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Printed by SciBulCom Ltd.,  
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## **EVALUATION OF DOSE FOR THE POPULATION DUE TO NATURAL RADIOACTIVITY OF UNCULTIVATED SOIL FROM THE SURROUNDING OF THE CITY OF SKOPJE**

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**Abstract.** Determination of natural radionuclides in the environment is important as relevant parameter for radiological risks estimations. The objective of this research was focused to define the activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in samples of uncultivated soil, taken from 13 locations in the surrounding of the Skopje city. Within the research performed, the obtained data indicated that the value of <sup>232</sup>Th and <sup>226</sup>Ra activity was within the range from 27.06 to 43.66 Bq kg<sup>-1</sup>, and from 20.38 to 39.44 Bq kg<sup>-1</sup>, respectively. As expected, the <sup>40</sup>K concentration in soil was significantly higher, ranged between 392.48 and 625.80 Bq kg<sup>-1</sup>. The concentrations obtained were the basis for calculation of other parameters that are fundamental for determination of the environmental radiation safety for the population. The obtained results from this study suggest that in the surrounding of Skopje there is no significant radiation risk for the population, i.e. the safe limits were not exceeded, pointing out the insignificant risk that arises from naturally occurring radionuclides in the environmental soil.

**Keywords:** soil radioactivity, natural radionuclides, gamma spectrometry, radiation risk index, radium equivalent.

### **AIMS AND BACKGROUND**

The understanding of environmental parameters and how radionuclides are transferred between environmental systems, may provide a foundation for projections that all criteria and norms of radiation security will be built on later. The migration of radionuclides in the environment does not depend on how radioactivity emerged, rather radionuclides are accumulated in specific parts of the environment depending on the characteristics and the processes that take place in the very environment.

Soil as the basic component of the biosphere, has a significant role in the distribution and transfer of radionuclides. The concentration of radionuclides that

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\* For correspondence.

naturally emerge in soil depends on the type of segments of which soil is formed<sup>1</sup>. All radionuclides that are initially released in the atmosphere deposit on the soil. The extent of natural radioactivity of the environment and the additional external exposure that is due to the gamma radiation primarily depend on the geological and geographical conditions. For this reason there are different levels of natural radioactivity in the soil for each region of the world. Hence, it is important to understand the behaviour of natural radionuclides in the environment, because such data can be used as accompanying parameters for the implementation of a radiological assessment<sup>2,3</sup>.

In every ecosystem it is necessary to make radio-environmental characterisation of soils in order to assess the concentrations of radionuclides in regard to natural basic levels, to assess the potential impact on the human health and to discover their spatial distribution<sup>4-6</sup>. The purpose of this research was focused on the determination of the concentrations of activity of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in the uncultivated soils from location in the surrounding of the city of Skopje. The obtained data were the foundation for the calculation of other parameters that are the basis for the determination of the radiation security in the environment, that is, in the population.

## EXPERIMENTAL

The soil samples were collected during the year of 2014 from different locations in the surrounding of the city of Skopje. The samples were obtained by clearing up the surface vegetation and by elimination of the dead organic material from the surface of the location, and then with a special sampler, taken from different depths of 0–5, 5–10 and 10–15 cm. Soil sampling is performed so that 2 samples have been taken from each location, which is in accordance with the recommendations from IAEA, according to which the number of samples should be 2 or 3.

All soil samples were left to dry separately on plastic plates on room temperature for several days. In order to eliminate moisture, the samples were heated in an electric oven at a temperature of 110°C. After drying, the samples were crushed, homogenised and stored in airtight plastic containers.

The samples were analysed on an instrument-gamma spectrometer (Canberra Packard) with a high-purity germanium detector. The analysis was conducted in containers that were hermetically sealed so <sup>222</sup>Rn produced from decay of <sup>226</sup>Ra will not result in a gas leak. Once time balance between the successors of <sup>238</sup>U and <sup>232</sup>Th series is ensured (21 days), these sealed samples were prepared for an analysis. The obtained spectra from the measurement were analysed by using the programme GENIE 2000. The specific activity of <sup>226</sup>Ra is calculated for an energy line of 186.1 (keV) and <sup>232</sup>Th through its decomposition descendant <sup>228</sup>Ac (second

in the decomposition sequence), that is, through its three gamma decomposing energy lines that emerge at 338.4; 911.07 and 968.9 keV.

The activities of  $^{40}\text{K}$  were determined for its  $\gamma$ -line of 1460 keV. The interval for the time of calculation (counting) was 65.000 s. The natural spectrum was registered immediately after or before the sample calculation.

The external index was calculated in order to assess the equivalent average of annual effective dose intended for the residents of every area.

$$H_{\text{eks}} = A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{k}}/4810,$$

where  $A_{\text{Ra}}$ ,  $A_{\text{Th}}$ ,  $A_{\text{k}}$  are the specific activities (Bq/kg) of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively.

The maximum value of  $H_{\text{ex}}$  that equals 1 corresponds to the upper limit of activity of an equivalent of radium (370 Bq/kg).

Considering that the distribution of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in uncultivated soil is not uniform, the radium equivalent of activity was calculated ( $\text{Ra}_{\text{eq}}$ ).

$$\text{Ra}_{\text{eq}} \text{ (Bq/kg)} = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.07A_{\text{k}}$$

where  $A_{\text{Ra}}$ ,  $A_{\text{Th}}$ ,  $A_{\text{k}}$  are the specific activities (Bq/kg) of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ , respectively.

In order to be able to define  $\text{Ra}_{\text{eq}}$ , it is assumed that 370 Bq/kg  $^{226}\text{Ra}$  or 259 Bq/kg  $^{40}\text{K}$  produce the same dosage strength at 1 m above the soil surface. The value of the radium equivalent of activity of 370 Bq/kg corresponds to the maximum allowed dose for a population of 1 mSv.

## RESULTS AND DISCUSSION

The presented results in Table 1 have mean values of measurements performed on soil samples taken from 3 depths, each with 2 samples.

On the basis of the performed research, the data show that the average value of activity of  $^{232}\text{Th}$  is within a range from 27.06 to 45.66 Bq kg<sup>-1</sup>. The activity of  $^{226}\text{Ra}$  is within the range from 20.38 to 39.44 Bq kg<sup>-1</sup>.

The concentration of activity of  $^{40}\text{K}$  in the soil at all locations has a size higher than the one of  $^{232}\text{Th}$  and  $^{226}\text{Ra}$  for all samples and it is within the range from 392.48 to 625.80 Bq kg<sup>-1</sup>. The mean values of the specific activities were determined from all depths for the same radionuclides where it was calculated that  $^{40}\text{K}$  has the highest specific activity which is 506.54 Bq/kg, then  $^{232}\text{Th}$  35.70 Bq/kg while a value of 28.30 Bq/kg is calculated for  $^{226}\text{Ra}$ .

The obtained data from the measured activities for the three different depths, for all locations from the surrounding of Skopje have been statistically processed and are presented in Table 2.

**Table 1.** Mean values of specific activities of values of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in samples of uncultivated soil

Locality	Specific activity (Bq/kg)		
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$
Petrovec	24.03±1.50	28.26±2.50	516.60±2.30
Belimbegovo	26.01±1.53	32.71±1.20	489.62±20.80
Aracinovo	33.79±1.82	45.66±4.51	540.24±19.00
Radisani	27.32±4.50	34.61±1.20	507.18±20.20
Cucer	23.86±1.30	33.89±3.30	450.80±21.80
Vizbegovo	36.95±2.55	38.11±2.51	605.28±45.22
Bardovci	39.44±4.30	42.91±4.50	625.80±48.20
Saraj	32.32±4.55	40.31±2.20	476.44±21.50
Nerezi	23.62±1.51	38.39±2.00	522.71±19.50
Lisice	20.38±1.33	34.66±2.20	438.25±45.00
Dracevo	22.97±2.00	28.56±4.30	431.44±21.00
Pintija	30.69±1.30	33.57±2.00	522.60±22.00
Batinci	27.12±1.31	35.96±2.30	488.56±19.50
Volkovo	23.70±1.52	27.06±2.40	392.48±20.00

**Table 2.** Descriptive statistics of the mean values of the specific activities of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  according to the depth of the samples of uncultivated soil

Parameter	$^{226}\text{Ra}$			$^{232}\text{Th}$			$^{40}\text{K}$		
	soil depth (cm)			soil depth (cm)			soil depth (cm)		
	0–5	5–10	10–15	0–5	5–10	10–15	0–5	5–10	10–15
Mean value (Bq/kg)	28.19	28.12	28.60	35.21	35.58	36.31	534.58	498.01	487.04
Median (Bq/kg)	26.26	26.72	27.56	34.67	35.10	35.29	527.12	498.65	472.21
Standard deviation (Bq/kg)	5.74	5.61	5.29	5.32	5.57	6.07	60.82	54.89	61.19
Minimum (Bq/kg)	19.20	19.63	20.31	25.49	26.66	26.22	447.00	399.02	403.13
Maximum (Bq/kg)	40.00	39.78	39.22	43.46	46.32	49.67	666.10	611.21	627.82
Ranking (Bq/kg)	20.80	20.15	18.91	17.97	19.66	23.45	219.10	212.19	224.69
Coefficient of asymmetry	0.65	0.64	0.54	-0.21	0.10	0.35	0.63	0.17	1.03
Flatness coefficient	-0.64	-0.46	-0.51	-1.01	-0.80	-0.29	-0.14	-0.32	0.44

The specific activity of  $^{40}\text{K}$  in the samples of uncultivated soil has an order of magnitude higher than the one of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . An important data in the determination of the activity of  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  is their time of half-life because it is different, i.e. the longer the half-life period, the higher the concentration of the radionuclide, which can be noticed in both types of tested soil.

Also the measured values of specific activity of the radionuclides  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  in all soils being tested, are within the world range indicated by international organisations. The mean values of specific activities at different depths



are presented in Figs 1–3. Due to the long half-life period of  $^{232}\text{Th}$ , the values are higher than the ones of  $^{226}\text{Ra}$  in all uncultivated soils in all sampling locations. In terms of the activity in regard to the depth, mutual relation has not been found between the activity and the depth for  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . The measured values of radioactivity indicate that it is accidentally distributed in different soil depths that are being analysed.

Compared to the calculated mean values for the specific activity of all three radionuclides, one can determine that the activity of all analysed natural radionuclides in uncultivated soil is significantly lower than the cultivated soil. This is due to the application of different fertilisers on agricultural soils, in specific recommended quantities, which may increase the radioactivity level in soils.



Fig. 1. Mean values of specific activities of  $^{226}\text{Ra}$  at different depths



Fig. 2. Mean values of specific activities of  $^{232}\text{Th}$  at different depths



Fig. 3. Mean values of specific activities of  $^{40}\text{K}$  at different depths

The values of index of radiation risk ( $H_{\text{eks}}$ ) and radium equivalent ( $\text{Ra}_{\text{eq}}$ ) are presented in Table 3. The mean value of the index of radiation risk  $H_{\text{eks}}$  is 0,31, which indicates that in the surrounding of Skopje, there is no significant radiation risk for the population.

Table 3. Index of radiation risk ( $H_{\text{eks}}$ ) and radium equivalent ( $\text{Ra}_{\text{eq}}$ )

Location	$H_{\text{eks}}$	$\text{Ra}_{\text{eq}}$
Petrovec	0.28	100.62
Belimbegovo	0.29	106.72
Aracinovo	0.38	136.89
Radisani	0.31	112.31
Cucer	0.29	103.87
Vizbegovo	0.37	133.81
Bardovci	0.40	144.60
Saraj	0.34	123.31
Nerezi	0.32	115.10
Lisice	0.28	100.61
Dracevo	0.26	94.01
Pintija	0.32	115.27
Batinci	0.31	112.74
Volkovo	0.25	89.87

The value of the external danger index obtained in this study, regardless of the location and the composition of the soil, does not exceed the security limits, pointing out the insignificant radiation danger that arises from earth radionuclides that emerge naturally.

From the presented results can be determined that the value of the radium equivalent of activity  $\text{Ra}_{\text{eq}}$  in the surrounding of Skopje is from 89.87 Bq/kg

(Volkovo) to 144.60 (Bardovci), and it is below the maximum recommended limit, i.e. 370 Bq kg<sup>-1</sup>, which corresponds to a dose of 1 mSv for population.

Compared to the other countries, the activity of the radium equivalent determined in our research is higher in Nigeria (132 Bq/kg) (Ref. 7), Bangladesh (126.72 Bq/kg) (Ref. 8), Saudi Arabia (140 Bq/kg) (Ref. 9, while lower than Malaysia (186 Bq/kg) (Ref. 10).

The value of radium equivalent  $Ra_{eq}$  and radiation risk index  $H_{eks}$  is lowest in the regions of the Persian Gulf in Iran and the Jordan cities – Ajloun, Amman, Irbid where the country is mostly sandy<sup>11</sup>.

## CONCLUSIONS

In this paper the activity concentrations of <sup>226</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K in the uncultivated soils of the Skopje city area has been determined. The soil samples were taken from 13 locations at depth of 5, 10 and 15 cm.

The value of <sup>232</sup>Th and <sup>226</sup>Ra activity is in the range from 27.06 to 43.66 Bq kg<sup>-1</sup>, and from 20.38 to 39.44 Bq kg<sup>-1</sup>, respectively. Concentration of <sup>40</sup>K in soils is significantly higher, ranging between 392.48 and 625.80 Bq kg<sup>-1</sup>. Also the mean values of the specific activities for the respective radionuclides from all depths were calculated.

The values of the specific activity and the calculated index of radiation risk ( $H_{eks}$ ) and radium equivalent ( $Ra_{eq}$ ) obtained in this study, regardless of the location and the composition of the soil, did not exceed the safety limits, pointing out the insignificant danger for radiation that arises from earth radionuclides that emerge naturally.

These results can be the foundation for a preparation of a radiological map of the studied area, as well as for an enrichment of the world data base. However, in order to be able to define more final conclusions about the risk of exposure of the population in this area, more systematic research of the very area is necessary.

The results can be used as reference values for current assessment of doses due to natural radioactivity in uncultivated soils in the surrounding of the city of Skopje.

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*Received 8 June 2016*

*Revised 26 July 2016*