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ANALYSIS OF ²²⁶Ra, ²³²Th ⁴⁰K AND ¹³⁷Cs IN SAMPLES OF SOIL FROM SOME AREAS OF REPUBLIC OF MACEDONIA BY USING GAMMA SPECTROMETRY

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ABSTRACT

Taking into consideration the importance of the distribution and transfer of radio nuclides in soil, an attempt was made in this work to determine the concentration of 226 Ra, 232 Th 40 K and 137 Cs in the same. The concentrations of activity in the gamma-absorbed dose rates of the terrestrial naturally occurring radio nuclides, as follows, 226 Ra, 232 Th and 40 K were determined in samples of soil collected from some parts of Republic of Macedonia, i.e. from three major cities in the Republic of Macedonia. The samples are taken by means of a special dosage dispenser which enables sampling of samples at a depth of 0-5 cm, 5-10cm and 10-15cm, thus disabling the sampling above these layers of soil.

An identification of radio nuclides and assessment of their activity has been performed by applying gamma spectrometry. The time of counting for each sample was 65000 s. in order to obtain statistically small mistake. The spectrums were analyzed by a commercially available software GENIE-2000 received from Canberra, Austria. The activity of soil had wide range of values: 20.3 to 82.9 Bq kg⁻¹for ²²⁶Ra, 16.1 to 82.5 Bq kg⁻¹for ²³²Th, 325 to 799.0 Bq kg⁻¹for ⁴⁰K and 9.1 to 24.3 Bq kg⁻¹for ¹³⁷Cs, respectively. The concentrations of these radio nuclides have been compared with the available data from the other countries. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions. Namely, the specific levels of terrestrial environmental radiation are related to the type of rocks from which the soils originate. The obtained data indicate that the average value of activity of ²³²Th is about higher than the one of ²²⁶Ra. The concentration of activity of ⁴⁰K in the soil has greater value than ³²Th and ²²⁶Ra in all soils.

The cause for the existence of ¹³⁷Cs in these soils are the nuclear explosions, waste radioactive materials and other incidents. It reaches the surface of the pedosphere by means of dry and wet flushing and depending on the physical and the chemical properties of the soil, as well as depending on the eventually applied agrotechnical and agrochemical measures, it connects, that is, it penetrates in the deeper soil layers.

Key words: gamma spectrometry, samples of soil, technique for monitoring fission.

INTRODUCTION

People have always been exposed to natural radiations which emanate from the interior and the exterior of the Earth. The exposure to ionizing radiations from natural sources occurs because of the radioactive elements that naturally occur in

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soil and in rocks, the cosmic rays which penetrate from space into Earth's atmosphere, and the internal exposure to radioactive elements through food, water and air.

Usually radionuclides are accumulated on the surface of the ground, and 85% of them are retained at a depth up to 5 cm.

Most of these isotopes originate from the atmosphere, which by means of dry and wet washing (falling), reach the surface of the pedosphere, where depending on the physical and the chemical properties of soil, as well as from the eventually applied agrotechnical and agrochemical

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measures, they are connected, that is, they penetrate into deeper soil layers. There are great differences in the vertical distribution of the radiological contamination of soil, and also between arable and non-arable agricultural surfaces.

At the same time, the mineral fertilization increases the radioactivity of soil, taking into consideration that some fertilizers, such as phosphorus ones, are extremely rich with radioactive matters, which belong to the products of the decay of uranium. (1,2)

Generally, it is clear that the natural radioactivity of the environment and the related external exposure due to gamma-radiation primarily depends on geological and geographical conditions and occurs at different levels of radionuclides in the soils in every region of the world. For this reason, in order to estimate the dose rate caused by radionuclides, their concentrations in soil need to be determined.

Lately, radioactive substances, due to their specific properties, have been subject to numerous research and they are one of the main problems for protection of soil.

Most attention is paid to the research of artificial radionuclides, and the natural radioactivity was less important, in terms of the research, as well as in terms of protection. (3)

However, in order to be able to monitor the artificial radioactivity in the soil, knowledge about the nature of the same is required.

Presence of 40 radioactive elements has been found in the soil, whose contribution is different (4), but it is considered that only three natural radioactive elements in the Earth crust are present with a concentration greater than 10ppm. These are potassium, thorium and uranium.(5).

The natural radioactivity in the soil samples originated from series of U and Th and natural K, so that usually the contents of ²³⁸U, ²³²Th and ⁴⁰K are being determined (6).

Especially significant effect on the level of natural radioactivity, have the different processes of formation of ore bodies which have been monitored in hydro-thermal processes(7)

The artificial radionuclides can also be present like ¹³⁷Cs, which results from the decay of weapon tests. The concentration of activity in the mass of natural radionuclides in the soil was reviewed in (UNSCEAR), for the purpose of further assessment of the dose of gamma radiation from natural sources since the natural radiation mostly contributes for the external dose in the world population (8).

The concentration of radioactivity importantly differs from a country to a country, so that the measurement of the natural radioactivity due to gamma rays from the dose rate is necessary in order to implement caution measures every time when it is considered that the dose is above the recommended limits. In order to assess the earth gamma dose rate for external intake, it is very important to assess the level of natural radioactivity in soils. An attempt was made in this work to determine the concentration of natural radionuclides and artificial radionuclides in the samples of soil collected from different locations, i.e. from three major cities in Republic of Macedonia. The concentrations of activity of ²²⁶Ra, ²³²Th ⁴⁰K and ¹³⁷Cs in the collected samples of soil have been assessed with gamma spectrometry

MATERIALS AND METHODS

Assessment of the natural levels of radioactivity with a technique for gamma spectrometry

The take of soil samples has been performed so that 5 samples have been taken from each location, for the given depths according to the recommendations of IAEA (9)

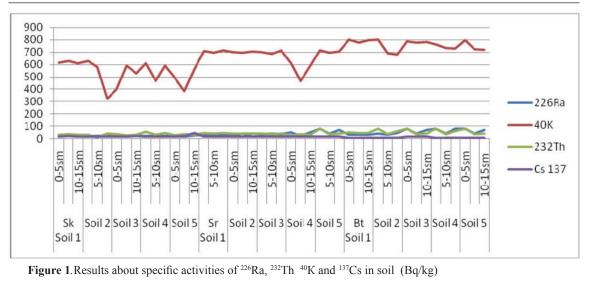
The samples have been taken by using a special dosage dispenser, which is made of a steel pipe with a length of 700mm, where borders are located, which allow sampling of the samples at a depth of 0-5cm, 5-10cm and 10-15cm, thus enabling sampling above these soil layers.

After removing the stones and organic materials, the samples were dried in a furnace at a temperature of about 100°C in a period of 1-2 hours, in order to remove the content of moisture and then they were crushed so that they can pass through a sieve mesh of 150 µm and in order to be homogenized. Then, a sample of (450 ± 0.05) g has been measured and in the end, a divided part of the prepared sample has been packed in a standard plastic dish (marinela) and after appropriate closing of the cover, the dish was sealed with a sealing tape and left for at least 2 weeks (> 7 half-life of ²²²Rn and ²²⁴Ra) before counting with gamma spectrometry in order to ensure that the daughter products of ²²⁶Ra to ²¹⁰Pb and of ²²⁸Th to ²⁰⁸Pb achieve balance with their appropriate radionuclides. The contents of radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K have been assessed by applying a system of gamma spectrometry with high resolution which consists of germanium with high purity (HPGe), coaxial detector (relative efficiency: 30%, active volume: 180 cubic centimeters with beryllium-ending window and FWHM: 2.0 keVat 1332 KeVfor ⁶⁰Co). The time of counting for each

sample was 65000 s. in order to obtain statistically small mistake. The spectrums have been analyzed by a commercially available software GENIE-2000 received from Canberra, Austria.

Table 1:Specific activity of radionuclides (Bq/kg)

Locations	Samples	²²⁶ Ra	⁴⁰ K	²³² Th	¹³⁷ Cs
Skopje Soil 1	0-5sm	24.2	612	35.9	19.6
380550 3011	5-10sm	24.1	626	38.3	21.8
	10-15sm	23.1	610	37.1	18.3
Soil 2	0-5sm	27.1	630	35.7	19.8
	5-10sm	25.0	583	16.2	19.8
	10-15sm	28.6	325	46.8	20.7
Soil 3	0-5sm	23.1	399	41.6	21.2
	5-10sm	23.5	590	29.1	20.3
	10-15sm	24.7	531	36.1	22.3
Soil 4	0-5sm	25.0	612	59.4	20.7
	5-10sm	26.2	468	33.1	21.3
	10-15sm	26.3	592	47.8	21.2
Soil 5	0-5sm	23.2	493	29.9	20.3
	5-10sm	20.3	385	38.3	20.3
	10-15sm	30.0	563	34.3	23.3
Strumica Soil 1	0-5sm	36.6	709	47.2	17.8
Strumea Son 1	5-10sm	36.0	695	46.4	19.3
	10-15sm	37.1	712	47.4	19.3
Soil 2	0-5sm	37.2	699	45.2	19.5
5011 2	5-10sm	22.5	695	46.4	20.4
	10-15sm	43.9	706	46.4	20.4
Soil 3	0-5sm	40.9	699	45.2	20.5
5011 5	5-10sm	43.6	684	38.3	20.3
	10-15sm	41.3	714	46.4	20.3
Soil 4	0-5sm	52.3	612	30.9	17.8
5011 4	5-10sm	23.6	468	40.3	17.8
	10-15sm	54.2	592	34.2	17.3
Soil 5	0-5sm	80.9	716	82.5	17.5
5011 5	5-10sm	43.6	693	38.2	19.8
	10-15sm	72.7	706	46.0	20.5
Ditala Sail 1	0-5sm	36.3	700	51.8	9.4
Bitola Soil 1	5-10sm	36.8	798	50.2	9.4
	10-15sm	37.1	794	50.6	9.2
S - 1 2	0-5sm	43.3	794	82.5	10.8
Soil 2	5-10sm	36.8	689	38.2	9.2
	10-15sm	47.1	680	64.2	9.2
Q - 1 2		80.8		82.5	9.5
Soil 3	0-5sm 5-10sm		786		
		43.6	775	38.2	19.3
S - :1 4	10-15sm	72.7	782 762	46.0	19.4
Soil 4	0-5sm	80.9		82.5	10.4 9.2
	5-10sm	43.6	734	38.2	
0.11.5	10-15sm	82.9	728	64.2	9.3
Soil 5	0-5sm	80.9	796	82.5	10.4
	5-10sm	43.6	725	38.2	9.5
A 1	10-15sm	72.7	720	46.0	9.3
Average value		41.7	655	46.6	17.9



RESULTS AND DISCUSSION

The soil that has been a subject of research was normally fertilized soil. The concentrations of activity of the samples of soil from the area that is being investigated, were individually determined for each depth.

He concentration of activity of ⁴⁰K in the soil has higher value than ³²Th and ²²⁶Ra for all soils, and this may be due to the presence of radioactivity in the fertilizers (10).

It can be assumed that the presence of ¹³⁷Cs is due to the accident in Chernobyl and also due to the low accumulation from different atmospheric tests of nuclear weapon in the neighboring countries.

Its distribution in the soil expresses the quantity

of nuclear decay and the erosive rate of soil. The presence of ¹³⁷Cs in soil is very important and it is a clear indicator that the area under study may have received certain radioactivity from decay, however, it is very difficult, even impossible, to precisely determine the source of this contamination since there are no data about the supplies of radio-cesium in the environment of the selected area before this study (11).

In the current study, it has been noted that the specific activity of these radionuclides differs from one soil to another depending on the geological or typographical character of the area. Besides this, it also depends on the type of the past agricultural activities and different minerals that are present in the soil.

Locations	²²⁶ Ra	⁴⁰ K	²³² Th	¹³⁷ Cs
R.Macedonia	41.76	654.74	46.57	17.87311
Serbia	36.00	596.00	36.00	
Croatia	43.00	423.00	37.00	
World average	32.00	420.00	45.00	

Table 2. Comparison of specific activities of radionuclides in soil samples at different locations (Bq/kg)

From these data, a conclusion can be made that the specific activity of natural radionuclides in Macedonia is similar to the medium value of these radionuclides in the surrounding countries, which have similar geological and pedological structure. We have no available data about specific activities of ¹³⁷Cs in Serbia and Croatia.

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