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ORIGINAL PAPER

Preliminary studies on the spatial distribution of artificial ^{137}Cs and natural gamma radionuclides in the region of the Ojców National Park, Poland

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Abstract. The aim of the research is to obtain preliminary information about the spatial distribution of gamma radionuclides in the soils taken from the Ojców National Park with emphasis on the behaviour of artificial radionuclides, with ^{137}Cs as a representative. The natural radionuclides ^{40}K, ^{226}Ra (uranium series), and ^{228}Th (thorium series), which are considered as background radiation, were also determined. In total, 18 soil samples were collected during the summer periods in 2015–2017, while the sampling points were selected with respect to differences in rainfall and local topography gradient. The method was based on gamma-ray spectrometry performed on high-purity germanium (HPGe) gamma detector (relative efficiency 34%). ^{137}Cs was mostly deposited in the top soil layers, with activity in the range of 27.9÷586.6 Bq·kg^{-1}. We found strong positive correlation of the ^{137}Cs activity with the soil organic matter content, and at the same time, its dependence on the rainfall amount. Consequently, the soil types and local climate can control the spatial distribution of ^{137}Cs on a small spatial scale. The quantity of natural radionuclides was highly similar in all samples with the following mean values: 38.0 Bq·kg^{-1} for ^{228}Th, 33.1 Bq·kg^{-1} for ^{226}Ra, and 479.9 Bq·kg^{-1} for ^{40}K.

Keywords: ^{137}Cs • gamma spectrometry • natural radioactivity • soil • rainfall • organic content

Introduction

There are many types of pollutants in the environment, and most of them can affect ecosystems. The paper is focused on artificial radionuclides, which are among these undesirable elements.

Natural radionuclides are widespread in the environment and are an external source of radiation that influences human body. Most of them are terrestrial in their origin and are included in the earth crust as minerals, e.g., thorium, uranium-radium, uranium-actinium series and potassium (^{40}K) [1]. Due to this, in many environmental studies, soil is considered as an indicator of environmental degradation. Soil plays the most important role as the element closest to the humans living on the Earth and is directly or indirectly related to conservation of nature [2, 3]. However, natural radiation is quite an independent phenomenon, which is considered as the level of the background radiation. Apart from natural radiation, there is also artificial radiation, which can affect us significantly [4]. Researchers focus mostly on radionuclides that come from nuclear accidents (e.g., Chernobyl disaster) or nuclear bomb tests (20th century) [5, 6]. The radioactive fission products are then released to the atmosphere and spread thereafter [7]. Therefore, radiological research is focused on an increased level of radioactivity. The

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most commonly traced element is ^{137}Cs, because of its special properties [8]. Radiocesium is one of the most efficient fission products, with a long half-life (30.05 years). It can be easily released as aerosol and transported over long distances [9]. Because of its chemical similarity to potassium (an element of almost all living cells), ^{137}Cs is hazardous to human health [10, 11].

After the accidents, researchers were challenged to discover and study the spreading mechanism of cesium. The global deposition mechanism was recognized and described in many publications. Firstly, after an event, cesium gets released to the atmosphere. A number of particles get to the stratosphere and are temporarily accumulated, while the others stay in the troposphere. The troposphere activity is washed out within weeks to months, but the particles that are released above the troposphere remain there much longer and are dispersed worldwide [12, 13]. Subsequently, after deposition, the activity is accumulated on the surface with diverse local spatial distribution [14].

While the global radioactive distribution pattern is fairly clarified, the local patterns are more unexpected. In any local area, a wide range of different connections can be detected. This paper aims to identify the local pattern of spatial distribution of ^{137}Cs and natural radionuclides in the Ojców National Park (Ojcowski Park Narodowy in Polish or OPN), one specific region of Poland. Human activity has been limited there since 1956 when the OPN was established. This date is close to the period in which the first global environmental input of ^{137}Cs was recorded in 1952 [15]. It is estimated that a measurable amount of cesium has appeared in soils since 1954 [16]. Therefore, the area is suitable for the present studies.

Materials and methods

Study area

The OPN is a small complex of ecosystems under strict legal protection. It is situated about 16 km from Krakow (a very urbanized area) but retains a mountainous character. The terrain is formed mostly of limestones, which are a source of unique landscape, with steep canyons, caves, and various stone formations [17].

The OPN territory occupies 2146 ha, including two river valleys (Dolina Prądnika, Dolina Sąspowska) and adjacent slopes. The terrain is covered by forest (70%), meadow (10%), and rocky valley. The topography is presented in Fig. 1.

The OPN limestones were created in the earliest age of the Late Jurassic epoch, namely, the Oxfordian stage. We can distinguish several limestone types, with stack rock mainly.

Due to the extremely varied terrain, the climate of the OPN is diverse and has features of a mountain climate. The area is characterized by a different degree of insulation and, hence, with significant daily differences in air temperatures and humidity [19]. In the higher parts of the Park, humidity is low due to the higher wind speed and the lack of areas that can retain moisture. On the other hand, the valleys, due to the shading of the terrain and lower temperatures, are characterized by higher humidity, which is similar to that in the high parts of the mountains.

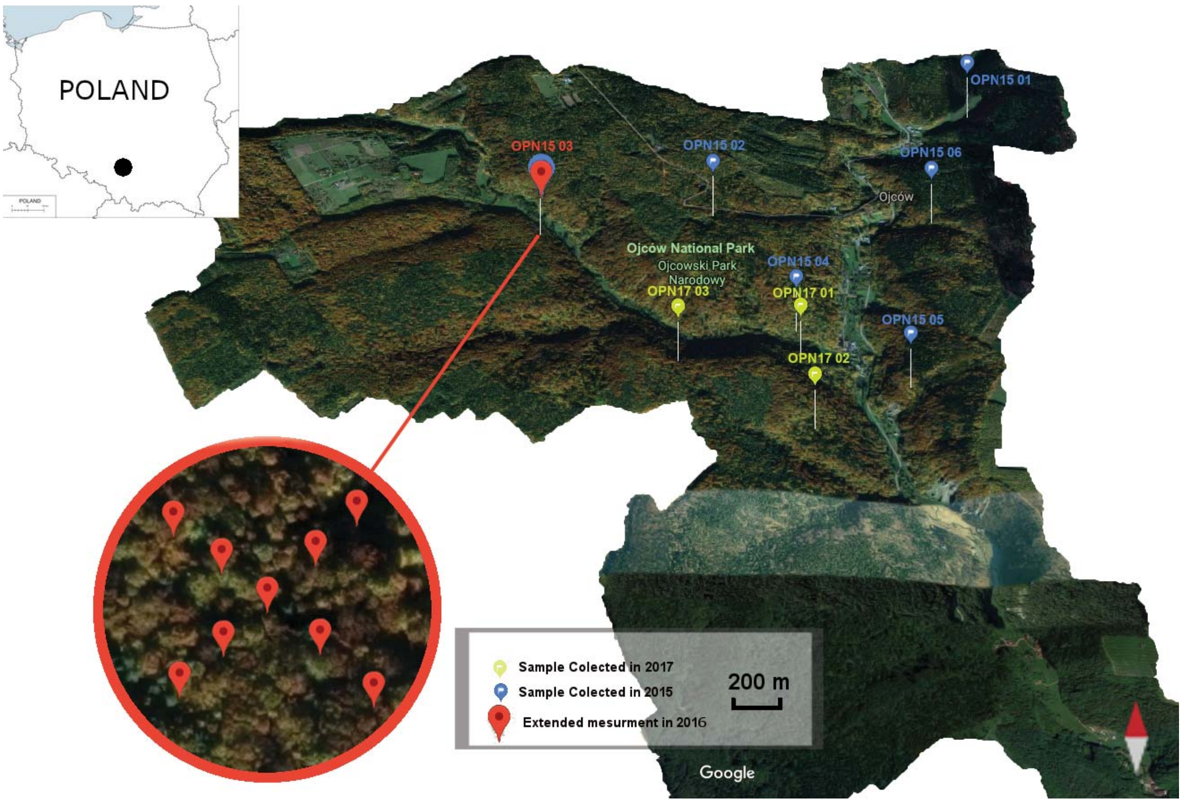

Fig. 1. Distribution of the measurement points in the Ojców National Park. Source: www.google.com/maps [18].

Precipitation is not uniform throughout the entire area either. The largest annual rainfall is observed on the northern slopes. Due to these reasons, up to 20 microclimates can be distinguished there [20].

In the OPN, there are mainly fertile soils formed from clays, limestones, and loess. These include rendzinas, podzolic soils, and brown soils with a relatively high content of organic matter. The north-facing slopes are covered by rendzinas. Podzolic soils are distributed mostly on the south-facing slopes [21].

Field sampling

The research was aimed at determination of natural radioactive series, ^{137}Cs, and ^{40}K in soil samples. In total, 18 soil samples were collected at various locations in the OPN. The research began in 2015, when six soil samples were collected. Then, in 2016, eight samples were collected, and other soil cores were taken in 2017.

A cylindrical probe, 10 cm in diameter and height was used, and the top 10 cm of soil core was taken up. The coordinates and altitudes of the locations were specified with Garmin satellite navigation system. Next, the material was transported to the laboratory and divided into three layers, marked as follows: A – the top 3 cm of the core; B – 3–6 cm of the core; C – the remainder of the material. Afterward, all the samples were prepared for measurement (dried, weighed, homogenized, and packed in a hermetic measuring vessel). After packaging, the samples were left for up to 1 month until an equilibrium of post--radon nuclides (in the uranium series) was retrieved.

Laboratory measurements

The activity was examined using the gamma-ray spectrometry technique. Each sample was measured for 72 hours using a broad-energy germanium detector (model BE3830; Canberra). The system consists also of low-background shielding (0.1 cm Cu; 0,5 cm Cd; 5.5 cm low-activity lead; 7.5 cm lead) and Canberra electronics equipment. The detector's relative efficiency was 34%. The absolute efficiency was identified using the following reference material: IAEA-447 (^{137}Cs and ^{40}K); IAEA-RGU-1 (uranium series gamma emitters); IAEA-RGTh-1 (thorium series gamma emitters).

The efficiency calibration was conducted for the following isotopes (characteristic gamma-ray quantum): ^{137}Cs (661.7 keV); ^{40}K (1460.8 keV); ^{226}Ra (1764.5, 1120.3, 609.3, 351.9, 295.2 keV); ^{228}Th (238.6, 583.1, 727.3, 2614.5 keV).

The activity was calculated according to the formula:

$$(1)\quad A\left[\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right]=\frac{N_s-N_b}{m\,[\mathrm{kg}]\cdot T\,[\mathrm{s}]\cdot p(E)\cdot\varepsilon(E)\cdot T_z}$$

where N_s – number of counts of sample, N_b – number of counts of background, m – mass of sample, T – time of measurement, $p(E)$ – probability of emission, $\varepsilon(E)$ – efficiency, and T_z – self-absorption coefficient.

The self-absorption coefficient was measured following the methodology described previously [22]. All measurements of ^{137}Cs were calculated on October 1, 2015, when the study started. The uncertainty of measurements includes the following: uncertainty of reference material activity, uncertainty of background measurement, uncertainty of efficiency measurement, and uncertainty of sample measurement.

Cesium activity was also expressed in terms of the surface units, Becquerel per square meter [Bq·m^{-2}]. The value was calculated using the following formula:

$$(2)\quad A_{\mathrm{total}}\left[\mathrm{Bq}\cdot\mathrm{m}^{-2}\right]=\frac{A_a\left[\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right]\cdot m_a\,[\mathrm{kg}]}{0.0085\left[\mathrm{m}^2\right]}+\frac{A_b\left[\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right]\cdot m_b\,[\mathrm{kg}]}{0.0085\left[\mathrm{m}^2\right]}+\frac{A_c\left[\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right]\cdot m_c\,[\mathrm{kg}]}{0.0085\left[\mathrm{m}^2\right]}$$

where: A – total activity of ^{137}Cs in the soil [Bq·m^{-2}]; A_a, A_b, and A_c – activities of ^{137}Cs in the given layers of the soil (a, b, and c, respectively); m_a, m_b, and m_c – total masses of the samples obtained in the given layers; 0.0085 m^2 – surface of the soil taken by the cylinder probe.

The organic matter content was also determined. Portions of the samples were heated to 600°C. The weight loss indicated the percentage of organic matter.

Results and discussion

The first year of study was devoted to general measurement activities. In 2015, six points, scattered over the entire national park, were selected. The locations were marked on a satellite 3D photograph (Fig. 1) to show the characteristics of the landscape. The results are presented in Table 1, and the uncertainty did not exceed 10%.

The measurement in 2015 showed that the activity at 'OPN15 03' point was the highest. In a study published about the climate of the OPN [20], increased rainfall was recognized at this point. Generally, ^{137}Cs is transported mostly by rainfall to the ground, and then it settles on the surface of soils, vegetation, and water reservoirs. Cesium from the vegetation could be absorbed by the leaves or washed down to the soils; the second option is more frequent. Therefore, rainfall variation can significantly affect the activity of the soil. This hypothesis can be confirmed with the help of composite ion exchangers [23]. Probes with sorbent are placed in the OPN. This allows for measurement of both the current deposition from the atmosphere and the differences in humidity distribution.

Natural radionuclides were at the same level of activity, and no special trends were noticed.

In 2016, measurement was executed in only one area, in which the activity was found to be increased

Table 1. Activity of selected radionuclides in soil samples taken from the Ojców National Park in 2015

Tag	Layer	Activity of ^{228}Th [Bq·kg^{-1}]	Activity of ^{226}Ra [Bq·kg^{-1}]	Activity of ^{40}K [Bq·kg^{-1}]	Activity of ^{137}Cs [Bq·kg^{-1}]	Total activity of ^{137}Cs [Bq·m^{-2}]
OPN15 01	A	42.46 ± 1.43	35.36 ± 1.70	499 ± 13.86	30.88 ± 0.91	3 344
	B	46.23 ± 1.35	35.21 ± 1.60	488 ± 12.20	45.55 ± 1.14	
	C	50.84 ± 1.31	35.90 ± 1.17	520 ± 16.91	43.87 ± 1.09	
OPN15 02	A	37.17 ± 1.71	25.58 ± 1.72	499 ± 15.47	88.83 ± 1.21	7 004
	B	33.88 ± 1.93	29.71 ± 1.67	534 ± 13.37	94.37 ± 1.34	
	C	42.86 ± 1.98	23.73 ± 1.41	471 ± 9.76	76.21 ± 1.15	
OPN15 03	A	41.24 ± 1.02	27.55 ± 1.04	382 ± 8.65	578.58 ± 3.12	16 508
	B	44.91 ± 1.41	33.88 ± 1.43	443 ± 7.95	327.90 ± 2.27	
	C	44.33 ± 1.54	32.93 ± 1.52	486 ± 13.50	64.07 ± 0.85	
OPN15 04	A	39.76 ± 1.55	27.89 ± 1.10	553 ± 13.82	37.25 ± 0.76	2 555
	B	41.38 ± 1.68	29.54 ± 1.11	555 ± 18.05	44.38 ± 0.89	
OPN15 05	A	43.46 ± 1.80	32.19 ± 1.11	529 ± 16.40	27.92 ± 0.74	2 643
	B	47.75 ± 1.35	31.47 ± 1.34	525 ± 13.14	27.08 ± 0.73	
	C	47.97 ± 1.36	32.91 ± 1.05	559 ± 11.58	27.32 ± 0.70	
OPN15 06	A	38.48 ± 1.07	29.24 ± 1.38	463 ± 10.49	73.49 ± 1.12	5 274
	B	43.31 ± 1.08	11.58 ± 0.88	506 ± 9.09	81.06 ± 0.92	
	C	35.62 ± 1.17	26.80 ± 0.97	439 ± 9.10	76.99 ± 0.81	

in the previous year. To locate the maximum activity, measurements were repeated at the same point, but with an extended measurement methodology. Samples were taken from the central point and then

Table 2. Activity of selected radionuclides in soil samples taken from the Ojców National Park in 2016

Description	Tag	Activity of ^{228}Th [Bq·kg^{-1}]	Activity of ^{226}Ra [Bq·kg^{-1}]	Activity of ^{40}K [Bq·kg^{-1}]	Activity of ^{137}Cs [Bq·kg^{-1}]
Central point	OPN OA	34.59 ± 1.46	31.26 ± 1.24	484.17 ± 6.95	204.48 ± 0.98
	OPN OB	36.53 ± 1.28	29.63 ± 1.06	501.04 ± 6.29	34.16 ± 0.41
	OPN OC	37.56 ± 1.33	31.37 ± 1.11	507.45 ± 6.54	8.67 ± 0.28
5 m to top of valley	OPN GA5	36.08 ± 1.32	29.31 ± 1.12	470.51 ± 6.15	281.93 ± 1.02
	OPN GB5	37.12 ± 1.54	30.44 ± 1.29	498.33 ± 7.49	48.17 ± 0.56
	OPN GC5	38.19 ± 1.70	30.56 ± 1.42	492.63 ± 8.16	29.19 ± 0.50
10 m to top of valley	OPN GA10	32.98 ± 1.61	29.46 ± 1.39	445.35 ± 7.34	586.62 ± 1.76
	OPN GB10	34.08 ± 1.18	28.92 ± 0.99	484.44 ± 5.69	137.38 ± 0.67
	OPN GC10	36.45 ± 1.32	28.81 ± 1.11	485.70 ± 6.51	37.95 ± 0.44
5 m to east side	OPN PA5	35.01 ± 1.71	29.04 ± 1.45	445.29 ± 7.64	524.74 ± 1.73
	OPN PB5	35.56 ± 1.47	30.58 ± 1.24	497.77 ± 7.06	141.82 ± 0.84
	OPN PC5	34.39 ± 1.16	29.57 ± 0.95	508.04 ± 5.65	60.96 ± 0.46
10 m to east side	OPN PA10	38.79 ± 2.29	31.94 ± 2.15	487.45 ± 11.46	108.65 ± 1.19
	OPN PB10	37.80 ± 2.18	28.31 ± 2.05	476.30 ± 10.75	78.81 ± 0.84
	OPN PC10	36.13 ± 1.75	29.45 ± 1.64	576.00 ± 8.80	29.44 ± 0.49
5 m to west side	OPN LA5	32.85 ± 1.91	30.84 ± 1.81	481.17 ± 9.60	189.68 ± 1.04
	OPN LB5	34.43 ± 2.75	26.99 ± 2.56	507.04 ± 13.85	31.92 ± 0.78
	OPN LC5	32.74 ± 1.79	33.34 ± 1.70	507.77 ± 9.16	9.65 ± 0.41
10 m to west side	OPN LA10	37.88 ± 1.96	28.97 ± 1.85	475.66 ± 9.79	152.65 ± 0.96
	OPN LB10	36.40 ± 2.21	30.05 ± 2.08	504.90 ± 11.20	86.33 ± 0.88
	OPN LC10	33.17 ± 2.27	29.82 ± 2.11	499.88 ± 11.32	68.47 ± 0.81
5 m to bottom of valley	OPN DA5	32.96 ± 2.50	26.60 ± 2.44	471.69 ± 12.47	467.10 ± 1.97
	OPN DB5	26.33 ± 2.66	26.06 ± 2.52	507.74 ± 13.47	90.31 ± 1.07
	OPN DC5	34.28 ± 2.57	33.52 ± 2.40	522.40 ± 13.27	64.47 ± 1.01
10 m to top of valley	OPN DA10	37.40 ± 2.20	29.15 ± 2.07	479.93 ± 10.92	129.25 ± 1.01
	OPN DB10	35.44 ± 2.23	30.61 ± 2.04	460.55 ± 10.80	52.57 ± 0.73
	OPN DC10	39.90 ± 1.88	30.64 ± 1.73	483.81 ± 9.29	31.31 ± 0.52

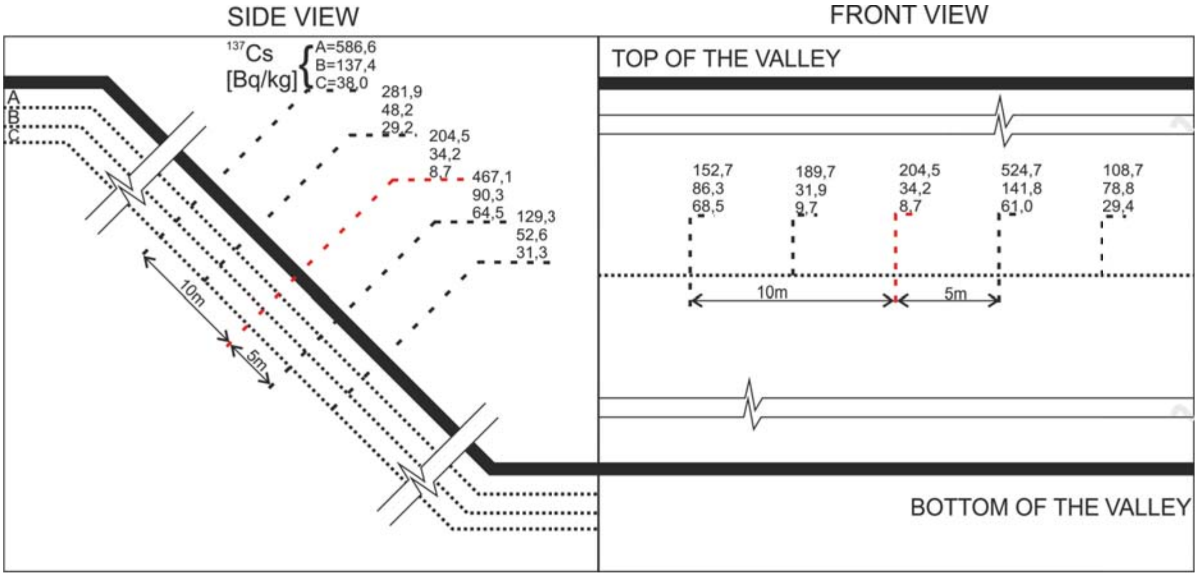

Fig. 2. Spatial distribution of ^{137}Cs activity on the slope of Sąspowska Valley (OPN).

Table 3. Activity of selected radionuclides in soil samples taken from the Ojców National Park in 2017

Tag	Activity of ^{228}Th [Bq·kg^{-1}]	Activity of ^{226}Ra [Bq·kg^{-1}]	Activity of ^{40}K [Bq·kg^{-1}]	Activity of ^{137}Cs [Bq·kg^{-1}]	Total activity of ^{137}Cs [Bq·m^{-2}]
OPN17 01A	47.3 ± 3.7	34.7 ± 3.1	358.9 ± 10.0	63.7 ± 0.9	2070
OPN17 01B	47.7 ± 3.5	34.8 ± 3.0	376.3 ± 9.4	66.0 ± 0.9	
OPN17 02A	28.8 ± 5.3	25.9 ± 3.7	415.4 ± 13.5	132.2 ± 2.0	
OPN17 02B	30.7 ± 4.9	28.1 ± 3.4	409.5 ± 12.7	270.8 ± 2.4	7098
OPN17 02C	35.3 ± 4.3	29.9 ± 2.6	465.1 ± 11.6	196.4 ± 1.9	
OPN17 03A	33.6 ± 3.7	28.9 ± 2.6	510.0 ± 10.6	71.5 ± 1.2	
OPN17 03B	37.3 ± 3.9	31.0 ± 2.8	497.9 ± 11.3	57.7 ± 1.2	3672
OPN17 03C	37.0 ± 3.2	29.7 ± 2.2	501.6 ± 9.0	49.9 ± 0.9	

from several other points located 5 and 10 m away in four different directions (toward the top of the slope, down the slope toward the bottom of the valley, to the east side, and to the west side). Table 2 shows the results.

The distribution does not show any clear dependence (Fig. 2). No horizontal or vertical correlations were observed. Other authors have observed that cesium is strongly associated with organic matter and its predominant migration is negligible [24]. Therefore, the radionuclide is hard to be washed and transported with surface water. The results of the present study seem to confirm this view.

In 2017, soil samples were taken from three other locations, which are shown in Fig. 1. The aim was to expand the grid to other locations in the Park. No other hotspots were detected. The results are presented in Table 3.

To check the dependence of the radionuclides, principal component analysis (PCA) was performed for all samples.

The characteristic eigenvalues and percentage of variance explained for the five extracted major components are shown in Table 4. As can be seen, there are only two initial major components whose eigenvalues are >1. Therefore, in accordance with Kaiser's criterion, our further considerations would include two major components only. These two initial major components account for almost 70% of the variances of variables.

In Fig. 3, a clear correlation between the activities of ^{228}Th and ^{226}Ra is seen. As shown in the figure, the concentrations of these radioisotopes are unrelated to organic matter content (they are orthogonal). It seems to be quite normal. They are of natural origin, come from the parent rock, and are associated with the inorganic part of the soil. The ^{137}Cs activity is correlated with the organic matter content and negatively correlated with the activity of natural ^{40}K. Cesium is well absorbed in the organic part of the soil, whereas potassium shows similarity to the inorganic part of the soil.

The radioactivity of natural series was similar at every point (relative standard deviation was <10%), and no specific variation was observed. The mean

Table 4. Characteristic eigenvalues and percentage of explained variance for principal component analysis

Component number	Eigenvalue	Percent of variance	Cumulative percentage
1	2.246010	44.920	44.920
2	1.253060	25.061	69.981
3	0.722165	14.443	84.425
4	0.507382	10.148	94.572
5	0.271388	5.428	100.000

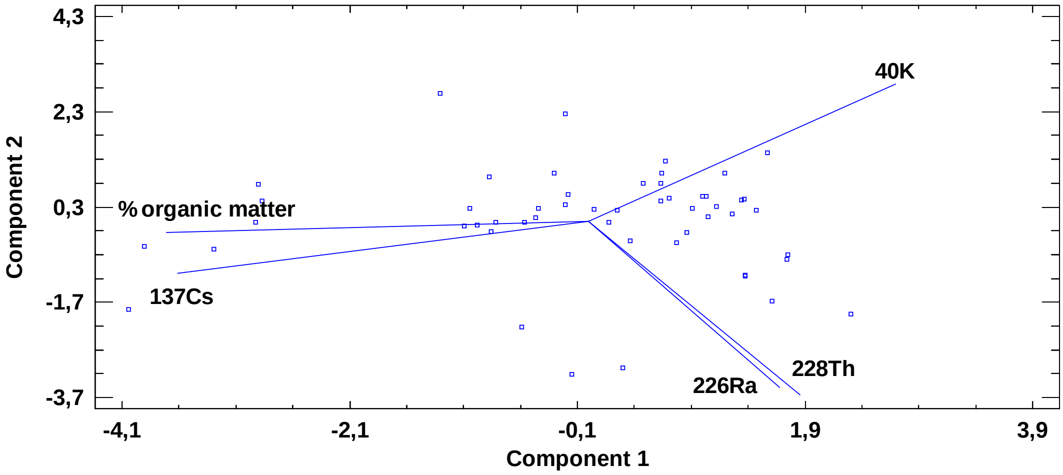

Fig. 3. The biplot with projection of the variables and cases onto the plane of the first two principal components of PCA.

values (range) of ^{226}Ra, ^{232}Th, and ^{40}K activities were, respectively, 33.1 Bq·kg^{-1} (25.9–51.1), 38.0 Bq·kg^{-1} (26.3–50.8) and 479.9 Bq·kg^{-1} (358.9–576.0). The values did not differ significantly from those given for the region of Poland in the Annual Report of the National Atomic Energy Agency [25].

Conclusions

Cesium-137 is strongly correlated with the organic matter content. If the soil is rich in organic matter, the cesium activity is increased. Based on this, we can distinguish humus-rich soil types, e.g., rendzinas, which should absorb more cesium. Concluding, organic matter has better ability for absorption of cesium. The same relationship is observed in other Polish protected areas, e.g., the Babia Góra National Park and the Tatra National Park [3, 4].

Apart from the organic matter content and the type of soil, local climate-related properties could be another factor conditioning the spatial distribution of ^{137}Cs in small areas. Studies of the regional microclimates could help indicate the area most exposed to artificial radionuclides. The amount of rainfall is one of the most important factors.

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