

Long-term monitoring of natural radionuclides in Uzhgorod city, Ukraine

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Abstract The goal of the present work is to determine the specific activity of terrestrial thorium, uranium and potassium in Uzhgorod city and to predict the values of specific activity over the whole area of Uzhgorod city. The following values for the average specific activity in 2006–2012 were found for different radioisotopes from the surface soils (in Bq kg^{-1}): $^{40}\text{K} = 304.0 \pm 12.3$; $^{232}\text{Th} = 24.2 \pm 2.1$; $^{238}\text{U} = 19.2 \pm 1.2$. The average value of the absorbed dose was equal to $36.3 \pm 2.3 \text{ nGy h}^{-1}$. The average value of the external hazard index was equal to 0.22. The average annual effective dose of natural radionuclides was equal to $4.5 \pm 0.3 \cdot 10^{-5} \text{ Sv}$.

Keywords Natural radionuclides · Specific activity · Absorbed dose · External hazard index · Annual effective dose

Introduction

The research into the background of natural radioactivity worldwide is aimed at cosmic rays and terrestrial radiation as presented in UNSCEAR [1]. Natural background radioactivity depends on geological and geographical conditions and is randomly distributed in soil in different regions of the world [2, 3].

Radionuclides from the ^{238}U and ^{232}Th decay series and ^{40}K are widely spread in the environment of the Earth's crust which generates a significant amount of background

radiation exposure of the world population. Measurement of these nuclides allows to determine the radioactive concentrations dependence on soil types, the external sources of radiation to humans and the background baseline of the natural radioactivity. The estimation of the total radiation dose for the world population has shown that about 96 % of this dose is from natural sources [4, 5].

Uzhgorod city is located in the western Ukraine, at the border with Slovakia and near the border with Hungary. The city is located at the foothills of the Carpathian Mountains in the east and in the west of the Danube lowland. The river Uzh which flows through Uzhgorod, starts from the western slopes of the Carpathians, its basin area is around 2000 km^2 . Podzolic and gray forest soil types prevail in the city.

The systematized long-term research in the urban areas of the Ukrainian Carpathians can be used as a reference for the ecological certification and classification of Uzhgorod surface soils, and also helps to study the geology of the area [6, 7]. Determination of the concentration of radionuclides in surface soil is essential for assessment of radiological risk to human health [8].

The present study is initiated to determine the contribution of the natural radionuclides ^{238}U , ^{232}Th , and ^{40}K in soil from 37 sampling locations in Uzhgorod within the period of 7 years using the gamma-ray spectrometry to compute activity and absorbed gamma dose rate in the air due to the presence of the terrestrial radionuclides in soil samples and also predict the values of specific activity over the whole area of Uzhgorod city.

Materials and methods

The samples from the surface soils were collected from 37 points (Fig. 1) in the Uzhgorod area during 2006–2012 years from the same sampling point every year. The global

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Fig. 1 Geographical position of Uzhgorod city and relevant sampling points

positioning and navigation system (GPS) was used to obtain information about the geographical positions of the sampling locations (Table 1).

Each sample was obtained from the 3–5 sub-samples collected from the 0–10 cm depth and merged in situ.

All samples were dried at the temperature of 60 °C during 48 h, sieved through the 2 mm strainer, weighted and placed/stored in the cylindrical polyethylene containers (0.5 dm³ volume) for the further measurements.

The measurements were carried out using high resolution gamma-spectrometry technique with the Ge(Li) detector DGDK-100 V. The detector was placed inside the massive 15 cm thick lead shield with the inner aluminum (3 mm thick), copper (8 mm thick) and cadmium (1 mm thick) sheets in order to reduce background radiation. The detector has an average gamma-ray photo-peak resolution of 3.5 keV for the 1332.5 keV (⁶⁰Co) line. The bulk standard γ -sources (⁴⁰K, ¹³⁷Cs, ¹⁵²Eu, ²³²Th) were used for the detector efficiency calibration.

The absolute efficiency of the detector is related to energy by the expression [9]:

$$\ln \varepsilon(E_\gamma) = -0.1909 \left(\frac{\ln E_\gamma}{E_0} \right) + \left(-0.2195 \left(\frac{\ln E_\gamma^2}{E_0} \right) \right) + 4421.445 \left(\frac{\ln E_\gamma^3}{E_0} \right) - \frac{0.0161}{E_a} \quad (1)$$

where $\varepsilon(E_\gamma)$ is the γ -spectrometric detector efficiency, E_γ is the γ -quanta energy (keV), $E_0 = 1$ keV and $E_a = 1.8442$ keV.

The minimum detectable activity (MDA) of the system was estimated with the 95 % confidence level using the following formula [10, 11]:

$$\text{MDA} = \frac{4.66 \times B^{0.5}}{t \times \gamma \times \varepsilon \times w} \quad (2)$$

where B is the background counts, t is the counting time (s), γ is the gamma emission probability, ε is the absolute detector efficiency at the particular gamma energy and w is the sample weight (kg).

The MDA with the 95 % confidence level (for the 650 g soil sample and the 20,000 s counting time) for the important radionuclides such as ²³⁸U (²²⁶Ra), ²³²Th, and ⁴⁰K are 6.38, 9.23, 36.85 Bq kg⁻¹, respectively.

All samples were measured with an identical geometry. The measurement time was equal to 20,000 s for each sample.

The typical gamma spectra of the sample and the background are shown in Fig. 2.

When the measurements were ongoing the spectrometer was controlled for the following parameters: channel drift, energy resolution and efficiency. The variation of these parameters did not exceed 1 % during the measurements.

The activity concentrations of ²³⁸U (²²⁶Ra) were calculated using the weighted average values of the γ -ray lines of ²¹⁴Bi.

Table 1 The GPS coordinates of the sampling locations

Sampling points	Geographical coordinate	
	Latitude	Longitude
1	48°38'44.38"	22°20'25.48"
2	48°36'55.26"	22°19'13.37"
3	48°37'55.42"	22°19'47.10"
4	48°37'1.31"	22°19'29.48"
5	48°37'3.96"	22°18'15.97"
6	48°37'11.67"	22°18'35.64"
7	48°38'34.39"	22°18'32.68"
8	48°38'13.65"	22°19'19.49"
9	48°38'49.10"	22°18'24.99"
10	48°37'28.86"	22°18'10.03"
11	48°38'55.20"	22°19'45.66"
12	48°37'7.30"	22°19'36.39"
13	48°37'56.72"	22°19'3.28"
14	48°37'8.50"	22°17'51.89"
15	48°39'17.66"	22°21'14.78"
16	48°37'27.67"	22°16'13.83"
17	48°37'27.75"	22°17'3.33"
18	48°36'53.61"	22°15'23.70"
19	48°38'52.80"	22°19'23.78"
20	48°35'53.87"	22°18'47.07"
21	48°36'36.06"	22°17'3.62"
22	48°36'24.41"	22°16'6.01"
23	48°37'23.89"	22°15'14.55"
24	48°38'31.81"	22°16'2.78"
25	48°38'15.83"	22°17'32.11"
26	48°39'12.27"	22°16'46.59"
27	48°37'34.85"	22°20'12.64"
28	48°35'51.82"	22°17'36.76"
29	48°38'48.31"	22°18'33.73"
30	48°37'51.80"	22°17'4.05"
31	48°36'44.98"	22°18'44.76"
32	48°37'0.55"	22°18'31.74"
33	48°37'32.71"	22°18'15.90"
34	48°37'34.94"	22°18'16.96"
35	48°37'33.24"	22°17'32.48"
36	48°37'20.15"	22°15'44.29"
37	48°35'55.23"	22°21'2.37"

In the case of the ²³²Th series, the activities were calculated by using the gamma-ray lines of ²²⁸Ac and ²⁰⁸Tl. The specific activity (Bq kg⁻¹) of the samples was calculated using the following formula:

$$A = \frac{C}{t \times \gamma \times \varepsilon \times w} \quad (3)$$

where *C* is the net peak area, *t* is the counting time (s), γ is the gamma emission probability, ε is the absolute efficiency

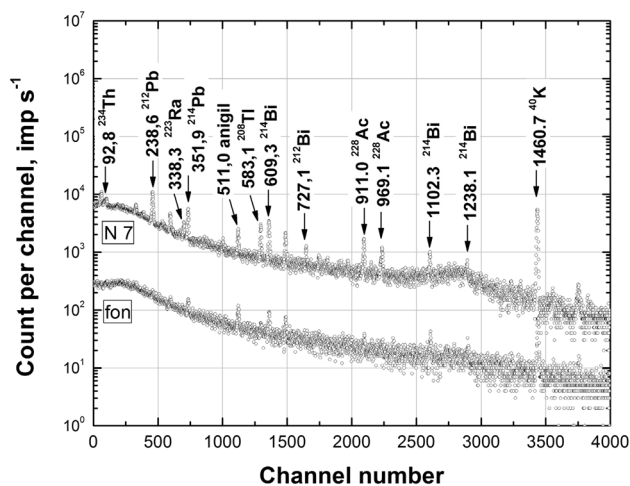


Fig. 2 Gamma spectra of the sample no. 7 and the background

of the detector at the particular gamma energy and *w* is the dry weight of the sample (kg).

The statistic error of the measured specific activity is in the range of 8–12 %.

The specific activity values were used in order to determine the absorbed doses.

The total air absorbed dose rate (nGy h⁻¹) at 1 m above the ground level due to the specific activities of natural radionuclides was calculated using the eq [1]:

$$D = 0.462A_U + 0.604A_{Th} + 0.042A_K \quad (4)$$

where *A_U*, *A_{Th}* and *A_K* are the specific activities [Bq kg⁻¹] of the ²³⁸U, ²³²Th, and ⁴⁰K radioisotopes respectively.

The values of the external hazard index of natural radionuclides were determined by the following formula:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (5)$$

The values of the external hazard index are usually less than 1, but in case of the artificial (technogenic) pollution they exceed 1 [1].

The level of the annual effective dose of natural radionuclides was determined by the formula [1]:

$$S_{ef} = D \times 24 \times 365 \times 0.7 \times 0.2 \quad (6)$$

where *D* is the absorbed dose [nGy h⁻¹].

Results and discussion

The following values of the average specific activity in 2006–2012 years were detected for different radioisotopes from the surface soils of the Uzhgorod area: ⁴⁰K = 304.0 ± 12.3 Bq kg⁻¹, ²³²Th (²²⁸Ac) = 24.2 ± 2.1 Bq kg⁻¹, ²³⁸U (²¹⁴Bi) = 19.2 ± 1.2 Bq kg⁻¹ (see Table 2).

Table 2 Activity concentrations (Bq kg^{-1}) of terrestrial radionuclides in the surface soil samples collected in Uzhgorod and world average value

	^{238}U	^{232}Th	^{40}K
Average	19.2 ± 1.2	24.2 ± 2.1	304.0 ± 12.3
Min.	15.1 ± 1.0	19.8 ± 1.7	235.8 ± 9.9
Max.	23.0 ± 1.3	28.9 ± 2.4	374.6 ± 15.5
Median	18.7 ± 1.1	23.6 ± 2.0	310.0 ± 11.7
World average value [1]	35	30	400

The average values of the specific activity and dose rate for ^{40}K , ^{232}Th , and ^{238}U of the surface soil samples in 2006–2012 are shown in Figs. 3, 4 and 5.

The values of the specific activity of the natural ^{40}K radionuclide and the members of the ^{232}Th and ^{238}U series were

used for constructing the experimental GIS maps (this geostatistical technique was applied to interpolate the values of the specific activity of the terrestrial radionuclide ^{40}K and the members of the ^{232}Th and ^{238}U series in the sampling points).

The geographic information mapping system may be determined as production of maps by using specific activity and computer program that calculates the interpolation values at the locations of the area under study [12]. The GIS maps of the average specific activity of the natural ^{226}Ra , ^{232}Th and ^{40}K radionuclides in the surface layers of the Uzhgorod soils were presented. For the GIS maps creation we have used the Quantum GIS software product, which is an open-source GIS system freely available on the internet for the general public.

The GIS maps of the specific activity for ^{40}K , ^{232}Th , and ^{238}U of the surface soil samples are shown in Figs. 6, 7 and 8.

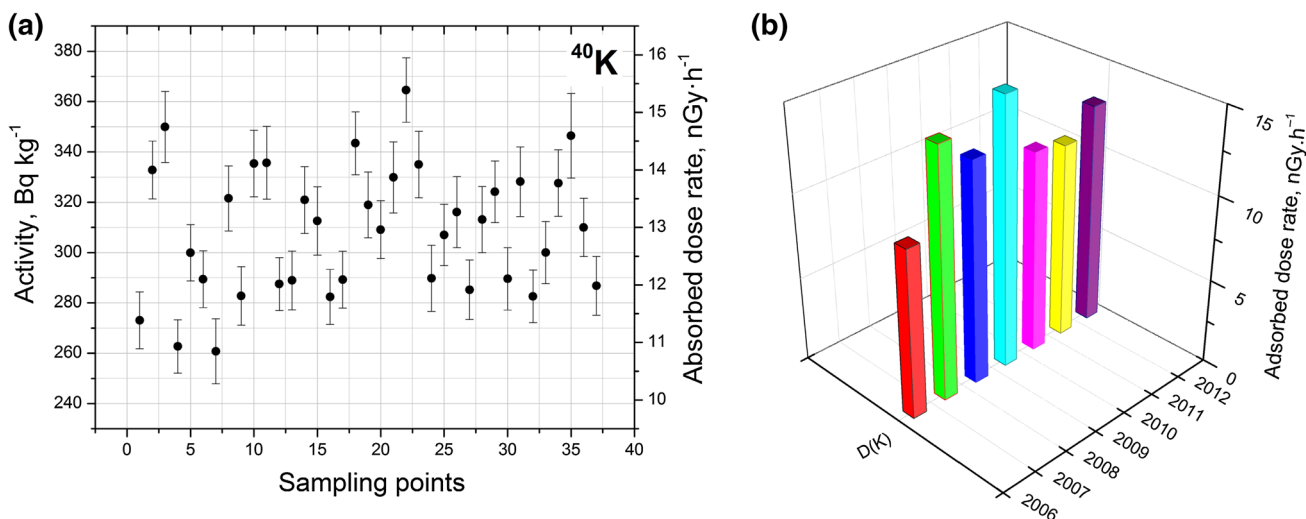


Fig. 3 The trend average values of activity (a) and dose rate data of ^{40}K in 2006–2012 (b)

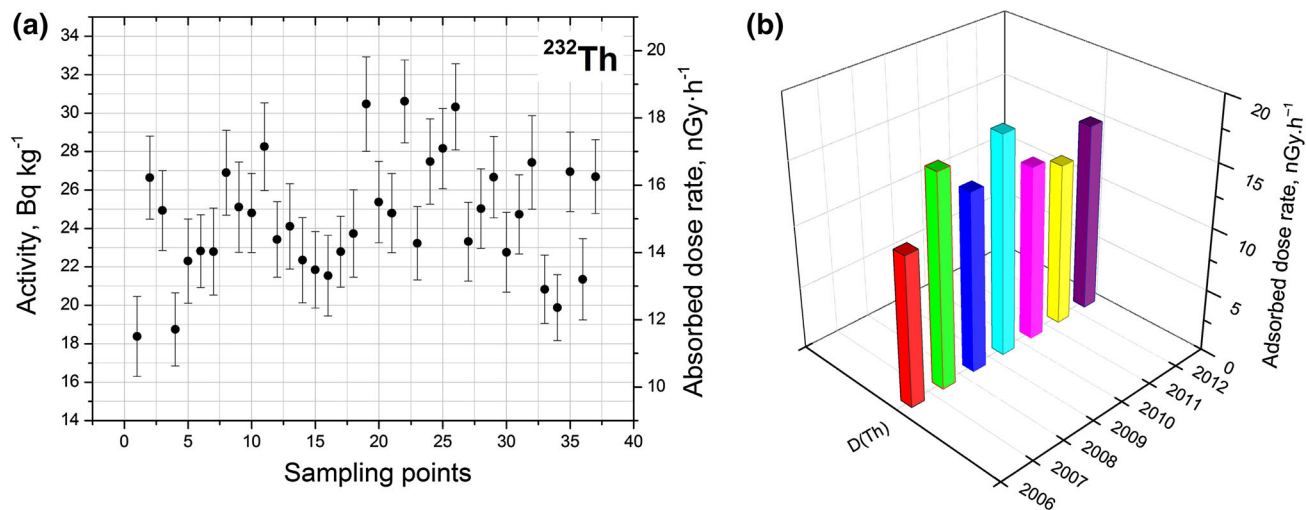


Fig. 4 The trend average values of activity (a) and dose rate data of ^{232}Th in 2006–2012 (b)

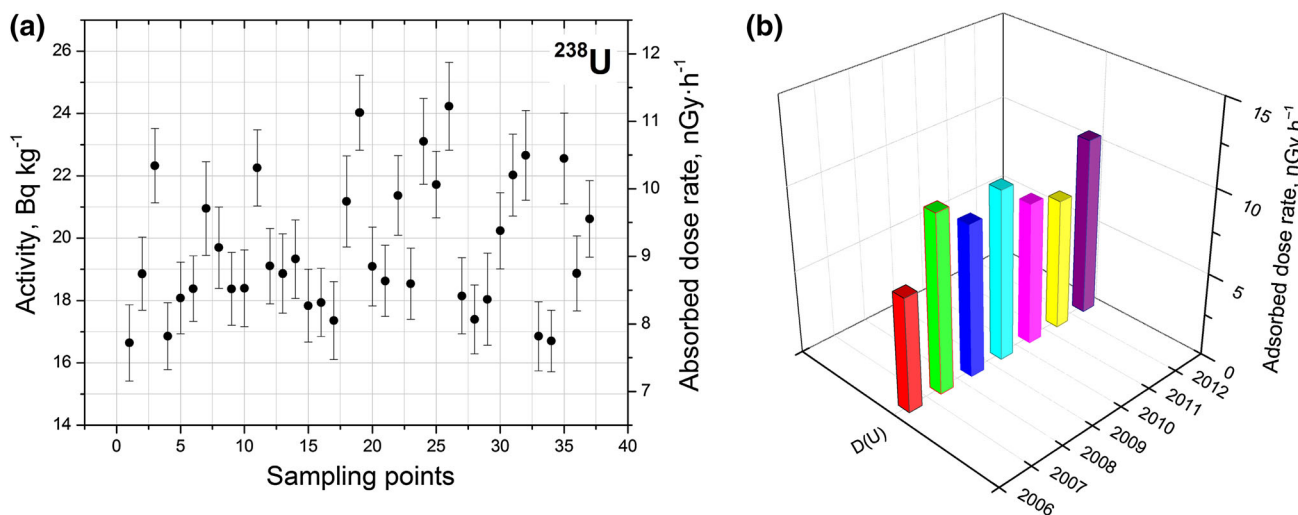


Fig. 5 The trend average values of activity (a) and dose rate data of ^{238}U in 2006–2012 (b)

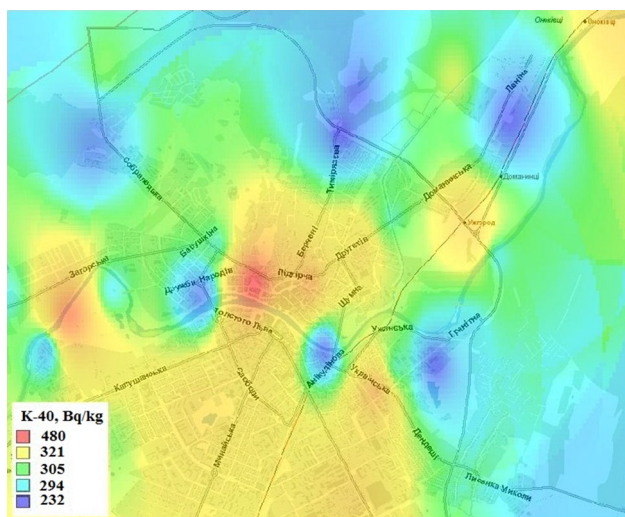


Fig. 6 GIS maps of the specific activity for ^{40}K

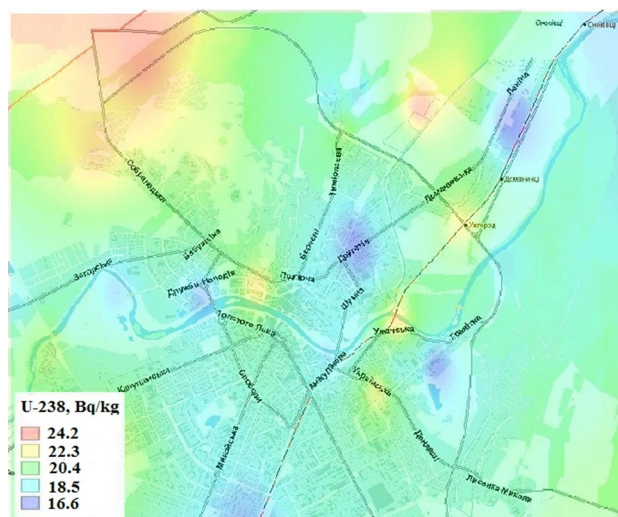


Fig. 8 GIS maps of the specific activity for ^{238}U

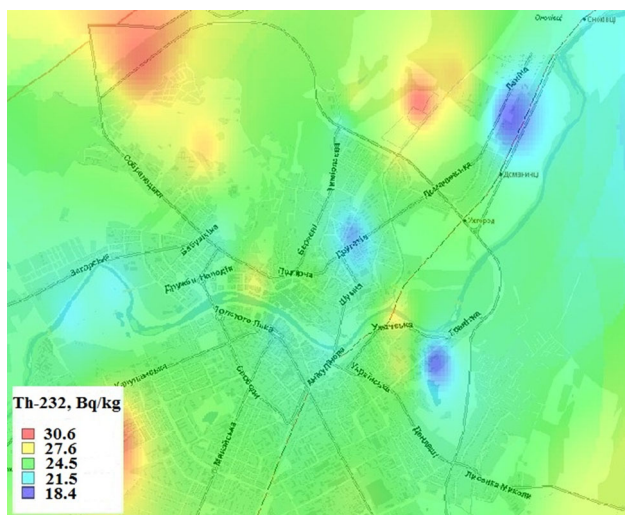


Fig. 7 GIS maps of the specific activity for ^{232}Th

The concentration of the radionuclides from the ^{238}U and ^{232}Th series and ^{40}K in soil of the area under study depends on the geology of the area and the surrounding mountains where the material was eroded and transported by rain water into the basin of the river Uzh. Distribution of ^{40}K in surface soil of Uzhgorod in relation to radionuclides ^{238}U and ^{232}Th series may be explained by the weathering in crystalline rocks of the Carpathians Mountains, potassium is readily dissolved therefore may have been removed from the rocks [13].

Due to the natural processes of soil formation, each type of soil is characterized by the particular distribution of natural radionuclides in the surface layer [14]. Podzolic soil types have lower specific activity of natural radionuclides ($^{40}\text{K} = 300 \text{ Bq kg}^{-1}$, $^{232}\text{Th} = 20 \text{ Bq kg}^{-1}$, $^{238}\text{U} = 18 \text{ Bq kg}^{-1}$) and gray forest soil types have higher specific activity of natural radionuclides ($^{40}\text{K} = 350 \text{ Bq kg}^{-1}$, $^{232}\text{Th} = 27 \text{ Bq kg}^{-1}$, $^{238}\text{U} = 21 \text{ Bq kg}^{-1}$) in Uzhgorod.

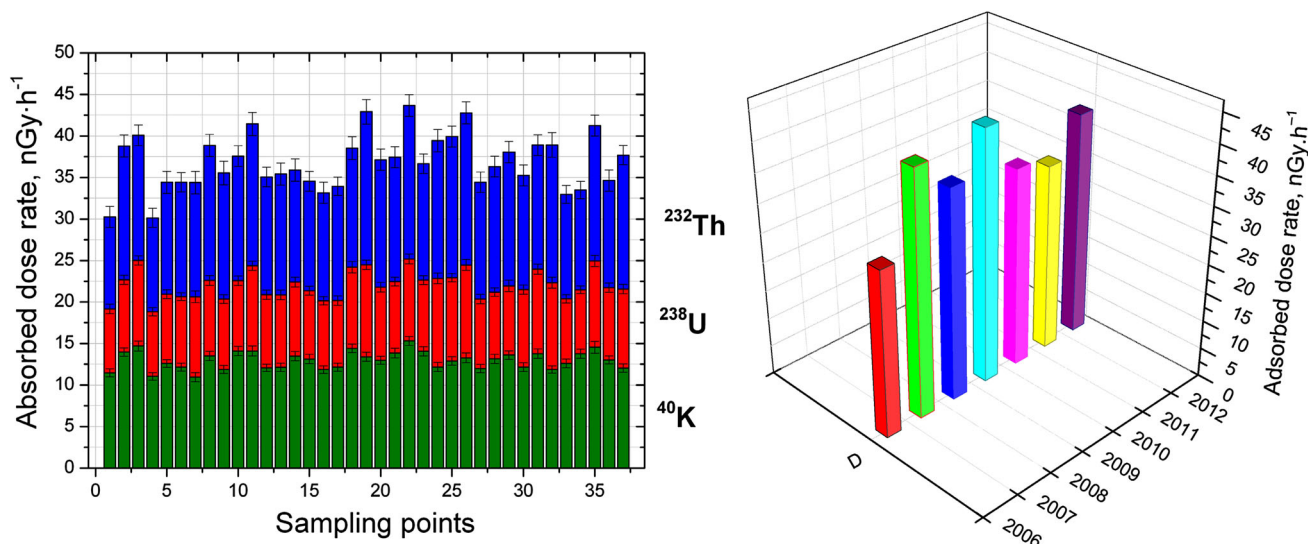


Fig. 9 Relative contribution to the total absorbed dose rate [nGy h⁻¹] of thorium (²³²Th), uranium (²³⁸U) and potassium (⁴⁰K) for all surface soil samples in Uzhgorod city

Table 3 The annual effective dose (10⁻⁵ Sv) of surface soil samples in Uzhgorod

Location	2006	2007	2008	2009	2010	2011	2012
Annual effective dose (10 ⁻⁵ Sv)	3.5 ± 0.2	5.2 ± 0.3	4.5 ± 0.3	5.3 ± 0.3	4.2 ± 0.3	4.0 ± 0.3	4.7 ± 0.3

The average values of the absorbed dose for the natural radionuclides ⁴⁰K, ²³²Th, ²³⁸U are shown in Fig. 9. The average value of the absorbed dose is 36.3 ± 2.3 nGy h⁻¹.

The values of the annual effective dose from natural radionuclides measured in Uzhgorod in 2006–2012 are shown in Table 3. The average annual effective dose of the natural radionuclides is 4.5 ± 0.3 10⁻⁵ Sv.

The average value of the external hazard index of the natural radionuclides for period 2006–2012 was calculated to be 0.22 that testifies to the absence of technogenic (artificial) pollution.

Table 4 The values of the absorbed dose, external hazard index and annual effective dose from natural radionuclides for the near-border countries

Location	D (nGy h ⁻¹)	H _{ex}	S _{ef} (10 ⁻⁵ Sv)	Ref.
Hungary	61	0.27	7.48	[15]
Poland	45	0.24	5.52	[16]
Slovakia	60	0.34	7.31	[1]
Romania	59	0.34	7.24	[17]
Uzhgorod, Ukraine	36.3 ± 2.3	0.22	4.5 ± 0.3	–
World average value	51	<1	6.9	[1]

The values of the absorbed dose, external hazard index and annual effective dose for natural radionuclides for the neighboring countries (Table 4) show that they are higher than those for Uzhgorod.

Conclusions

The results confirm that the average values of ²³⁸U (19.2 ± 1.2 Bq kg⁻¹) and ²³²Th (24.2 ± 2.1 Bq kg⁻¹) are slightly less than the world average values of ²³⁸U (35 Bq kg⁻¹) and ²³²Th (30 Bq kg⁻¹). The present ⁴⁰K average activity (304.0 ± 12.3 Bq kg⁻¹) is lower than the world average value (400 Bq kg⁻¹) [1]. Consequently, these values can be used as a reference for ecological certification and classification of Uzhgorod surface soils.

The present average gamma dose rate in the air is 36.3 ± 2.3 nGy h⁻¹ which does not vary significantly from place to place within the natural background area. The calculated annual effective dose with the average value (4.5 ± 0.3 10⁻⁵ Sv) is lower than the world average value (6.9 10⁻⁵ Sv) [1].

Geostatistical technique was applied to interpolate the values of the specific activity of the terrestrial radionuclide ⁴⁰K and the members of the ²³²Th and ²³⁸U series in the sampling points.

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