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Assessment of Natural Radioactivity and Radiological Hazards in Soil Samples from Una National Park, Bosnia and Herzegovina

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Abstract

This paper evaluates natural environmental radioactivity and assesses potential radiation hazards from primordial radionuclides ^{226}Ra, ^{232}Th and ^{40}K across various surface areas within Una National Park, Bosnia and Herzegovina. This study represents the first systematic radiological survey conducted inside the park boundaries. Using high-resolution gamma-ray spectrometry, the mean specific activity concentrations were determined to be 55 ± 3 Bq kg^{-1} for ^{226}Ra, 42 ± 1 Bq kg^{-1} for ^{232}Th, and 399 ± 18 Bq kg^{-1} for ^{40}K. Based on these baseline empirical data, the ambient absorbed dose rate in air reached an average value of 67.48 nGy h^{-1}, which corresponds to a mean outdoor annual effective dose equivalent of 0.083 mSv y^{-1}. The calculated radium equivalent activity showed a mean value of 145.96 Bq kg^{-1}, while the external and internal hazard indices registered safe averages of 0.39 and 0.54, respectively. Finally, the average excess lifetime cancer risk was found to be $0.29 \cdot 10^{-3}$, aligns perfectly with the current global baseline average. These findings demonstrate that despite minor geologically-driven elevations, the protected area remains radiologically safe for the general public and tourism.

Keywords

Natural radioactivity, Gamma spectrometry, Activity concentrations, Environmental risk assessment, Una National Park

Introduction

Human exposure to background radiation is driven by a combination of cosmic, terrestrial, and internal sources. Within the lithosphere, primordial radionuclides are incorporated into rocks and soil profiles, where they undergo successive radioactive decay series, continuously emitting ionizing radiation into the surrounding environment [1]. The human population is directly exposed to radiation from natural radionuclides as a result of external exposure, or by incorporating these radionuclides into the body by inhalation or ingestion as a result of internal exposure.

The biggest part of the total dose contribution in the form of natural sources comes from terrestrial sources [2]. Natural radionuclides of terrestrial origin are everywhere present in the environment. Just those natural radionuclides with a very large half-lives, and their decay products, exist in considerable quantities. The external radiation exposure is mostly caused by ^{226}Ra (or by ^{238}U), ^{232}Th and ^{40}K in soil and rocks on the Earth´s crust [3].

The most widespread natural radionuclide from soil is ^{40}K, with average content about to 0.003%. This radionuclide has a half-life of $T_{1/2} = 1.28 \cdot 10^9$ years disintegrates by beta decay to stable isotope ^{40}Ar (89%) and electron capture to also stable isotope ^{40}Ca (11%). The primary decay chains are headed by long-lived isotopes, such as ^{232}Th ($T_{1/2} = 1.39 \cdot 10^{10}$ years), which transforms through a sequence of alpha and beta emissions to stable ^{208}Pb. At the same time, the^{238}U series ($T_{1/2} = 4.47 \cdot 10^9$ years), remains a primary contributor to soil radioactivity, leading to the formation of ^{226}Ra before terminating at stable ^{206}Pb [4].

This research aims to determine the activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected from various locations of Una National Park, Bosnia and Herzegovina using gamma ray spectrometry and also absorbed dose rate, annual effective dose equivalent, radium equivalent activity, external and internal hazard indices and average excess lifetime cancer risk are rated. The measurements from this article are compared with the measurements performed in the Republic of Srpska, central Bosnia and North-Eastern Bosnia [5, 6, 7]. The results of this study will compile a basic set of data to assess population exposure and contribute to the mapping of natural radionuclide concentrations in surface soils in Bosnia and Herzegovina [8].

Materials and Methods

Study area

Una National Park is located in the north-western part of Bosnia and Herzegovina, mainly in the area of Bihac municipality and one small part in the municipalities of Bosanski Petrovac and Drvar [9]. Una National Park stretches along the Croatian border and it is approximately 25 kilometres away from Plitvice Lakes National Park and 120 kilometres from the Adriatic Sea (Fig. 1). The Park comprising valley of the river Una, the canyon of the river Unac (right tributary of river Una) to the river Krka in the west and the orographic slopes of the mountains Pljesevica, Grmec and Osjecenica [10].

Geology of the study area

Una National Park features a 20 cm to 2 m thick humus layer overlying deep deposits of limestone, calcarenite, and marly limestone [10]. The underlying geological structure consists of microcrystalline calcite with a chemical composition dominated by calcium carbonate and

significant water content, as detailed in the analysis [10].



Fig. 1 The location of Una National Park

Sample collection and preparation

Between July and December 2025, we collected topsoil samples from 80 different sites across the region, focusing on the upper 10 cm layer. The sampling campaign was distributed as follows: 25 samples from Bihać, 15 from Kulen Vakuf, 10 from Martin Brod, 10 from the Ćelije–Orašac area, 10 from Štrbački Buk, and 10 from Ripač. Every sample was taken strictly following the standard guidelines recommended by the International Atomic Energy Agency (IAEA) [11]. In the laboratory, we cleaned each sample by hand to remove any organic debris,

roots, or small pebbles. The soil was then left to dry completely in an oven at 100 °C. Once dried, the samples were crushed into a fine powder and transferred into tightly sealed plastic containers (with an average sample weight of around 250 g). We stored these sealed containers for at least 30 days before testing. This waiting period is necessary to let ^{226}Ra and ^{232}Th reach a state of radioactive equilibrium with their short-lived decay products.

Measurement of ^{226}Ra, ^{232}Th and, ^{40}K activity concentration

Gamma spectrometric measurements for ^{226}Ra, ^{232}Th and, ^{40}K activity concentration were conducted using the portable GAMMA-RAD 5 spectrometer. This device combines a complete NaI scintillation probe with a digital pulse processor, a shaping amplifier, and a multi-channel analyzer (MCA), all linked to a PC via USB for data processing. The detector functions within an energy range of 10 to 3000 keV, featuring an energy resolution of under 7% FWHM at 662 keV and below 5% FWHM at 1332.5 keV [8].

To calibrate the system, we used standard check sources supplied by Spectrum Techniques LLC (USA), specifically ^{137}Cs (with its peak at 662 keV) and ^{60}Co (with peaks at 1173.2 keV and 1332.5 keV). Ambient background radiation was suppressed by placing the detector inside a customized shielding chamber consisting of a 20 mm thick plastic inner layer and a 10 mm thick outer lead plate jacket. Positioned dead-center within this shield to minimize any interference from scattered radiation, each soil sample was measured for a counting time of 10 hours. Afterward, we processed the acquired spectra offline using the Sodigam software package, which automatically fits the relevant photopeaks. This analytical step allowed us to extract the net photopeak count rates and compute the specific activity per unit mass for each identified radionuclide [8]. From this analysis, photopeak count rates and the specific activity (activity per unit mass) of each detected radionuclide were determined. Specific activity concentrations (A_{Ei}, Bq·kg^{-1}) or each radionuclide i at a distinct photopeak energy E were evaluated using the following relation [1]:

$$A_{E_i} = {N_E}\Big/{\varepsilon_E \cdot I_\gamma \cdot m \cdot t} \quad (1)$$

Where N_E denotes the net photopeak area under the designated energy E, ε_E is the absolute detection efficiency at that specific energy, I_γ represents the fractional gamma-ray emission probability (gamma yield), m is the dry sample mass (kg), and t is the spectrum acquisition time (s).

The specific activity of ^{40}K was directly monitored via its single1461 keV photopeak. For the ^{232}Th decay series, the activity was derived from the weighted mean of the progeny photopeaks: ^{212}Pb (239 keV), and ^{228}Ac (911 keV and 969 keV). Similarly, ^{226}Ra activity was quantified based on the prominent gamma lines of its short-lived daughters: ^{214}Pb (351.9 keV) and ^{214}Bi (1120 keV and 1764 keV).

Absorbed dose rate calculation

To evaluate the environmental radiation risk, the raw activity concentrations were converted into ambient absorbed gamma dose rates in air (D, nGy h^{-1}) at a height of 1 m above the ground surface. Assuming a state of secular equilibrium within the terrestrial decay series (^{238}U up to ^{210}Pb and ^{232}Th up to ^{208}Pb), the conversion coefficients of 0.462, 0.604, and 0.0417 nGy h^{-1} per Bq·kg^{-1} were adopted for ^{226}Ra, ^{232}Th, and ^{40}K, respectively [1, 14]. The total absorbed dose rate was calculated using Eq. (2):

$$D(\text{nGy h}^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (2)$$

criteria where A_{Ra}, A_{Th}, and A_K represent the respective specific activities (Bq·kg^{-1}) of the radionuclides. Minor radiological contributions from anthropogenic or trace primordial nuclides (such as ^{137}Cs and ^{235}U) were considered negligible. Given that approximately 98% of the total gamma dose from the ^{238}U chain originates from the ^{226}Ra sub-series, potential disequilibrium between uranium and radium does not significantly bias the final dose estimations [15].

Estimate the annual effective dose equivalent

The long-term biological impact of radiation exposure on the local population was evaluated by calculating the outdoor annual effective dose equivalent (E_{out}, mSv y^{-1}). This calculation accounts for a standard adult conversion factor of 0.7 Sv·Gy^{-1} (translating the absorbed dose in air to human effective dose) and an outdoor occupancy factor of 0.2, assuming that individuals spend approximately 20% of their time outdoors [2]. The annual effective dose was computed via Eq. (3) [16, 17]:

$$E_{out}(\text{mSv y}^{-1}) = D(\text{nGy h}^{-1}) \cdot 8760\ (\text{h y}^{-1}) \cdot 0.2 \cdot 0.7(\text{Sv Gy}^{-1}) \cdot 10^{-6} \quad (3)$$

where 8760 represents the total duration of a calendar year in hours.

Radium equivalent activity

Since the distribution of ^{226}Ra, ^{232}Th, and ^{40}K in the soil profiles of Una National Park is inherently non-uniform, the radium equivalent activity (Ra_{eq}, Bq kg^{-1}) was utilized as a unified index to assess the collective external and internal radiological hazard. This index mathematically normalizes the specific activity of the mixture, operating under the criterion that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K induce an identical gamma dose output [18]. The index was formulated according to Eq. (4) [19]:

$$Ra_{eq}(\text{Bq kg}^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (4)$$

Here, A_{Ra}, A_{Th}, and A_K are the respective specific activities of ^{226}Ra, ^{232}Th, and ^{40}K. While defining Ra_{eq} according to Eq. (4). The coefficients are based on the assumption that 370 Bq kg^{-1} ^{226}Ra or 259 Bq kg^{-1} ^{232}Th or 4810 Bq kg^{-1} ^{40}K yield the same gamma dose rate.

Radiological hazard indices

To quantify the potential radiation risks associated with the soil samples and ensure public safety, both external (H_{ex}) and internal (H_{in}) hazard indices were evaluated. These dimensionless indices serve as a tool to determine whether the radiation exposure from the terrestrial radionuclides stays below the internationally accepted public safety limit of 1 mSv·y^{-1} [20].

The external hazard index (H_{ex}) assesses the risk level stemming from direct exposure to outdoor gamma radiation emitted by the local soil. It is mathematically formulated using Eq. (5) [21]:

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

Concurrently, the internal hazard index (H_{in}) is utilized to account for the additional respiratory risk caused by the inhalation of radon gas ^{222}Rn and its short-lived alpha-emitting daughters. This index is particularly relevant for evaluating health risks in spaces where radon can accumulate, and it is calculated according to Eq. (6) [22]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (6)$$

In both relations (5) and (6), A_{Ra}, A_{Th}, and A_K represent the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, measured in the samples. For a safe environment, both calculated indices must yield values lower than unity ($H \leq 1$).

Excess lifetime cancer risk

The Excess Lifetime Cancer Risk (*ELCR*) is a critical parameter used in environmental dosimetry to project the probability of an individual developing cancer over their lifespan due to chronic radiation exposure. This estimation directly builds upon the calculated outdoor annual effective dose equivalent, integrating both average life expectancy and a standardized radiological risk coefficient.

To evaluate the likelihood of fatal cancer induction from the received dose, the International Commission on Radiological Protection (ICRP) recommends a risk factor (*RF*) 0.05 Sv^{-1} for the general population. The ELCR is mathematically derived using Eq. (7) [22, 23]:

$$ELCR = E_{out} \times DL \times RF \quad (7)$$

In this formula E_{out} represents the annual effective dose equivalent ($mSv \cdot y^{-1}$), while (*DL*) denotes the average duration of human life, which is standardly assumed to be 70 years in these models. The resulting value is dimensionless and provides a baseline for determining whether the cumulative exposure in the study area falls within globally acceptable radiological safety margins. [24].

Results and discussion

Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K

The specific activity concentrations of the examined primordial radionuclides of ^{226}Ra, ^{232}Th and ^{40}K across different zones of the Una National Park are summarized in Table 1. Looking at the individual nuclides, the activity concentrations for ^{226}Ra fluctuated between 44 to 65 Bq kg^{-1}, yields an overall average value of 55 Bq kg^{-1}. The spatial distribution shows that the lowest activity (44 Bq kg^{-1}) was recorded in the soil beds of Martin Brod, whereas the highest value (65 Bq kg^{-1}) was found in the Ćelije–Orašac sector.

Table 1 Activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples in Una National Park, Bosnia and Herzegovina

Sample location	Number of samples	Average activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Bihac	25	54 ± 3	41 ± 1	367 ± 17
Martin Brod	10	44 ± 2	40 ± 1	376 ± 17
Kulen-Vakuf	15	54 ± 3	43 ± 1	420 ± 19
Celije-Orasac	10	65 ± 3	43 ± 1	391 ± 18
Strbacki Buk	10	55 ± 3	43 ± 1	416 ± 19
Ripac	10	56 ± 3	44 ± 1	424 ± 19
Average		55 ± 3	42 ± 1	399 ± 18
Median		55 ± 3	43 ± 1	403 ± 18
Minimum		44 ± 2	40 ± 1	367 ± 17
Maximum		65 ± 3	44 ± 1	424 ± 19
Standard deviation		6.7	1.5	24.4
Geometric mean		54.3	42.3	398.4
Frequency distribution		Normal	Normal	Normal

For^{232}Th, the measured values were much more uniform, narrow-ranging from 40 to 44 Bq kg^{-1} with a calculated mean of 42 Bq kg^{-1}. The baseline minimum (40 Bq kg^{-1}) was again detected at Martin Brod, while the maximum concentration (44 Bq kg^{-1}) appeared in the Ripač samples. Regarding ^{40}K, the concentrations showed wider variations, spreading from a minimum of 367 Bq kg^{-1} in Bihać to a peak of 424 Bq kg^{-1} in Ripač, maintaining a general average of 399 Bq kg^{-1}. Evaluating the inter-radionuclide dynamics, the mean activity ratio ^{226}Ra to ^{232}Th was found to be 1.31, confirming a clear dominance of radium over thorium in the park's topsoil. Additionally, the calculated specific activity ratios for ^{226}Ra/^{40}K and ^{232}Th/^{40}K were found to be low, marking mean values of 0.13 and 0.10, respectively.

To better understand these values, we compared our results with international data and baseline levels from neighboring regions, as summarized in Table 2. The average ^{226}Ra concentration obtained in this study (55 Bq kg^{-1}) noticeably exceeds both the worldwide average (35 Bq kg^{-1}) and the European benchmark (41 Bq kg^{-1}), while also sitting higher than the averages reported for adjacent countries. Similarly, the mean ^{232}Th concentration (42 Bq kg^{-1}) stands above the global (30 Bq kg^{-1}) and continental (31 Bq kg^{-1}) marks, although it matches well with the values documented in the Republic of Srpska. Conversely, the ^{40}K radioactivity level (399 Bq kg^{-1}) is almost identical to the global reference baseline (400 Bq kg^{-1}), slightly lower than the European average (465 Bq kg^{-1}), and shows a strong similarity to data from Bulgaria. These elevated concentrations of ^{226}Ra and ^{232}Th in the soil profiles of Una National Park are closely tied to the regional geology. The area is dominated by weathered carbonate rocks and specific limestone matrices, which naturally tend to accumulate trace amounts of primordial uranium and thorium series [25].

Table 2 Natural radionuclide (^{226}Ra, ^{232}Th and ^{40}K), content of soil in some countries [2] in comparison with their content in Una National Park, Bosnia and Herzegovina (Bq kg^{-1})

Location	^{226}Ra	^{232}Th	^{40}K
Una National Park	55	42	399
Central Bosnia	66	40	470
NE Bosnia	41	32	331
R. of Srpska	44	41	649
Croatia	43	37	423
Montenegro	29	24	246
Slovenia	41	35	374
Bulgaria	46	30	400
Hungary	33	28	370
Slovenia	41	35	374
Europe average	41	31	465
World average	35	30	400

Absorbed dose rates and outdoor annual effective dose equivalent

The calculated values for the ambient absorbed dose rates alongside the corresponding outdoor annual effective dose equivalents are detailed in Table 3. The total absorbed gamma dose rate, which combines the radiation outputs of ^{226}Ra, ^{232}Th, and ^{40}K in the monitored soil strata, varied between 60.17 nGy h^{-1} to 72.31 nGy h^{-1}. This results in an overall average of 67.48 nGy h^{-1}, a value that noticeably exceeds the global terrestrial benchmark 57 nGy h^{-1} documented by UNSCEAR. On the other hand, the outdoor annual effective dose equivalent calculated for the local population shifted within a narrow margin from 0.074 to 0.088 mSv y^{-1}, with an average value of 0.083 mSv y^{-1}. These values are well below the safety limit of 1 mSv y^{-1}, as established by UNSCEAR safety reports [2].

Table 3 Absorbed dose rate and outdoor annual effective dose equivalent of soil samples in Una National Park, Bosnia and Herzegovina

Sample location	D (nGy h^{-1})	E_{out} (mSv y^{-1})
Bihac	65.10	0.080
Martin Brod	60.17	0.074
Kulen-Vakuf	68.43	0.084
Celije-Orasac	72.31	0.088
Strbacki Buk	68.73	0.084
Ripac	70.13	0.086
Average	67.48	0.083
Median	68.58	0.084

Minimum	60.17	0.074
Maximum	72.31	0.088
Standard deviation	4.23	0.005
Geometric mean	67.36	0.082
Frequency distribution	normal	normal

Hazard indices

The estimated radiation hazard indices, specifically the radium equivalent activity, the external and internal hazard parameters, and the projected excess lifetime cancer risk are collectively summarized in Table 4.

Table 4 Hazard indices of soil from Una National Park, Bosnia and Herzegovina

Sample location	Ra_{eq} (Bq kg^{-1})	H_{ex}	H_{in}	$ELCR$ ($\times 10^{-3}$)
Bihac	141.12	0.38	0.53	0.28
Martin Brod	130.15	0.35	0.47	0.26
Kulen-Vakuf	147.83	0.40	0.54	0.29
Celije-Orasac	156.56	0.42	0.60	0.31
Strbacki Buk	148.52	0.40	0.55	0.29
Ripac	151.57	0.41	0.56	0.30
Average	145.96	0.39	0.54	0.29
Median	148.12	0.40	0.55	0.29
Minimum	130.15	0.35	0.47	0.26
Maximum	156.56	0.42	0.56	0.31
Standard deviation	9.24	0.02	0.04	0.02
Geometric mean	145.71	0.39	0.54	0.29
Frequency distribution	normal	normal	normal	normal

Similarly, the external hazard index fluctuated between 0.35 and 0.42, with a regional average of 0.39. The internal hazard index followed a comparable pattern, ranging from 0.47 up to a maximum of 0.60, yielding an overall mean of 0.54. Because both indices stay well below the critical threshold value of 1, the outdoor environment does not present any elevated threat from alpha or gamma exposure. Lastly, the calculated excess lifetime cancer risk (*ELCR*) fell within the range of 0.26×10^{-3} to 0.31×10^{-3}, maintaining a steady average of 0.29×10^{-3}. This final metric is highly consistent with the typical baseline values and standard world averages recommended in the UNSCEAR guidelines [2].

Conclusions

This study establishes the first comprehensive baseline database for natural environmental radioactivity and population exposure across the territory of Una National Park. Notably, this research represents a pioneering radiological assessment within the national parks of Southeast Europe, making its findings highly valuable for both the local community and regional conservation strategies. The spatial monitoring reveals that while the average activity concentrations of ^{226}Ra and ^{232}TH exceed global and European baseline averages due to the local carbonate geology, the cumulative radiation risks remain well within safe environmental limits. The ambient absorbed gamma dose rates stand slightly above the worldwide reference level, yet the resulting annual effective dose equivalent for the population is comfortably below international safety margins. Furthermore, all calculated radiological hazard parameters—including radium equivalent activity, external and internal indices, and the excess lifetime cancer risk—confirm that the natural radiation levels within Una National Park are completely normal.In conclusion, the outdoor environment of the park poses no radiotoxicity or health hazards to either seasonal visitors or the resident population. These results can comfortably serve as a reliable reference point for future environmental mapping and similar radiological surveys across protected areas in the Balkan region.

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