NATURAL RADIOACTIVITY LEVELS IN SOME VEGETABLES COMMONLY USED IN THE CITY OF SKOPJE (MACEDONIA)

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1. INTRODUCTION

Radionuclides can originate from a variety of sources and processes; either those occurring naturally, such as primordial or cosmogenic formation, or through human activities, such as release from nuclear weapons testing or accidents (1). Understanding the range of radionuclides in the diet and their respective activity concentrations is necessary to be able to quantify the risk of exposure. Different researchers have demonstrated that plants, vegetables, shrubs, fungi, and algae can take in, keep up and store radionuclides(2). Natural radionuclides by means of migration in the soil reach the plant crops and contribute to the total radiation load in the population. Artificial radionuclides behave in a similar way, and over the past half-century there has been worldwide contamination of food chains with radionuclides produced during nuclear weapon tests in the atmosphere (3).

The health hazard of potassium-40 is associated with cell damage caused by the ionizing radiation that results from radioactive decay, with the general potential for subsequent cancer induction.

Considering that they can cause side effects on the human organism, the assessment of contamination with radionuclides in nutrition is an important aspect of modern food safety. Some vegetables are able to incorporate great amounts of radioactive matters in their tissues, without visible and provable changes, however their consumption can cause serious impairments and disease in human. It can certainly have a negative effect on the development and growth of plants when it comes to greater intensity of radioactive radiation. However, it has been found that the understanding of the behavior of natural radionuclides in the environment is very important, because such information can be used as the related parametric values for radiological assessments (4).

2. MATERIALS AND METHODS

2.1. Sampling and preparation

Forty two vegetable samples were collected from local markets selected according to the level of consumption of the population in Macedonia. The food samples were washed with normal water, as used for human nutrition, measured and divided into small portions, dried in a stove at a temperature of 80°C for 48 hours and grounded in powder(5,6).

All samples were packed in a plastic (polyethylene) cylindrical containers with a volume of approximately 500 cm3 and the containers were completely sealed for at least 4 weeks in order to enable radioactive equilibrium to be reached (3).

2.2. Gamma Spectrometry

2.2.1. Instrumentation

An analysis of natural radionuclides by means of γ-ray spectrometer was performed for the vegetable samples, 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 232^{nd} 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.

2.3. Measurements and Calculation

2.3.1. Absorbed dose rate in the air (D).

People are exposed to radiation from natural radionuclides that are present in the soil. The level of radiation is closely related to the radioactive elements and minerals in the respective regions. The dose of gamma radiation that the population receives during their lifetime is very important. The absorbed rate in air for radionuclides with a height of 1 m above the ground is calculated based on the guidelines indicated below (1).

$$D(nGy/h)=0.462A_{Ra}+ 0.604\ A_{Th}+0.042\ A_k \qquad (1)$$

where A_{Ra}, A_K and A_{Th} accordingly are the average activity concentrations of ^{226}Ra, ^{40}K and ^{232}Th, expressed in Bq/kg.

2.3.2. Calculation of the external hazard index (H_{ex}).

The external hazard index is calculated with an equation proposed by (7).

$$H_{ex} = C_{Ra}/370+ C_{Th}/259+ C_K/4810 \qquad (2)$$

where H_{ex} is the external hazard value, C_{Ra}, C_{Th} and C_K are the mean activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of this index must be less than unity, thus keeping the radiation hazard insignificant. The maximum value of H_{ex} = 1, corresponds to the upper limit of Ra_{eq} (370 Bq kg^{-1}).

2.3.3. Calculation of radium equivalent activity.

Considering that we do not have equal distribution of 226Ra, 232Th and 40K in the soil samples, by using the radium equivalent (Raeq) we can define the exposure to radiation, in order to compare the specific activity of the radionuclides (7).

The radium equivalent activity could be defined as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the specific activities (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. By defining the Ra_{eq} according to the previous equation, it is assumed that 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4810 Bq kg^{-1} ^{40}K produce equal gamma dose rate (8).

3. RESULTS AND DISCUSSION

Table 1 shows the concentration of natural radionuclides measured in vegetable samples that are frequently consumed in Macedonia.

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

Sampling sites	A±SD ($Bq\cdot kg^{-1}$) ^{226}Ra	A±SD ($Bq\cdot kg^{-1}$) ^{232}Th	A±SD ($Bq\cdot kg^{1}$) ^{40}K
Potatoes (n=7)	0.35±0.1.0	2.74±0.05	108.15±2.00
Tomatoes (n=7)	0.52±0.05	1.17±0.03	99.00±2.00
Cucumber (n=7)	0.24±0.10	4.22±0.12	69.70±1.00
Spinach (n=7)	3.24±0.60	2.41±0.50	85.35±1.50
Cabbage (n=7)	0.88±0.05	3.90±0.08	99.86±1.54
Radish (n=7)	0.15±0.05	0.70±0.03	115.54±2.40

n= number of vegetable

The results presented in the table are average values from the measurements performed on vegetable samples taken from different markets, with 7 samples each.

From the obtained results, it is clear that the concentration of radium, thorium and potassium differed significantly in different vegetable samples depending on the amount and the type of fertilizer used. The data show that the average value of the activity of ^{232}Th is higher than the one of ^{226}Ra which may be due to the longer half-life of ^{232}Th compared to ^{226}Ra. The use of fertilizers containing potassium has contributed to the unequal distribution of ^{40}K in the soil and thus in vegetables. However, ^{40}K is the only radionuclide found in significant quantities in all types of vegetable samples. All found radionuclides that are naturally present are considered to have nominal concentrations.

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

Sampling sites	D(nGy/h)	H_{eks}	Ra_{eq}
Potatoes (n=7)	6.35	0.033	11.56
Tomatoes(n=7)	5.02	0.032	9.53
Cucumber(n=7)	5.58	0.031	9.92
Spinach(n=7)	6.53	0.036	13.61
Cabbage(n=7)	6.95	0.058	12.84
Radish(n=7)	5.34	0.027	9.80

The values of the radiation risk index H_{eks} are within the range from 0,027 to 0,058, which shows that there is no radiation risk for the population. The radium equivalent activity (Raeq) was calculated with Equation (3). Based on the UNSCEAR report, the threshold value of the radium equivalent activity (Raeq) must be less than 370 Bq/kg.(1)

From Table 2 it is observable that the values of the radium equivalent activity are within the range from 9,53 to 13,61. On the basis of UNSCEAR(1) the world average value of the absorbed dose (D) is

60 nGy/h According to our findings, the values of the absorbed dose, the rate was from 5,02 to 6,95 nGy/h Still, this is lower than the average world value indicated by UNSCEAR (1).

The values obtained in this study, regardless of the type of vegetable, did not exceed the safety limits, pointing out the insignificant danger of radiation arising from naturally occurring terrestrial radionuclides.

The results for natural radioactivity were compared with the results for different countries in the world which are presented in Table 3.

Table 3. Comparison of activity concentration (Bq/kg) with the data in literature.

Sampling sites	^{226}Ra	^{232}Th	^{40}K
Nigeria	-	20.8	232 (9)
Iran	-	2.20-10.56	108.99 (9)
Turkey	2.41	10.54	491-2324 (10)
Pakistan	6.53	2.37-7.20	189.9-410 (11)
India	0.01-1.16	0.02-1.26	45.9-649 (12)
Egypt	1.50	-	235-507 (13)

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