Original Scientific paper
10.7251/AGRENG2303096A
UDC 539.16:[631.4:633.11](497.7)

ASSESSMENT OF NATURAL RADIOACTIVITY LEVELS AND RADIATION HAZARDS IN AGRICULTURAL SOIL AND TRANSFER IN WHEAT IN THE REGION OF NORTH MACEDONIA

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ABSTRACT

Bad agricultural practices, such as the excessive use of fertilizers, often lead to the accumulation of radioactive elements in the soil and further in the crops.This research aims to determine the activity concentration levels of radionuclides of 226Ra, 232Th, and 40K in soil and wheat samples. At the same time, an assessment was made of the radiation hazard indices from natural radioactivity as well as the transfer from soil to the wheat of the studied radionuclides. A total of 92 soil and wheat samples were collected in the studied area (the western part of the country). The samples were measured on an instrument - a gamma spectrometer (Canberra Packard) using the GENIE 2000 program. In this study, it was observed that the specific activity of 40K, 226Ra, and 232Th in the soil is not uniform and probably differs depending on the geological or typographical characteristics of the area as well as the applied agricultural techniques and agrochemicals. The average activity concentrations of 226Ra, 232Th, and 40K in the soil samples were 35.86 ± 1.55 Bq/kg, 37.40 ± 2.86 Bq/kg, and 165.78 ± 4.05 Bq/kg, respectively, while the average values for wheat were 0.63 ± 0.20 Bq/kg, for 226Ra 0.40 ± 0.08 Bq/kg, for 232Th and for 40K 61.18 ± 3.50 Bq/kg. The calculated mean values of radium equivalent activity Raeq, Dout, and Din were 102.98 Bq/kg, 46.72 nGy/h, and 77.93 nGy/h respectively. The soil-to-wheat transfer factor of naturally occurring radionuclides was in the following order: 40K > 226Ra > 232Th. This shows that there is no risk of radiation contamination in the tested wheat soils and there is no considerable risk to human health.

Keywords: *radionuclides, soil, wheat.*

INTRODUCTION

Researchers explore the natural radiation of the environment and the radioactivity in soils to conduct background checks and detect environmental radioactivity (Radhakrishna et al., 2012). Radioactivity levels can be used in order 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 consist of naturally occurring series such as 238 U, 232 Th, and the 40 K series which are usually long-lived and have a half-life of more than 100 million years (UNSCEAR, 2000). The indicated radionuclides can be found in all elements of the environment and are present in different amounts in soil, water, air, vegetables, animals, and the human body itself (IAEA 1989). Soil is the first link of the ecological chain soil-food-animals-man and for this reason, it has an important role in the distribution and transfer of radionuclides. Long-lived natural radionuclides can be transferred to plants, considering that the main part of the minerals that make up the plant's body comes 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 a few tens of becquerels (Bq) to several hundreds of becquerels per kilogram. Some plants are able to incorporate large amounts of radioactive substances into their tissues without visible and demonstrable changes, however, their consumption can cause serious damage and diseases in human organisms. Of course, it can have a negative impact on the growth and development of plants if it is a matter of a stronger intensity of radioactivity. The absorption of radionuclides from the soil by plants (wheat) is usually described with a transfer factor (TF) (Harb et al., 2014) which is an important parameter in the assessment of environmental safety. With the increase in the world population, the need for larger amounts of food increases, which prompts many countries to use phosphate fertilizers in crop production in order to increase their annual yields. Fertilizers contain radionuclides, the decay series of uranium and thorium, as well as potassium. Very often farmers use artificial fertilizers without expert assessment of the content of present radionuclides. Therefore, this research was conducted aiming to investigate the levels of radioactivity as a result of the natural radionuclides of 226 Ra, 232 Th, and 40 K in soils and wheat, and also to estimate the soil-to-wheat transfer factor. The study was conducted in the Republic of North Macedonia, which, due to its favorable geographical position and mild continental and Mediterranean climate, contributes to the production of numerous cereals, especially wheat. The daily consumption of wheat as the basic cereal crop in the diet of the population in the Republic of North Macedonia was the basis for this research.

MATERIAL AND METHODS

Sampling and preparation

The process of preparation of all samples was carried out in accordance with the recommendations of the International Atomic Energy Agency. The samples were collected in 2022 at approximate depths of 10 cm and were placed in polythene bags in order to avoid sample contamination. They were appropriately labeled for facilitated identification. The soil samples were first left to dry in the sun for 2 days in order to remove moisture (Changizi et al., 2013). Then, the soil was cleaned of stones, gravel, and debris and adequate drying was ensured at a temperature of 105°C. Then, the samples passed through a mesh (1 mm) to obtain homogeneous samples, and were placed in a 500 ml plastic container, known as Marinelli beaker. The containers were tightly closed and stored for more than 10 days in order to allow the parent radionuclides in the samples to reach secular equilibrium with their respective progeny (Tawalbeh et al., 2011). After this, the final step was radiometric measurement and analysis.

The wheat samples were carefully cleaned and sieved in order to obtain homogenized samples. To remove moisture, they were dried in an electric oven at 100°C. For measurements, the samples were packed in 500ml plastic containers (Marinelli beakers) and tightly closed, and then they were stored to reach secular equilibrium.

Instrument

An analysis of natural radionuclides by means of γ-ray spectrometer was performed for the soil and wheat, i.e. gamma spectrometry method was applied. We used a high-purity HPGe detector (Canberra Packard), with a volume of 180 cm2. It is a detector that was protected with lead with a relative efficiency of 30%, an operating voltage of 3000 V and a resolution of 2 keV at 1332.5 keV. The duration of the measurement was 108000 sec and the background radiation spectra were collected so that we could obtain a net count rate. The high voltage for the detector was provided by a preamplifier, which was connected to an amplifier with a computer-based channel through ADC (analog to digital converter). The software package Canberra Genie-2000 was used during the analysis. Performance calibration was performed with a mix standard source, originating from the Czech Meteorological Institute, an inspectorate for ionizing radiation. The activity of 226Ra was determined from the gamma lines associated with low half-life daughters of 214Bi (609.31, 1120.29 and 1794.49 keV) and 214Pb (351.93 keV). The activity of the 232nd is determined by the gamma lines 338.4, 911.2 and 969.1 keV of 227Ac and its decay products, while K40 was determined through the Gamma line at 1460.8 keV.

Determination of radiological index parameters

The radiation exposure resulting from terrestrial radionuclides in the examined soils and wheat plantations in North Macedonia can be determined in terms of some parameters as given below:

Radium equivalent activity (Raeq)

The Radium equivalent (Raeq) activity is a radiological risk index that is used to assess the harmful effects of ionizing radiation on health. It is assumed that 370 Bq·kg-1 of 226Ra, 259 Bq·kg-1 of 232Th and 4810 Bq·kg-1 of 40K produce the same gamma-ray dose rate. It is calculated by using the following ratio (Rahman et al. 2008):

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

where A_{Ra}, $A_{Th,}$ A_k – specific activities ($Bq·kg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of the radium equivalent activity of 370 $Bq·kg^{-1}$ corresponds to the maximum allowed dose for a population of 1 mSv.

Outdoor absorbed gamma dose rate (D_{out})

The outdoor absorbed dose rate Dout at 1 m above the ground level is determined using equation (Belivermis et al., 2010): postulating that the effects of the other artificial and natural radionuclides are low and could be neglected.(UNSCEAR, 2008)

$$D_{out}\ (nGy / h) = 0{,}462\ A_{Ra} + 0{,}604\ A_{Th} +0{,}0417\ A_{k}$$

Where D is the dose rate in nGy h−1 and A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Indoor absorbed gamma dose rate (D_{in})

The primordial radionuclides are causing to indoor exposures, the conversion factors (0.92 nGy/h/Bq/kg) for ^{226} Ra, (1.1 nGy/h$^{/}$ Bq/kg) for ^{232} Th, and (0.081 nGy/h/Bq/kg) for ^{40} K are used to estimate the D_{in} due to the presence of gamma-ray dose indoors using the following formula.(Clouvas et al., 2004).

$$D_{in}(nGy.h–^{1}) = 0.926A_{Ra} + 1.1A_{Th} + 0.0096A_{K}$$

Outdoor annual effective dose rate.

The outdoor terrestrial gamma radiation value is needed to calculate the outdoor annual effective dose equivalent (AEDE). Annual effective dose should be calculated to assess the health effects of the absorbed dose by using a conversion coefficient (0.7 Sv Gy−1) to transform absorbed dose in air to the effective dose received by humans, with an outdoor occupancy factor (0.2), which is equivalent to an outdoor occupancy of 20% and 80% for the indoors. The annual bill of effective dose received by the population can be calculated using (Cevik et al., 2008)

$$AEDR_{out}\ (mSvy^{-1}) = D\ (nGy\ h^{-1}) \times 8760\ h \times 0.7\ (Sv\ Gy^{-1})\times 0.2 \times 10^{-6}$$

where (nG/h) is the total air absorbed dose rate in the outdoors; 8760 h is the number of hours in one year; 0.2 is the outdoor occupancy factor; 0.7 $Sv\ Gy^{-1}$ is the conversion coefficient from absorbed dose in air to effective dose received by adults; 10^{-6} is the conversion factor between nano- and millimeasurements (Kalaitzis et al., 2019).

Indoor annual effective dose rate ($AEDR_{in}$)

About 80% of people stay indoors most of the time, therefore indoor exposure becomes more important if one considers the duration of exposure. E_{in} is an amount of dose that is taken by human beings, and it can be calculated from the indoor absorbed dose using the dose conversion factors; the conversion coefficient from the absorbed dose rate (0.7) and indoor occupancy factor (0.8) the time staying in the indoor during the year. The indoor annual effective dose rate ($AEDR_{in}$) can be mathematically represented as the following equation. (Mohammed et al., 2008)

$AEDR_{in}$ (mSvy^{-1}) = D (nGy h^{-1}) × 8760 h × 0.7 (Sv Gy^{-1})×0.8 × 10^{-6}

Transfer factor

The ratio of the concentrations of the primordial radionuclide present in the wheat to that of the corresponding soil is commonly called the Bioaccumulation Factor or Transfer Factor (TF). The transfer factor (TF) for food according to IAEA (1989) is defined as: (IAEA 1989)

$TF = dry$ wheat $Bq.kg{-}1\ /dry\ soil\ Bq.kg{-}1$

The main factors that affect the absorption and transfer of radioactive elements in plants are the soil properties, the present chemical substances, the pH factor and the soil texture/type (Ugbede and Osahon, 2021). In a situation where TF is lower than one, it means that the radionuclide is only absorbed by corn but not accumulated alternatively, TF greater than one means that the radionuclide has been accumulated by a plant.

RESULTS AND DISCUSSION

Table 1. shows the activity concentrations of 226 Ra, 232 Th, and 40 K in soil samples. In general, the radionuclides of interest, namely 226 Ra, 232 Th, and 40 K were found in all analyzed soil samples. The specific activity of 40 K in all agricultural soil samples was greater than the specific activities of 226 Ra and 232 Th. The average value of the activity concentration of 40 K in this study was lower than the recommended world average value (412 Bq/kg), which is recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation Sources (UNSCEAR, 2008). The use of large amounts of artificial fertilizers increases the activity concentration of radionuclides. The world average activity concentrations for soil samples for 226Ra and 232Th are 32 Bq/kg and 45 Bq/kg. In this study, the mean concentration of 226Ra was 35.86 Bq/kg, and compared to the world average values for soil, it is slightly higher, while the average concentration of 232Th was 37.40 Bq/kg, which is lower than the recommended value of 45 Bq/kg (UNSCEAR, 2008).

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

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=5)	34.36±4.12	42.42±4.23	316.60±2.30
S2 (n=4)	24.71±0.05	31.26±2.02	102.50±2.50
S3 (n=5)	41.04±0.01	40.95±3.02	269.70±4.00
S4(n=4)	29.65±0.20	35.41±2.50	125.35±3.50
S5 (n=4)	30.02±1.20	33.90±1.50	167.55±2.55
S6(n=5)	58.62±0.05	48.05±2.50	115.54±5.50
S7 (n=4)	42.92±1.00	45.19±4.00	137.02±5.90
S8 (n=5)	32.56±6.09	39.47±4.20	148.85±4.50
S9 (n=6)	33.58±1.52	36.38±2.50	142.87±5.25
S10 (n=4)	31.23±1.31	41.04±2.15	131.87±4.53
Average	35.86±1.55	37.40±2.86	165.78±4.05

Table. 2. Mean values of specific activities (A) of values of 226Ra, 232Th, and 40K in wheat

Sampling sites	A±SD (Bq kg^{-1}) ^{226}Ra	A±SD (Bq kg^{-1}) ^{232}Th	A±SD (Bq kg^{1}) ^{40}K
WS1(n=5)	0.70±0.10	0.58±0.05	74.70±2.00
WS2 (n=4)	0.34±0.10	0.26±0.02	66.50±2.50
WS3 (n=5)	<MDA	0.25±0.02	44.70±3.00
WS4(n=4)	0.85±0.20	0.65±0.02	85.15±4.50
WS5 (n=4)	0.52±0.20	0.21±0.50	77.15±2.55
WS6(n=5)	0.62±0.15	0.25±0.03	85.04±5.50
WS7 (n=4)	0.47±0.20	<MDA	37.02±3.90
WSS8 (n=5)	0.56±0.09	0.47±0.08	78.85±3.50
WS9 (n=6)	0.78±0.52	0.71±0.02	84.90±3.05
WS10 (n=4)	0.83±0.31	0.28±0.05	51.87±4.50
Average	0.63±0.20	0.40±0.08	61.18±3.50

Table 2. shows the activity concentrations of 226 Ra, 232 Th, and 40 K for 46 studied wheat samples. The activity concentration of 226Ra varies from 0.34±0.10 Bq.kg-1 (WS2) to 0.85±0.20 Bq.kg-1 1 (WS4) with an average value of 0.63±0.20Bq.kg-1. The activity concentration of 232Th varies from 0.21±0.50 Bq.kg-1 (WS5) to 0.71±0.02 Bq.kg-1 (WS9) with an average value of 0.40±0.08 Bq.kg-1, however, no 232Th was detected in any sample (WS7). Furthermore, the activity concentration of 40K was found in all samples with a minimum value of 37.02±3.90 Bq.kg-1 (WS7) and a maximum value of 85.15±4.50 Bq.kg-1 (WS6), with an average value of 61.18±3.50 Bq.kg-1. Moreover, the detection of 226Ra in wheat samples was expected, since it is a daughter product in the decay series of 238U which is usually presented in the environment. It can be determined that 40K has significantly higher concentrations than those for 226Ra and 232Th due to its

high solubility in water and high mobility in soil (Kumar, et al., 2008). The average values of the activity concentrations of 226 Ra, 232 Th, and 40 K for the wheat samples in this study were lower than the acceptable values recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation Sources (UNSCEAR,2000). The higher recorded values of 40 K in wheat samples within this study are similar to the findings observed in the literature. (Pulhani et al., 2005), (Akhtar et al., 2006), (Changizi, et al., 2013a).

Table 3. The radiological hazard parameters due to the natural radioactivity in soils of the wheat-plantation fields of Macedonia

Sampling sites	Raeq (Bq.kg)	D (nGy / h)	D_{in}(nGy.h–^{1})	$AEDR_{out}$ (mSvy^{-1})	$AEDR_{\mathbf{in}}$ (mSvy^{-1})
S1(n=5)	117.82	54.69	81.46	0.067	0.26
S2 (n=5)	67.21	34.56	57.24	0.042	0.17
S3 (n=7)	118.46	54.93	85.62	0.067	0.27
S4(n=7)	89.50	40.29	66.65	0.049	0.19
S5 (n=7)	90.21	41.31	66.68	0.050	0.20
S6(n=7)	135.41	60.90	108.23	0.074	0.29
S7 (n=7)	117.13	52.82	90.75	0.064	0.26
S8 (n=7)	99.41	45.03	74.98	0.055	0.22
S9 (n=7)	95.60	37.97	72.47	0.046	0.18
S10 (n=7)	99.14	44.69	75.31	0.054	0.22
Average	102.98	46.72	77.93	0.056	0.22

In order to determine the uniformity of the distribution of natural radionuclides of 226 Ra, 232 Th, and 40 K in soils, in relation to the radiation exposure due to these radionuclides, Raeq is used (UNSCEAR, 2000). The Raeq values in soil samples were within the range of 67.21 Bq/kg to 118.46 Bq/kg with an average value of 102.98 Bq/kg. These values are lower than the maximum permissible limit (370 Bq/kg) recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation Sources (UNSCEAR,2000). Furthermore, the mean value of Dout is lower than the recommended value of 57 nGy/h (UNSCEAR,1998), while the mean value of Din was 77.93 nGy/h. The calculated values of AEDRout and AEDRin are shown in Table 3. The values of AEDRout and AEDRin are within the range from 0.042 to 0.074 mSvy^{-1} per year with an average value of 0.056 mSvy^{-1} per year and from 0.17 to 0.29 mSvy^{-1} year with an average value of 0.22 mSvy^{-1} per year, respectively. The reported values of AEDRout and AEDRin in this study were lower than the world average values of 0.07 and 0.41 mSvy^{-1} per year, respectively, provided by (UNSCEAR, 2000).

Table 4. Soil-to-wheat transfer factors of ^{226}Ra, ^{232}Th, and ^{40}K

Sampling sites	^{226}Ra	^{232}Th	^{40}K
WS1	0.020	0.013	0.230
WS2	0.013	0.008	0.640
WS3	0	0.006	0.165
WS4	0.028	0.018	0.679
WS5	0.017	0.006	0.460
WS6	0.010	0.005	0.730
WS7	0.010	0	0.270
WS8	0.017	0.001	0.529
WS9	0.023	0.019	0.594
WS10	0.026	0.006	0.393
Average	0.016	0.0082	0.469

The soil-to-plant transfer factor is considered one of the most important required parameters to assess environmental safety (Essiett et al., 2005). The values of the transfer factor (TF) for wheat ranged from 0.010 to 0.028, from 0.001 to 0.019 and from 0.165 to 0.730 for 226Ra, 232Th and 40K respectively as presented in Table 4. These values are lower than the standard values for 226Ra, 232Th and 40K respectively, proposed by IAEA (IAEA,1994). The lowest average values of the transfer factor are found in WS7, maybe the soil there is alkaline and insoluble precipitates are formed that reduce the availability of radionuclides for plants, or because of the content of organic substances in the soil itself (Abu-Khadra et al., 2005). In regard to thorium, its lower content in wheat is due to its insolubility and low specific activity. The ions of this element can bind so tightly to soil particles that they remain immobile and are not absorbed by plants (Eisenbud, 1987). In addition, the higher transfer factor for 40K may be due to the continuous accumulation of 40K by means of root uptake where K is an essential macronutrient for the metabolism and is taken up by plants from the soil in varying amounts (Kritsananuwat et al., 2017).

CONCLUSION

This study presented the specific activity of the radionuclides 226Ra, 232Th and 40K by using gamma-ray spectroscopy in different types of soil and wheat from areas of North Macedonia. The average values of the activity concentration of naturally occurring radionuclides in the samples were lower than the world average values. Furthermore, the calculated average values of the radiological index parameters were similar to the findings observed in the literature. The soil-to-wheat transfer factor for the natural radionuclides 226 Ra, 232Th, and 40K has the following order 40 K > 226 Ra > 232Th. The amount of organic fertilizers and chemical fertilizers that are being used to increase soil fertility and plant production affect the stable content of elements in the soil, and simultaneously they are the reasons for the differences in the values of natural radioactivity in the studied samples. Overall, it was confirmed that the levels of natural radioactivity in the collected soil and wheat samples are not within the range of health risk because in all parameters of the calculated radiological index they were below the international permissible limits. These baseline data will help us to assess any variations in the radioactivity levels due to any unexpected events such as nuclear reactor accidents and/or nuclear weapon tests or due to the anthropogenic activities within the study area. The results of the study are used as a reference for future assessment.

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