RAD Conference Proceedings, vol. 1, pp. 71-74, 2016
www.rad-proceedings.org

THE NATURAL RADIONUCLIDES IN SOILS OF SUBOTICA (SERBIA): DISTRIBUTION AND CORRESPONDING GAMMA DOSE RATES

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Abstract. *The activity concentrations of ^{40}K, ^{226}Ra and ^{232}Th from 50 locations on the territory of Subotica, North Serbia, were determined by gamma ray spectrometry. Based on the activity concentrations of investigated radionuclides, the absorbed gamma dose rate in the air was calculated. The mean values of activity concentrations were found to be 290 Bq/kg for ^{40}K, 20 Bq/kg for ^{226}Ra and 18 Bq/kg for ^{232}Th. The total absorbed gamma dose rate varied between 24 and 46 nGy/h. The mean value of 32 nGy/h was lower than the world average value.*

Key words: *Radium, thorium, potassium, exposure, dose*

DOI: 10.21175/RadProc.2016.17

1. INTRODUCTION

Natural radionuclides in soil generate a significant component of the background radiation exposure of the population [1]. The natural radioactivity in soil mainly comes from the uranium and thorium decay series and potassium [2]. Obviously, the actual level of radiation caused by the radionuclide content of rocks and soil varies widely from place to place and the actual background contribution to the external gamma dose rate at a given location can be determined only by measurements. Thus, the dose rate depends on the geological location [3].

Numerous studies have been conducted to assess the radiation exposure due to these radionuclides and the results obtained are exploited to enrich the world's data bank, greatly needed for evaluating worldwide average values of radiometric and dosimetric quantity [4]. Investigation of soil radioactivity receives particular attention worldwide. These results serve as reference information to assess any changes in background level due to any artificial influences on the environment. Baseline data of this type are of importance in making estimations of population exposures. Concentrations in soil and their mutual relationships are presented.

The aim of this work is to determine the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples collected from the territory of Subotica, the area with specific geological settings, using gamma ray spectrometry and to calculate the corresponding absorbed gamma dose rate in the air.

1.1. Study area

Subotica is the most northern city in the Republic of Serbia located on the latitude of 46°5'55" North and the longitude of 19°39'47" East. By the 2011 census, it has 105,681 inhabitants and it is the second largest city in the Autonomous Province of Vojvodina and the sixth in the Republic of Serbia. Subotica and its surroundings have a continental climate with average annual temperature of 11.4 °C. The city is located in the Panonian Plane on 1,008 km^2 [5].

Subotica and its nearest surroundings are built out of loess and loess formations but their thickness is much smaller than in the most parts of Vojvodina, even though both are parts of the edge of the former Pannonian Sea. Sandy soil dominates in Subotica. This sand has two important characteristics: it is free of carbonate, on the surface it has a strong capability of water absorption (it is wet on a relatively low depth, even in summers). Its geomorphological characteristics are homogeneity and plains. Major features of this terrain are subsidence of sediments from Pleistocene and Holocene that are mostly spread through the creation of Paleozoic and Mesozoic formations. The whole Pannonian Basin was underwater which indicates a very intensive subsidence of marine and freshwater formations. By the step-by-step paving of the Sarmatian Sea and the recent part of Tethys, the ocean land was formed in this part of Europe where these weakly tied segments came to the surface where they were exposed to glacier, water or wind erosion during ice and semi-ice phases [6].

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2. MATERIALS AND METHODS

The samples of undisturbed soils were collected from 50 locations in urban area of Subotica (about 60 km^2) during 2013-2015. From each location, 3-4 subsamples were collected by the template method [7], up to the depth of 10 cm, and then mixed (put together) into one composite sample Samples were dried at 105 °C to a constant weight and then homogenized. The homogenized samples were placed in 0.5 L Marinelli beakers. The beakers were hermetically sealed and kept aside for about a month to ensure the equilibrium between ^{226}Ra and its daughters before being taken for the gamma spectrometric analysis.

The measurements were performed using p-type high-purity germanium detector ORTEC-AMATEC, (relative efficiency 49% and resolution 1.95 keV, for ^{60}Co at 1.33 MeV, 8192 channels) shielded with 10 cm lead internally lined with 2 mm copper foil. The weight of each sample was approximately 0.5 kg. The activity of each sample was measured for 60 ks. A mixed calibration source MBSS 2 (total activity 38 kBq on 16 august 2010 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co,^{60}Co,^{137}Cs, ^{113}Sn, ^{85}Sr,^{88}Y,^{203}Hg) from the Czech Metrological Institute was used for efficiency calibration in the same geometry as the measured samples.

For the purpose of quality assurance, independent checks on calibration were performed using two standard reference materials from the International Atomic Energy Agency (IAEA-385) and the Environmental Measurement Laboratory, United States Department of Energy (QAP-9803). The counting errors were 5-10%. The activity of ^{226}Ra was evaluated from the gamma ray of 609.3 keV of ^{214}Bi and 351.9 keV of ^{214}Pb, while 911.2 and 969.1 keV gamma-ray lines emitted by ^{228}Ac and 238.6 keV emitted by ^{212}Pb was used to determine ^{232}Th. The activity of ^{40}K was determined using its 1460.8 keV gamma-ray line, taken into account all systematic and random uncertainties. The minimum detectable activity for each radionuclide was determined from the background radiation spectrum for the same counting time as for soil samples and was estimated to be 0.4 Bq/kg for ^{226}Ra, 0.8 Bq/kg for ^{232}Th and for 2.0 Bq/kg ^{40}K.

Gamma Vision 32 was used to process the obtained spectra [8]. The statistical evaluation of the obtained results was performed using the statistical package *SPSS* 10.0 for *Windows*[9].

2.1. Absorbed gamma dose rate

The external gamma dose rate in the air at 1 m above ground level was calculated from the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil assuming that other radionuclides, such as ^{137}Cs, ^{90}Sr and the ^{235}U series, can be neglected as they contribute very little to the total dose from the environmental background. The calculations were performed according to the following equation (1):

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.042A_K \quad (1)$$

where A_{Ra}, A_{Th}, and A_K are activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively [10-12].

3. RESULTS AND DISCUSSION

Descriptive statistics for activity concentrations of primordial radionuclides in soil samples collected in the Subotica area is presented in Table 1.

Table 1. Descriptive statistics of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for all analyzed soil samples

Parameter	Activity concentration (Bq kg^{-1})		
	^{226}Ra	^{232}Th	^{40}K
Mode	19	15	350
Mean	20	18	290
Median	19	18	290
St. deviation	5	3	63
Minimum	12	13	260
Maximum	33	23	390

The mean activity concentrations for ^{226}Ra ^{232}Th and ^{40}K were lower than the averages for Serbia, i.e. 33 Bq/kg, 36 Bq/kg 510 Bq/kg for these radionuclides, respectively [10], which is the consequence of the difference in geological structures of the study area and the whole territory of Serbia. The territory of Serbia includes a great variety of rock complexes (magmatic, sedimentary, metamorphic) which are markedly different in age, genesis, mineral content, petrochemical and geochemical characteristics and thus in the content of radionuclides. The activity concentrations of analyzed radionuclides are similar to those obtained nearby Palic (310 Bq/kg for ^{40}K, 19.9 Bq/kg for ^{226}Ra and 23.5 Bq/kg for ^{232}Th) [13] Activity concentrations of analyzed radionuclides in soil are lower than those reported for the neighboring Hungary {^{40}K (97-701) Bq/kg, ^{226}Ra (17-93) Bq/kg and ^{232}Th (15-55) Bq/kg} [2].They are of the same order of magnitude as activity concentrations in the soil of Csongrad, the city in Southeast Hungary (about 130 km far from Subotica) with similar geological substrate {^{40}K (276-453) Bq/kg, ^{226}Ra (14-44) Bq/kg, ^{232}Th (14-35) Bq/kg} [14].

The values of total gamma dose rates varied between 24 and 46 nGy h^{-1} with the mean value of 32 nGy h^{-1}. According to UNSCEAR (2000) [2], the dose rate in air outdoors from terrestrial gamma rays in normal circumstances is approximately 58 nGy h^{-1} with average ranges by country from 1 to 1100 nGy h^{-1}. The mean value for soils analyzed in this study is about 40% lower than the world average. The contribution to the total absorbed gamma dose rate by ^{226}Ra, ^{232}Th and ^{40}K was 29%, 34% and 37%, respectively. The contribution of each radionuclide to the total dose depends primarily on the soil type and geological background of each investigated location. The range and the mean values of the absorbed gamma dose rate due to ^{226}Ra, ^{232}Th and ^{40}K obtained for Subotica are compared with values reported for cities worldwide (Table 2).

Table 2. Absorbed gamma dose rates in cities from different parts of the world compared with those obtained in the present study

City	D (nGy h^{-1})
Bangalore, India [15]	117 (61-202)
Csongrad, Hungary [14]	38
Debrecen, Hungary [14]	24
Faisalabad, Pakistan[16]	73 (62-79)
Istanbul, Turkey[17]	49 (15-71)
Palic, Serbia[13]	36
Horgos, Serbia[13]	32
Belgrade, Serbia[18]	60 (25-90)
Alkharje, Saudi Arabia [19]	24 (15-30)
Tafila, Jordan [20]	40(22-63)
Tripoli, Libya [21]	23 (20-24)
Windhoek, Namibia [22]	56 (41-70)
Zacatecas, Mexico [23]	45 (28-67)
Zahedan, Iran [24]	158 (16-300)
Subotica,this study	32 (24-46)

It can be seen that the mean value of the gamma dose rate observed in the present study (32 nGy h^{-1}) lies within the values reported worldwide. It is comparable with those reported for the cities of Palic and Horgos [13], which are close to Subotica. The obtained results were similar to cities that have a dominant sandy soil composition, such as Alkharje, Saudi Arabia [19] and Tripoli, Libya [21]. These differences in the terrestrial radioactivity and the associated external exposure due to gamma radiation, are caused by geological and geographical specificities of investigated areas.

5. Conclusions

The mean values of activity concentrations of analyzed radionuclides were found to be 290 Bq/kg for ^{40}K, 20 Bq/kg for ^{226}Ra and 18 Bq/kg for ^{232}Th. The mean calculated value of the total absorbed gamma dose rate of 32 nGy/h (24-46 nGy/h) was lower than the world average. The obtained activity concentrations and total gamma dose rates are lower than the world average values. The results obtained in this study may be used for the preliminary estimation of the population exposure due to natural radionuclides. Further investigation is needed before definite conclusions on this issue are drawn.

***Acknowledgement**: This work was supported by the Ministry of Education and Science of the Republic of Serbia (Project No. III 43009).*

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