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PRELIMINARY INVESTIGATION OF NATURALLY OCCURRING RADIONUCLIDES IN SOME SPICES USED IN ALBANIA

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Abstract. *We are using everyday spices in food as pigment taste, flavor of foods or in human diet and some of them have great benefits for our health and body. In Albania the type of spices in food has been increased in recent years and these vary from country to country, depending on the type of soil and how they are grown. Thus, the aim of this current study attempts to determine the level of radioactivity in different types of spices, which are consumed by people living in the city of Tirana in Albania, where is concentrated the largest number of the population and to estimate their effective dose to the human body. Samples of spices are collected randomly in some different markets in Tirana city, which may be produced in Albania or imported. The activity concentration of natural radionuclides of ^{40}K, ^{226}Ra and ^{232}Th were measured in twenty types of spices. A high-resolution HPGe detector was employed to perform the measurements. The obtained results indicate that ^{40}K, ^{226}Ra and ^{232}Th was detected in all selected samples for study, whereas the presence of artificial radionuclide of ^{137}Cs was found only in two spices samples. ^{40}K activity concentration varies from 173.72 ± 9.34 Bq kg^{-1} to 849.47 ± 39.36 Bq kg^{-1}. The range of activity concentration of ^{226}Ra varies from 5.15 ± 0.52 Bq kg^{-1} to 21.01 ± 1.80 Bq kg^{-1}. The activity concentration of ^{232}Th varies from 2.04 ± 0.31 Bq kg^{-1} to 21.90 ± 1.78 Bq kg^{-1}. The estimated Average Annual Committed Effective Dose (AACED) due to ingestion of these spices varies from 5.61 ± 0.29 μSv y^{-1} to 10.91 ± 0.56 μSv y^{-1}. All these values are far below than the world average value dose for individual of 290 μSv y^{-1} for all foods reported by UNSCEAR 2000. This indicates that no risk is expected by the intake of spices samples in food. The obtained data provide us the baseline levels of natural radioactivity and background information for future research on foodstuff for radiological protection of the human.*

Keywords: *Spices, ingestion dose, HPGe gamma-ray spectrometry*

1. Introduction

Spices come from the bark, roots, dried seeds, fruit of plants and trees. We are using everyday spices in food as pigment taste, flavor of foods widely used ingredient in food preparation and processing or in human diet and some of them have great benefits for our health and body. Benefits of spices are in preventing and treating of diseases such as cancer, aging, metabolic, neurological, cardiovascular, and inflammatory diseases [1]. Spices are also used in food industry, where many foods isolated with spices inside after preparations, have antimicrobial activity against many of the common microorganisms that affect the food quality and shelf life, so we can use these foods later [2]. Preparation and use of spices in the meals has many various beneficial effects as well. In this way, effects of spices can stimulate the secretion of saliva, promote the digestion, prevent from cold, to have immunity from influenza, and reduce nausea and vomiting [3].

Nowadays, humans around the world are using in their food over 100 varieties of spices, coming from different countries or produced locally. Countries from Asia are known mainly for production of spices, such as cinnamon, pepper, different kinds of seeds, cloves, ginger, etc. In Europe grows mostly spices of basil, bay leaves, bay laurel, celery leaves, chives, dill, different kind of peppers, etc., while in America grows mainly, pepper, nutmeg, ginger, allspice, and sesame seed are mostly produced [4].

In Albania the type of spices used in food has been increased in recent years and these vary from country to country, depending on the type of soil and how they are grown. Some types of spices are grown in Albania, while some others are imported abroad. As we know, radionuclides are found everywhere in the air, water, soil and therefore in the drinks and foods. During and after World War II, many nuclear bomb tests in atmosphere led to a contamination in water, air, soil and by fallout of the Northern Hemisphere with different kind of radionuclides, such as isotopes of cesium, strontium, plutonium etc. Environmental contamination was then more heavy after the reactor fire of Chernobyl in April of 1986 [5]. Thus, the control of radioactivity in air, water, soil and food is an important task of every state.

The aim of this current study is to determine the level of radioactivity in different types of spices which are consumed by people living in the city of Tirana in Albania, where is concentrated the largest number of the population and to estimate the Average Annual Committed Effective Dose (AACED), due to the ingestion of radionuclides to the human body. It has

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been estimated that about, at least one eighth, of the average annual dose due to natural sources is caused by the intake of food [6].

Samples of spices are collected randomly in some different markets in Tirana city, which are produced in Albania or imported. The activity concentration of natural radionuclides of ^{40}K, ^{238}U (^{226}Ra) and ^{232}Th and artificial radionuclide of ^{137}Cs were measured in twenty types of spices. A high-resolution HPGe detector was employed to perform the measurements.

^{40}K, ^{238}U and ^{232}Th are long-lived naturally occurring radionuclides, but also ^{137}Cs has a great impact on human lifetime and enter in human body through the food spices in our case. These radionuclides accumulate in different parts of human body, ^{40}K accumulates in muscles, ^{238}U in lungs and kidney, ^{232}Th in lungs, liver and skeleton tissues and ^{137}Cs to be distributed in the soft tissues, especially muscle tissue. These radionuclides are dangerous when activity concentration becomes high due to half-life and chemical behavior. So, ^{40}K is radiotoxic as well as nutritionally important element, ^{238}U is both radiotoxic as well as chemically toxic, ^{232}Th and ^{137}Cs are mainly radiotoxic [7].

In the case when activity concentration of these radionuclides is large quantities, in particular organs, this radiation produces damages, biochemical and genetic changes. Therefore, also deliver radiation dose accumulated in these organs will be higher. All of these will increase the probability of weakening the immune system, developing different types of diseases associated with cancer risk and increasing the mortality rate. In this way, we are interesting to investigate exposure of the general public due to intake of spices in Albania, to establish baseline values and to control the exposure of the public from natural and artificial radionuclides, due to the consumption of spices.

2. Materials and methods

2.1. Sampling and sample preparation

We collected samples of spices that are most often used in food in some markets of the city of Tirana. Samples were open in air for drying on trays for a period of one week and then in oven were dried at a temperature of about 100°C for 2 to 4 hours until constant mass was obtained and to remove as much as possible moisture. All samples examined were powdered to average particle size lower than 1 mm, and then their mass is accurately weighed. Each of them is placed in a Marineli beaker with a volume of 500 ml, hermetically sealed so that we do not have radon gas ^{222}Rn leaks. Isolated Mrineli beaker are kept for about 30 days until the secular equilibrium of ^{222}Rn is reached with the daughter nuclei produced after decay chain and then the measurements are performed.

2.2. Sample analysis

Measurement of activity concentrations of radionuclides of samples was studied by gamma spectrometry and were performed by using a p-type High Purity Germanium (HPGe). Our detector is Model GX4018-7500SL (by Canberra Industry) which was incorporated with digital spectrum analyzer, DSA-1000 and has equipped with a carbon epoxy window. Relative efficiency of detector is 40% and Full Width at Half Maximum (FWHM) is 1.8 keV for 1332. 5 keV from gamma emission of ^{60}Co. In order to minimize gamma radiation by the surrounding environment HPGe was shielded with 10 cm lead, 1.6 and 1 mm copper-cadmium foils. The detector was cooled in liquid nitrogen at −196 °C (77K) provided in a 25 liter Dewar. HPGe detectors have a very good energy resolution, in this way, even complex spectra can be analyzing without using processes of chemical separation. Therefore, two properties of resolution and sensitivity, are more important for this kind of detectors. Analyzing of spectra for spice samples in this study, are performed by used software Genie 2000 (V3.2.1). Counting time interval for each spice sample was the same by 86400 seconds, respectively. Energy calibration is performed using some point sources and absolute efficiency calibration in every photopeak is performed in the energetic range from 30 keV to 2000 keV, using software Laboratory Sourceless Calibration Software (LabSOCS) [8]. The absolute efficiency uncertainties vary in energetic range from 4% at high energies to 10% at low energies. The efficiency calibration curve was validated by using reference material supplied by the International Atomic Energy Agency (IAEA) and through the international participation in IAEA Worldwide Proficiency Test for environmental radionuclides [9]. The activity concentration and uncertainties of natural and artificial radionuclides of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs in the food spices were determined after background correction.

The activity concentrations of the spice samples were determined by main energy peaks of every radionuclide or daughter products in secular equilibrium. In the case of ^{40}K activity concentration was found by using energy key line of 1460.82 keV, with intensity gamma emission 10.55%.

The activity concentration of ^{226}Ra was estimated by averaging activities of ^{214}Pb and ^{214}Bi. Energy lines for activity calculated of ^{214}Pb are in 241.99, 295.22 and 351.93 keV, with intensities of 7.27%, 18.41% and 35.60%, while energy lines for ^{214}Bi are 609.31 keV and 1120.29 keV, with intensities of gamma emission of 45.49% and 14.91%. The activity of ^{232}Th was estimated from energy lines of 338.32 and 911.20 keV of ^{228}Ac, with intensities 26.20% and 11.40%. Activity concentration of artificial radionuclide ^{137}Cs, was estimated from photopeak of 661.66 keV, with intensity 84.99%. The intensities of gamma emissions for every energy line were taken from library Nuclide-LARA [10].

The activity concentration A of every radionuclide, in each main energy line, were estimated by the using of formula (1):

$$A = \frac{N_{net}}{\varepsilon(E_\gamma) \cdot I_\gamma \cdot t \cdot m} \tag{1}$$

and is expressed in Bq kg^{-1}, while N_{net} is the net peak area for the radionuclide in the peak energy, $\varepsilon(E_\gamma)$ is the measured counting efficiency of the detector in every peak, I_γ is the intensity of the gamma line from the radionuclide, t is the counting live time and m is the dried sample mass in kilogram [11].

Minimum Detectable Activity (MDA) were calculated by using Currie formula as following [12].

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

where C_B is the background counts in the corresponding peak, while the other terms entering the formula are mentioned above.

3. Results and discussion

3.1. Activity concentrations in food spice samples

The values of activity concentration of natural radioactivity of radionuclides of ^{40}K, ^{226}Ra, ^{232}Th and artificial radionuclide of ^{137}Cs were found. In the Table 1, results of activity concentrations of natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th in twenty spices samples collected in the markets of city Tirana are presented. The activity concentrations for every radionuclide are given in Bq kg^{-1} and the uncertainty in all calculated values is within ±1σ. The results indicate that natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th were present in all selected spice samples, whereas the presence of artificial radionuclide of ^{137}Cs was found only in two spices samples.

Range of activity concentrations for ^{40}K is from 173.72 ± 9.34 Bq kg^{-1} for Sri Lanka cinnamon spice, which has the lowest value to 849.47 ± 39.36 Bq kg^{-1} for Turmeric spice sample, which has the highest value. Range of activity concentration of ^{226}Ra is from 5.15 ± 0.52 Bq kg^{-1} for Dill, which has the lowest value spice to 21.01 ± 1.80 Bq kg^{-1} for Bay laurel spice, which has the highest value. The activity concentration of ^{232}Th varies from 2.04 ± 0.31 Bq kg^{-1} for Mixed spices 2 to 21.90 ± 1.78 Bq kg^{-1} for Sri Lanka cinnamon spice. The values calculated for the artificial radionuclide of ^{137}Cs are 0.53 ± 0.21 Bq kg^{-1} in Oregano spice and 0.41 ± 0.10 Bq kg^{-1} in Bay laurel spice, while in all other spice samples are below MDA.

Table 1. Activity concentration of radionuclides of ^{40}K, ^{226}Ra and ^{232}Th were measured in twenty types of spices.

No.	Sample code	Activity Concentration (Bq kg^{-1} ± 1σ)		
	English name	^{40}K	^{226}Ra	^{232}Th
1	Black pepper	457.97±20.31	12.75±1.34	6.32±0.95
2	Curry	178.93±8.65	7.66±0.79	10.84±1.63
3	Ginger	612.83±26.51	7.63±0.80	6.56±0.98
4	Turmeric	849.27±36.94	15.90±1.67	5.20±0.78
5	S. L. cinnamon	173.72±9.34	12.28±1.30	21.90±1.78
6	Cloves	518.17±23.54	15.32±1.60	7.89±1.18
7	Chia	231.94±10.64	11.92±1.26	3.62±0.54
8	Cayenne pepper	720.82±31.27	17.07±1.80	4.00±0.60
9	Mix spices 1	411.84±18.55	13.73±1.43	2.79±0.42
10	Mix spices 2	429.51±19.24	11.81±1.22	2.04±0.31
11	Garlic	337.15±15.36	14.74±1.50	3.52±0.53
12	Celery	674.02±29.32	11.70±1.21	4.46±0.67
13	Parsley	674.94±29.45	15.08±1.60	5.46±0.82
14	Dill	812.75±34.57	5.15±0.52	4.46±0.67
15	Winter savory	420.44±18.89	9.87±1.05	5.48±0.82
16	Oregano	564.68±24.76	9.22±0.73	4.21±0.63
17	Rosemary	516.19±22.63	14.54±0.85	3.89±0.58
18	Peppermint	587.05±25.60	18.30±1.32	5.60±0.84
19	Bay laurel	247.41±12.41	21.01±0.95	5.65±0.85
20	Basil	742.85±31.77	11.20±0.52	5.78±0.87

If we look at a comparison between activity concentration values, the highest values are those of natural radionuclides. Activity concentration of isotope of ^{40}K is much higher compare with three other radionuclides of ^{226}Ra, ^{232}Th and ^{137}Cs.

The activity concentration of ^{40}K is expected to be higher, due to using of potassium in the soil of fertilizers by farmers for many years. Also, transfer factor of ^{40}K from soil to spices that come from the bark, roots, dried seeds, leaves, fruit of plants and trees, is higher than some natural radioisotopes [13]. But, ^{40}K is important element in biological processes that occur in living organisms and it is controlled in plants and humans, in this way, potassium is remain constant by metabolic process. In the previous studies, we have found activity concentration of twenty medicinal and herbal plants, mostly produced in Albania [14]. Their values showed relatively a low level of the activity concentrations of ^{232}Th and artificial radionuclide of ^{137}Cs. In that study, activity concentration for ^{40}K ranged between 133.54 and 839.96 Bq kg^{-1}, where the highest activity concentration was found in Chamomile sample. The most values of activity concentration for ^{226}Ra varies from 5.28 to 25 Bq kg^{-1}, where the highest activity concentration was found in Sage sample. For ^{232}Th activity concentration ranged between 4.22 and 8.16 Bq kg^{-1}, while almost all samples except two of them shows a for ^{232}Th, these values are < MDA. In the case of ^{137}Cs the mostly of values varies from 0.40 to 1.55 Bq kg^{-1} and the highest activity concentration was found in Mistletoe sample, while 14 of 20 samples showed that for ^{137}Cs these values were < MDA. If we see the range of activity concentration for radionuclides of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs, found in the previous study for medicinal and herbal plants, it is comparable to spices.

3.2. Average annual committed effective dose from ingested spice foods

The activity concentration of natural radioactivity of radionuclides of ^{40}K, ^{226}Ra, ^{232}Th, and artificial radionuclide of ^{137}Cs were found above. We found activity concentration for each radionuclide, showed the range of values as well as those spices that had the lowest and highest values and discussed them. In this way, we have determined the level of radioactivity in different types of spices which are consumed by people living in the city of Tirana. Now we will estimate the Average Annual Committed Effective Dose (AACED) in food spice samples, due to the ingestion of radionuclides to the human body, which is the aim of this work.

From calculated values of the activity concentrations in the spice samples, the AACED due to ingestion of natural and artificial radionuclides were estimated by expression as following:

$$AACED = C_r \cdot DCF \cdot A \qquad (3)$$

where C_r (in kg/year) is the consumption rate of food spices in a year. In our study, the consumption rate values for spices foods are assumed an average 1 kg/year for an adult person according to UNSCEAR 2000 report [15]. DCF (in Sv Bq^{-1}) is the internal dose conversion factor for ingestion, for each radionuclide, that we have calculated activity concentration, which are: for ^{40}K dose conversion factor is 6.2×10^{-9} Sv Bq^{-1}, for ^{226}Ra DCF is 2.8×10^{-7} Sv Bq^{-1}, for ^{232}Th is 2.3×10^{-7} Sv Bq^{-1} and for artificial radionuclides of ^{137}Cs is 1.3×10^{-8} Sv Bq^{-1} respectively for an adult [16]. The average annual committed effective dose in spices foods due to the ingestion of ^{40}K, ^{226}Ra, ^{232}Th, and ^{137}Cs are presented in

Table 2. Range of values of AACED is from 5.61 μSv y^{-1} to 10.91 μSv y^{-1}. The lowest value of AACED 5.61 μSv y^{-1} that was found in Chia spice sample and highest values of 10.91 μSv y^{-1} was found in Turmeric spice sample.

Table 2. Average annual committed effective d*ose (AACED in μSv y^{-1})* by radionuclides of ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs were found in twenty types of spices.

No.	Sample code	AACED (μSv y^{-1})
1	Black pepper	7.86 ± 0.45
2	Curry	5.75 ± 0.44
3	Ginger	7.44 ± 0.36
4	Turmeric	10.91 ± 0.56
5	S. L. cinnamon	9.55 ± 0.55
6	Cloves	9.32 ± 0.55
7	Chia	5.61 ± 0.38
8	Cayenne pepper	10.17 ± 0.56
9	Mix spices 1	7.04 ± 0.43
10	Mix spices 2	6.44 ± 0.37
11	Garlic	7.03 ± 0.45
12	Celery	8.48 ± 0.41
13	Parsley	9.66 ± 0.52
14	Dill	7.51 ± 0.30
15	Winter savory	6.63 ± 0.37
16	Oregano	7.06 ± 0.29
17	Rosemary	8.16 ± 0.31
18	Peppermint	10.05 ± 0.45
19	Bay laurel	8.72 ± 0.34
20	Basil	9.07 ± 0.32

Range of values for the activity concentration of natural radionuclides from Table 1 is wide, from 2.04 Bq kg^{-1} for ^{232}Th to 849.27 Bq kg^{-1} for ^{40}K, while range of AACED is small, from 5.61 μSv y^{-1} to 10.91 μSv y^{-1}. Higher values of the activity concentration are for natural radionuclide of ^{40}K, but dose conversion factor DCF for this radionuclide is very smaller than the others radionuclides of ^{226}Ra and ^{232}Th. The artificial radionuclide of ^{137}Cs has lowest values of the activity concentration and small dose conversion factor, so the dose contribution from this radionuclide in the two spices Oregano and Bay Laurel, is also very small. The estimated average annual committed effective dose (AACED) due to ingestion of these spices varies from 5.61 μSv y^{-1} to 10.91 μSv y^{-1}. Average of AACED for all spices is about 8.12 μSv y^{-1} and higher value is 10.91 μSv y^{-1}, therefore, these values are far below than the world average value dose of 290 μSv y^{-1} for all foods reported by UNSCEAR 2000 [15]. In previous study, we have estimated AACED, due to ingestion from some medicinal and herbal plants used in Albania, and range it was from 4.84 μSv y^{-1} to 34.13 μSv y^{-1}, but consume rate was assumed 2 kg/year [14]. Values of AACED for medicinal and herbal plans and spices in food are somehow approximates, but more important is then these values are very lower than the world average value.

4. Conclusion

In this paper our interest has been to know the level of radioactivity in some spices regardless of the purpose of use. Also, to estimate the average annual committed effective dose in spices foods due to the ingestion of natural radionuclides of ^{40}K, ^{226}Ra, ^{232}Th and artificial radionuclide of ^{137}Cs and therefore, to indicate if there is risk by the consumption imported and locally produced of spices samples in food.

The obtained results indicate that ^{40}K, ^{226}Ra and ^{232}Th was detected in all selected samples of study, whereas the presence of artificial radionuclide of ^{137}Cs was found only in two spices samples. The activity concentration varies from 173.72 ± 9.34 Bq kg^{-1} to 849.47 ± 39.36 Bq kg^{-1} for ^{40}K, from 5.15 ± 0.52 Bq kg^{-1} to 21.01 ± 1.80 Bq kg^{-1} for ^{226}Ra and from 2.04 ± 0.31 Bq kg^{-1} to 21.90 ± 1.78 Bq kg^{-1} for ^{232}Th.

The values found for ^{137}Cs are 0.53 ± 0.21 Bq kg^{-1} in Oregano and 0.41 ± 0.10 Bq kg^{-1} in Bay laurel, while in all other samples are below MDA.

The estimated Average Annual Committed Effective Dose (AACED) due to ingestion of these spices varies from 5.61 ± 0.29 μSv y^{-1} to 10.91 ± 0.56 μSv y^{-1}. All these values are far below than the world average value dose for individual of 290 μSv y^{-1} for all foods reported by UNSCEAR 2000. However, spices were of no special problem because the consumption rate of spices is relatively low in comparison with the consumption rate in a year. Low dose values are by fact that mostly of spices are imported from the Middle East, Far East and Mediterranean countries, which is not affected by the fallout from Chernobyl or nuclear test.

This indicates that no risk is expected by the intake of spices samples in food, that we are using every day in the city of Tirana. The obtained data provide us the baseline levels of natural and artificial radioactivity, a background information for future research on foodstuff, for radiological protection of the human and safety of our health and body, by the consumption of spices foods.

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