BioRisk

BioRisk 24: 103–117 (2026)
DOI: 10.3897/biorisk.24.178091

Research Article

Investigation of the radioactivity of sands collected from Bulgarian Black Sea coast

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Abstract

Radionuclide concentrations in coastal sand samples from the Bulgarian Black Sea coast were analyzed using gamma-spectrometry. The average activity concentrations for ^{238}U, ^{232}Th, and ^{40}K were found to be 11.62, 6.82 and 396.55 Bq/kg, respectively, all of which are lower than global averages. Additionally, the technogenic radionuclide ^{137}Cs was detected in 11 samples, with an average concentration of 3.16 Bq/kg. Radiation hazard coefficients were calculated, and their average values were also lower than the global benchmarks. To explore the relationships between radionuclides and radiological variables, multivariate statistical methods such as Pearson correlation, principal component analysis, and cluster analysis were used. These findings contribute to a more comprehensive understanding of the radiological condition of the environment along Bulgarian beaches.

Key words: Beach sediments, Bulgaria Black See, gamma ray spectrometry, multivariate statistical analysis, radiological hazards

Introduction

The Bulgarian Black Sea coastline has a total length of approximately 354 km, 200 km of which about are composed of sandy beach areas. The coastline begins at cape Kartal, located at the Romanian border, and extends to the mouth of the Rezovska River at the Turkish border. These beach areas are primarily composed of sand deposits formed by the weathering and erosion of igneous or metamorphic rocks in the surrounding environment, with natural radionuclides often present in these sediments. Studies focusing on the concentrations and distribution of radionuclides in sands provide valuable insights into the presence of various radioactive elements in the region. Natural radionuclides contribute to both external radiation exposure when people are outdoors and internal exposure through inhalation or ingestion (Örgün et al. 2007; Alshahri 2017). The health risk associated with radiation exposure depends on the radiation dose received by individuals (Hu et al. 2010). People are constantly exposed to natural background radiation, originating from radionuclides in the Earth's crust (soils, rocks), water, air, food, and cosmic radiation. Approximately 80% of the total radiation dose a person receives annually originates from terrestrial radionuclides present in soils and rock formations (IAEA 1996). Among the most widespread natural radionuclides in the environment, which

OPEN ACCESS

Academic editor: Josef Settele
Received: 5 December 2025
Accepted: 28 March 2026
Published: 30 April 2026

Citation: Arhangelova N, Salim S (2026) Investigation of the radioactivity of sands collected from Bulgarian Black Sea coast. BioRisk 24: 103–117. https://doi.org/10.3897/biorisk.24.178091

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are abundant in the Earth's crust include ^{40}K, as well as radionuclides from the ^{238}U and ^{232}Th decay series (UNSCEAR 1993).

Therefore, soil, water, vegetation, sands, and building materials of natural origin become major sources of both external and internal radiation exposure to humans. Given this, studies related to dose rate assessment and radiological indices are crucial, as they provide valuable information regarding the lifetime cancer risks.

The Black Sea basin is considered one of the most polluted marine environments in terms of artificial radioactivity (Buesseler and Livingston 1996; Egorov et al. 1999; Polikarpov et al. 2008; Thébault et al. 2008; Egorov et al. 2010; Dovlete et al. 2021). The radiological contamination of the Black Sea, has been investigated within several international and national projects initiated by the countries bordering the Black Sea. Various regulatory documents define the acceptable levels of contamination for Black Sea ecosystems (Ordinance No. 8 2001; Nuclear Regulatory Agency 2002; Ordinance No. H-4 2012).

In this study, the quantitative contents of radionuclides found in sand samples collected from beaches along the Bulgarian Black Sea coast were investigated. The data obtained for the specific activities of the individual radionuclides were used for calculating radiation hazard indices, creating GIS maps to visualize spatial distributions, and performing statistical analyses.

Materials and methods

Semple collection and preparation

Sand samples were collected from 33 locations distributed along the entire Black Sea coastline, from Durankulak to Rezovo. The GPS coordinates and altitudes of the sampling points are presented in Fig. 1.

The collected samples were cleaned and dried in a laboratory dryer at a temperature of up to 60 °C. Approximately 0.10 kg of each sample were placed in cylindrical containers suitable for gamma spectrometric analysis.

Gamma ray spectrometric analysis

The analysis of the samples was conducted using a gamma spectrometric system consisting of a semiconductor detector with the following parameters: crystal volume of 60 cm^3; operating voltage of 1 kV; relative efficiency of 4.5% for the ^{137}Cs line (E = 661.66 keV). To reduce the influence from background radiation in the measurements, a low-background shielding was applied. Several layers of lead, aluminum, cadmium, and polyethylene of different thickness were used (Zhechev A 1996). The processing of the obtained spectra was performed using the ANGES program.

The results obtained from the gamma spectrometry were used in the identification of radionuclides in the sand samples and determination of their specific activities. The activities of the detected radionuclides were calculated using the following relation:

$$A = \frac{S}{\varepsilon I t_L} \quad (1)$$

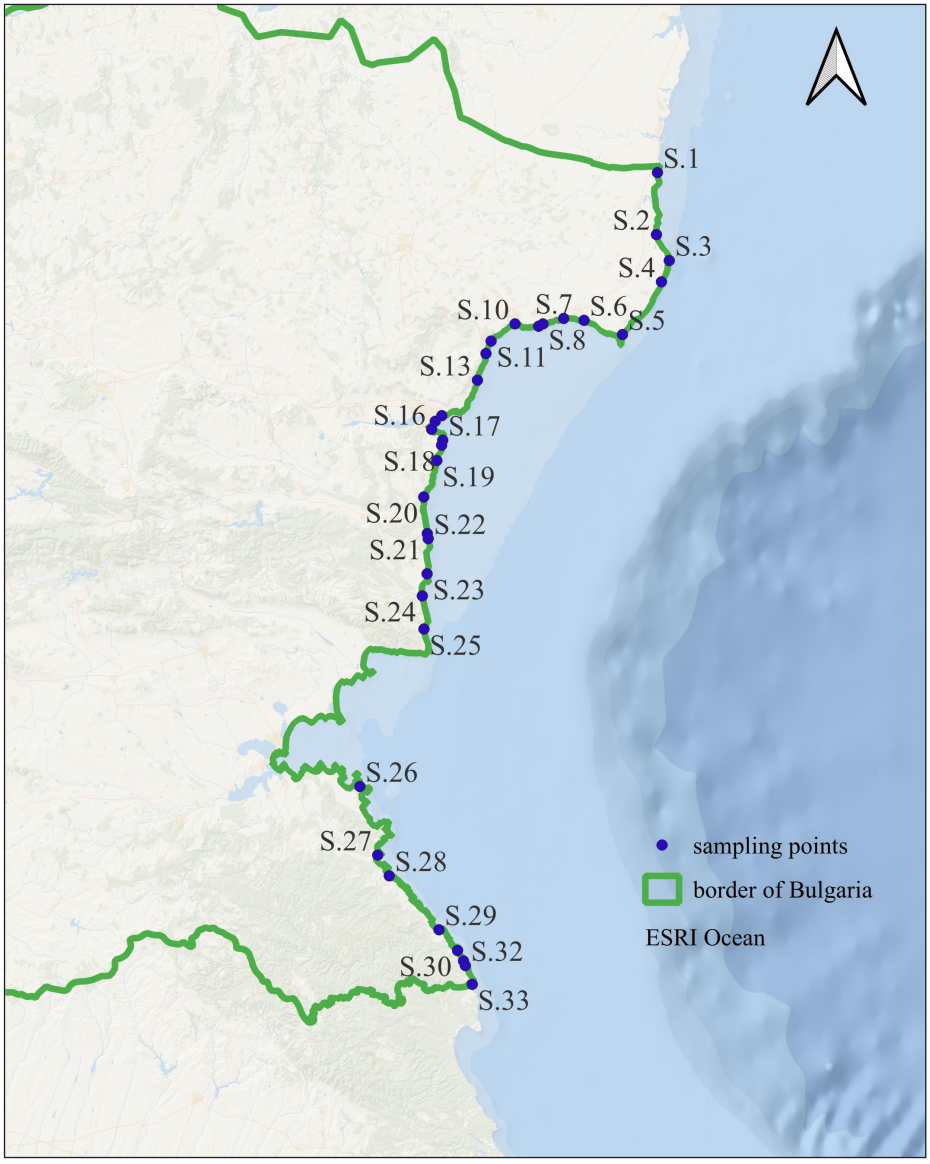

Figure 1. Map of sampling points. Map data ESRI Ocean.

where A is the activity of the radionuclide in Bq; S is the net peak area; I is the intensity; ε is the detector efficiency, and t_L is the acquisition live time.

The radiation safety assessments for the population and the evaluation of the radioecological situation are associated with several well-established radiation hazard indices, including the Radium Equivalent Activity (Ra_{eq}), Gamma Index (I_γ), External (H_{ex}) and Internal (H_{in}) Hazard Indexes, Outdoor absorbed dose rate (D_{out}), Annual outdoor individual effective dose (E_{out}), Excess Lifetime Cancer Risk (ELCR), Absorbed Dose Rate in Air (ADRA) and Annual Effective Dose Rate (AEDR). Data for some of these indices can be found in reports from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and publications from the 1970s. In recent years, there has been an increasing use of indices associated with the quantity of radionuclides entering the human body, due to both outdoor and indoor exposure. According to UNSCEAR reports, the radioactivity from natural sources in the environment is estimated to be 2.4 mSv/y, while the contribution from anthropogenic sources is assessed to be 0.8 mSv/y (UNSCEAR 2000; Dhawal et al. 2013).

Radiation hazard indices

Radium Equivalent Activity – Ra_{eq}

Ra_{eq} can be determined using the formula:

$$Ra_{eq} = \left(\frac{1}{370} A_{sp}\left(^{238}U\right) + \frac{1}{259} A_{sp}\left(^{232}Th\right) + \frac{1}{4810} A_{sp}\left(^{40}K\right) \right) .370 \quad (2)$$

where A_{sp}(^{238}U), A_{sp}(^{232}Th), A_{sp}(^{40}K), are the specific activities in Bq/kg for ^{238}U, ^{232}Th and ^{40}K, respectively. It is assumed that the same gamma dose rate in the sample corresponds to 370 Bq/kg for ^{238}U, 259 Bq/kg for ^{232}Th, and 4810 Bq/kg for ^{40}K (Beretka and Mathew 1985).

Gamma Index (radiation hazard index) – I_γ

The I_γ index is calculated using the specific activities of ^{40}K, ^{232}Th and ^{238}U divided by corresponding coefficients for ^{40}K, ^{232}Th and ^{238}U. The resulting value, according to the world average cited in UNSCEAR (2000) should be less than one (NEA-OECD 1979; Beretka and Mathew 1985).

$$I_\gamma = \frac{1}{150} A_{sp}\left(^{238}U\right) + \frac{1}{100} A_{sp}\left(^{232}Th\right) + \frac{1}{1500} A_{sp}\left(^{40}K\right) \quad (3)$$

External and Internal Hazard Indexes – H_{ex} and H_{in}

These indices are calculated using formulas (4) and (5) (UNSCEAR 1988), assuming that the decay rate of ^{226}Ra for external exposure is taken as 370 Bq/kg and for internal exposure as 185 Bq/kg (Beretka and Mathew 1985).

$$H_{ex} = \frac{1}{370} A_{sp}\left(^{238}\mathrm{U}\right) + \frac{1}{259} A_{sp}\left(^{232}Th\right) + \frac{1}{4810} A_{sp}\left(^{40}K\right) \leq 1 \quad (4)$$

$$H_{in} = \frac{1}{185} A_{sp}\left(^{238}U\right) + \frac{1}{259} A_{sp}\left(^{232}Th\right) + \frac{1}{4810} A_{sp}\left(^{40}K\right) \leq 1 \quad (5)$$

Outdoor absorbed dose rate – D_{out}

As an index of radiological hazard, the outdoor absorbed dose rate is also calculated. Due to the relatively uniform distribution of ^{238}U, ^{232}Th, and ^{40}K at a height of 1 meter above the ground surface, the outdoor absorbed dose rate can be estimated using (NEA-OECD 1979):

$$D_{out} = 0.436 A_{sp}\left(^{238}U\right) + 0.599 A_{sp}\left(^{232}Th\right) + 0.0417 A_{sp}\left(^{40}K\right) \quad (6)$$

Annual outdoor individual effective dose – E_{out}

The assessment of the annual individual effective dose received outdoors can be made by considering the conversion coefficients from the absorbed dose rate in air to the effective dose, as well as the outdoor occupancy factor. According to UNSCEAR reports, a conversion coefficient of 0.7 Sv/Gy is used for the absorbed dose in air to the effective dose received by adults (UNSCEAR 1993; UNSCEAR

2000). It is assumed that individuals spend approximately 20% of their time outdoors, thus the outdoor occupancy factor is 0.2. Therefore, the annual individual effective dose E_{out} (mSv/y) outdoors can be calculated using formula (7).

$$E_{out}\left[\frac{mSv}{y}\right] = D_{out}\left[\frac{nGy}{h}\right] + 8760[h] \times 0.2 \times 0.7\left[\frac{Sv}{Gy}\right] = D_{out} \times 1.226[\mu Sv] \quad (7)$$

Excess Lifetime Cancer Risk – ELCR

The risk of lifetime cancer can be determined as the probability of developing cancer due to radiation exposure levels. This factor can be calculated by considering the values obtained for the mean annual effective dose, average lifespan (DL), and the risk factor (RF), which, according to the ICRP (International Commission on Radiological Protection) (1991), is 0.051 Sv for stochastic effects in the population. The average lifespan for Bulgaria, according to data from the National Statistical Institute, is 73 years.

$$ELCR_{out} = E_{out} D_L R_F \quad (8)$$

Absorbed Dose Rate in Air – ADRA

For the calculation of ADRA, the specific activities of ^{40}K, ^{232}Th, and ^{238}U are used along with conversion factors (9). The cited average global value for ADRA in publications by UNSCEAR 2000 is 84 nGy/h (UNSCEAR 2000).

$$ADRA = 0.461 A_{sp}\left(^{238}U\right) + 0.623 A_{sp}\left(^{232}Th\right) + 0.0417 A_{sp}\left(^{40}K\right) \quad (9)$$

Annual Effective Dose Rate – AEDR

The annual effective dose in nGy/h can be converted into AEDR in mSv/y using the following relationship:

$$AEDR\left[\frac{mSv}{y}\right] = T \cdot Q \cdot ADRA \cdot 10^{-6} \quad (10)$$

where T is the time during a year (8760 hours), Q is the conversion factor of 0.7 Sv/Gy, which converts the absorbed dose rate in the air to human effective dose received. The world mean value for ADRA cited in UNSCEAR (2000) publications is 0.48 mSv/y.

Statistical analysis

Geostatistics is used in the analysis and forecasting of values related to spatial or spatio-temporal phenomena and processes. The statistical theory allows spatial correlations to be used to interpolate values at locations where Statistical theory enables the utilization of existing spatial relationships for the interpolation of values at locations where field sampling is not possible. This has led to an increasing popularity and applicability of geostatistical analysis in environmental sciences, particularly in assessing the levels of various pollutants that pose a threat to the environment or human health.

Result and discussion

After performing the gamma spectrometric analysis on the sand samples, radionuclides from the Uranium-radium and Thorium families, as well as the naturally occurring radionuclide ^{40}K, were detected. The analyses were based on gamma lines of daughter radionuclides such as ^{214}Pb, ^{214}Bi, ^{228}Ac, and ^{208}Tl, which are in secular equilibrium with their respective parent isotopes. The analysis of ^{40}K was conducted using its gamma line with an energy of 1460.80 keV and a quantum yield of 10.67%. Additionally, the anthropogenic radionuclide ^{137}Cs was detected in some of the samples. The specific activity results for ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs are presented in Figs 2, 3, 4, 5.

The specific activities of ^{238}U range from 6 to 28 Bq/kg, with the lowest value of 5.80 ± 0.87 Bq/kg observed in sample S33 (Rezovo Beach), and the highest value of 28.30 ± 4.25 Bq/kg recorded at the Irakli Beach (S25). According to the UNSCEAR (2000) report, the world average for ^{238}U is 35 Bq/kg (Fig. 2).

The measured specific activities for ^{232}Th range from 3 to 12 Bq/kg, with the exception of S25, where the calculated specific activity is 19.06 ± 2.86 Bq/kg. According to UNSCEAR (2000), the world average for ^{232}Th is 30 Bq/kg (Fig. 3).

The values obtained for the specific activities of ^{40}K in the sand samples vary from 200 to 700 Bq/kg. The highest value was recorded for sample S14, collected from Rapongi Beach in Varna, while the lowest was found in the village of Tyulenovo (S4). According to UNSCEAR (2000), the average world concentration of ^{40}K in beach sands from regions with normal radioactivity is 370 Bq/kg (Fig. 4).

The anthropogenic radionuclide ^{137}Cs was detected in eleven of the samples, with concentrations ranging from 1.4 to 3.4 Bq/kg. The highest value was recorded for the sample from Kavarna Beach (S6). Concentrations of ^{137}Cs in sands (dry weight) in other regions of the Bulgarian Black Sea coast in recent years have varied between 0.8 and 12.3 Bq/kg (Fig. 5) (IAEA 2004a, 2004b).

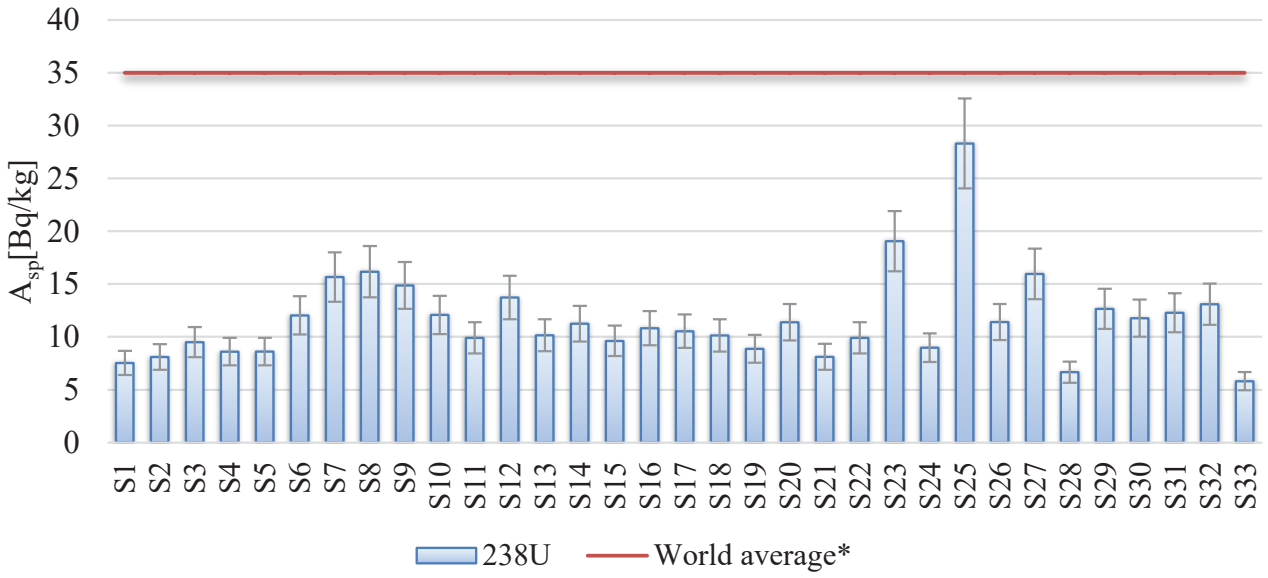

Figure 2. Graphical representation of the specific activities of ^{238}U in sand samples. World average* according UNSCEAR (2000).

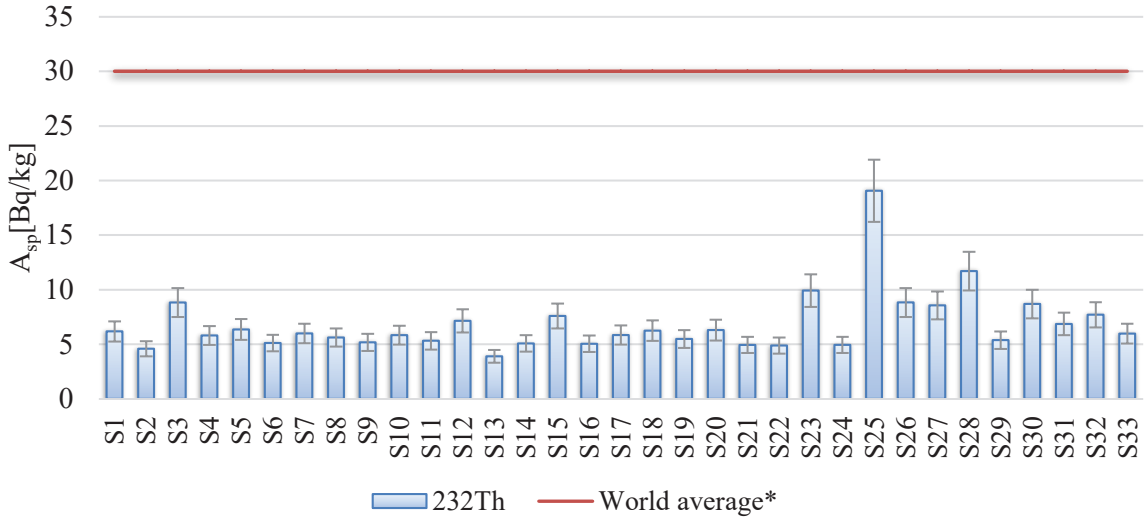

Figure 3. Graphical representation of the specific activities of ^{232}Th in sand samples. World average* according UNSCEAR (2000).

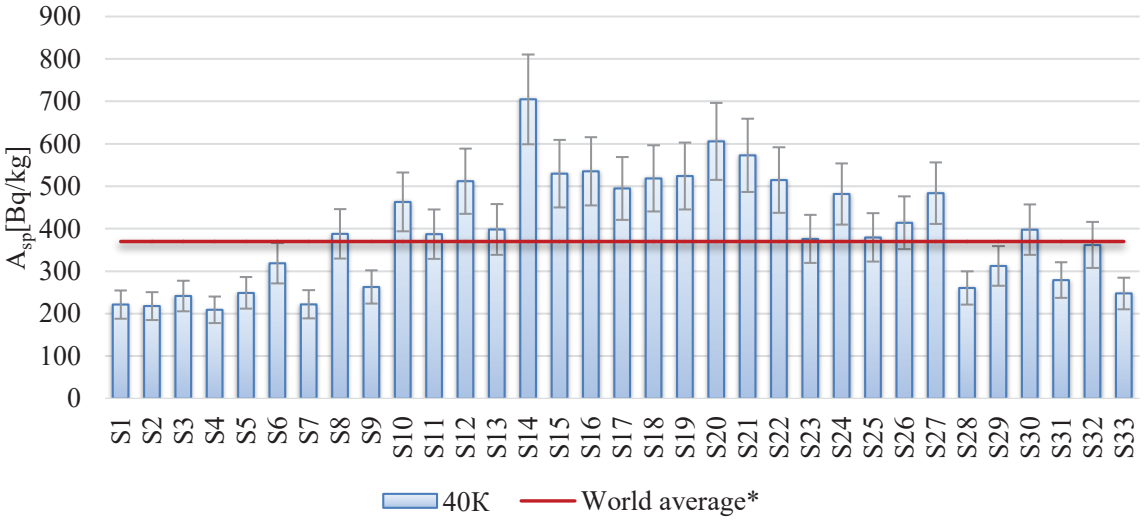

Figure 4. Graphical representation of ^{40}K specific activities in sand samples. World average* according UNSCEAR (2000).

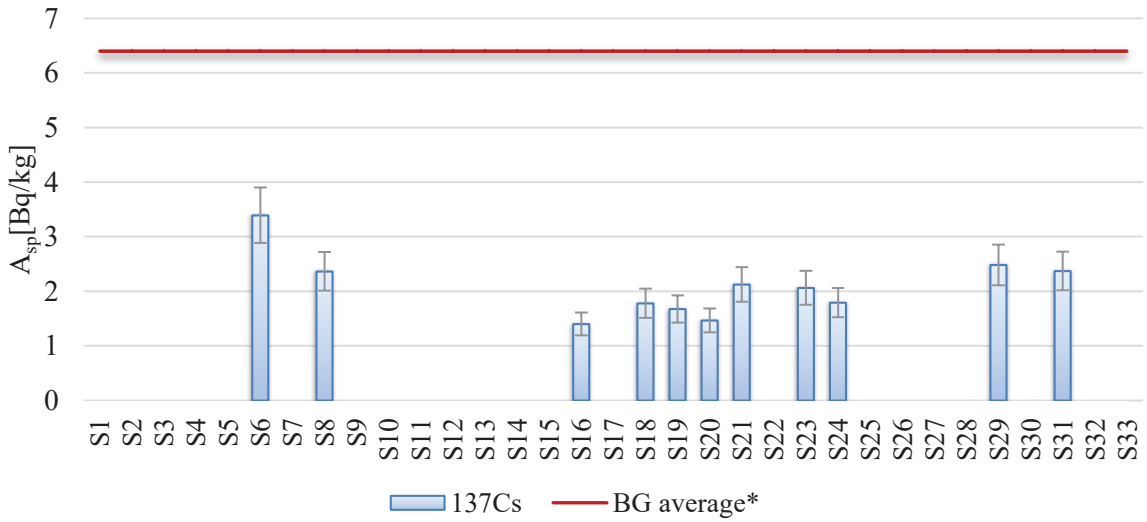

Figure 5. Graphical representation of ^{137}Cs specific activities in sand samples. * according to IAEA (2004a, 2004b).

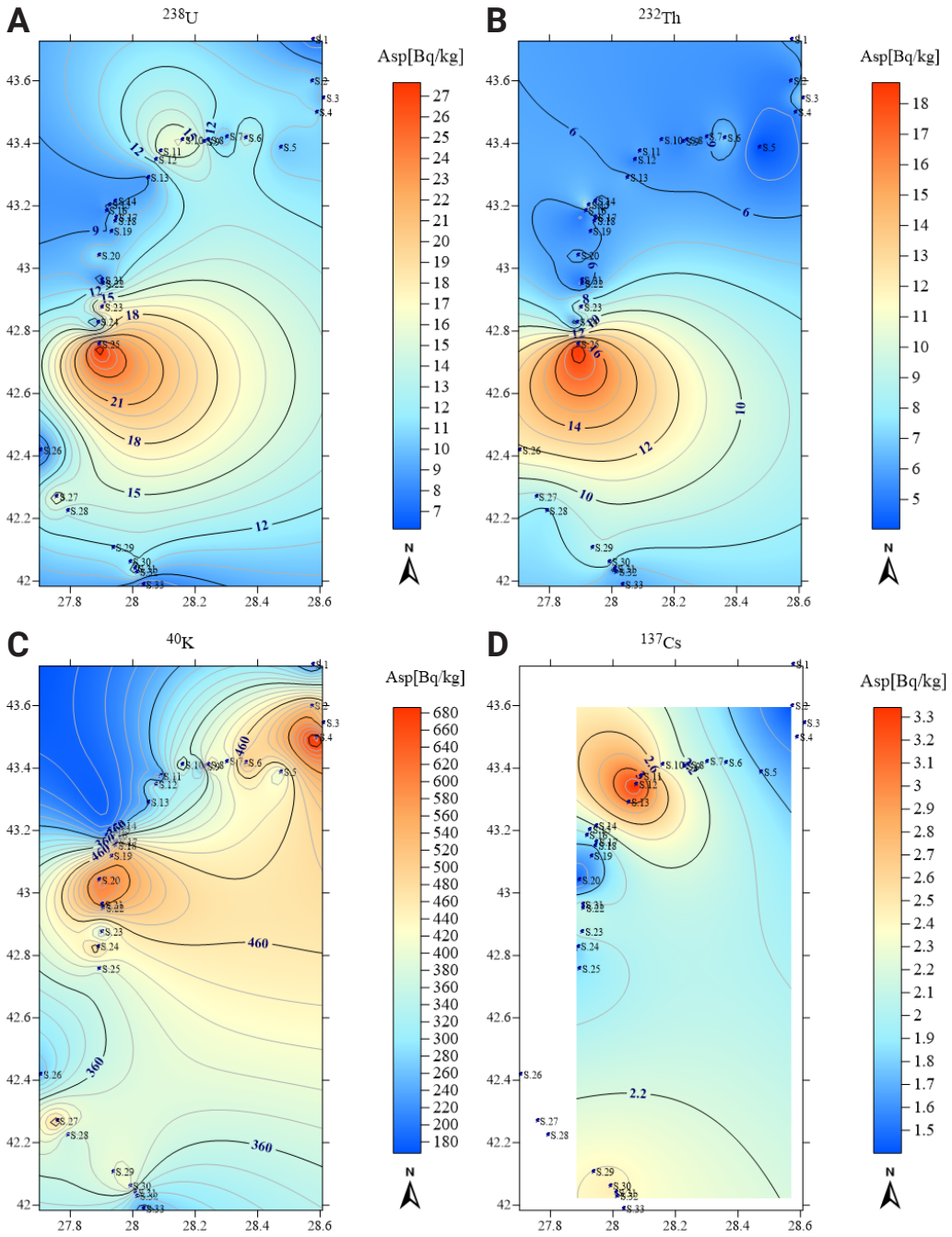

Figure 6. Distribution maps of **A.** ^{238}U; **B.** ^{232}Th; **C.** ^{40}K; **D.** ^{137}Cs of sand samples.

Using the calculated specific activities, a geostatistical analysis of the isotope distribution along the Bulgarian Black Sea coast was performed. Maps of the detected radionuclides, based on GPS coordinates of the sampling points, were generated using SURFER 20.1.195 software and are presented in Fig. 6.

Spatial analysis revealed the following:

- The distribution of ^{238}U was fairly uniform, with the highest concentration observed in the area of Irakli Beach (sampling point S25) (Fig. 6A).
- The specific activity distribution of ^{232}Th showed minimal values in the Northern Black Sea. High values were recorded in the region of Irakli Beach (S25), where the only "hot spot"(maximum value) is observed. (Fig. 6B);

- Hotspots (areas of maximum concentration) for ^{40}K were detected in the village of Tyulenovo (S4) and the Rai campsite area, Kamchiya (S20). The lowest values were recorded near the Rai campsite, with a gradual increase in concentrations observed in the southern part of the Bulgarian Black Sea coast (Fig. 6C).

The highest concentrations of ^{137}Cs were observed at Kranevo village (S12) and the central beach of Golden Sands (S13). The lowest levels were found in the Ray campsite area (Fig. 6D). A gradual increase in values is noted in the southern part of the Bulgarian Black Sea coast (Fig. 6D).

The mean, minimum, and maximum values of the calculated radiation risk coefficients for the samples, alongside world average values, are summarized in Table 1.

Table 1. Data for ADRA, AEDR, Ra_{eq}, E_{out}, H_{ex}, H_{in}, D_{out}, I_γ and ELCR of sand samples.

Sample	*ADRA*, nGy/h	*AEDR*, mSv/y	Ra_{eq}, Bq/kg	E_{out}, mSv/y	H_{ex}, Bq/kg	H_{in}, Bq/kg	D_{out}, nGy/y	I_γ, mSv	*ELCR* x 10^{-3}
Mean	26.32	0.16	51.90	0.03	0.14	0.17	25.69	0.41	0.11
Min	15.69	0.10	31.45	0.02	0.08	0.11	15.37	0.25	0.07
Max	40.74	0.25	84.77	0.05	0.23	0.31	39.58	0.63	0.18
World average	84.00[b]	0.48[a]		-			56.00[b]		0.29[b]
Limit			370.00[c]		1.00[d]	1.00[d]		1.00[b]	

Limits and World averages have been taken from (a): UNSCEAR (1993); (b): UNSCEAR (2000); (c) NEA-OECD (1979) and (d) ICRP 60 (ICRP 1991).

Spatial distribution maps were generated based on the calculated radiation risk coefficients and the GPS coordinates of the sampling locations.

An uneven spatial distribution of radiological indices was observed across the study area (Fig. 7). The density distribution of the individual radiation hazard indices at sampling sites was relatively homogeneous. Distribution maps derived from field data highlight several hot spots exhibiting elevated activity levels of radiological indices ADRA, AEDR, Ra_{eq}, E_{out}, H_{ex}, H_{in}, ELCR, I_γ and D_{out}, notably at Tyulenovo village (sampling point S4), Ray Camping, Kamchia (S20), and Primorsko, South Beach (S27). The lowest values were recorded at Rezovo Beach.

Statistical analysis

Statistical analysis of the data was performed using IBM SPSS STATISTICS 19 software. The results of the calculated statistical parameters are presented in Table 2.

Frequency distributions of the specific activities of detected radionuclides were obtained and are graphically represented in Fig. 8, allowing for the following interpretations of their percentage distribution:

- Specific activities of ^{238}U in the sand samples ranged from 5.8 to 28.3 Bq/kg, with the majority (84%) falling between 7 and 16 Bq/kg (Fig. 8A).
- For ^{232}Th, 87% of the specific activities fell within the 5 to 10 Bq/kg range (Fig. 8B).
- For ^{40}K, 70.5% of the specific activities were between 240 and 550 Bq/kg (Fig. 8C).
- For ^{137}Cs, 75% of the specific activities ranged from 1.4 to 2.5 Bq/kg (Fig. 8D).

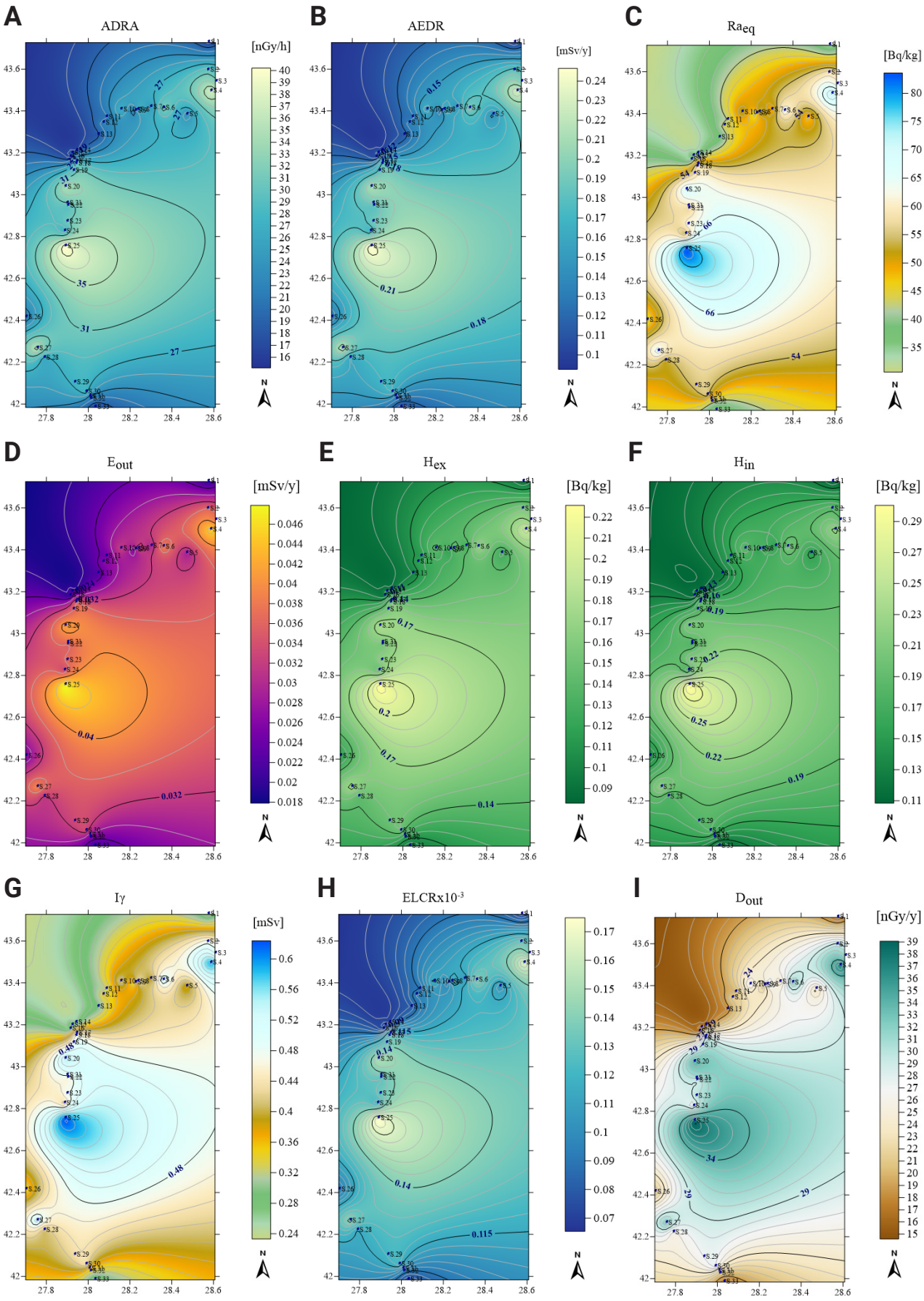

Figure 7. Distribution map of **A.** ADRA; **B.** AEDR; **C.** Ra_{eq}; **D.** E_{out}; **E.** H_{ex}; **F.** H_{in}; **G.** I_{γ}; **H.** ELCR; **I.** D_{out} of sand samples.

Table 2. Data of the statistical analysis of the concentrations of the radionuclides.

Variables	^{238}U	^{232}Th	^{40}K	^{137}Cs
Minimum	5.80	3.90	209.21	1.40
Maximum	28.30	19.06	704.89	9.73
Median	10.82	5.98	378.95	2.12
Mean	11.62	6.82	396.55	3.16
Std. Deviation	4.19	2.78	131.82	2.69
Mode	9.91	5.84	209.21	1.40
Skewness	2.14	2.95	0.26	2.06
Kurtosis	7.00	11.32	-0.77	3.08
Pearson correlation	0.57	0.90	0.40	1.16

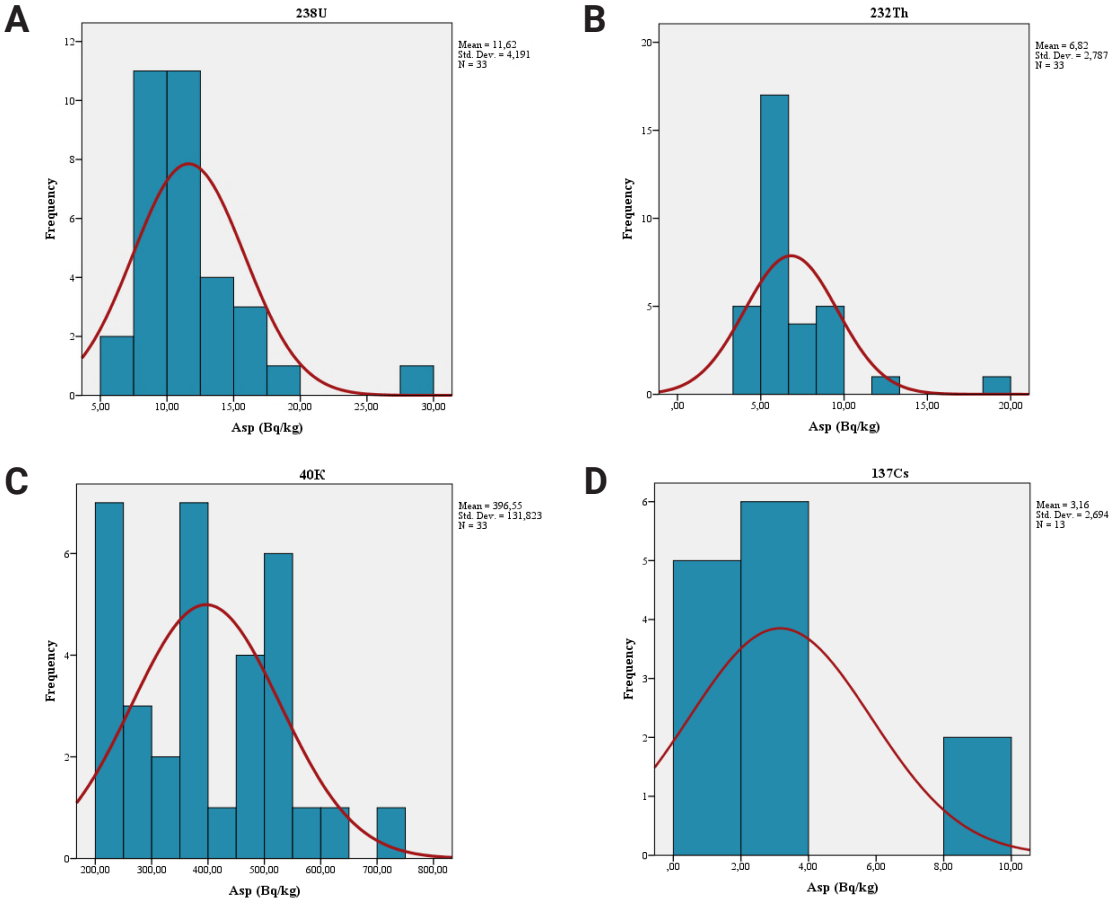

Figure 8. Frequency distribution of radionuclides **A.** ^{238}U; **B.** ^{232}Th; **C.** ^{40}K; **D.** ^{137}Cs in sand samples.

Equation Editor has to be used for formulae with ample space around them.

Kurtosis values were calculated for each radionuclide to assess the distribution type of the data. The kurtosis values can serve as indicators of the distribution shape. For seven of the radionuclides, including ^{238}U and ^{232}Th, the calculated kurtosis exceeded 3, indicating a leptokurtic distribution with a positive excess kurtosis. In contrast, ^{40}K exhibited a negative kurtosis value, corresponding to a platykurtic distribution, which reflects a low concentration of values around the mean for the respective sample set. The kurtosis value for ^{137}Cs was approximately 3, suggesting a distribution close to normality.

Median, mean, and mode values were used to compute the skewness of the data relative to a normal distribution (Table 2). Skewness is a measure of the asymmetry of the probability distribution. The distributions of ^{238}U, ^{40}K, ^{232}Th, and ^{137}Cs showed positive (right) skewness.

To evaluate the relationships among the radiological variables, a Pearson correlation test was performed. Table 3 presents the correlation coefficients for twelve radioactive variables. All values were positive except those involving ^{232}Th and ^{40}K, indicating linear relationships among the parameters. Two coefficients fell within the weak correlation range (0 to 0.3), corresponding to the relationships between ^{238}U and ^{40}K, and ^{232}Th and ^{238}U with E_{out}. Moderate correlations (0.3 to 0.5) were observed for two relationships: ^{238}U with I_γ and with E_{out}. Nine coefficients indicated significant correlations (0.5 to 0.7), ten indicated high correlations (0.7 to 0.9), and thirty-five coefficients demonstrated very high correlations (0.9 to 1.0). Overall, 83% of the correlation coefficients fell within the ranges of high and very high correlation between the parameters.

A hierarchical cluster analysis was performed on 12 radiological parameters. The results, shown as a dendrogram in Fig. 9, classify the system's objects into groups based on similarity.

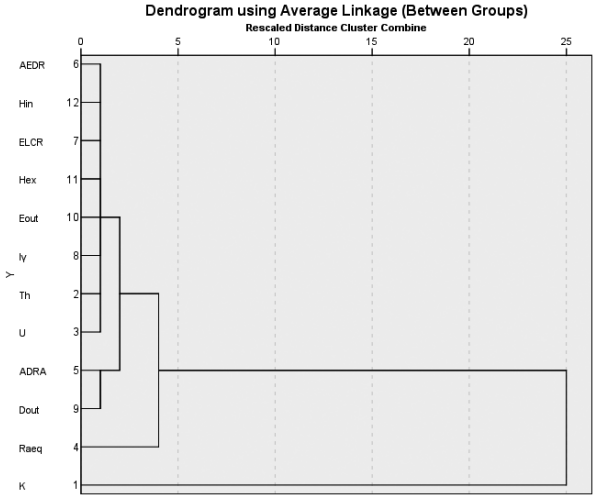

Figure 9. Hierarchical cluster analysis of radionuclides and radiological parameters in Black Sea sand samples.

Table 3. Pearson correlation coefficients.

	^{238}U	^{232}Th	^{40}K	AEDR	Ra_{eq}	H_{ex}	H_{in}	I_γ	ELCR	D_{out}	E_{out}	ADRA
^{238}U	1.00											
^{232}Th	0.64	1.00										
^{40}K	0.04	-0.12	1.00									
AEDR	0.53	0.35	0.85	1.00								
Ra_{eq}	0.58	0.44	0.80	0.99	1.00							
H_{ex}	0.59	0.45	0.79	0.99	1.00	1.00						
H_{in}	0.74	0.56	0.66	0.95	0.97	0.97	1.00					
I_γ	0.49	0.35	0.86	0.99	0.99	0.99	0.94	1.00				
ELCR	0.50	0.34	0.86	0.99	0.99	0.98	0.94	0.99	1.00			
D_{out}	0.50	0.35	0.86	0.99	0.99	0.99	0.95	1.00	0.99	1.00		
E_{out}	0.43	0.28	0.84	0.94	0.93	0.92	0.87	0.94	0.93	0.94	1.00	
ADRA	0.53	0.36	0.84	1.00	0.99	0.99	0.95	0.99	0.99	1.00	0.93	1.00

Cluster I consist of two subclusters. The first includes Ra_{eq}, while the second comprises ten radiological factors – AEDR, H_{ex}, H_{in}, E_{out}, I_{γ}, ELCR, ADRA, D_{out}, ^{232}Th and ^{238}U. Within the second subcluster, ADRA and D_{out} are grouped closely together. Similarly, AEDR, AEDR, H_{ex}, H_{in}, E_{out}, I_{γ} and ELCR form a cluster, indicating similarity among these parameters. Cluster II (one of the two main clusters) is distinctly separated and contains only ^{40}K. The cluster analysis reveals that the concentrations of ^{232}Th and ^{238}U are more closely associated with all radiological parameter levels in the study area, suggesting that this radionuclide does not play a significant role in the formation of natural radioactivity in the investigated locations.

Conclusions

A radiological analysis was conducted on sand samples collected from the Bulgarian Black Sea coast. The obtained data allowed for a qualitative and quantitative assessment of the radionuclides detected in the samples. Distribution maps of radionuclides along the Bulgarian Black Sea coast were generated, and radiological indices related to the safety of beach use in Bulgaria were calculated. The obtained values fall within the permissible limits according to internationally published data from UNSCEAR, NEA-OECD, and ICRP.

The observed dependencies and correlations confirm the relationships between the natural radionuclides found in the samples and the calculated radiation indices. The radionuclide ^{232}Th and its decay products exert the least influence on the radiological parameters. These results provide a more comprehensive understanding of the radiological status of the environment along the Bulgarian beaches.

Acknowledgements

The measurements were made with the financial support of project RD-08-113/05.02.2025, of Konstantin Preslavsky University of Shumen.

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Additional information

Conflict of interest

The authors have declared that no competing interests exist.

Ethical statement

No ethical statement was reported.

Artificial Intelligence (AI) use

The authors accept full responsibility for the content of the manuscript, including the disclosure of any use of AI.
No AI tools were used in the preparation of this manuscript.

Funding

Part of this work has been sponsored by Grant No.РД-08-108/2024 from Konstantin Preslavsky University of Shumen, Bulgaria.

Author contributions

Author Contributions: Conceptualization, N.A. and S.S.; methodology, N.A. and S.S.; software, N.A. and S.S.; validation, N.A. and S.S.; formal analysis, N.A. and S.S.; investigation, N.A., and S.S.; resources, N.A. and S.S.; writing original draft preparation, N.A. and S.S.; writing review and editing, N.A. and S.S.; visualization, N.A. and S.S. All authors have read and agreed to the published version of the manuscript. Authorship was limited to those who have contributed substantially to the work reported.

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Data availability

All of the data that support the findings of this study are available in the main text.