Depth distribution of selected geogenic radionuclides (\(^{40}\)K, \(^{226}\)Ra, \(^{232}\)Th) and anthropogenic \(^{137}\)Cs in an undisturbed forest soil in East Slovenia

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To reinforce Slovenian radioecological survey information, 20 soil profiles divided into four depth increments of 10 cm were collected in an undisturbed forest in east Slovenia (46°44’N, 16°7’E) to establish the natural (\(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th) and anthropogenic (\(^{137}\)Cs) level of radioactivity. Using gamma spectrometry and depending on the depth increment, the average activity concentration of radionuclides ranged from 0.5 to 70 Bq kg\(^{-1}\) for \(^{137}\)Cs, from 535 to 703 Bq kg\(^{-1}\) for \(^{40}\)K, from 49 to 52 Bq kg\(^{-1}\) for \(^{226}\)Ra and from 54 to 62 Bq kg\(^{-1}\) for \(^{232}\)Th. Based on top soil data (0-10 cm), the radium equivalent activity (\(Ra_{eq}\)) is 167 ± 7 Bq kg\(^{-1}\), while absorbed dose rate (\(D\)) due to external gamma radiation of 79 ± 3 nGy h\(^{-1}\) is higher than the world population-weighted average, reported as 58 nGy h\(^{-1}\) by UNSCEAR.

Keywords: \(^{137}\)Cs, \(^{40}\)K, \(^{226}\)Ra, \(^{232}\)Th, Radium equivalent activity, External gamma dose rate, Radionuclides

1 Introduction

Measurements of natural background radiation and anthropogenic radionuclides in the terrestrial environment, especially in soil, have been carried out in many countries for several decades to establish baseline data on radiation level\(^{1}\). The total received dose due to ionizing radiation in the outdoor environment is the sum from the following sources: (i) naturally occurring radionuclides (NOR) from geogenic sources (uranium, thorium and potassium), (ii) cosmic radiation and (iii) anthropogenic or artificial radionuclides. The landscape distribution of radionuclides is affected by weathering and erosive processes and depends on their origin, their physico-chemical behaviour and geographical conditions. The natural radioactivity in soil can be used as background information by which possible man-made contamination can be detected and quantitatively determined\(^{2}\).

The greatest part of the external irradiation for humans comes from terrestrial radionuclides (e.g. \(^{40}\)K, \(^{226}\)Ra, \(^{232}\)Th). Generally, \(^{40}\)K is an important component and the relevance of this long-living natural geogenic radionuclide is linked to its considerable uptake by plants\(^{3}\). Effectively \(^{40}\)K is on average responsible for 98% of the gamma emission of the ‘primordial’ radionuclides present in the earth\(^{4}\). The anthropogenic fission product \(^{137}\)Cs which was introduced to the environment during past nuclear aerial weapon tests (late 1950s-early 1960s) as well as from the Chernobyl accident (1986), does also contribute significantly to ambient radioactivity. Based on its environmental behaviour, the \(^{137}\)Cs is also the most well-known fallout radionuclide soil tracer which has been tested and validated worldwide to assess soil erosion processes\(^{5}\), and a preliminary study has been conducted by the authors in Slovenia to evaluate the base-line level\(^{6}\) of \(^{137}\)Cs.

To assess the external exposure due to gamma emitters, the radium equivalent activity (\(Ra_{eq}\)) and the absorbed dose rate (\(D\)) are considered as the most widely used international parameters in radiological risk assessment\(^{7,8}\). Hence, the radioecological effects of NOR and anthropogenic radioisotopes on human health have been investigated to evaluate the total received dose of ionizing radiation in the outdoor environment. Effectively, the human exposure to NOR can be calculated through an annual effective dose (in µSv y\(^{-1}\)) considering the total external gamma dose rate and a specific indoor occupancy factor of the population\(^{7,9}\).

According to existing literature so far, information on the concentrations of natural and anthropogenic gamma radionuclides and the associated absorbed...
dose rates is scarce in Slovenia\textsuperscript{10}. The purpose of this study was to contribute to the radioecological survey information in Slovenia by: (i) establishing a background level of the following naturally occurring radionuclides: \textsuperscript{40}K, \textsuperscript{226}Ra and \textsuperscript{232}Th and the anthropogenic \textsuperscript{137}Cs and (ii) providing information on the terrestrial (NOR) and anthropogenic (\textsuperscript{137}Cs) gamma radiation dose rate in soils of the study area.

2 Experimental Details

2.1. Study site and soil sample analysis

A 2 hectares flat oak forest in Šalamenci located in East Slovenia at the beginning of the Pannonian plains (46°44′N, 16°6′E) was selected for evaluating the background radioactivity level. The investigated site is located on the edge of the alluvial Pleistocene terrace\textsuperscript{11}, consisting of silty clay sediments deposited by Ledava river and the subsidiary affluent. Developed on non-carbonated substratum, the Šalamenci forest soil is a Haplic Stagnosol, strongly acidic developed over 70 cm deep. It was, further, characterized as a silt loam with high organic matter content and with bulk density increasing from 1 to 1.5 g cm\textsuperscript{-3} from top soil to 40 cm depth.

In this undisturbed forest, according to a 40×30 m grid, 20 soil cores were collected along a regular grid at 4 different depth increments (0-10, 10-20, 20-30 and 30-40 cm). After pre-treatment of the soil layers (drying and sieving through 2 mm diameter mesh), the samples were analysed for radionuclide activities (\textsuperscript{\textsuperscript{\textsuperscript{40}}K, \textsuperscript{226}Ra, \textsuperscript{232}Th and \textsuperscript{137}Cs) using a HPGe coaxial gamma detector. Calibration of equipment, analysis and quality control of the measurements were performed following IAEA standard procedure\textsuperscript{12}.

2.2 Radium equivalent activity, NOR and \textsuperscript{137}Cs external gamma dose rate calculation

(i) The radium equivalent activity (Ra\textsubscript{eq} in Bq kg\textsuperscript{-1}), also called the radiation hazard index used to compare the specific activity of matrix/materials, is based on the assumption that 370 Bq kg\textsuperscript{-1} of \textsuperscript{226}Ra, 259 Bq kg\textsuperscript{-1} of \textsuperscript{232}Th and 4810 Bq kg\textsuperscript{-1} of \textsuperscript{40}K produce the same gamma dose rate. Ra\textsubscript{eq} was calculated using the following equation\textsuperscript{13}:

\[
Ra_{eq}(\text{Bq kg}^{-1}) = C_{Ra} + 1.43C_{Th} + 0.07C_{K}\quad \ldots(1)
\]

where \(C_{Ra}, C_{Th}, C_{K}\) are the soil activity concentrations (in Bq kg\textsuperscript{-1 dry weight}) of \textsuperscript{226}Ra, \textsuperscript{232}Th and \textsuperscript{40}K, respectively.

(ii) The absorbed dose rate in the air at 1 m above ground level from external exposure to gamma rays from naturally occurring radionuclides – \(D\) in nGy h\textsuperscript{-1} – was calculated using dose coefficients and the equation proposed by UNSCEAR:

\[
D(\text{nGy h}^{-1}) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_{K}\quad \ldots(2)
\]

where \(C_{Ra}, C_{Th}, C_{K}\) are the soil activity concentrations (in Bq kg\textsuperscript{-1 dry weight) of \textsuperscript{226}Ra, \textsuperscript{232}Th and \textsuperscript{40}K, respectively.

For the anthropogenic radionuclide (\textsuperscript{137}Cs), the external gamma dose rate was estimated using the dose rate conversion factors obtained from computational codes, which is simulated by Monte Carlo methods. Using the Monte Carlo N-Particle code (MCNP) of Los Alamos\textsuperscript{14}, this dose rate can be evaluated as:

\[
D(\text{nGy h}^{-1}) = 0.11761C_{Cs}\quad \ldots(3)
\]

where \(C_{Cs}\) is the specific soil activity (in Bq kg\textsuperscript{-1 dry weight) of \textsuperscript{137}Cs. This dose rate estimate has been used for \textsuperscript{137}Cs soil profiles that decrease exponentially with depth, as is the case in most undisturbed, forest terrains\textsuperscript{15}.

3 Results and Discussion

3.1 Activity levels of the naturally occurring radionuclides and anthropogenic \textsuperscript{137}Cs

Depending on the depth increment, the average activity concentration of radionuclides ranged from 0.5 to 70 Bq kg\textsuperscript{-1} for \textsuperscript{137}Cs, from 535 to 703 Bq kg\textsuperscript{-1} for \textsuperscript{40}K, from 49 to 52 Bq kg\textsuperscript{-1} for \textsuperscript{226}Ra and from 54 to 62 Bq kg\textsuperscript{-1} for \textsuperscript{232}Th. The average activity concentration of NOR and \textsuperscript{137}Cs for each soil layer is presented in Table 1. From these results, it can be noticed that: (i) the \textsuperscript{137}Cs content decreases exponentially with depth; (ii) \textsuperscript{40}K slightly increases with depth (similar results were also found in forest soil in Germany\textsuperscript{16}); and (iii) activity concentrations of \textsuperscript{226}Ra and \textsuperscript{232}Th are almost constant with depth.

The world average activity concentrations reported for the naturally occurring radionuclides in top soil is 412 Bq kg\textsuperscript{-1} with a range in reported values from 140 to 850 for \textsuperscript{40}K, 32 Bq kg\textsuperscript{-1} with a range from 17 to 60 for \textsuperscript{226}Ra, and 45 Bq kg\textsuperscript{-1} with a range from 11 to 64 for \textsuperscript{232}Th. Comparing our results with the UNSCEAR world average data, the activity concentration of all NOR (i.e. \textsuperscript{40}K, \textsuperscript{226}Ra and \textsuperscript{232}Th) in the Slovenian forest soil is above the world average value even though still within the world range. Higher activity concentrations of natural radionuclides in our study (Table 1) were found as well as in comparison with the results from Jankovic \textit{et al}.\textsuperscript{8} which presented an average value of
536, 47, 41 Bq kg\(^{-1}\) for \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th in Serbian soils, respectively. Moreover, the activity concentrations of NOR in this forest are in the range of data observed in Serbia and Kosovo which were 190 to 2400 Bq kg\(^{-1}\) for \(^{40}\)K, 10 to 14700 Bq kg\(^{-1}\) for \(^{226}\)Ra and 16 to 150 Bq kg\(^{-1}\) for \(^{232}\)Th (Ref.17). High elevated concentration value of the naturally occurring radionuclides was found in Ikaria, an island in the Eastern Aegean Sea of Greece\(^{18}\). The results of mass activity of radionuclides in soils from that study ranged from 238 to 1439 Bq kg\(^{-1}\) for \(^{40}\)K, from 17 to 422 Bq kg\(^{-1}\) for \(^{226}\)Ra and from 5 to 296 Bq kg\(^{-1}\) for \(^{232}\)Th. Their results were relatively high comparing to ours, especially the maximum value as this area is a geothermal spring.

As reported in Table 1, the results concerning the \(^{137}\)Cs content can be summarised as following: most of the \(^{137}\)Cs is present in the top soil with 98% accumulated in the first 20 cm, the total areal inventory reached 7300 ± 2500 Bq m\(^{-2}\) and approximately 45% of this current inventory is due to the Chernobyl contribution\(^{6}\) that occurred in 1986.

### 3.2 Radium equivalent activity and external gamma dose rate of the forest site

Based on top soil data (0-10 cm) of mass activities of the naturally occurring radionuclides (Table 1), the radium equivalent activity (Ra\(_{eq}\)) was evaluated at 167 ± 7 Bq kg\(^{-1}\) (mean ±SD).

The absorbed dose rate \((D)\) can be calculated from the mass activities of \(^{226}\)Ra, \(^{232}\)Th and \(^{40}\)K in the soil, for this the activity concentrations in the 0-10 cm layer were used (Table 1), giving a value of 79 ± 3 nGy h\(^{-1}\) (mean ±SD). This estimated value is approximate, because of the assumptions of secular equilibrium in the decay chains and homogeneity in the soil. The \(^{40}\)K mass activities increase slightly with depth, however due to the significant attenuation of gamma rays from the lower levels, this change with depth would result in only a small influence on the actual \(D\) value at the surface. The \(D\) value at the surface of this forest soil is about 40% higher than the global average external exposure rate from terrestrial gamma radiation\(^{8}\) established at 58 nGy h\(^{-1}\) by UNSCEAR.

The contribution to the external dose rate of the NOR due to \(^{232}\)Th is the most important in this forest soil with 42%, while \(^{226}\)Ra and \(^{40}\)K contribute to 30% and 28% of the \(D\) value, respectively. On the other hand, the assessment of external gamma dose rate for \(^{137}\)Cs varies with depth increment from 8.2 ± 3.9 nGy h\(^{-1}\) (top soil; 0-10 cm) to 0.06 ± 0.03 (30-40 cm), the first 10 cm contributing to more than 90% to the total dose produced by \(^{137}\)Cs. According to the protocol proposed by Golikov et al\(^{19}\), a similar value of 10 nGy h\(^{-1}\) has been obtained for the \(D\) value linked to the \(^{137}\)Cs content of the total soil profiles (0-40 cm).

### 4 Conclusions

From the selected naturally occurring radionuclides (i.e. \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th) in soils from the study site in Slovenia, only the vertical profile activity of \(^{40}\)K exhibits a heterogeneous distribution with an increase of its content with depth. Vertical distribution of the anthropogenic \(^{137}\)Cs in this forest soils showed an exponential decrease with depth, as expected in undisturbed forest, giving an average total areal inventory of 7300 ± 2500 Bq m\(^{-2}\) (mean±SD; \(n = 20\)). This radio-caesium background activity value will be used in a future investigation to assess soil degradation magnitude in the neighbouring agricultural field using the \(^{137}\)Cs method.

Based on activity levels of the NOR the external gamma dose rate of the top soil (79 ± 3 nGy h\(^{-1}\)) in this forest is higher (approx. 40%) than the world average of 58 nGy h\(^{-1}\) reported by UNSCEAR.
The $^{137}\text{Cs}$ anthropogenic top soil contribution to the total external dose rate represents only an addition of 10% to the external dose rate of the NOR.

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