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GAMMA-SPECTROMETRIC CHARACTERISATION OF RECLAIMED SOILS FROM THREE MINING-AFFECTED AREAS IN BULGARIA

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Abstract

Reclaimed soils formed on mine spoils may modify the local natural radiation background because of differences in parent material, texture, and sorption properties. The present study evaluates the activity concentrations and distribution patterns of natural and anthropogenic radionuclides in reclaimed soils from three mining-affected areas in Bulgaria: Maritsa Iztok, Chukurovo, and Asarel-Medet. Gamma-ray spectrometry (HPGe detector; BDS EN ISO 18589-3) and selected physico-chemical analyses (pH, cation exchange capacity, organic carbon, and particle-size composition) were conducted on horizon-wise samples from the main soil profiles (Profiles 1–5). Natural radionuclides (U-238 series, Th-232 series, and K-40) exhibited object-specific patterns consistent with geogenic and textural controls. Spearman correlation and principal component analysis identified a geogenic/texture component dominated by U-238, Ra-226, and Th-232, which were positively associated with fine fractions and cation exchange capacity, and an opposing K-40 trend linked to coarser material. Pb-210 was more closely associated with sorption-related soil properties, suggesting combined control by texture, pH, and the sorption complex. Cs-137 was predominantly below the detection limit, with measurable values mainly in Maritsa Iztok and Chukurovo (West), and no consistent depth-related pattern indicative of substantial downward migration. The external hazard index (H_{ex}) remained below 1 in all studied sites, indicating no radiological concern regarding external exposure.

Key words: gamma spectrometry, reclaimed soils, natural radionuclides, Cs-137, Spearman correlation, principal component analysis

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Introduction. The terrestrial natural radiation background is controlled mainly by the geochemical composition of rocks and soils and by radionuclides from the U-238 and Th-232 decay series and K-40 [1]. In soils, radionuclide distribution is further shaped by soil-forming processes, geochemical interactions, sorption and profile redistribution [1, 2]. These controls may vary strongly in reclaimed soils on mine spoils because parent materials, texture and sorption capacity are often highly heterogeneous [2].

Anthropogenic radionuclides in soils originate mainly from atmospheric nuclear weapons testing and major nuclear accidents (Chernobyl, 1986; Fukushima Daiichi, 2011) [1, 3]. Cs-137 is widely used as an indicator of anthropogenic contamination and a tracer of radionuclide behaviour in soils [3]. In Bulgaria, post-Chernobyl observations show persistent Cs-137 with site-specific differences in inventories and vertical distribution [4]. In addition to post-Chernobyl studies, a related Bulgarian investigation by Kobilarov et al. [5] examined the spatial distribution of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in surface soils from the Bansko–Razlog region and reported marked spatial variability of radionuclide activities, including an increase in the ^{238}U/^{232}Th ratio near former uranium-mining areas. Unlike that regional surface-soil mapping approach, the present study focuses on horizon-wise reclaimed soils from mining-affected areas and evaluates radionuclide distribution in relation to texture and sorption-related soil properties.

Mining and mineral processing can increase the presence of naturally occurring radioactive material (NORM) and elevate the local radiation background [6]. This study investigates the activity concentrations and distribution of natural and anthropogenic radionuclides in reclaimed soils from three mining-affected areas in Bulgaria: Maritsa Iztok, Chukurovo, and Asarel-Medet. The hypothesis is that geogenic and textural factors primarily control the distribution of the U-238 series, Th-232 series, and K-40, while sorption-related properties exert a stronger influence on Pb-210 and Cs-137 behaviour.

Materials and methods. ***Study areas and sampling design.*** Three mining-affected areas in Bulgaria were investigated: Maritsa Iztok (East spoil heap), Chukurovo, and Asarel-Medet. Five representative reclaimed-soil profiles were opened: two in Maritsa Iztok (arable, humus-covered sector, Profile 1; forest, non-humus sector, Profile 2), two in Chukurovo (West spoil heap, Profile 3; Mlada gvardiya spoil heap, Profile 4), and one in Asarel-Medet (north-western spoil heap, Profile 5). Profiles were described by genetic horizons using standard field procedures and harmonised soil description terminology [8, 9], and sampled horizon-wise in accordance with ISO guidance [10, 11] to $\sim$ 1 m depth or to the lithological limit (0–100 cm for Profiles 1–3, 0–90 cm for Profile 4, and 0–70 cm for Profile 5). Samples for laboratory analyses were air-dried, homogenised, and sieved [12]. All measured variables were obtained from horizon-wise samples; for descriptive comparison among study objects, results are additionally presented as object-level aggregated values (mean; min–max across horizons).

Gamma spectrometry. Gamma-emitting radionuclides were determined by high-resolution gamma spectrometry (CANBERRA DSA 1000, HPGe detector, 30% relative efficiency, 1.8 keV resolution) according to BDS EN ISO 18589-3:2024, with counting times of 19–24 h per sample. K-40 was measured directly at 1461 keV, whereas U-238, Th-232, and Ra-226 were evaluated via daughter products under secular equilibrium; Cs-137 was determined at 661.6 keV. The system was calibrated using a certified gamma-emitting radionuclide standard. Expanded uncertainty was calculated at 95% confidence ($k = 2$). The external hazard index (H_{ex}) was calculated according to Montes et al. [7]:

$$H_{\mathrm{ex}} = \frac{A_{\mathrm{Ra}}}{370} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_{\mathrm{K}}}{4810}.$$

Physico-chemical analyses. Soil pH (H_2O and 1 mol L^{-1} KCl; 1:5 soil:solution) was determined according to ISO 10390 [13]. Effective cation exchange capacity (CEC, T) and base saturation (V) were determined by the Ganev and Arsova method [14]. Particle-size distribution was determined by the Kachinskii method (sieving and sedimentation) [15] and is reported as sand, silt, and clay (normalised to 100%); correspondence with ISO 11277 classes was considered in interpretation [16]. Soil organic carbon (OC) was determined by modified Walkley–Black [17], and alkali-extractable carbon (reported here as DOC) by combined alkaline extraction (NaOH + sodium pyrophosphate) [18, 19].

Statistical analyses. Spearman's rank correlation analysis was used to evaluate relationships between radionuclide activities and selected physico-chemical and texture indicators. Principal component analysis (PCA) was applied to z-standardised variables (mean = 0, SD = 1) using horizon-level observations to explore multivariate patterns. For comparative interpretation among study objects, horizon-wise measurements were additionally summarised as object-level mean values with min–max ranges (descriptive synthesis only). Values of Cs-137 below the detection limit were reported as < LOD; for correlation and PCA, censored values were substituted by LOD/2 (LOD = 1 Bq·kg^{-1}; substitution value = 0.5 Bq·kg^{-1}). Because many Cs-137 values were < LOD, interpretation of depth-related distribution patterns was based on horizon-level observations (profile/horizon data) rather than on object-level aggregated values.

Results and discussion. ***Profile description and physico-chemical gradients.*** Distinct physico-chemical gradients are observed among the investigated sites, as shown in Table 1. Soil pH ranges from near-neutral to alkaline in Maritsa Iztok (arable) (pH(KCl) 7.38) to strongly acidic in Chukurovo (Mlada gvardiya) (pH(KCl) 3.57), with intermediate values in Maritsa Iztok (forest) and Chukurovo (West), and a broad range in Asarel-Medet (Table 1). Base saturation is very high in Maritsa Iztok (arable), remains high in Maritsa Iztok (forest) and Chukurovo (West), and is lower in Chukurovo (Mlada gvardiya), while Asarel-Medet exhibits greater variability (Table 1). Cation exchange capacity is highest

Table 1

Physico-chemical properties and simplified particle-size composition by object (mean; min–max). Sand, silt and clay were aggregated and normalised to 100%

Object	pH (KCl)	Base sat., V (%)	CEC, T	OC (%)	DOC	Sand (%)	Silt (%)	Clay (%)
Maritsa Iztok (arable)	7.38 (7.21–7.75)	99.90 (99.90–99.90)	33.23 (27.90–43.90)	0.84 (0.23–2.04)	0.40 (0.11–0.89)	26.21 (21.95–31.68)	36.90 (34.16–39.03)	36.89 (34.16–39.02)
Maritsa Iztok (forest)	5.96 (5.80–6.10)	97.10 (96.10–97.80)	37.40 (32.60–40.30)	1.56 (0.97–2.54)	0.68 (0.46–1.04)	9.78 (4.56–17.35)	45.10 (39.16–47.72)	45.12 (39.20–47.70)
Chukurovo (West)	6.36 (5.50–7.00)	99.20 (97.60–99.80)	12.03 (10.00–16.00)	0.83 (0.49–1.69)	0.25 (0.13–0.54)	35.58 (1.55–51.30)	34.47 (30.20–46.79)	29.95 (10.91–44.15)
Chukurovo (Mlada gvardiya)	3.57 (3.55–3.60)	79.43 (75.20–82.00)	16.07 (13.00–18.20)	4.02 (2.28–5.21)	2.06 (1.38–2.50)	33.69 (19.97–39.89)	34.06 (29.73–37.71)	32.25 (30.39–34.43)
Asarel-Medet	4.47 (3.00–5.50)	85.37 (56.60–99.80)	15.53 (10.50–21.50)	0.51 (0.24–0.88)	0.14 (0.06–0.29)	93.04 (89.43–96.48)	3.98 (2.64–6.30)	2.98 (1.88–4.27)

in the Maritsa Iztok soils, indicating greater sorption capacity compared to the Chukurovo and Asarel-Medet materials (Table 1).

Organic matter indicators also differentiate the objects: OC and DOC are highest in Chukurovo (Mlada gvardiya), intermediate in Maritsa Iztok (forest), and lowest in Asarel-Medet (Table 1).

The simplified particle-size composition reveals pronounced textural contrasts, ranging from a sand-dominated substrate in Asarel-Medet (mean sand approximately 93%) to fine-textured material in Maritsa Iztok (forest), where silt and clay each constitute about 45%. Maritsa Iztok (arable) and Chukurovo (Mlada gvardiya) display more balanced proportions of sand, silt, and clay, while Chukurovo (West) exhibits the greatest heterogeneity across horizons (Table 1). These gradients form the basis for interpreting radionuclide distribution and covariation.

Radionuclide distribution: object-level summaries and profile-level contrasts. Object-level summaries (Table 2) show that the measured natural radionuclides (U-238, Ra-226, Th-232 and K-40) are generally within the ranges reported for Bulgarian soils in the UNSCEAR compilation [1]. Among the studied objects, the highest mean U-238 and Ra-226 activities occur in Maritsa Iztok (forest), whereas K-40 is highest in Chukurovo (Mlada gvardiya) and Asarel-Medet, consistent with differences in parent material and particle-size composition (Tables1 and 2) [1].

Table 2

Activity concentrations by object (Bq·kg^{-1}), mean (min–max). Cs-137 < LOD values were replaced by LOD/2 (0.5 Bq·kg^{-1}) for summary statistics

Object (site)	Pb-210	U-238	Ra-226	Cs-137	Th-232	K-40
Maritsa Iztok (arable)	53.83 (40–65)	58.17 (30–91)	68.67 (54–79)	8.33 (0.5–22)	49.83 (46–60)	573 (460–730)
Maritsa Iztok (forest)	80.3 (56–102)	97.2 (73–115)	110 (93–140)	9.55 (0.5–52)	62.1 (56–73)	399 (280–520)
Chukurovo (West)	47.5 (36–62)	47.17 (36–71)	67.17 (43–97)	2.00 (0.5–6)	43.0 (37–48)	674 (585–760)
Chukurovo (Mlada gvardiya)	37.0 (26–44)	38.83 (32–48)	49.0 (44–59)	0.5 (0.5–0.5)	48.67 (43–52)	858 (800–910)
Asarel-Medet	26.0 (15–44)	29 (22–36)	35.5 (17–49)	0.5 (0.5–0.5)	33.25 (21–43)	875 (453–1090)

At the profile scale, a within-site comparison at Maritsa Iztok shows higher activities in Profile 2 (and associated horizon samples) than in Profile 1 (approximately 1.5× for Pb-210, 1.7× for U-238, 1.6× for Ra-226, and 1.2× for Th-232). This contrast is consistent with the finer, clay-richer material and stronger radionuclide fixation in the non-humus sector (Tables 1 and 2).

For the anthropogenic radionuclide Cs-137, many values are below the detection limit (< LOD) (Table 2). In the present dataset, detectable Cs-137 occurs mainly in Maritsa Iztok and Chukurovo (West), whereas Chukurovo (Mlada gvardiya) and Asarel-Medet are dominated by < LOD values (Table 2), consistent with long-term decay and site-specific retention conditions reported for Bulgaria [4]. Comparable Bulgarian evidence from surface soils was reported by Kobilarov et al. [5], who found marked spatial variability of ^{137}Cs, ^{40}K and the ^{238}U/^{232}Th ratio in the Bansko–Razlog region. However, unlike that regional mapping study, the present work provides profile-scale information for reclaimed soils and links radionuclide distribution to texture and sorption-related soil properties. Because of the high frequency of < LOD values, depth-pattern interpretation was based on horizon-level observations, which do not indicate a consistent pattern of substantial downward migration across the analysed profiles. Instead, detectable Cs-137 occurs mainly in surface to near-surface layers, and its distribution remains heterogeneous among profiles and horizons [4, 20].

The external hazard index (H_{ex}) ranges from 0.39 to 0.61 and remains below 1 in all investigated soils, indicating no radiological concern regarding external exposure under the studied conditions [7].

Controls: correlation matrix and PCA. The Spearman correlation matrix (Fig. 1) reveals two primary control patterns. First, a geogenic component is

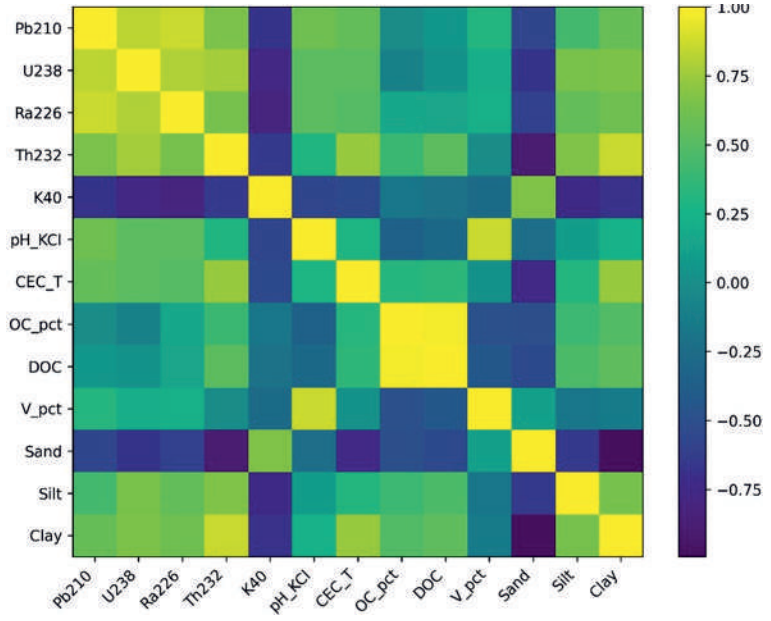


Fig. 1. Spearman correlation matrix for radionuclides and selected soil variables based on horizon-wise data from Profiles 1–5. Cell colours indicate the direction and strength of Spearman's rank correlation coefficient (warm colours, positive correlations; cool colours, negative correlations), and colour intensity reflects the magnitude of ρ. Only relationships with $|\rho| \geq 0.6$ are shown. Cs-137 values below the detection limit were replaced by LOD/2 (0.5 Bq·kg^{-1}) for the analysis

demonstrated by strong positive co-variation among U-238, Ra-226, and Th-232, indicating shared control by parent material and mineral composition. In contrast, K-40 exhibits an opposing trend relative to the U–Th group and is more closely associated with the coarser fraction, suggesting a distinct mineral source and a stronger association with K-bearing phases in the reclaimed materials (Fig. 1).

Second, variables describing the sorption-related soil domain (especially OC and the fine fraction) show coherent relationships, indicating joint effects of organic matter and texture on retention processes. Pb-210 is more closely linked to this sorption-related domain than the U–Th series, supporting the role of soil properties in its retention within the soil matrix (Fig. 1). Overall, Fig. 1 supports a distinction between radionuclides dominated mainly by parent material/mineralogical controls (U-238, Ra-226, Th-232, and partly K-40) and variables reflecting sorption-related soil functioning, with Pb-210 occupying an intermediate position.

The principal component analysis (PCA) biplot (Fig. 2) supports this structure. Principal component 1 (PC1) represents a combined geogenic-textural gradient, distinguishing horizons with higher U-238, Ra-226, Th-232, and clay content from those associated with K-40 and coarser material. Principal component 2

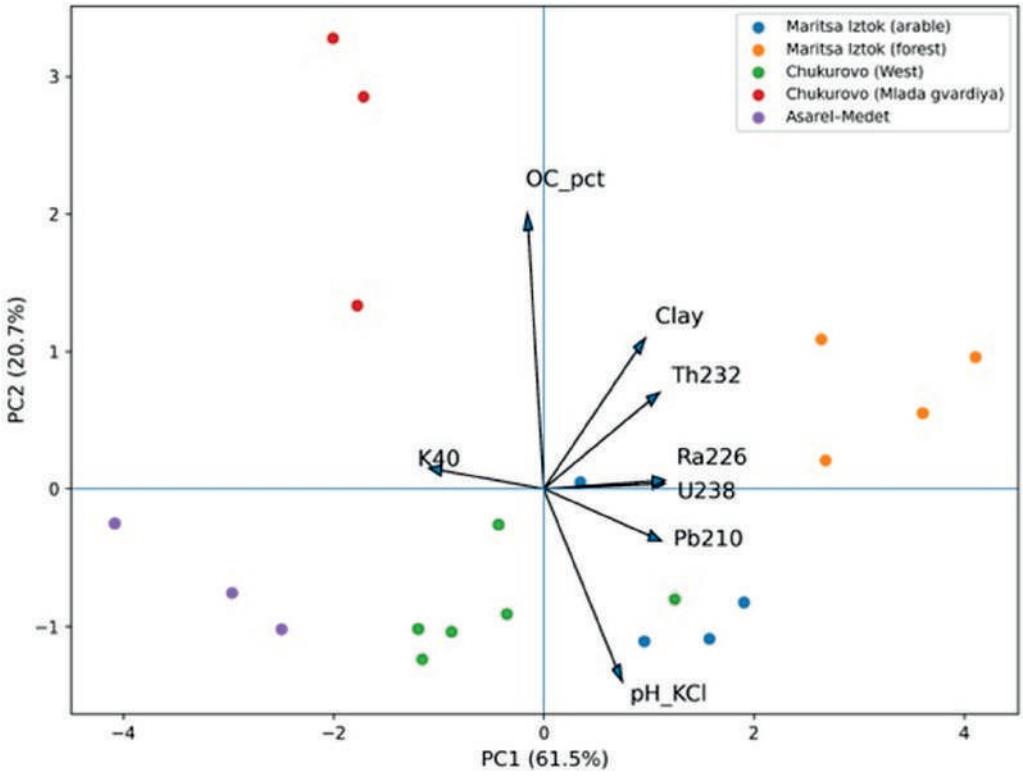


Fig. 2. PCA biplot of z-standardised radionuclide and soil variables based on horizon-wise observations from Profiles 1–5. Points represent individual horizons and are grouped by study object; arrows show variable loadings, with arrow direction indicating increasing values and arrow length reflecting the relative contribution of each variable to the ordination. PC1 represents mainly a geogenic-textural gradient, whereas PC2 reflects an organic/sorption-related gradient

(PC2) reflects an organic/sorption axis, primarily influenced by organic carbon content (OC) (Fig. 2). In the score space, Maritsa Iztok (forest) is positioned towards the U-238/Ra-226/Th-232-clay direction, consistent with finer texture and higher sorption capacity, while Asarel-Medet is located on the opposite side, reflecting predominantly sandy material and low clay content. The two Chukurovo sites occupy intermediate positions, with Mlada gvardiya extending along the OC-driven axis due to elevated organic matter under strongly acidic conditions (Fig. 2).

Conclusions. Reclaimed soils from Maritsa Iztok, Chukurovo and Asarel-Medet show strong contrasts in physico-chemical conditions and texture, ranging from alkaline, high-CEC materials to strongly acidic, organic-rich horizons and very sandy substrates.

The U-238, Ra-226, and Th-232 radionuclides co-vary and are mainly controlled by geogenic factors and texture/CEC, whereas K-40 shows an opposite pattern, consistent with different mineralogical control.

Pb-210 is more closely associated with sorption-related soil properties (pH, fine fraction, and CEC), serving as an intermediate indicator between geogenic and sorption-related controls.

Cs-137 is mostly below the detection limit in several objects and shows no consistent depth-related pattern indicative of substantial downward migration within the studied profiles, indicating a limited present anthropogenic signal in these reclaimed soils.

The external hazard index (H_{ex}) remains below 1 in all investigated soils, indicating no radiological concern regarding external exposure under the studied conditions.

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