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Radioactivity of soils enriched with pyrogenic artefacts in the land of Pernik city, Bulgaria

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Abstract

This article aims to study the radioactivity of soils enriched with pyrogenic artefacts and the possible enhancement of radioactive background and the dose load on the population as a result of coal mining and electricity generation in mine-energetic region Pernik, Bulgaria. Content of major radionuclides responsible for radiation loading – ^{238}U, ^{232}Th, ^{40}K, their progenies ^{226}Ra and ^{210}Pb, and technogenic ^{137}Cs was determined in five soil types representative for the region. Data show that the activity (Bq kg^{-1}) of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in studied soils slightly fluctuates around average values in Bulgarian soils, accepted as background levels in this study. The activity of ^{210}Pb and ^{137}Cs was also comparable to that established in other uncontaminated soils. The mixing of materials and artefacts containing pyrogenic carbon (coal-clayey substrate occurring in the unproductive coal strata and slag produced in the local thermal power plant) during the formation of reclaimed soils increases the content of studied radioactive elements, but the highest levels are found in soils (Cambisols) enriched with soot. Thus, a typical anthropogenic enhancement of radiation background was observed but it is also not hazardous to the population. Gamma radiation emanating from studied soils is below the recommended values and vary from 0.23 to 0.57 for external hazard index, and from 0.29 to 0.61 mSv y^{-1} for outdoor annual effective dose. The established interlink between ^{210}Pb activity and pyrogenic carbon content show that ^{210}Pb could be used as a marker of the recent deposition of pyrogenic carbon formed during the coal combustion and other activities emitting pyrogenic carbon-containing aerosols.

1. Introduction

Soils and coal usually contain natural radionuclides in trace amount (Sheppard, 1980; Naidenov and Zaharinov, 2012). Still, coal mining can lead to enhancement of natural radioactivity by emitting fine coal dust. Coal burning in thermal power plants (TPPs) may generate products with radioactivity several times higher than that of coal (Tadmor, 1986; Lazarova et al., 2020). Together with particular matter they can fallout on the topsoil of both, nearby soils and those located at greater distances (Papp et al., 2002; Flues et al., 2002). Such accessary products increase the radiation background level and total dose load on the population (Hasan et al., 2014).

TPP "Republika" has been identified as one of the biggest polluters in Pernik district, Bulgaria (Pernik Municipality, 2012). The contribution of heaps emerged as a result of brown coal extraction in Pernik is also significant but naturally occurring radionuclides are still the major source of radiation (Tufail, 2012). More than 80 % of the radiation dose received by mankind is due to natural radiation sources (USNRC, 2010). Natural radioactivity is associated mainly to primordial radionuclides, including the isotope ^{40}K and the radioactive series ^{238}U and ^{232}Th.

Therefore, this study aims to determine the radioactivity of soils enriched with pyrogenic artefacts and the possible enhancement of radiation background and the dose load on the population as a result of coal mining and electricity generation in mine-energetic region Pernik, Bulgaria.

2. Materials and methods

2.1. Objects of the research

Soils, representative of the prevailing soil types in the land of Pernik city, Bulgaria, were studied (Fig. 1).

The soil study was carried out in accordance with the classical pedological methods for morphogenetic diagnostics (FAO, 2006) and monitoring approach (BIS, 2019).

Fig. 1. Location of soil profiles

Profile 1 is representative of reclaimed soils, dark grey, heavy sandy-clayey, shallow (Yolevski and Hadjiyanakiev, 1976). They are classified as Pantogarbic Technosol (Eutric, Loamic, Gleyic, Endohyperartefactic, Endohyposulfidic) according to the World Reference Base (IUSS Working Group WRB, 2015). The profile is located in the ridge of the „Maxim“ heap formed by mixing of brown coal, sandy clays and marls, which have occurred in the unproductive coal strata of the mine "St. Anna" and slag from TPP "Republika". It was reclaimed in 1974 with forest vegetation. Currently, the introduced species (Ilinkin and Dimitrova, 2019) are preserved only on the northern slope. The soil reaction varies from medium acid to very slightly alkaline (pH 4.5–7.3) and the soil texture is sandy clay loam to clay loam (clay content 51.3–67.6%). Content of total organic carbon (TOC) (8.63–20.11%) and pyrogenic carbon (PyC) (9.6–18.9%) is very high.

For more complete assessment of the radioactive status of the heap soil samples from 4 points were taken from 0–20 and 20–40 cm depth (Fig. 2). The fourth point is located in a burned slop due to the self-ignition of coal during the summer season, when the soil temperature reaches 80°C (Kuzev et al., 2000).

The analysed coal sample is compiled of well-preserved coal fragments (with unchanged structure, anthracite color and polished surfaces) occurring in Profile 1.

Profile 2 is representative of cinnamon forest soils, smolnitsa-like, deep (WRB name: Endocalcic Chernic Phaeozem (Bathyclayic, Profundihumic)). Soil is located in a forest meadow in the middle part of "Voynikovets" Park. The parent soil-forming materials are carbonate sandy-clayey sediments. The pH varies from slightly alkaline to alkaline (7.7–8.9), textural class changes from clay to heavy clay (clay content: 51.3–66.1%), TOC – 1.01 to 3.93% and PyC – 0.28–0.65%.

Profile 3 is located in smolnitsa, typical, chernozem-like, moderately eroded. The corresponding WRB soil name is Calcic Vertisol (Mollic). The profile is situated in an arable field, sown with maize. Parent rocks are carbonate sandy-clayey sediments. Clay content fluctuates between 58.2% and 66.7%, TOC – from 0.52 to 1.95%, PyC – 0.10–0.39% and pH from slightly to moderately alkaline (8.0–8.5).

Profiles 4 and 5 are located in typical cinnamon forest soils, moderately eroded, occupying the southern part of the city. Profile 4 characterizes the uncultivated variety and can be classified as Endorhodic Hypereutric Cambisol (Bathybrunic, Bathyclayic, Ochric, Endoprototechnic, Reductic). Profile 5 (Rhodic Hypereutric Cambisol (Bathybrunic, Bathyclayic, Ochric, Epiprototechnic)) is located 100 m away from Profile 4 in a field sown with barley. Both soils are enriched with soot. In the meadow analogue (Profile 4) soot is at a greater depth (more than 30 cm), while in Profile 5 it occurs from the surface. The cultivated Cambisol is characterized by more alkaline reaction (pH 7,1–7,2) and lower TOC content (0.67–1.12%) than uncultivated (pH 6.6–6.7 and TOC 0.45–1.74%).

Fig. 2. Location of sampling points in Maxim heap

2.2. Methods of research

The soil samples were homogenized, dried at 80°C, sieved through a 2 mm mesh. Content of natural and technogenic radionuclides was determined by gamma-spectrometric analysis. A multi-channel analyser DSA 1000 (CANBERRA) with HPGe detector, 30% efficiency and 1.8 keV resolution were used. Samples were measured in 450 ml Marinelli Beaker sample containers. The spectrum was analysed by GENIE-2000 software with measurement uncertainties less than 10%. Typical counting times were 19–24 h. The measurements were performed according to BDS EN ISO 18589-3 standard (BIS, 2018):

- of technogenic origin: ^{137}Cs (661.6 keV full energy peak) and;
- naturally present: ^{238}U – by the gamma lines of ^{234}Th (63.5 and 92 keV), ^{235}U (185.7 keV), ^{226}Ra – by the gamma line at 186,2 keV after correction for ^{235}U contribution in the full energy peak at 185.7 keV, ^{210}Pb (46.5 keV), ^{232}Th – by the gamma line of ^{228}Ac at 911 keV and ^{40}K (at 1461 keV).

The efficiency calibration of the system was performed using standard containing lead-210 with density 1 g/cm^3. All soil samples were measured in the same geometry and at the same counting conditions as the calibration cocktail. For the analysis of attenuation correction of ^{210}Pb a procedure described by Długosz-Lisiecka and Bem was performed (Długosz-Lisiecka and Bem, 2013). Activity ratios of two pairs of γ-lines ^{228}Ac 911 keV/209 keV and ^{214}Bi 609keV/^{214}Pb 295 keV were calculated. The values obtained were used as an index for evaluating the attenuation correction factors of ^{210}Pb activity in soil samples.

The procedure for PyC measurement includes prior triple treatments of soil samples (1.5 g dried and grounded to 2 mm) to remove inorganic forms of carbon (with 3 M HCl), followed by dichromate extraction (with 0.1 M $K_2Cr_2O_7$ and 2 M H_2SO_4 for 60 hours in a water bath at 55°C) of soluble and extractable organic substances to separate the fraction of chemically stable pyrogenic carbon (by rinsing and centrifugation of the samples). PyC content of oven-dried samples (60°C) was determined by Elemental automatic analyzer EuroEA 3000 (Tsolova, 2022).

2.3. Assessment of radiation hazard

Gamma-ray radiation hazard caused by radionuclides ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K is evaluated by two indexes.

Outdoor annual effective dose (E_{out})

This index estimates the possible impact of gamma radiation emanating from studied soils and is calculated by Eq. 1 (UNSCEAR, 2000):

$$E_{out} = (0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K) \times 6.136 \times 10^{-3} \quad (1)$$

The recommended value of E_{eff} is 0.7 mSv y^{-1} if a person spends 100% of his time in the given territory.

External hazard index (H_{ex})

The risk of additional dose load on population as a result of soil radioactivity is evaluated by the external hazard index calculated according to eq. 2 (Pourcelot et al., 2003):

$$H_{ex} = A_U/370 + A_{Th}/259 + A_K/4810 \le 1 \quad (2)$$

where A_U, A_{Th} and A_K are the activities in Bq kg^{-1} of ^{238}U, ^{232}Th and ^{40}K, respectively.

3. Results and discussion

Activities of natural radionuclides and radiocaesium measured in soils and coal samples are presented in Table 1. Data show that the specific activity of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K varies around their average value in Bulgarian soils (UNSCEAR, 2000), accepted as background level in this study (Table 1; Fig. 3).

Table 1
Activity (Bq kg^{-1}) of radionuclides in studied soils and coal

Horizon	Depth, cm	^{210}Pb	^{238}U	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Profile 1 – Pantogarbic Technosol							
Acu	0–20	38.0±8.0	51.0±10.0	50.0±10.0	40.0±4.0	420.0±20.0	4.0±0.5
C1u	20–40	35.0±6.0	48.0±10.0	54.0±10.0	38.0±4.0	490.0±20.0	2.0±0.5
C2u	40–60	44.0±6.0	45.0±10.0	53.0±10.0	40.0±4.0	406.0±20.0	2.0±0.3
C3u	60–80	50.0±10.0	36.0±8.0	42.0±8.0	30.0±3.0	300.0±15.0	<1.0
C4u	80–100	46.0±8.0	43.0±8.0	50.0±10.0	34.0±3.0	340.0±15.0	<1.0
C5u	100–120	55.0±10.0	40.0±8.0	51.0±10.0	35.0±3.0	350.0±15.0	<1.0
Average content		**44.7**	**43.8**	**50.0**	**36.2**	**384.3**	**2.7***
Grid points							
Point 1	0–20	64.0±10.0	48.0±8.0	56.0±10.0	38.0±3.0	380.0±20.0	16.0±2.0
	20–4	48.0±8.0	60.0±10.0	60.0±10.0	40.0±4.0	420.0±20.0	2.0±0.5
Point 2	0–20	34.0±6.0	45.0±8.0	48.0±8.0	35.0±3.0	406.0±20.0	10.0±1.0
	20–40	33.0±6.0	36.0±6.0	51.0±10.0	40.0±4.0	314.0±15.0	6.0±0.5
Point 3	0–20	46.0±10.0	34.0±8.0	40.0±10.0	30.0±4.0	420.0±20.0	7.0±0.5
	20–40	35.0±6.0	36.0±6.0	48.0±8.0	33.0±3.0	430.0±20.0	4.0±0.5
Point 4	0–20	36.0±6.0	40.0±8.0	44.0±8.0	40.0±4.0	500.0±20.0	0.8±0.1
	20–40	34.0±6.0	37.0±8.0	40.0±8.0	36.0±4.0	500.0±20.0	2.0±0.2
	40–60	40.0±8.0	42.0±8.0	40.0±8.0	36.0±3.0	500.0±20.0	2.0±0.2
Average content		**41.1**	**42.0**	**47.4**	**36.4**	**430.0**	**5.5**
Profile 2 – Endocalcic Chernic Phaeozem							
Ahk	0–5	62.0±10.0	24.0±5.0	34.0±6.0	38.0±3.0	400.0±20.0	22.0±2.0
A2ck	5–20	32.0±6.0	22.0±5.0	36.0±6.0	40.0±4.0	380.0±20.0	13.0±1.0
A3ck	20–40	22.0±4.0	28.0±5.0	35.0±6.0	44.0±4.0	420.0±20.0	5.0±0.5
A4ck	40–60	25.0±4.0	24.0±5.0	39.0±7.0	42.0±4.0	440.0±20.0	<1.0
A5ck	60–80	28.0±4.0	22.0±4.0	38.0±7.0	44.0±4.0	440.0±20.0	<1.0
A6ck	80–100	30.0±6.0	22.0±4.0	32.0±6.0	44.0±4.0	400.0±20.0	<1.0
A7ck	100–120	16.0±3.0	27.0±4.0	29.0±6.0	37.0±4.0	350.0±20.0	<1.0
Average content		**30.7**	**24.1**	**34.7**	**40.7**	**404.3**	**13.3**
Profile 3 – Calcic Vertisol							
Apk	0–15	38.0±4.0	240±5.0	380±7.0	41.0±4.0	440.0±20.0	7.0±0.4
A1k	15–30	28.0±4.0	25.0±4.0	35.0±7.0	41.0±4.0	460.0±20.0	1.8±0.2
BCck	30–45	17.0±4.0	24.0±4.0	32.0±6.0	32.0±4.0	320.0±20.0	<1.0
C1c	45–60	21.0±4.0	29.0±6.0	33.0±6.0	31.0±3.0	330.0±20.0	<1.0
C2c	60–80	25.0±5.0	26.0±5.0	32.0±6.0	33.0±3.0	360.0±20.0	<1.0
Average content		**25.8**	**25.6**	**34.0**	**35.6**	**382.0**	**4.4**
Profile 4 – Endorhodic Hypereutric Cambisol							
Ah	0–10 (7)	66.0±7.0	60.0±7.0	62.0±8.0	55.0±5.0	590.0±20.0	6.0±1.0
AB	10 –20	54.0±6.0	56.0±7.0	64.0±8.0	60.0±5.0	600.0±20.0	4.0±1.0
Bu	20–60	40.0±8.0	60.0±8.0	80.0±9.0	60.0±5.0	610.0±20.0	<1.0
B1u	60–75	50.0±6.0	65.0±6.0	70.0±6.0	64.0±5.0	615.0±20.0	<1.0
Average content		**52.5**	**60.3**	**69.0**	**59.8**	**603.8**	**5.0**
Profile 5 – Rhodic Hypereutric Cambisol							
Apku	0–10	56.0±7.0	50.0±6.0	52.0±6.0	52.0±5.0	530.0±20.0	8.0±1.0
ABpu	10–30	45.0±5.0	58.0±6.0	54.0±5.0	60.0±5.0	560.0±20.0	5.0±1.0
Bu	30–50	38.0±6.0	60.0±6.0	45.0±5.0	60.0±5.0	580.0±20.0	2.0±1.0
Average content		**46.3**	**56.0**	**50.3**	**57.3**	**556.7**	**5.0**
Coal	*average*	35.0±6.0	31.0±6.0	35.0±6.0	24.0±2.0	200.0±10.0	<1.0
***Average in Bulgarian soils*[9]**			***40***	***45***	***30***	***400***	
***World average values*[9]**			***35***	***35***	***30***	***400***	

* The average was calculated with values > 1 Bq kg^{-1}

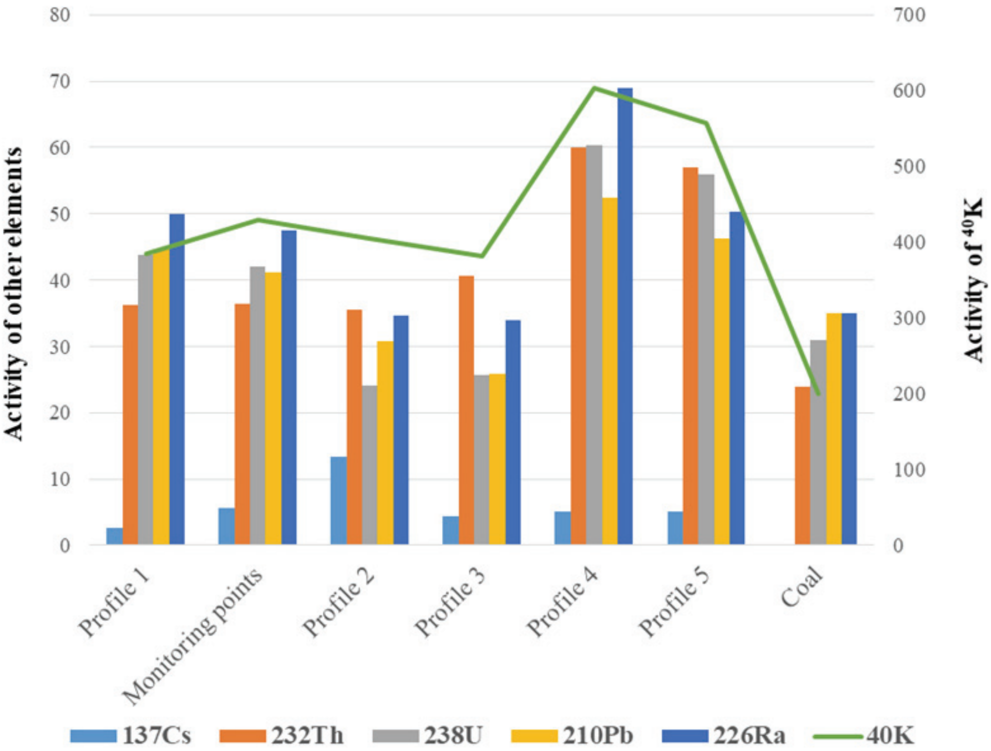

Fig. 3. Average activity of radionuclides in the studied soils and coal

Some of the lowest activities, especially of uranium are found in limy soils – Endocalcic Chernic Phaeozem (profile 2) and Calcic Vertisol (profile 3) where carbonates presumably facilitate its migration out of profile (Vandenhove et al., 2007). The highest values of all measured activities are found in soils enriched with soot – Cambisols (profiles 4 and 5). According to the calculated averages only the activity of ^{232}Th in all soils is higher than the average values in Bulgarian and other globally distributed soils, while the activities of the remaining radionuclides varies in both directions.

An idea of modern distribution of studied radioelements ensures the comparison with data (Table 2) summarized by Osburn (1965). According to them, ^{40}K content is almost unchanged compared to concentrations in soils of the pre-industrial era (1965), in contrast to the content of uranium and thorium, which is increased, although the average values do not give a good idea of variations in concentrations which are generally large in natural objects such as rocks and soils.

The average activity of studied radioactive elements (Table 1; Fig. 3) in reclaimed soils (Profile 1) was approximately 1.5 times higher than in coal. This trend is probably due to the higher radioactivity of slag obtained as a waste product in the TPP "Republika" (also added to reclaimed soils). Thus a typical anthropogenic enhancement of radioactive background was observed.

Table 2.
Content of natural radioactive elements in the lithosphere and soils

Element	Average content		
	Lithosphere	Soils, 1965	Studied soils
^{40}K (%)	3.1	1.6	1.4
U (μg/g)	3.0	1.0	3.3
Th (μg/g)	8.0	6.0	10.4

The activities of radionuclides measured in coal from the region of Pernik were close and comparable to data on coal from other regions of Bulgaria (Lazarova et al., 2020) and the world (UNSCEAR, 2000).

The ecological impact of a radioactive element depends on factors determining its mobility and bioavailability. Soil reaction, which is one of the most important factors of mobility, influences the behaviour of radioactive elements in studied natural soils according to the calculated correlations. The dependence is inversely proportional (r = –0.92) and with increasing of pH decreases the activity of radioisotopes. As far as ^{238}U activity in calcareous soils is concerned the carbonates allowing formation of soluble uranyl carbonate complex ion which can migrate out of profile (Forkapic et al., 2017) are more important than pH.

Clay fraction favours accumulation of ^{238}U (r = 0.79), ^{226}Ra (r = 0.70), ^{232}Th (r = 0.71) and ^{40}K (r = 0.68) by many possible mechanisms but generally interlinks established in this paper coincide with findings of Forkapic et al. (2017). Other correlations indicate that ^{137}Cs is typically attached to the TOC (r = 0.88) and pyrogenic carbon, while ^{210}Pb – mainly to pyrogenic carbon (Fig. 4). The weak relationship between ^{238}U, ^{226}Ra and ^{210}Pb also indicates that ^{210}Pb is not only a product of radioactive decay (r = 0.64). Guogang and Torri (2007) found that ^{210}Pb activity in ash from coal power plants can reach 143 ±6 Bq kg^{-1} and this can change ^{210}Pb amount in soils. Still ^{210}Pb content in studied soils

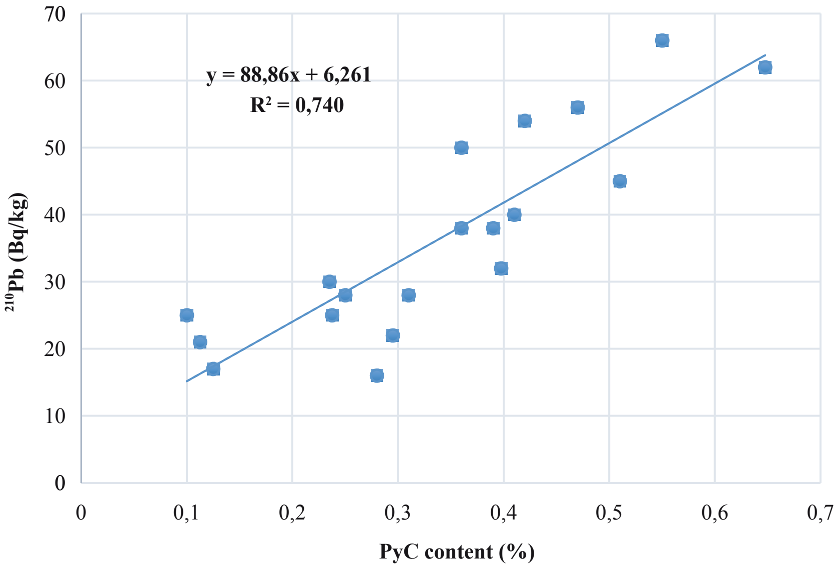

Fig. 4. Correlation between PyC and ^{210}Pb content in studied soils

falls in the typical range pointed out by Prakash et al. (2018) including IAEA-326 and IAEA-327 soil reference materials.

The established correlation between content of PyC and ^{210}Pb (Fig. 4) shows that ^{210}Pb can be used as an indicator of the current accumulation of pyrogenic carbon, as the half-life of ^{210}Pb is ~ 22 years. Another finding can support this assumption – soils may contain higher amount of ^{210}Pb ($^{210}Pb_{ex}$) than the equilibrium concentration with ^{226}Ra (Pb_{supp}) as a result of wet or dry atmospheric deposition (Benmansour et al., 2014) and this could be used to prove the soil additional enrichment with recently produced PyC. This relationship will be further verified due to the self-extinguishing radiation of ^{210}Pb (Bonczyk, 2013) which may reduce the correlation significance.

The content of ^{137}Cs was comparable to that in other regions of Bulgaria (Yordanova et al., 2014) affected by the global fallout after Chernobyl accident (1986 year) and therefore heterogeneously distributed. Another reason for radiocaesium presence in the atmosphere, respectively in soils is biomass burning (Bourcier et al., 2010) and this link should also be taken into consideration in radiocaesium assessment.

The calculated external hazard index shows that studied soils and coal do not present a radiological danger to the population of Pernik city (for all studied soil and coal samples H_{ex} ranges from 0.23 to 0.57). The additionally calculated index – outdoor annual effective dose (E_{out}), which each person can receive as a result of gamma radiation from studied soils is also below the recommended value of 0.7 mSv y^{-1}.

4. Conclusions

The activity of ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb and ^{137}Cs in studied soils is comparable to established in other uncontaminated soils. The mixing of materials and artefacts containing pyrogenic carbon (coal-clayey strata and slag produced in the local thermal power plant) in reclaimed soils increases the radioactivity, but the highest levels are found in soils enriched with soot (Cambisols). Thus, a typical anthropogenic enhancement of radiation background was observed but it is also not hazardous to the population. Gamma radiation emanating from studied soils is below the recommended values and vary from 0.23 to 0.57 for external hazard index, and from 0.29 to 0.61 mSv y^{-1} for outdoor annual effective dose.

Data show that ^{210}Pb could be used as a marker of the recent deposition of pyrogenic carbon formed during the coal combustion or other activities emitting PyC-containing aerosols.

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