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ASSESSMENT OF RADIOACTIVITY AND RADIOLOGICAL HAZARD FROM NATURAL RADIONUCLIDES CONTAINED IN RICE FROM NORTH MACEDONIA

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Abstract

Studies on radioactivity in consumable food are becoming increasingly important from a safety perspective as it is necessary to estimate the dose of ingestion by the public. For this reason, the focus of this study was on determining the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K. Thirty-eight rice samples were collected during 2024 from rice fields (city of Kochani) in the Republic of North Macedonia and the samples were analyzed by using a high-purity germanium (HPGe) detector for assessment of natural and artificial radioactivity. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K of the tested samples were 2.17±0.70, 1.78±0.72, and 48.48±1.92 Bq/kg, respectively. No artificial radionuclide was found in any of these samples. The total absorbed dose (D) was 4.10 D (nGy h-1), while the average value of the radium equivalent activity in all samples was 8.10 Bq kg-1, which was less than the maximum permitted value of 370 Bq kg-1. The values of Hex, Hin and $I\gamma$ for the samples were 0.02, 0.027 and 0.064, which are less than one in all samples indicating the harmlessness of the samples. The average activity concentrations of ^{226}Ra, ^{40}K, and ^{232}Th (Bq kg-1) in the samples were used to calculate the annual effective dose equivalent (AEDE), whose average value for all samples was 0.005(μSv/y). The average calculated cancer risk due to the intake of the tested radionuclides found in the rice samples was estimated at 0.017, which is within the range of acceptable risk values recommended by USEPA. The results of this study indicate that the intake of radionuclides due to consumption of the mentioned food does not have a harmful effect on public health. These data would be useful for establishing a baseline for the concentrations of natural radioactivity in food products consumed in the Republic of North Macedonia.

Keywords: *gamma spectrometry, radiation risk, rice, cancer risk.*

Introduction

Researchers study the natural environmental radiation and radioactivity in soils to conduct background checks and detect environmental radioactivity (Angeleska et al., 2023). Radioactivity levels can be used to estimate public dose rates and radioactive contamination, and to predict changes in environmental radioactivity caused by nuclear accidents, industrial activities and other human activities (UNSCEAR,2000). Primordial radionuclides comprise of natural series such as ^{238}U, ^{232}Th, and the ^{40}K series, which are usually long-lived and have half-lives of more than 100 million years (UNSCEAR, 2000). These radionuclides can be found in all elements of the environment and are present in varying amounts in soil, water, air, vegetables, animals, and the human body itself (IAEA, 1989). Long-lived natural radionuclides can be transferred to plants, considering that the main part of the minerals that make up the plant body originates from the soil. Therefore, primarily the physical and chemical characteristics of the soil are the main parameters that determine the amount of accumulated radioactive substances in plant organs. The levels of radionuclides in plants usually vary from several tens of Becquerel (Bq) to several hundred Becquerel per kilogram. Some plants are capable of incorporating large quantities of radioactive substances into their tissues without visible and demonstrable changes; however, their consumption can cause serious damage and diseases in human organisms. With the increase in the world population, the need for a larger amount of food increases, which encourages many countries to use phosphate fertilizers in plant production to increase their annual production. Fertilizers also contain radionuclides that are the decay series of uranium and thorium, as well as potassium. Farmers are increasingly using artificial fertilizers without expert assessment of the content of present radionuclides in the fertilizers. Therefore, there is a global interest in human radiation exposure due to intake of radionuclides from food (Alrefae, 2012). Rice is one of the most commonly consumed types of food in the world. Hence, studies on the radioactivity of rice have been carried out in different regions around the world. Rice also has an important share in the Macedonian diet, especially in the Kochani region, which is known for its rice fields. Therefore, this research was conducted to investigate the levels of radioactivity as a result of the natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in rice, and to estimate the total absorbed dose, radium equivalent activity, external and internal hazard index, and gamma index. Based on the average concentrations of the examined radionuclides, an annual effective dose (AEDE) was determined, as well as the risk of cancer due to the intake of the examined radionuclides. The research was conducted in the Kochani region (Republic of North Macedonia), which, due to its favorable geographical location and mild continental and Mediterranean climate, contributes to the production of larger quantities of rice, which is often used in the diet of the population. Gamma spectrometry was used as a technique, while the analysis of the spectral data was carried out with the Genie 2000 software developed by CANBERRA.

Materials and methods

The study area, sampling and preparation

For the purpose of the study, a total of 38 rice samples were collected from 10 different locations. These locations were selected based on the availability of fields in the Kochani region. Rice plants were randomly selected from each location during harvest. All collected samples were cleaned of soil deposits by hand and then transferred to a polyethylene bag, sealed, and labeled in order to avoid cross-contamination. The samples were then transferred to beakers (Marinelli), sealed with adhesive rubber, weighed, and left to stand for several days to enable secular equilibrium between radon (^{222}Rn) and ^{226}Ra nuclides (Panghal et al., 2018). The prepared samples were analyzed with a high-purity germanium (HPGe) detector.
Experimental set-up of gamma ray spectroscopy

The research is focused on determining the radioactivity level in rice by using a high-resolution HPGe detector. The gamma-ray spectrometry technique was used to determine the radioactivity of the tested samples. The spectrometer consisted of an HPGe detector, model 3020 (Canberra Packard, Meriden, CT, USA), with an active volume of 180 cm2, a relative efficiency of 30%, an operating voltage of 3000 V, and a resolution of 2 keV at 1332.5 keV. The detector was enclosed in a massive lead shield with a thickness of 12 cm and an inner high-purity copper sheath of 2 mm. The analysis of the spectral data was carried out with the Genie 2000 software developed by CANBERRA, and the contribution of background radiation during the counting was subtracted during the spectral analysis. The activity of ^{226}Ra was determined from the gamma lines associated with the short half-life daughters of ^{214}Bi (609.31, 1120.29 and 1794.49

keV) and ^{214}Pb (351.93 keV). The 232 activity was determined with the 338.4, 911.2, and 969.1 keV gamma lines of ^{228}Ac and its decay products. The gamma line at 1460.8 keV was used to determine the activity of ^{40}K (Hauwa et al., 2018).
The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K was calculated by using the following relation

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

Where, N is the clean surface of the peak value accumulated from a specific radionuclide in the analysis of a specific sample (number of readings), N_0 is the clean surface of the peak value accumulated from the spot of a specific radionuclide without an analysis of the 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), ε is detector efficiency for a given energy (for a specific peak), γ is intensity of gamma transition in radioactive decay for a respective radionuclide (%), and m is sample mass (kg).

Results and discussion

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

Table (1) shows the activity concentrations of the examined natural radionuclides in rice samples in the Kochani region.

Table 1. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in rice

Sampling sites	A±SD (Bq kg^{-1}) ^{226}Ra	A±SD (Bq kg^{-1}) ^{232}Th	A±SD (Bq kg^{1}) ^{40}K
WR1(n=3)	4.02±2.20	2.90±1.50	65.53±2.50
WR2(n=2)	2.71±0.05	1.26±0.02	42.50±4.50
WR3 (n=4)	2.24±0.01	0.91±0.02	89.70±2.00
WR4 (n=2)	2.65±0.20	1.41±0.50	45.35±2.50
WR5 (n=5)	1.02±2.20	<MDA	47.55±1.55
WR6 (n=4)	2.06±0.12	<MDA	26.11±2.50
WR7 (n=3)	1.09±0.25	0.19±1.00	33.02±3.00
WR8 (n=1)	3.14±1.00	0.11±0.04	44.12±2.00
WR9 (n=1)	1.03±1.20	6.18±1.40	97.62±0.50
WR10 (n=3)	2.18±1.05	<MDA	47.32±3.00
WR11(n=4)	2.00±1.05	<0.03	24.71±2.00
WR12 (n=1)	3.91±0.11	1.24±1.30	40.50±0.50
WR13 (n=1)	2.13±0.09	<0.03	58.13±1.50
WR14 (n=1)	1.19±0.55	<MDA	28.56±0.25
WR15 (n=3)	1.17±0.35	<MDA	36.35±0.55
Average	2.17±0.70	1.78±0.72	48.48±1.92
(UNSCEAR, 2000)	0.08	0.003	-

Table 1 shows that the activity concentration of ^{226}Ra was within the range from 1.02 Bq/kg (R5) to 4.02 Bq/kg (R1) with a mean value of 2.17 Bq/kg, for the concentration of ^{232}Th it was determined that there were samples with values lower than MDA and the mean value for this

radionuclide is 1.78 Bq/kg and the ^{40}K activity concentration was within the range from 24.71 (R11) to 97.62 Bq/kg (R9) with a mean value of 48.48 Bq/kg. The average activity concentration in this study for ^{232}Th is lower than the world average value of 45 Bq/kg, the activity concentration of ^{226}Ra is lower than the world average of 32 Bq/kg, and the activity concentration of ^{40}K is also lower than the world average of 412 Bq/kg (UNSCEAR, 2008). The findings of this study indicate that the levels of natural radionuclides in the analyzed rice samples are within acceptable limits.
Almost everyone who has investigated the radionuclide content of rice has found that the activity concentration of ^{40}K radionuclides in rice tends to be higher than that of ^{226}Ra and ^{232}Th, as shown in Table 1. The increased concentration of ^{40}K compared to other radionuclides can be attributed to its prevalence in soil and the widespread application of nitrogen phosphate (Solehah and Samat, 2017).

Radiological Parameters
Gamma Absorbed Dose Rate (D)
The total absorbed dose rate (nGy/h) outdoor at 1 m above the ground due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K was calculated by using the following equation (Yassin et al., 2016)
$D(nGy/h)=0.462A_{Ra}+ 0.604\ A_{Th}+0.042\ A_k \quad (2)$
where A_{Ra}, A_K and A_{Th} accordingly are the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, expressed in Bq/kg

Radium-equivalent activity (Raeq)
The radium-equivalent activity (Raeq) is commonly used as an important radiological index to provide the exact activity levels of evaluated radionuclides in the samples due to their uneven distribution
It is calculated by using equation (3) (Abdu et al., 2024)

$$Raeq\ (Bq/kg) = A_{Ra}+1.43A_{Th}+0.07\ A_K \quad (3)$$

External hazard index (H_{eks})
In order to assess the equivalent average of the annual effective dose imposed to the residents of each area, the external hazard index for the soil samples was calculated.

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

A_{Ra}, A_{Th}, A_k-specific activites ($Bq\cdot kg^{-1}$) ,$^{226}Ra$$^{232}Th$ and ^{40}K, respectively (Abdu et al., 2024)

Internal hazard index (Hin)
Radon and its short-lived products are also hazardous to the respiratory organs. Hence, the internal exposure to radon and its short-lived products is quantified by the internal hazard index and is expressed mathematically as (Laith et al., 2015)

$$Hin = A_{Ra}/185+ A_{Th}/259 + A_k/4810\leq 1 \quad (5)$$

Gamma Index (Iɣ)
The gamma index (Iɣ) for rice samples was calculated by using the following equation (Laith et al., 2015)

$I\gamma = A_R/150 + A_{Th}/100 + A_k/1500 \leq 1 \quad (6)$

The annual effective dose equivalent (AEDE) was calculated by using equation (7):

$$AEDE\ (\mu Sv/y) = D \times DCF \times OF \times T \quad (7)$$

where D is the absorbed dose rate in air (nGy/ h), DCF is the dose conversion factor (0.7 Sv/Gy), OF is the outdoor occupancy factor (0.2), T is the time (8760 h/y) (UNSCEAR ,2000)
The **Excess Lifetime Cancer Risk** was calculated by using equation (8):

$$ELCR = AEDE \times DL \times RF \ (8)$$

where DL is duration of life (70 years) and RF is risk factor (Sv-1). For stochastic effects, ICRP 90 uses values of 0.05 for the public (Abdulkarim,et al., 2023)

Table 2. Average values of activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in rice

Radionuclides	Bq kg – 1
^{26} Ra	2.17±0.70
^{232}Th	1.78±0.72
^{40}K	48.48±1.92

Table 3. The radiological hazard parameters due to the natural radioactivity in rice fields of the Kochani region in Macedonia

Sample	D (nGy h^{-1})	Ra_{eq} (Bq kg^{-1})	Hex	Hin	$I\gamma$	AEDE (μSv/y)	ELCR
WR (1-15)	4.10	8.10	0.02	0.027	0.064	0.005	0.017

The absorbed dose in this study has an average value of 4.10 nGy/h (table 3), which is lower than the maximum permitted value of 51 nGy/h reported by (UNSCEAR,2000), emphasizing the negligible radiation hazard arising from naturally occurring terrestrial radionuclides. The values of radium equivalent activity in all rice samples varied with a mean value of 8.10 Bq kg-1 which is far below the internationally accepted value of 370 Bq kg-1. On the other hand, the values of Hex and Hin are also below the recommended value of one, respectively (UNSCEAR, 2003). The average value of Iγ (0.064) for all rice samples is lower than the critical value. The annual effective dose equivalent (AEDE) was calculated with an average value of 0.005 μSv y-1, which is lower than the recommended value. The estimated world average for AEDIndoor is 0.34 mSv/y, respectively, as recommended by (UNSCEAR,2000). The average activity concentrations of natural radioactivity in rice from this study were compared with those from similar studies carried out in other countries. Variations in distribution behavior across studies may be due to differences in location, soil and physical properties of rice, climatic conditions, and fertilizers used for crop production (Mellawati et al., 2018).

Table 4. Comparison between the natural radioactivity levels in rice samples in this study and some other studies

Study location	^{226}Ra	^{232}Th	^{40}K	Refer
Malaysia	2.8	7.5	89.3	Asaduzzaman et al., 2015
Iraq	5.94	2.65	16.84	Abdulridha et al., 2019
Namibia	0.39	0.28	13.00	Onjefu et al., 2019
India	3.07	34.3	120.8	Shanthi et al., 2009
Iran	37	-	-	Fathabadi et al., 2017
Nigeria	54.29	63.70	235.81	Muhammada et al., 2024
Saudi Arabia	1.08	1.19	83.08	Al-Zahrani, 2016
Macedonia	2.17	1.78	48.48	

Conclusion

It was found that the values of the specific radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in different rice samples are within the internationally permitted range.
All calculated values for the assessment of the radiological hazard are lower than the global average values. All tested rice samples are considered safe since there is no increase in the concentrations of natural radionuclides according to the internationally permitted limits.
However, it is necessary to conduct other similar studies on other basic food products in order to create basic data on consumed food products, as well as for the preparation of a radiological map in Macedonia.

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