

ASSESSMENT OF NATURAL AND ARTIFICIAL RADIOACTIVITY LEVELS IN SOME SEEDS COMMONLY USED IN ALBANIA

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ABSTRACT

All types of foodstuffs contain an amount of radioactivity which can enter into the human body by the ingestion pathway. If radionuclides are deposited in the human body and concentrations of them was increase from foodstuffs, these may cause illness and a number of health hazards. Fifteen samples of different types of seeds commonly used in Albania were collected from the local markets of the city of Tirana. For this reason, the aim of this study was focused in the measurement and analysis of the activity concentrations of natural and artificial radionuclides in the seed samples. The measurements were performed by using the High Purity Germanium (HPGe) detector and the activity concentration for radionuclides of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs were determined by gamma-ray spectrometry method. The range of activity concentration of natural radionuclides was found to be from 0.93 ± 0.18 to 10.34 ± 1.17 Bq kg⁻¹ for ²²⁶Ra, < MDA to 2.20 ± 0.24 Bq kg⁻¹ for ²³²Th and 28.35 ± 2.08 to 446.36 ± 19.04 Bq kg⁻¹ for ⁴⁰K, respectively. The activity concentration of the artificial radionuclide of ¹³⁷Cs was detected only in four seed samples with highest value of 0.65 ± 0.22 Bq kg⁻¹. The average values of activity concentration for ²²⁶Ra and ⁴⁰K were found 5.35 Bq kg⁻¹ and 178.17 Bq kg⁻¹. The highest values of activity for ²³²Th and ¹³⁷Cs were found 2.20 Bq kg⁻¹ and 0.65 Bq kg⁻¹. The results of activity concentration of this study were compared with international reference values and the other literatures. The average values of activity concentration for seed samples were found lower than the worldwide average value, which is defined by UNSCEAR 2000 report. Therefore, the results obtained in this study for radioactivity levels show that the consumption of these seeds are safe and no harmful health effects are expected for peoples living in Albania.

Keywords: Radioactivity, Radionuclide, Gamma-ray spectrometry, Activity concentration, Seed.

INTRODUCTION

The naturally occurring radioactivity are main sources contributor's of natural radiation exposure to the human peoples around the world. The radionuclides by the natural series such as ²³⁸U, ²³²Th, and radionuclide of ⁴⁰K with a half-life of hundred million years are more important radionuclides and have main contribution of natural radiation, because these radionuclides are everywhere, in earth, atmosphere, water and foodstuffs (UNSCEAR, 2000). However, a contribution come from radiation of artificial radionuclides released into the environment by human activities, as nuclear power plants, nuclear tests, nuclear accidents, industrial and medicinal waste and the other

factors. Nuclear tests after 1945, the Chernobyl accident in 1986 and Fukushima in 2011, released many artificial radionuclides into the environment with high levels of activity concentration, but most of them have short half-life and are decayed, thus more important by the artificial radionuclide is cesium ¹³⁷Cs with long half-life 30.05 years, relatively (IAEA, 2006). In the environment naturally or artificially radionuclides as ⁴⁰K, ²²⁶Ra, ²³²Th and ¹³⁷Cs were taken into account in mostly of studies (UNSCEAR, 2000). All types of foodstuffs contain an amount of radioactivity which can enter into the human body by the ingestion and inhalation pathway, also from the surrounding environment. Soils and plants are the primary pathway of natural radionuclides entering into different types of seeds and after ingestion, them entering directly or indirectly to the human body through the food chain (Lordford et al, 2013). Transfer factors of these radioactive nuclides of ⁴⁰K, ²²⁶Ra, ²³²Th and ¹³⁷Cs from soil to food products were found as 0.17, 0.07, 0.16 and 0.23, respectively. The radionuclides of ¹³⁷Cs and ⁴⁰K have high transfer factor, but abundance of ⁴⁰K it is much higher into in the environment (Cengiz et al, 2021). Many studies of natural radioactivity concentration in seeds have been performed in several countries, while no information on the radioactivity levels in Albania, (Spahiu et al, 2019). Therefore, the aim of this study was focused in the measurement and analysis of the activity concentrations levels of natural and artificial radioactivity in the seed samples commonly used in Albania. The measurements were performed by using the High Purity Germanium (HPGe) detector and the activity concentration were determined by gamma-ray spectrometry method.

MATERIALS AND METHODS

Sampling and sample preparation

Fifteen samples of different types of seeds commonly used in Albania were collected from the local markets of the city of Tirana. In fact, mostly of them are used in all other regions of Albanian. All samples were open in air for drying on trays for several days and then in oven were dried at a temperature of 80°C (± 5°C) for about 14 hours until constant mass was obtained and as much as possible moisture was removed (IAEA, 1989). Some of them are kept in the form of grains, while others in the form of flour and then their mass is accurately weighed. Each of seed sample were put in a 250 ml plastic Marinelli beaker, hermetically sealed to prevent the escape of radon gas and kept for a period of a month in order to achieved an approximate secular equilibrium between ²²⁶Ra and its daughters ²¹⁴Pb and ²¹⁴Bi, also between ²³²Th and its daughters ²¹²Pb and ²²⁸Ac, before the measurements to have been performed.

Measurements and sample analysis

The measurements were performed by using the High Purity Germanium (HPGe) detector and the activity concentration for radionuclides of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs were determined by gamma-ray spectrometry method. The measurements of each seed samples were performed by HPGe detector Model GX4018-7500SL (Canberra Industries USA), which is connected with Multichannel Analyzer (MCA) of 8192 channels. Detector it's a p-type germanium equipped with a carbon epoxy window to protect the upper part of the detector. This detector has an energy resolution (FWHM) of 1.8 keV for 1.33 MeV γ -energy of ⁶⁰Co and 40% relative efficiency on peak. In order to minimize the gamma-ray background from surrounding or the other factors, the detector was shielded with 10 cm lead, 1.6 copper and 1 mm cadmium foils. Gamma-ray spectrometry method was used and for analyzing of spectra were performed from Genie 2000 (V3.2.1) software from Canberra Industries. The system was running freely, with an empty Marinelli beaker for 3 days live time, to evaluate the background spectrum. The Marinelli beaker contains sample was placed over cup of the detector and for each sample counting time interval was 86400 s, to have a better statistical of counts. After energy calibration was performed using different point sources, absolute efficiency was calibrated in the energetic range from 50 keV to 2000 keV using a mathematical model by Laboratory Sourceless Calibration Software (LabSOCS). The range of absolute efficiency uncertainties was calculated from 10% at low energies to 4% at high energies (Shyti, 2019 & Shyti et al, 2019).

The activity concentration (A) expressed in Bq kg⁻¹ was calculated by the following formula (1):

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

where N is the net peak area of peak at energy E_{γ} after subtracted from background, $\varepsilon(E_{\gamma})$ is the absolute efficiency in corresponding peak, P_{γ} is the gamma-ray yield per decay, t is the counting live time and m is the dried sample mass in kilogram (Dovlete et al, 2004). The activity concentration of radionuclides of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in

the seed samples were determined by using gamma-ray peaks, respectively. In case of ^{226}Ra , the activity concentration was calculated by averages of activities of decay product of ^{214}Pb and ^{214}Bi , in energy peaks 295.2 and 351.9 keV for ^{214}Pb , while for ^{214}Bi these peaks are 609.3 keV and 1120.29 keV. The activity concentration of ^{232}Th was calculated by activity of ^{228}Ac in energy peaks 338.4 and 911.2 keV. The activity concentration of ^{40}K was determined by using key line of energy 1460.8 keV and activity of artificial radionuclide ^{137}Cs was calculated from key line energy 661.7 keV. Gamma emission yields and gamma lines energy were taken from the Nuclide-LARA library (Bé, et al, 2008).

Minimum detectable activity (MDA) were performed by using Currie formula as following (Currie, 2004).

$$MDA = \frac{2.71 + 4.65\sqrt{C_B}}{\varepsilon(E_\gamma) \cdot P_\gamma \cdot t \cdot m} \quad (2)$$

In the formula (2), C_B is the background count rate under the corresponding peak and the other terms were mentioned in formula (1).

RESULTS AND DISCUSSION

Activity concentrations in the seed samples

The values of activity concentration of ^{226}Ra , ^{232}Th , ^{40}K and artificial ^{137}Cs radionuclides were calculated from formula (1) for seed samples with uncertainty ($\pm 1\sigma$) and are listed in Table 1. Minimum Detectable Activity (MDA), calculated from formula (2) for radionuclides of ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs were found for each of them 4.66, 0.03, 0.11 and 0.01 Bq kg⁻¹, respectively. The range of activity for radionuclide of ^{226}Ra was found from 0.91 ± 0.11 to 10.34 ± 1.17 Bq kg⁻¹, with average value 5.35 Bq kg⁻¹. The highest value of activity was found for Oat grain and lowest value for Barley grain. In case of ^{232}Th activity was below MDA for three samples of Kidney white beans, Bulgur wheat and Rice and highest value was 2.20 ± 0.24 Bq kg⁻¹ for Corn flour sample. Activity of ^{40}K ranged between 28.35 ± 2.08 to 446.36 ± 19.04 Bq kg⁻¹, where the highest activity concentration was found in Bean sprouts and lowest was for Rice, while average value of activity for ^{40}K was found 178.17 Bq kg⁻¹. Radionuclide of ^{137}Cs was detected in four seed samples as it showed in Table 1. The activity for artificial radionuclide of ^{137}Cs was below MDA for most of them, where highest value was found 0.65 ± 0.22 Bq kg⁻¹ for Kidney white beans.

Table 1. Activity concentration of radionuclides of the ^{226}Ra , ^{232}Th , ^{40}K and ^{137}Cs for 15 seed samples.

No.	Sample	Activity concentration (Bq kg ⁻¹)			
		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
1	Green pea	4.99 ± 0.23	1.48 ± 0.16	359.03 ± 15.49	<MDA
2	Barley grain	0.91 ± 0.11	1.43 ± 0.15	115.29 ± 5.51	<MDA
3	Beige beans	4.24 ± 0.18	2.09 ± 0.22	303.08 ± 12.96	<MDA
4	Kidney white beans	2.54 ± 0.21	<MDA	363.16 ± 15.66	0.65 ± 0.22
5	Bulgur wheat	7.03 ± 0.56	<MDA	81.22 ± 4.01	<MDA
6	Wheat grain	0.93 ± 0.18	1.62 ± 0.18	117.30 ± 2.46	0.35 ± 0.04
7	Wheat flour	7.89 ± 0.50	1.21 ± 0.14	31.36 ± 5.41	<MDA
8	Bean sprouts	8.66 ± 0.64	1.32 ± 0.16	446.36 ± 19.04	<MDA
9	Corn grain	6.92 ± 0.57	1.36 ± 0.16	70.33 ± 3.54	<MDA
10	Corn flour	9.36 ± 0.78	2.20 ± 0.24	35.88 ± 2.60	<MDA
11	Rice	2.56 ± 0.16	<MDA	28.35 ± 2.08	0.58 ± 0.21
12	Oat grain	10.34 ± 1.17	2.10 ± 0.18	105.23 ± 5.59	<MDA
13	Rye grain	2.11 ± 0.15	1.42 ± 0.16	112.63 ± 5.28	0.46 ± 0.06
14	Lentil Greece	6.84 ± 0.56	1.19 ± 0.13	255.60 ± 10.63	<MDA
15	Lentil Canada	4.92 ± 0.44	0.52 ± 0.08	247.70 ± 10.72	<MDA

The activity concentration have relatively a low level for ^{232}Th and artificial ^{137}Cs radionuclides. ^{40}K has highest values of activity for all samples and this was to be expected because it is naturally high abundance in environmental samples, also from soil fertilization. Moreover, transfer factors of ^{40}K are considerably higher than those for ^{226}Ra and ^{232}Th because of the high solubility of ^{40}K in water and its high mobility in soil (Cengiz et al, 2021). The results

of this study for activity concentrations for each radionuclides are comparable or lower than results by the other studies, which more measurements were performed and referred (Cengiz et al, 2021). Also, our results of activity concentration are lower than the worldwide average values of 35 Bq kg⁻¹, 30 Bq kg⁻¹ and 400 Bq kg⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K, defined by UNSCEAR 2000 report (UNSCEAR, 2000).

CONCLUSIONS

In this study, natural and artificial radioactivity levels in fifteen seed samples commonly used in were investigated. The measurements were performed by a (HPGe) detector and the activity of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs were determined by gamma-ray spectrometry method. The range of activity for natural radionuclides of ²²⁶Ra, ²³²Th, ⁴⁰K was found to be from 0.91 ± 0.11 to 10.34 ± 1.17 Bq kg⁻¹ for ²²⁶Ra, < MDA to 2.20 ± 0.24 Bq kg⁻¹ for ²³²Th and 28.35 ± 2.08 to 446.36 ± 19.04 Bq kg⁻¹ for ⁴⁰K, respectively. The artificial radionuclide of ¹³⁷Cs was detected only in four seed samples with highest value of activity 0.65 ± 0.22 Bq kg⁻¹. The average values of activity concentration for ²²⁶Ra and ⁴⁰K were found 5.35 Bq kg⁻¹ and 178.17 Bq kg⁻¹. The highest values of activity for ²³²Th and ¹³⁷Cs were found 2.20 Bq kg⁻¹ and 0.65 Bq kg⁻¹.

The results on this paper for activity concentrations for each radionuclides are comparable or lower than results by the other studies. Also, the results of activity concentration are lower than the worldwide average values of 35 Bq kg⁻¹, 30 Bq kg⁻¹ and 400 Bq kg⁻¹ for ²²⁶Ra, ²³²Th and ⁴⁰K, defined by UNSCEAR 2000 report.

Therefore, the results obtained for radioactivity levels show that the consumption of these seeds are safe and no harmful health effects are expected for peoples living in Albania.

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