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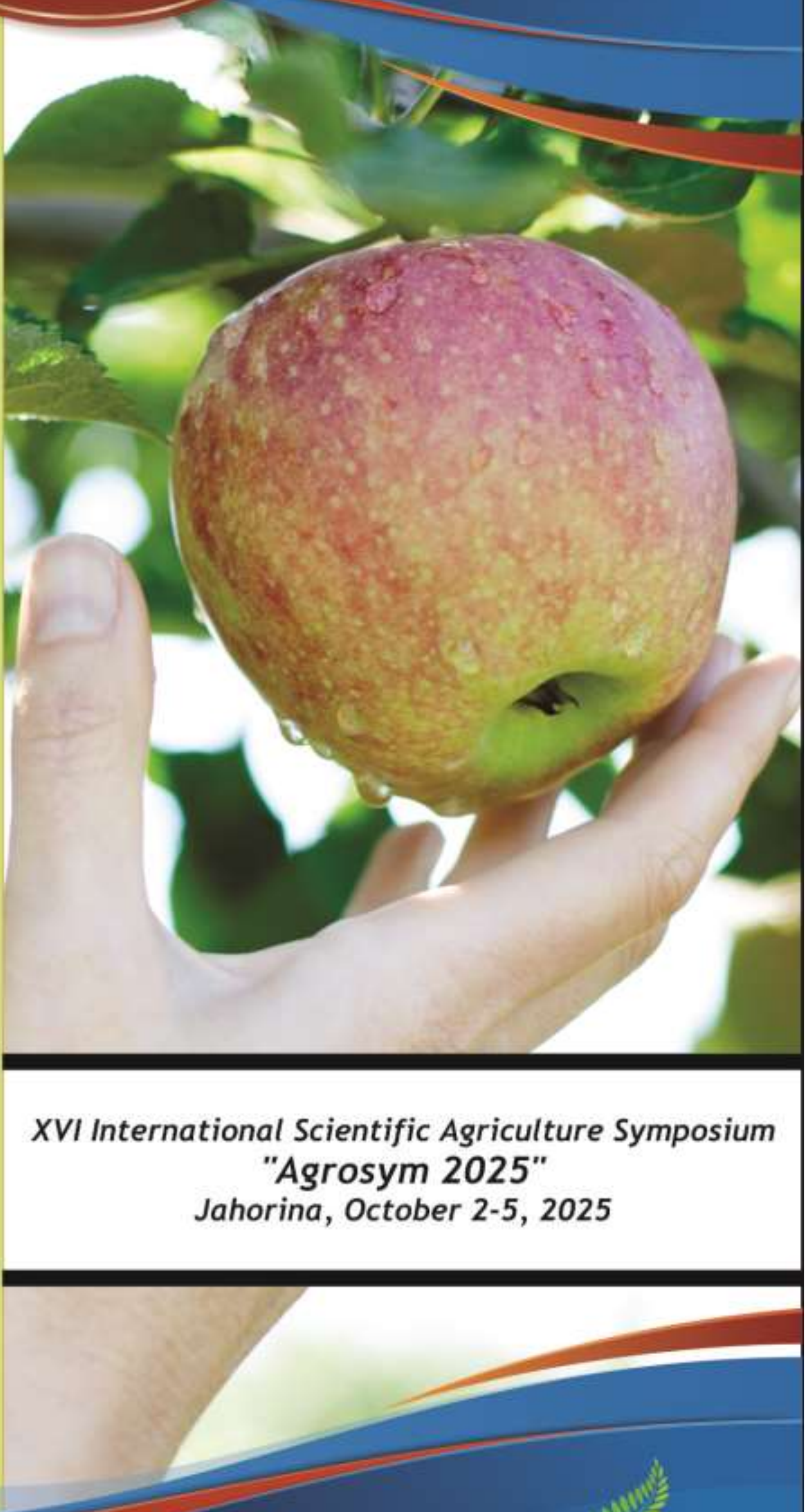

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"Agrosym 2025"
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ASSESSMENT OF NATURAL RADIOACTIVITY LEVELS IN AGRICULTURAL SOIL AND TRANSFER IN RICE IN THE KOCHANI REGION, NORTH MACEDONIA

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Abstract

Soil can transfer radionuclides to plants which are subsequently consumed by humans. Sometimes their levels are so high that they are a concern for human health. Rice has an important share in the Macedonian diet, especially in the Kochani region, which is famous for its rice fields. Therefore, the main goal of this study was to measure the natural radioactivity in agricultural soil and rice crops in the Kochani region and to study the interaction between soil and rice by calculating the transfer factor (TF). The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were determined by using gamma spectrometry, i.e. a high purity germanium (HPGe) gamma-ray spectrometer. In agricultural soils, the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were 33.72 Bq kg^{-1}, 37.62 Bq kg^{-1} and 152.90 Bq kg^{-1}, while the mean activity concentrations of rice were 1.99 Bq kg^{-1}, 2.51 Bq kg^{-1}and 49.44 Bq kg^{-1}, respectively. The world’s mean soil values of ^{226}Ra, ^{232}Th and ^{40}K are 35, 30 and 400 Bq kg^{-1}, respectively, hence these obtained values are lower compared to those obtained from research. This may be due to improved agricultural practices in the country and the geology of the area itself. However, we found that the radioisotopes have a heterogeneous distribution which must be caused by the nature and soil types in the area for which this study was conducted. However, the radioisotopes showed a heterogeneous distribution, likely influenced by the nature and soil types in the region. The mean transfer factors (TF) were very low for all samples, indicating a slow transport of these radionuclides to the rice grains. The results confirm that rice grown in these areas is safe for consumption. Further research on the TF across different rice varieties and geographical areas would allow for the development of predictive models for potential radiation exposure from rice consumption and help identify locations requiring additional monitoring.
All mean transfer factors (TF) for the samples were very low, implying a very slow transport of these radionuclides to the rice grains. The obtained results make rice grown in these areas safe for consumption. By studying the TF among different rice varieties and geographical areas, researchers will be able to develop models to predict possible radiation exposure from rice consumption and locate activities or areas that require additional attention.

Key words: *HPGe Detector, Natural radiation, Soil, Rice, Transfer factor (TF).*

Introduction

The main radionuclides that produce radiation are ^{40}K, ^{238}U and ^{232}Th (IAEA, 2004). 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. Soils and plants are the primary pathway for natural radionuclides to enter various types of seeds and, after ingestion, they directly or indirectly enter the human body through the food chain. (Spahiu et al., 2023). Soil is the first link in the ecological chain soil-food-animals-humans and therefore it has an important role in the distribution and transfer of radionuclides. The soil has a unique property of long-term accumulation and retention of a natural radionuclide introduced to it from external sources (Muhammad et al., 2024; Angjeleska et al., 2023). Radionuclides present in the environment are transferred to plants by – (i) penetration from the soil through the roots and (ii) direct absorption through the aerial parts of the plants. The former depends on the concentrations of radionuclide activity in the soil, and the latter on the rate of deposition of radionuclides from the atmosphere. One of the critical victuals for determining the intake of radionuclides by humans is rice, which is a dominant staple food worldwide (Uchida et al., 2009). According to the Food and Agriculture Organization (FAO) of the United Nations, national rice production has expanded dramatically during this period, increasing by an incredible 2.5 times, from 15 million tons in 1971 to 37.8 million tons in 2021(FAO, 2023). For the majority of the population in the Republic of North Macedonia, rice is an essential component of their diet and is mostly consumed in the form of white rice. The transfer factors of radionuclides have been studied for different components of food chains (corn, wheat), however studies regarding the transfer of radionuclides in rice are very limited. Therefore, the goal of this study was to measure and analyse the activity concentrations of natural radioactivity in soil and rice as well as the transfer factor from soil to rice.

Materials and Methods

Soil and rice samples, study area and preparation

A total of 44 soil and rice samples (22 rice and 22 soil samples) were collected from 10 different locations in the Kochani region. Throughout the entire area of study, an attempt was made to ensure that a sufficient number of representative samples were collected. At each of the locations of study, rice plants were randomly selected during the harvest period. As it is known, unlike other plants, rice has a root system that penetrates only to the top few inches of the soil (Asaduzzaman et al., 2015). Thus, soil samples were taken at a depth of 5cm from each location, which coincides with the rooting zone of the rice plant. A representative soil sample was prepared by removing non-representative components, such as stones, pebbles, leaves and roots, and then the mass of the sample was measured after it had been dried for several days at room temperature and had reached a stable mass, and was transferred to a Marinelli beaker with 500 g of sample. After the separation of the rice grain, the samples were sieved to exclude any extraneous materials and then a Marinelli beaker was filled with 500 g of rice powder from each sampling location for subsequent analysis.

Experimental setup

The measurements were carried out by using a high purity germanium (HPGe) detector while the activity concentrations were determined by the gamma-ray spectrometry method. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples were determined by using a high purity germanium (HPGe) detector with a relative efficiency of 30% and a multichannel analyzer (8192 channels).

The low background detector has a relative efficiency of 30% and an energy resolution of 1.67 keV full width at half maximum (FWHM) on the 1332 keV peak of 60 Co. The activity of ^{226}Ra in the samples was determined from the intensities of the 351.9 keV ^{214}Pb and 609.3 keV ^{214}Bi gamma lines, while for ^{232}Th, 911.21 keV and 968.97 keV ^{228}Ac gamma lines were used, with an intensity of 26.6% and 17.4%, respectively. In addition, 1460.70 keV was used for ^{40}K, respectively.

Activity calculation

The specific activity (A) is determined according the equation

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

Where, N is clean surface of peak accumulated from a specific radionuclide in the 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 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 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

The activity concentrations in 22 soil samples that were determined by using the HPGe detector are shown in (Table 1).

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

Sampling sites	A±SD (Bq kg^{-1}) ^{226}Ra	A±SD (Bq kg^{-1}) ^{232}Th	A±SD (Bq kg^{1}) ^{40}K
S1(n=4)	22.36±0.12	32.12±3.23	212.64±2.40
S2 (n=3)	24.01±1.05	30.11±2.00	111.50±2.50
S3 (n=1)	31.04±1.01	40.07±3.02	124.70±3.00
S4(n=1)	49.22±2.20	33.17±2.00	125.17±3.50
S5 (n=3)	31.17±1.05	34.77±2.50	147.32±2.55
S6(n=4)	38.13±1.05	46.03±2.50	115.54±5.50
S7 (n=1)	42.92±1.00	45.72±4.00	137.02±5.90
S8 (n=1)	37.53±2.09	39.82±4.20	148.85±4.50
S9 (n=1)	29.18±1.52	34.38±2.50	142.87±5.25
S10 (n=3)	32.22±1.31	40.04±2.05	131.87±4.53
Average	33.72±1.24	37.62±2.80	152.90±3.96

Table 2. Activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th 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
R1(n=4)	2.06±0.12	<MDA	91.03±0.50
R2 (n=3)	1.09±0.25	<MDA	26.11±2.50
R3 (n=1)	3.14±1.00	0.11 ± 0.04	44.12±2.00
R4(n=1)	1.03±1.20	6.18±1.40	97.62±0.50
R5 (n=3)	2.18±1.05	<MDA	47.32±3.00
R6(n=4)	2.00±1.05	<0.03	24.71 ± 2.00
R7 (n=1)	3.91±0.11	1.24±1.30	40.50 ± 0.50
R8 (n=1)	2.13±0.09	<0.03	58.13±1.50
R9 (n=1)	1.19±0.55	<MDA	28.56±0.25
R10 (n=3)	1.17±0.35	<MDA	36.35±0.55
Average	1.99±0.57	2.51±0.91	49.44±1.33

Note: MDA – Below detection limit

Table 1 shows that the average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the soil samples are 152.90 ± 3.96, 33.72 ± 1.24 and 37.62 ± 2.80 Bqkg^{-1}, respectively. The world's mean soil values of ^{226}Ra, ^{232}Th and ^{40}K are 35, 30 and 400 Bq kg^{-1}, respectively (Dabayneh et al., 2008). These values are lower compared to other values in literature which may be due to improved agricultural practices in the country and the geology of the area itself. (Alaamer, 2008; Jibiri et al., 2011). From Table 2, the average activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th in the rice samples are 49.44±1.33, 1.99±0.57 and 2.51±0.91. The concentration of radium in rice is usually regulated by the metabolic properties of the rice varieties, the amount of radium present in the soil and the amount available to the rice plant (Gupta et al., 2016). Of all examined locations, S4 had the highest average activity concentration of ^{226}Ra in the soil, despite the fact that the comparable rice samples from that area had the lowest average activity concentration. A possible explanation of this could be variations in the rice plant varieties and their radium uptake characteristics (Gupta et al., 2016). It was observed from the study that there was a slightly higher increase of ^{232}Th compared to ^{226}Ra in the soil samples. Typically, ^{226}Ra is rapidly leached and migrates with soil water, while ^{232}Th undergoes a significant erosion process and is absorbed into the soil in situ. (Hossain et al., 2024). This could be one of the reasons for the aforementioned behaviour. ^{40}K had the highest concentration at all locations in soil and rice samples. pH and soil type influenced the accumulation of ^{40}K at most of the studied locations. (Hossain et al., 2024)

Transfer factor

In order to quantify the transfer of radionuclides in the food chain in radioecological research, a measure known as the soil-to-plant transfer factor (TF) is often used. Using the measured activity concentrations of radionuclides, the soil-to-plant transfer factor for rice was computed by using the relationship shown below IAEA (Technical Reports Series No. 472, IAEA, 2010).
TF = Concentration of radionuclide in rice (Bq/kg) / Concentration of radionuclide in soil (Bq/kg dry soil mass) (2)
This ratio depicts the quantity of radionuclide expected to enter a plant from soil (Blanco Rodríguez et al., 2002). The TF can vary significantly based on the element types and environmental factors such as soil and plant types, rainfall frequency, sunlight intensity, temperature or seasonal variations, etc. (Adams et al., 2005). Moreover, the main factors that influence on the absorption and transfer of radioactive elements are considered to be the plant species, soil properties, chemical substances present, pH factor and soil texture/type (Ugbede et al., 2021). However, the soil-to-grain TF is crucial for estimating rice grain concentrations and potential human ingestion (Fujimura et al., 2012). This parameter is also very important because it can be used to predict radionuclide concentrations in the used crops and to estimate human dose consumption.
The calculations of radionuclide TF from soil to rice are presented in the Table 3 below.

Table 3. The transfer factor (TF) values of radionuclides from soil to rice
Note: NM- no radiation detected

Sampling sites	^{226}Ra	^{232}Th	^{40}K
WS1	0.09	NM	0.43
WS2	0.04	NM	0.23
WS3	0.10	0.02	0.35
WS4	0.09	0.19	0.78
WS5	0.07	NM	0.32
WS6	0.05	NM	0.21
WS7	0.09	0.02	0.29
WS8	0.05	NM	0.39
WS9	0.04	NM	0.20
WS10	0.03	NM	0.28
Average	0.06	0.07	0.34

The corresponding TF values for all radionuclides are calculated by using the equation (2) and they are presented in Table 3. The transfer factors of ^{226}Ra, ^{232}Th and ^{40}K in soil and food in the region of study were 0.06, 0.07 and 0.34, respectively.
The results show that in this area, the potassium content is the most observable in the TF. This is due to the fact that potassium is an important element for crop fertilization. Potassium is important for plant growth to adapt to environmental stresses. It is vital for plant development and adaptation to natural pressure. Consequently, potassium has the highest transfer factor value in contrast to radium and thorium. When it comes to ^{232}Th and ^{226}Ra, it is evident that they show low mobility in soil due to their strong affinity for clay minerals and organic matter.
Finally, the absorption and transport systems in plant roots have a finite capacity, and when soil radionuclide concentrations exceed this capacity, the transfer efficiency decreases. Our results differ significantly from the published TF value of the International Atomic Energy Agency (IAEA) for clay soil to rice for ^{226}Ra (0.00025-0.0029, average: 0.00057) (Blanco Rodríguez et al., 2002)
The TF values in this study were compared to other studies where the TF values of natural radionuclides (^{40}K, ^{226}Ra and ^{232}Th) were lower than some reported cases. (Ugbede et al., 2021; Asaduzzaman et al., 2015; Saeed et al., 2012).
ND indicates that no TF radiation was detected in (WS1 WS2 WS5 WS6 WS8 WS9 and WS10). Soil sorption reduces the availability of the radionuclide for absorption. Similar chemical characteristics allow radionuclides to be selectively absorbed or excluded (Gupta et al., 2016). Studies suggest that the TF is a site-specific parameter and is not influenced by a single factor (Karunakara et al., 2013). Additional factors that influenced the TF were soil strength, crop cultivation techniques, length of growing season, and root spreading characteristics in the soil. The results show that in this area, the potassium content is most observable in the TF. This is due to the fact that potassium is an important element for crop fertilization. Potassium is important for plant growth in order to adapt to environmental stresses. Potassium is vital for plant development and adaptation to natural pressures. Consequently, potassium has the highest transfer factor in contrast to radium and thorium. When it comes to ^{232}Th and ^{226}Ra, it is evident that they show low mobility in soil due to their strong affinity for clay minerals and organic matter (Wei-Hsiang et al., 2025). Finally, the absorption and transport systems in plant roots have a finite capacity, and when soil radionuclide concentrations exceed this capacity, the transfer efficiency decreases

Conclusion

The results show that the agricultural practices used in the area of study did not have a significant impact on the soil activity concentrations of ^{40}K, ^{232}Th and ^{226}Ra before planting and at harvest. All mean transfer factors (TF) for the samples were very low and this implies a very slow rate of radionuclide uptake from soil to rice plants. Based on the presented data, it was concluded that the consumption of rice grown on the studied soil is considered safe i.e. the study indicates that the soil remains suitable for rice cultivation. The calculated results may be useful in assessing the radiation dose exposure due to naturally occurring radioactive material contents in the soil samples. The study also helps to provide results regarding the safety of the rice consuming population in Macedonia by proactively studying the factors that support the uptake of radionuclides in rice. The results themselves provide valuable information and are used as a basis for establishing a database of TF in soil to grain.

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