±3.00
Apple(n=7)	0.65±0.20	1.41±0.50	25.35±3.50
Rice (n=7)	6.02±2.20	3.90±1.50	67.55±2.55
Wheat (n=7)	0.62±0.05	0.25±0.03	155.54±5.30
Corn (n=7)	1.92±1.00	2.19±1.00	37.02±5.90
Pea (n=7)	12.56±6.09	9.47±4.20	78.85±7.50
Red Lentil (n=7)	3.58±1.52	2.38±1.01	142.87±10.25
Milk (n=7)	1.83±0.31	1.04±0.15	31.87±4.53
Average	2.85±1.15	2.48±0.84	83.52±5.46

The specific activity due to ^{226}Ra, ^{232}Th and ^{40}K in different types of food was measured as presented in Table 1. The specific activity of ^{226}Ra is found within the range from 0.24 ± 0.01 Bq kg^{-1} to 12.56 ± 6.09 Bq kg^{-1}, the one of ^{232}Th from 0.25 ± 0.03 Bq kg^{-1} to 9.47 ± 4.20 Bq kg^{-1} and the specific activity of ^{40}K was within the range from 25.35 ± 3.50 Bq kg^{-1} to 142.87 ± 10.25 Bq kg^{-1}. There is a variation in the specific activity of radionuclides in different food samples where it can be seen that in wheat, the concentration value of ^{40}K is higher than in other food products. The highest concentration level of the ^{226}Ra and ^{232}Th radionuclides were in pea and red lentils, respectively. The activity concentration of ^{226}Ra in cereal crops (wheat and rice) was higher than the UNSCEAR reference values of 0.08 Bq/kg (UNSCEAR, 2000). The source of contamination of ^{226}Ra, ^{232}Th and ^{40}K of rice and wheat is considered to be due to the absorption of soil by the plant roots and irrigation water through the root and also the irrigation period of wheat and rice is longer. The obtained average activity concentration in milk for ^{40}K was 31.87 Bq kg^{-1}, which is lower when compared to other results (ICRP,1996).

Table 2. Obtained values from the absorbed dose rate in the air (D), the radiation risk index (H_{eks}), the radium equivalent (Ra_{eq}), and AEDE.

Sample	D (nGy h^{-1})	Ra_{eq} (Bq kg^{-1})	Hex	AEDE (µSv y^{-1})
Potatoes	5.21	9.66	0.02	25.55
Tomatoes	5.35	9.68	0.02	26.24
Beans	3.58	6.46	0.01	17.56
Apple	2.20	4.43	0.01	10.79
Rice	7.94	16.31	0.04	38.95
Wheat	6.91	11.85	0.03	33.89
Corn	3.74	7.64	0.02	18.34
Pea	14.79	31.61	0.08	72.55
Red Lentil	9.03	16.89	0.04	44.29
Milk	2.78	5.54	0.01	13.63
Average	6.15	12.07	0.03	30.17

In this study, the values of the absorbed dose rate D (nGy h^{-1}) did not exceed the safety limits, emphasizing the negligible radiation hazard arising from naturally occurring terrestrial radionuclides. The radium equivalent activity values in all food samples vary with a mean value of 12.07 Bq kg^{-1} which is far below the internationally accepted value of 370 Bq kg^{-1}. On the other hand, the values of the hazard index varied from 0.01 to 0.08 with an average value of 0.03. The annual effective dose equivalent has been calculated from 10.79 µSv y^{-1} to 44.29 µSv y^{-1} with an average value of 30.17 µSv y^{-1} respectively which is less than the recommended value of the IAEA, which is 1000 µSv y^{-1}.

Table 3. Estimated radiation hazard indices and ingestion effective dose in food at the markets in North Macedonia.

Sample	^{226}Ra	^{232}Th	^{40}K	Sum
Potatoes	0.0051	0.019	0.025	0.044
Tomatoes	0.0088	0.012	0.028	0.048
Beans	0.0005	0.001	0.003	0.004
Apple	0.008	0.013	0.006	0.027
Rice	0.013	0.006	0.031	0.050
Wheat	0.013	0.004	0.070	0.087
Corn	0.040	0.036	0.017	0.093
Pea	0.028	0.016	0.003	0.047
Red Lentil	0.008	0.004	0.007	0.019
Milk	0.001	0.008	0.006	0.015

Table 3 shows the results of the effective dose of ingestion in mSv y^{-1} for an adult person due to specific activity of ^{226}Ra, ^{232}Th and ^{40}K in food samples calculated by using the equation (4). The sum of the effective ingestion doses varies from 0.004 mSv y^{-1} (in the beans sample) to 0.093 mSv y^{-1} (in the corn sample). The average effective ingestion dose due to ^{40}K was higher than due to ^{232}Th and ^{40}K as a result of the increased dose conversion ratio for radionuclide ingestion. This indicates that the effective ingestion dose in all food samples is lower than the permitted limits of 1 mSv y^{-1} recommended by the ICRP.

CONCLUDING REMARKS

The activity of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides was determined for the most available food products consumed in the Republic of North Macedonia. It was determined that the measured values are within the world range as published in this literature. The mean activity values of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides in 66 food products were identified as 2.85±1.15 Bq kg^{-1}, 2.48±0.84 Bq kg^{-1} and 83.52±5.46 Bq kg^{-1}, respectively. All calculated values for radiological hazard assessment are lower than the global average values. The dose of different components reduces in the following order corn>wheat>rice>vegetables>milk It should be noted that the geology of the region and the food processing methods have a major impact on the ^{40}K concentration. This study established a map of basic information about the future studies on radiation levels and the distribution of radionuclides in food products in the Republic of North Macedonia. The results of the study will also be used as a reference for future assessment.

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ОЦЕНКА НА РАДИОАКТИВНАТА ОПАСНОСТ ОД РАЗЛИЧНА ХРАНА КОЈА Е НАЈЧЕСТО КОРИСТЕНА ВО РЕПУБЛИКА СЕВЕРНА МАКЕДОНИЈА

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Резиме

Консумирањето храна што содржи радионуклиди е особено опасно. Ако некој внесе или вдиши радиоактивна честичка, таа продолжува да го зрачи телото сѐ додека останува радиоактивна и останува во телото. Сепак, студиите за радиоактивноста на потрошната храна имаат значење бидејќи е неопходно да се процени дозата за голтање на јавноста. Поради сето ова, фокусот на ова истражување беше на одредување на концентрациите на активност на ^{226}Ra, ^{40}K и ^{232}Th. Четириесет и девет примероци во три категории зеленчук, житарки (ориз, пченица, пченка) и млеко беа собрани од локалните пазари (Град Скопје) во Република Северна Македонија и беа анализирани со употреба на германиум со висока чистота (HPGe) детектор за проценка на природна и вештачка радиоактивност. Просечните концентрации на активност на ^{226}Ra, ^{40}K и ^{232}Th од тестираните примероци беа 2,85±1,15, 2,48±0,85 и 80,64±5,45 Bq kg^{-1}, соодветно. Во ниту еден од овие примероци не беше пронајден вештачки радионуклид. Просечната вредност на активноста на еквивалент на радиум во сите примероци беше 12,56 Bq kg-1, што беше помало од максималната дозволена вредност од 370 Bq kg^{-1}. Вредностите на надворешните индекси на опасност за примероците од зеленчук, житарки и млеко варираат со просечна вредност од 0,11, што е помалку од една во сите примероци што укажува на нештетноста на примероците. Просечните концентрации на активност на ^{226}Ra, ^{40}K и ^{232}Th (Bq kg^{-1}) во примероците беа искористени за пресметување на стапката на апсорбирана доза чија средна вредност за сите примероци на храна беше 6,16 Bq kg^{-1}. Утврдено е дека измерените вредности се во рамките на глобално прифатените вредности, односно се доста пониски од податоците во литературата. Овие податоци би биле корисни за утврдување на основната линија за концентрациите на природна радиоактивност во прехранбените производи што се консумираат во Република Северна Македонија.

Клучни зборови: *природна радиоактивност, храна, гама спектрометрија, ризик од зрачење*