

Assessment of the influence of soil properties on ^{137}Cs accumulation in Of horizon in forest soils

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Abstract. The work focuses on assessment of soil physicochemical parameters influence on ^{137}Cs accumulation in Of soil horizon. Besides organic matter content and pH, the parameters related to sorption properties and mobile ions concentration were considered. The data were transformed using Box-Cox formula. To find mutual relationships between variables cluster analysis (CA) and principal components analysis (PCA) were used. It was found that the transformed physicochemical parameters in Of horizon are more or less related with each other but no linear or nearly linear relationships between ^{137}Cs activity and physicochemical parameters were found.

Key words: ^{137}Cs • forest soil • physicochemical parameters • cluster analysis (CA) • principal component analysis (PCA)

Introduction

The global fallout, resulting from nuclear tests, particularly intensified in 1963–1964, as well as the Chernobyl fallout of 1986 brought about the appearance of, among others, ^{137}Cs and ^{134}Cs (post-Chernobyl) isotopes in the environment.

In Poland, areas of increased radiocesium activity resulting from the Chernobyl fallout were observed. The highest activities were noted in the south-western Poland, in the eastern part of the Silesian and Foreland Lowland. In this zone, several smaller areas were distinguished, among which the most important is the so-called Opole Anomaly [6, 17], where areas of ^{137}Cs activity exceeding 34 kBq/m^2 [3] were still present in 2000.

Twenty three years after the fallout, increased activity of ^{137}Cs is still recorded, mainly in woodlands and wastelands. Because of its specificity, woodland soils are characterized by permanent system of genetic horizons present in the soil profile in which each horizon is distinguished by a relatively uniform color, consistency, granulation, chemical composition, quantity and quality of organic matter, and other properties. Because of this factor, soils of woodland complexes are ideal to follow migrations, changes of the content and spatial distribution of radioisotopes.

Contrary to farmlands, in woodlands the migration of ^{137}Cs results from physical and chemical characteristics of soil, weather conditions and type of vegetation. It was found out that 99% of radioactive fallout is still

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in the soil, especially in the upper layers. Woodland soil has several mineral and organic layers with different composition, physical and chemical properties that influence sorption capacity, sorption type and sorption preferences.

The distribution of ^{137}Cs in soil in a given area is influenced by initial fallout as well as horizontal and vertical transfer in soil. Radioisotope activity in vertical profile is the measure of radionuclide concentration changes depending on depth. The factors that determine changes of radionuclide activity in soil are: weather conditions, moisture content in soil dependent on the duration of fallout, soil structure and rate with which water might infiltrate [4, 5, 23]. Weak but significant correlations were found between the ^{137}Cs deposition densities and altitude and annual precipitation rate. A higher correlation coefficient of $r = 0.65$ was found for the correlation between the ^{137}Cs deposition densities and the climatic de Martone index [15].

The physicochemical soil parameters are also very important. They determine the nature of migrating cesium cations interaction with mineral and organic parts of soil. ^{137}Cs accumulation in soil is mainly based on chemisorption and follows the general principles of metals sorption in natural or artificial ion exchangers [2] and depends mainly on the charge of an ion and its size, including its hydration sphere. Experiments proved that the ^{137}Cs ion exchange depends upon the mineral particle size, structure, the ionic strength of the soil solution and the presence of the competing ions, especially K^+ and NH_4^+ [20]. It was found that clay components of the organic horizons in forest soils caused decrease in the available ^{137}Cs fraction [8]. Though organic soil horizons do not contain enough clay to immobilize cesium, many authors [1, 9–13, 22, 29–31], reported the highest ^{137}Cs activities just in upper, rich in organic matter soil horizons. A hypothesis was also suggested that organic matter modifies the adsorption properties of clay minerals in soil [12]. It was found that low pH values improve cesium uptake from soil by plants [21, 27, 28], but decreases velocity of the Cs cation transfer [27]. Type of clay and sand content in mineral horizons may also determine Cs transport parameters [7, 14, 26].

In our recent paper [32] we have described influence of soil horizon physicochemical properties on ^{137}Cs accumulation assessed by multiple linear regression. Our results supposed, that migration of Cs^+ in soil is affected mainly by horizon's acidity, presence of minerals and ion exchangeable substances. The important conclusion was that cation migration in soil is usually influenced by more than a single horizon parameter. Consideration values of two or more parameters would provide information necessary for ^{137}Cs activity prediction in a certain soil horizon. However, no significant linear relationships between ^{137}Cs activity and physicochemical properties in Of horizon were found. It is possible that in this horizon the relationships are more complicated than the linear ones. In this paper we show more detailed analysis of the problem of physicochemical parameters influence on ^{137}Cs activity in Of horizon of forest soil. Existence of structures in data was examined using methods of CA. To investigate interrelation between variables the PCA was used.

Methods

The samples were taken in the Polish-Czech border areas from the Opole Anomaly, and areas in the Czech Republic, which are the extension of the Opole Anomaly. They were taken in the vicinity of at least 20 year old trees, not less than 100 m from roads. On the ground around the trees grew at most few forest bed plants, it was covered mainly by fallen tree leaves. The samples of soil profiles were collected between end of autumn and early spring.

The measurement of ^{137}Cs activity in samples of woodland soil was carried out by means of a gamma-spectrometer with a germanium detector HPGe (Cannberra) of high resolution: 1.29 keV (FWHM – full width at half maximum) at 662 keV and 1.70 keV (FWHM) at 1332 keV. Relative efficiency: 21.7%. Energy and efficiency calibration of the gamma spectrometer was performed with the standard solutions type MBSS 2 (Czech Metrological Institute, Praha), which covers an energy range from 59.54 keV to 1836.06 keV. Geometry of calibration source was Marinelli ($447.7 \pm 4.48 \text{ cm}^3$), with density $0.985 \pm 0.01 \text{ g/cm}^3$, containing ^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{113}Sn , ^{85}Sr , ^{88}Y and ^{203}Hg . Geometry of samples container was Marinelli, 450 cm^3 . Measuring process and analysis of spectra were computer controlled with use of the software GENIE 2000. The radiation spectrum was recorded for 24 h. The measurement uncertainty did not exceed 3%.

The following physicochemical properties of soil horizons were determined, using methods generally applied in soil science [16, 24]:

- soil acidity – by the potentiometric method in 1 M KCl (pH_{KCl}) at soil/solution ratio 1:10 (mineral soil) or 1:1.25 (organic soil) and in distilled water (pH_{w}) (1:10 or 1:2, respectively),
- organic matter content (OM),
- hydrolytic acidity (Hh) – by the Kappen method in 1 M CH_3COONa ,
- total basic positive ions (S) – by the Kappen method in 0.1 M HCl;
- electrical conductivity (σ).

Additionally, sum T of hydrolytic acidity and total basic positive ions was computed.

The area on which the samples were collected was not contaminated uniformly. It was expected that location of the place from which sample was taken affects its total ^{137}Cs activity. To omit or, at least, to diminish the effect of unequal initial soil contamination the relative activities a_r were calculated. ^{137}Cs activities in consecutive horizons of the soil sample were added and then the activity of each horizon was divided by the sum calculated previously. It was expected, that the relative activities would be more proper for assessment of the influence of soil parameters on ^{137}Cs accumulation than the activities on their own.

It was found that distributions of our data were significantly different from the normal one. Because some of the methods applied for the results interpretation required data distributed at least nearly normally, our data were transformed using the Box-Cox formula. For statistical computations, the R language [25] was utilized. R is a free software environment for statistical computing and graphics.

To gain information about possible structures in our data clustering methods were utilized. These methods allow to assign objects to different groups, so that the data in each subset share some common trait. For our computation functions provided by the “cluster” library of R were used. Functions available in this library were described by Kaufman and Rousseeuw [19]. For each variable, data were standardized by subtracting the variable’s mean value and dividing by the variable’s mean absolute deviation, and then all of the pairwise dissimilarities between observations in the data set were computed. Dissimilarities were euclidean distances between points computed in space of standardized variables.

An overview of cluster existence in data is shown on dendrograms – a tree diagrams illustrating the arrangement of the clusters. Diagrams were constructed using “agnes” and “diana” functions from “cluster” library. The first of the functions uses an agglomerative nesting procedure. At the beginning, all elements (points) are apart, each one of them is a small cluster by itself. At the first step, the two closest (the least dissimilar) objects are joined to form first cluster. In next step the dissimilarities (distances, d) between the new cluster and the remaining elements are computed. The second cluster consists of the first one and the element closest to it. Joining to clusters and new distances, computing is repeated until one, big cluster appears. Function “diana” realizes the inverse procedure. At each step, a cluster is splitted into two smaller ones until all clusters contain only a single element.

Taking into account single point i belonging to cluster A , the $a(i)$ parameter can be defined as average dissimilarity of i to all other points of A . For any other cluster B different from A , the average dissimilarity $d(i,B)$ of i to all points of B can be calculated. The lowest value of $d(i,B)$ can be found and it is denoted as $b(i)$. The $s(i)$ parameter is described by the following relationship:

$$(1) \quad s(i) = \frac{b(i) - a(i)}{\max[a(i), b(i)]}$$

For each i -th single point in cluster A it is $s(i) = 0$. The $s(i)$ values are limited in the range from -1 to 1 . If $s(i)$ is close to 1 , the i -th point assignment to cluster A is well justified. When $s(i)$ is about 0 , it is not clear at all whether i should be assigned to cluster A or to B . The negative, close to -1 values of $s(i)$ suppose bad assignment of i -th point to cluster A . The $s(i)$ values can be computed for all points in data set. For a given number of clusters k , the mean value s can be computed. Interpretation of computations results was based on s parameter values. The silhouette coefficient (SC) is defined as the maximal value of all s computed for $k = 2, 3, \dots, n - 1$, where n is the number of points. Subjective interpretation can be assigned to silhouette coefficient value. For SC lower than 0.26 , no substantial structure in data can be supposed. Weak and probably artificial structures can be observed for SC in the range $0.26 - 0.50$. A reasonable structure is supposed for SC between 0.51 and 0.7 and a strong structures for $SC > 0.71$.

The aim of our computations was to find reliable cluster structures in data. To gain this aim two ap-

proaches were introduced. The first of them involved the selection of clustering variables and the second one was to find the best number of clusters.

To find physicochemical parameters which might produce clusters in data occurring as a result of their interaction with ¹³⁷Cs activities, additional data sets were created. Each of them was composed of column containing transformed a_r values and columns containing transformed values of physicochemical parameters combinations. Data sets composed of 2 up to 8 variables were used in computations (127 data sets were examined).

We presumed that a big number of clusters would be difficult to interpret, therefore it did not exceed 8. For each data set and for assumed number of clusters, the s parameter values were computed. Our interpretation of its values was the same like for the SC parameter. In computations the partition around medoids (PAM) algorithm (function “pam” in library “cluster”) was used.

PCA is widely used in multivariate analysis. This method involves calculation of the eigenvalue or singular value decomposition of covariance or correlation matrix computed from data. This method allows to reduce the dimensionality of a data set consisting of a large number of interrelated variables or help to identify new meaningful underlying variables. Original variables are transformed to new ones, called the principal components (PC). The components of eigenvectors are called loadings and the values in PCs vectors are scores. The PCs are uncorrelated and are usually ordered so that the first few retain most of the variation present in all of the original variables [18]. Variance of scores is used to select the PCs that contain most of data variation. Graphical representation of principal components variances can be shown on a scree plot. Bars in this plot show variances of scores sorted in descending order. On the plane of 2 PCs the vectors representing projections of eigenvectors could be drawn. Normalization of eigenvector length to unity keeps all of its components in a circle of unit radius in the variables factor map. Length of a variable representing vector shows the contribution of this variable to the 2 PCs considered. Angles between directions of vectors may also deliver information about mutual relationships. Long, almost parallel vectors suppose good covariability between appropriate variables, while perpendicular vector directions may indicate lack of covariability. Examination of relationships between variables, represented initially by short vectors, can be moved to other PC coordinates. Usually, only a few of the PCs with biggest variances can be considered in data interpretation.

Results and discussion

In Table 1 activities and relative activities of ¹³⁷Cs and physicochemical properties of Of horizon samples are presented.

Table 2 shows the main statistical parameters of the results obtained. In this table min is the lowest value in data, Q_1 is lower quartile, Q_2 is median, Q_3 is upper quartile, max is the highest value, x is arithmetic mean, SD is standard deviation, RSD is relative standard de-

Table 1. Activities and relative activities of ^{137}Cs and physicochemical properties of Of horizon samples

No	a (kBq/kg d.m.)	a_r	pH _w	pH _{KCl}	OM (%)	σ ($\mu\text{S/cm}$)	Hh (mmol/kg)	S (mmol/kg)	T (mmol/kg)
1	3.97	0.662	6.07	5.19	91.3	431	100.6	95.4	196.0
2	1.21	0.214	3.90	3.70	67.2	154	74.0	30.2	104.2
3	2.33	0.350	6.15	5.23	76.3	239	82.3	90.0	172.3
4	1.96	0.365	6.28	5.21	78.9	240	65.4	55.0	120.4
5	1.90	0.377	6.04	5.15	79.2	244	37.8	63.8	101.6
6	1.57	0.275	6.14	5.25	73.2	258	35.9	59.8	95.7
7	0.63	0.185	6.01	5.11	67.4	157	34.9	58.7	93.6
8	1.06	0.278	6.11	5.21	77.4	265	35.3	58.7	94.0
9	1.56	0.255	4.44	3.92	76.4	267	52.0	24.0	76.0
10	2.05	0.448	4.03	3.32	77.4	266	72.0	16.4	88.4
11	2.54	0.390	4.66	4.00	79.3	287	93.2	75.5	168.7
12	1.96	0.358	3.58	3.04	79.5	294	86.0	1.2	87.2
13	2.51	0.459	3.62	3.10	77.5	296	78.0	78.7	156.7
14	1.49	0.333	4.18	3.28	72.9	281	72.0	50.3	122.3
15	0.60	0.260	4.74	4.06	69.8	140	73.0	9.3	82.3
16	0.79	0.435	4.24	4.10	70.1	121	22.8	17.6	40.4
17	0.84	0.535	3.98	3.76	70.5	183	22.5	16.2	38.7
18	0.53	0.653	4.20	4.01	68.7	180	23.5	16.3	39.8
19	0.34	0.364	4.10	3.87	67.4	132	23.4	16.7	40.1
20	0.97	0.250	3.60	2.70	71.2	167	42.4	16.7	59.1
21	1.40	0.434	4.03	3.39	78.7	153	62.1	59.2	121.3
22	1.01	0.357	4.21	3.52	77.7	159	51.4	16.7	68.1
23	1.06	0.463	4.34	3.67	78.9	158	64.2	15.3	79.5
24	0.08	0.647	6.07	5.19	61.2	163	34.8	61.2	96.0
25	0.09	0.700	6.02	4.87	63.6	169	57.6	44.0	101.6
26	0.08	0.449	6.38	5.22	63.4	168	34.4	37.6	72.0
27	0.10	0.485	6.13	5.34	65.1	174	38.0	47.8	85.8
28	0.05	0.261	4.92	4.36	59.2	176	66.8	39.8	106.6
29	0.03	0.113	4.91	4.27	55.1	177	61.2	25.8	87.0
30	0.13	0.487	5.12	4.15	74.1	154	68.4	42.2	110.6
31	0.04	0.228	5.05	4.03	75.6	155	64.0	40.8	104.8
32	0.06	0.446	4.55	3.58	65.3	159	77.6	15.6	93.2
33	0.71	0.355	4.31	3.60	77.8	167	52.7	16.9	69.6
34	2.31	0.701	3.61	2.59	73.8	264	170.4	15.8	186.2
35	1.34	0.291	3.89	2.82	71.8	166	139.2	19.0	158.2
36	2.31	0.641	3.81	2.57	80.3	265	166.8	19.8	186.6
37	1.06	0.275	4.14	3.64	80.0	240	77.8	11.8	89.6
38	1.11	0.259	3.95	3.88	67.0	201	72.5	33.1	105.6
39	1.87	0.363	3.95	3.88	79.3	201	72.4	32.2	104.6

viation (the x and SD values ratio), g_1 and g_2 are skewness and kurtosis of distribution, respectively.

The values of skewness and kurtosis shown in Table 2 indicate deviations of variables distribution from

the normal one. To avoid misinterpretation of statistical computations results our data were transformed using the Box-Cox formula. If the original random variable is x and the transformed one is x' , the Box-Cox for-

Table 2. Statistical parameters of relative ^{137}Cs activity and physicochemical parameters of horizon samples. In rows 1–7 the units of parameters are the same like in Table 1, respectively. Parameters in rows 8–10 have no units

	a_r	pH _w	pH _{KCl}	OM	σ	Hh	S	T
Min*	0.113	3.58	2.57	55.1	121	22.5	1.2	38.7
Q_1 *	0.275	4.01	3.55	67.4	159	37.9	16.7	80.9
Q_2 *	0.364	4.34	3.92	73.8	177	64.2	32.2	95.7
Q_3 *	0.461	6.02	4.99	78.3	261	75.8	56.8	115.5
Max*	0.701	6.38	5.34	91.3	431	170.4	95.4	196.0
\bar{x} *	0.395	4.76	4.05	72.8	207	65.6	37.1	102.7
SD*	0.150	0.95	0.84	7.20	64	34.3	23.9	40.8
RSD	0.379	0.200	0.208	0.099	0.307	0.523	0.646	0.397
g_1	0.52	0.51	0.10	-0.24	1.18	1.37	0.68	0.65
g_2	-0.55	-1.36	-1.10	0.01	1.61	2.11	-0.54	-0.16

* data from [32].

Table 3. Values of the *w* parameters found for relative cesium activities and physicochemical parameters of Of soil horizon [32]

	<i>a_r</i>	pH _w	pH _{KCl}	OM	σ	Hh	S	T
<i>w</i>	0.378	-1.521	0.567	1.800	-0.948	0.000	0.415	0.348

mula can be expressed by the following relationships (Eq. (2a)–(2b)):

$$(2a) \quad x' = (x^w - 1) / w \quad \text{for } w \neq 0$$

and

$$(2b) \quad x' = \log x \quad \text{for } w = 0$$

The exponents *w* found for each variable value were utilized for *x'* calculation. Table 3 shows the values of *w* parameter found previously [32].

To verify transformation to normality effectiveness for the transformed variables the confidence levels of Shapiro-Francia (α_{SF}) and Lilliefors (α_L) tests were computed. Normal distributions of *a_r'* ($\alpha_{SF} = 0.362$, $\alpha_L = 0.626$), OM' ($\alpha_{SF} = 0.074$, $\alpha_L = 0.154$), Hh' ($\alpha_{SF} = 0.114$, $\alpha_L = 0.089$), T' ($\alpha_{SF} = 0.097$, $\alpha_L = 0.093$) were confirmed. Normality of σ' ($\alpha_{SF} = 0.056$, $\alpha_L = 0.017$) and S' ($\alpha_{SF} = 0.103$ and $\alpha_L = 0.035$) distributions was somewhat suspicious because of low α_L values. Distributions of pH_w' ($\alpha_{SF} = 0.014$ and $\alpha_L = 0.003$) and pH_{KCl}' ($\alpha_{SF} = 0.047$ and $\alpha_L = 0.029$) could not be assumed to be normal. The transformed variables were used in further computations, though doubtful normality of transformed acidity may weaken relevance of conclusions drawn from computations.

Graphical representation of transformed variables are shown in a box plot (Fig. 1). Each column in data frame was standardized using the equation:

$$(3) \quad x'_{is} = \frac{x'_i - x'}{SD'}$$

where *x'_{is}* is the standardized value of *x'_i*, *x'* is the transformed variables mean value and SD' is their standard deviation. Lower base of the rectangle is a lower quartile, upper base is an upper quartile and a hori-

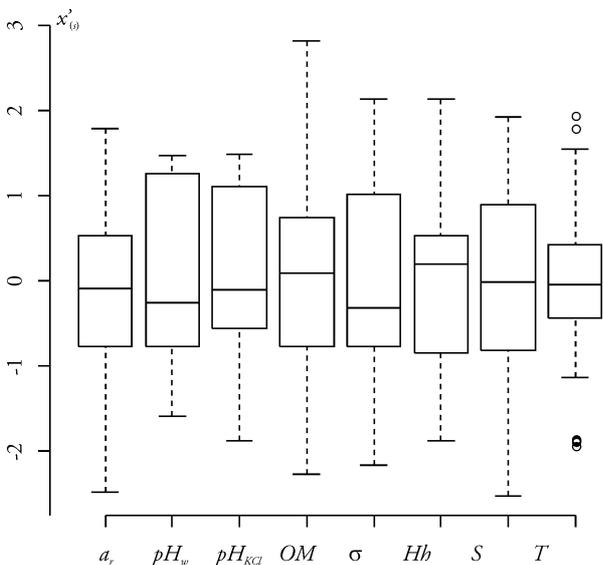


Fig. 1. Box plots of transformed and standardized values of *a_r'* and physicochemical soil parameters.

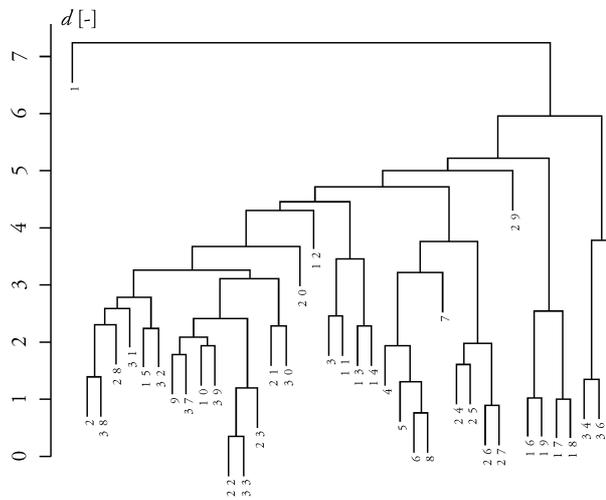


Fig. 2. Dendrogram constructed by agglomerative nesting procedure.

zontal line dividing the rectangle is a median. Whiskers are formed by connecting the formed box with short horizontal lines drawn for quantile *q* = 0.95 (upper whisker) and quantile 0.05 (lower whisker).

It should be noticed that variables distributions in Fig. 1 are not considerably different from each other. The distributions are nearly symmetrical in respect of medians, whose values are close to 0.

To assess cluster existence in data the agglomerative and divisive methods were employed. Figure 2 shows the dendrogram constructed by agglomerative nesting procedure for all of parameters considered, i.e. *a_r'*, pH_w, pH_{KCl}, OM', σ' , Hh', S' and T'. Number of a branch on dendrogram corresponds to sample number in Table 1.

Figure 3 shows the dendrogram constructed for the same parameters as the one shown in Fig. 2 but using the divisive method.

It should be noticed that cluster composition and their order depend on method utilized for construction. But some clusters, particularly the small ones,

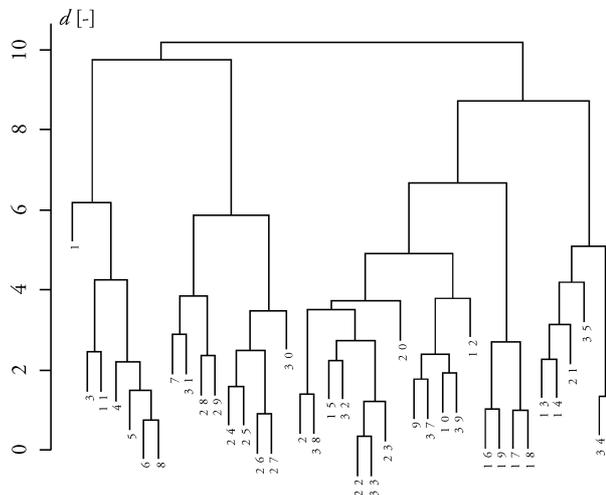


Fig. 3. Dendrogram constructed by divisive clustering.

Table 4. Correlation coefficients between transformed variables

	a'_r	pH'_w	pH'_{KCl}	OM'	σ'	Hh'	S'
pH'_w	-0.015						
pH'_{KCl}	-0.046	0.937					
OM'	0.222	-0.145	-0.172				
σ'	0.151	0.000	-0.016	0.568			
Hh'	0.053	-0.310	-0.474	0.365	0.436		
S'	0.060	0.616	0.604	0.129	0.304	-0.024	
T'	0.139	0.073	-0.074	0.391	0.574	0.829	0.505

Table 5. Results of eigenvalues decomposition and statistical parameters of ordered PCs

	PC1	PC2	PC3	PC4	PC5	PC6	PC7	PC8
a'_r	0.144	0.062	0.841	-0.509	-0.072	-0.033	0.023	-0.045
pH'_w	-0.243	0.532	-0.029	-0.058	0.063	<u>-0.479</u>	-0.648	-0.013
pH'_{KCl}	<u>-0.307</u>	0.512	0.027	0.078	-0.031	-0.292	0.742	-0.004
OM'	0.400	0.097	0.329	0.589	0.601	-0.115	0.005	0.020
σ'	0.416	0.237	0.093	<u>0.409</u>	-0.766	-0.004	-0.072	-0.045
Hh'	0.516	-0.030	-0.320	<u>-0.314</u>	0.089	<u>-0.384</u>	0.128	-0.600
S'	0.042	0.553	-0.076	-0.088	0.160	0.723	-0.062	-0.356
T'	0.473	<u>0.280</u>	-0.258	<u>-0.332</u>	0.087	0.028	0.067	0.713
s^2_{PC}	2.837	2.569	1.034	0.761	0.388	0.354	0.041	0.015
s^2_{PCr}	0.355	0.321	0.129	0.095	0.049	0.044	0.005	0.002
s^2_{PCc}	0.355	0.676	0.805	0.900	0.949	0.993	0.998	1.000

have the same structures independently of clustering method used. It is observed for four element clusters (e.g. elements 4, 5, 6, 8 and 9, 10, 37, 39 and 16, 17, 18, 19), as well as for two element clusters (e.g. 3, 11 and 24, 25, and 26, 27 and 34, 36). Structure of bigger clusters depends strongly on whether agglomerative or divisive procedure was used. Strong influence of clustering method utilized on resulting structures is probably an effect of overall weak clustering in data.

More detailed analysis was performed using the PAM method. But in all of data sets no reliable structures in variables containing the a'_r parameters were revealed. The biggest s values computed for a number of clusters from 2 to 8 were limited in the range 0.43–0.47. But this observation confirms possible usefulness of methods based on the assumption of common relationship between parameters. In the first step, the correlations coefficients r between transformed variables were computed. In Table 4 the r values are shown.

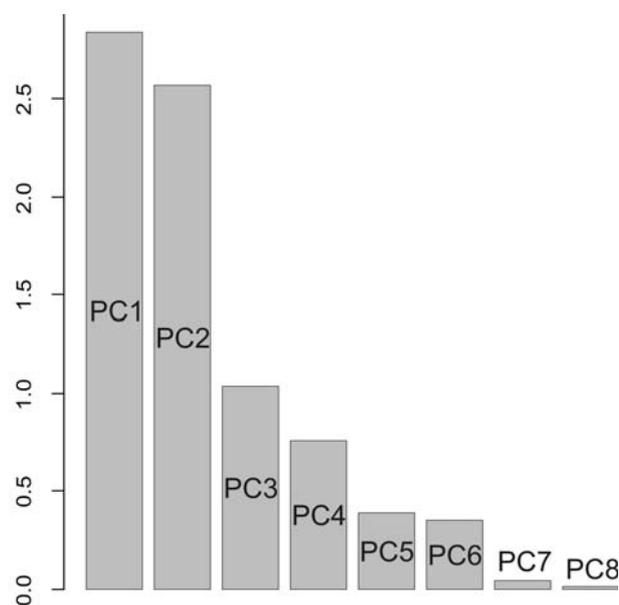
No significant correlation between a'_r and physico-chemical parameters were found. The best correlation exists between pH'_{KCl} and pH'_w and between Hh' and T' . The S parameter is well correlated both with pH'_{KCl} and pH'_w .

Table 5 shows the results of eigenvalues decomposition of the data standardized using formula shown in Eq. (2). PC1 to PC8 are principal component loadings, s^2_{PC} is the variance of the principal components scores, s^2_{PCr} is the ratio (proportion) of the PC's variance and sum of all variances, s^2_{PCc} is the cumulative proportion of PC's variance. Bold style was used to mark components whose absolute values are not smaller than 0.75 of the biggest PC component absolute value. The PC components whose absolute values are smaller than 0.75, but bigger than 0.5 of the biggest PC component absolute value were underlined.

Variances of scores are shown in the scree plot in Fig. 4.

The scree plot suppose that the first six components, containing above 99% of the total data variability, could be considered in evaluation of mutual relationships in data. However, the data from Table 4 show that the first four PCs cover 90% of the total data variability. Without a significant loss of information only the first four components can be considered. The PC1 component is determined by Hh' , T' , σ' and OM' parameters. It might be supposed that PC1 describes sorption properties and ion concentration in soil horizon. The PC2 component is determined by acid-base soil properties. The third component includes ^{137}Cs activity. No other parameters are considerably represented in PC3. Organic matter content and ^{137}Cs activity are the main elements in PC4.

In Fig. 5 the variables factor map of PC1 and PC2 is shown.

**Fig. 4.** Scree plot of score variances.

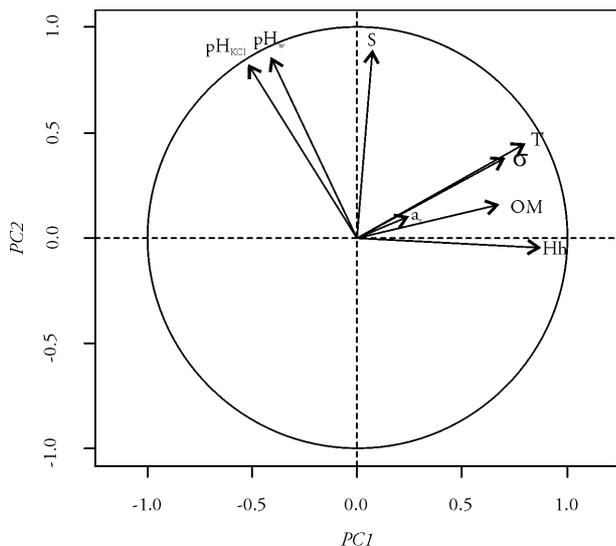


Fig. 5. Variables factor map of PC1 and PC2 principal components.

On the plain formed by the most variate principal components the long vectors representing most of the parameters are present. The shortest vector represents a_r . Lack of covariation of a_r and other parameters in this projection can be concluded.

In Fig. 6 the variables factor map of PC2 and PC3 is shown.

In the plane determined by PC2 and PC3 vector representing a_r , is long, but nearly perpendicular to directions of S and vectors representing soil acidity. It supposes lack of covariability between this variables.

In Fig. 7 the variables factor map of PC3 and PC4 is shown.

The longest vector in Fig. 7 represents a_r . Vectors representing the remaining parameters are shorter than a_r and nearly perpendicular to its direction. No relationship between a_r and other parameters can be supposed.

Analysis of computation results suppose no significant relationships between ¹³⁷Cs activity and Of horizon physicochemical parameters. However, this conclusion is not ultimate. Values of RSD in Table 2 show that

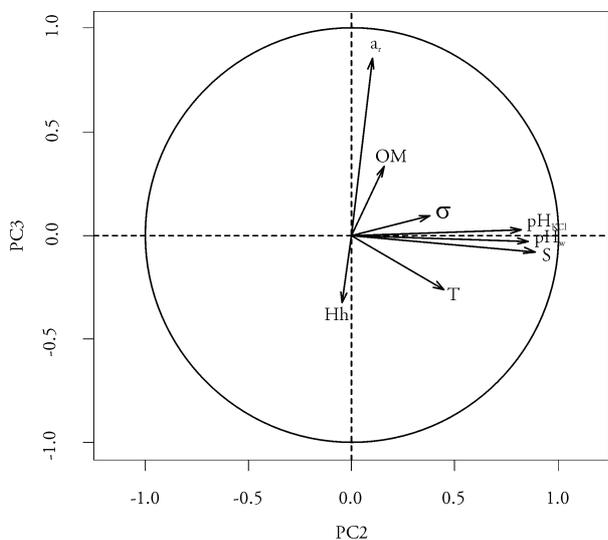


Fig. 6. Variables factor map of PC2 and PC3 principal components.

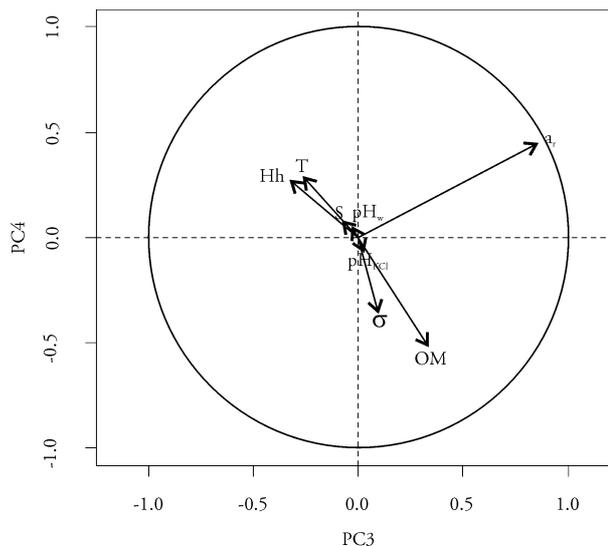


Fig. 7. Variables factor map of PC3 and PC4 principal components.

variability in a_r is rather big comparing to variabilities in some other parameters. A certain horizon property can actually influence Cs⁺ accumulation, but because of low parameter variation this effect might not be observed. Particularly, if a relationship between parameters values is weak or moderate, the influence of explanatory variable might be masked by random deviations in its value. May be for this reasons the influence of acidity or organic matter content on ¹³⁷Cs accumulation have to be considered as statistically insignificant.

There is no good evidence for clustering in the data. It might be supposed that the soil samples were collected at places in which additional factors did not significantly affect horizon's properties. Since PCA results depend on covariances or correlations values, interpretation of the results obtained for clustered data may lead to confusing conclusions. If points representing results of measurements are grouped, e.g. in 2 clusters, then the computed correlation coefficient might be high enough to accept false conjecture on linear dependence between variables. None or weak clustering in our data reduced risk of PCA results misinterpretation.

Conclusions

No clear clustering tendencies in our data were found. Points representing the data formed rather continuous structure in space of the parameters investigated. No reliable linear or linearizable relationships between ¹³⁷Cs activity and physicochemical soil parameters were found. Utilization of PCA for the experimental results interpretation led to similar conclusions. However, in spite of Box-Cox transformation some of the data (particularly soil acidity) remained not normally distributed. As an effect of non-normality, the results of our computations and the conclusions drawn could be affected strongly enough to conceal an existing, but weak relationship between ¹³⁷Cs activity and physicochemical soil parameters.

The transformed physicochemical parameters in Of horizon are more or less related with each other. As

it could be expected the pH^{w} and pH^{KCl} parameters were the most correlated ones. But because of Box-Cox transformation relationships between physicochemical parameters were essentially not linear.

References

- Beli M, Sansone U, Menegon S (1994) Behaviour of radiocaesium in a forest in the eastern Italian Alps. *Sci Total Environ* 157:257–260
- Berg MT, Shuman LJ (1995) A three-dimensional stochastic model of the behavior of radionuclides in forest. *Ecol Modelling* 83:359–372
- Biernacka M, Koczyński A, Sosińska A (2004) Systematic measurements of gamma radiation background and of radioactive contamination of the ground. In: Report of Central Laboratory of Radiological Protection 2002–2003. CLOR, Warsaw, pp 64–66
- Block J, Pimpl M (1990) Cycling of radiocesium in two forest ecosystems in the state of Rhineland-Palatinate. Elsevier, London and New York, pp 450–458
- Bunzl K, Schimmack W (1989) Effect of the microbial biomass reduction by gamma-irradiation on the sorption of Cs-137, Sr-85, Ce-139, Co-59, Cd-109, Zm-65, Ru-103, Tc-103 and I-131 by soil. *Radiat Environ Biophys* 27:165–176
- Communique II.411.K.S.96 (1996) Soil contamination in communes of Opole Voivodship. Państwowa Inspekcja Ochrony Środowiska/Wojewódzki Inspektorat Ochrony Środowiska, Opole, Poland (in Polish)
- Cremers A, Elsen A, De Preter P, Maes A (1988) Quantitative analysis of radiocaesium retention in soil. *Nature* 335:247–249
- De Brouwer S, Thiry Y, Myttenaere C (1994) Availability and fixation of radiocaesium in a forest brown acid soil. *Sci Total Environ* 143:183–191
- Dołhańczuk-Śródka A, Majcherczyk T, Ziembik Z, Smuda M, Waclawek M (2006) Spatial ^{137}Cs distribution in forest soil. *Nukleonika* 51;S2:S69–S79
- Dołhańczuk-Śródka A, Ziembik Z, Majcherczyk T, Smuda M, Waclawek M, Waclawek W (2006) Factors influencing perpendicular and horizontal translocation of ^{137}Cs in forest environment. In: Pachocki K (ed) Chernobyl – 20 years later: environment and food contamination, health effects. Nuclear power in Poland: pros and cons. Polskie Towarzystwo Badań Radiacyjnych, Zakopane, pp 427–435 (in Polish)
- Dołhańczuk-Śródka A, Ziembik Z, Waclawek M, Waclawek W (2006) Research of radiocaesium activity in the Opole Anomaly area and in the Czech Republic. *Environ Eng Sci* 23;4:642–649
- El-Reefy HI, Sharshar T, Zaghoul R, Badran HM (2006) Distribution of gamma-ray emitting radionuclides in the environment of Burullus Lake: I. Soils and vegetations. *J Environ Radioact* 87:148–169
- El Samad O, Zahraman K, Baydoun R, Nasreddine M (2007) Analysis of radiocaesium in the Lebanese soil one decade after the Chernobyl accident. *J Environ Radioact* 92:72–79
- Giannakopoulou F, Haidouti C, Chronopoulou A, Gasparatos D (2007) Sorption behavior of cesium on various soils under different pH levels. *J Hazard Mater* 149:553–556
- Handl J, Sachse R, Jakob D *et al.* (2008) Accumulation of ^{137}Cs in Brazilian soils and its transfer to plants under different climatic conditions. *J Environ Radioact* 99:271–287
- ISO (1995) Soil quality – sampling. Part 4: Guidance on the procedure for investigations of natural, near natural and cultivated sites. ISO/DIS 10381-4. International Standard Organization, Geneva
- Jagiela J, Biernacka M, Grabowski D, Henschke J (1996) Changes in environmental radiological situation in Poland during 10 years period after Chernobyl breakdown. Państwowa Inspekcja Ochrony Środowiska, Warsaw (in Polish)
- Jolliffe IT (2002) Principal component analysis. Springer Verlag, New York
- Kaufman L, Rousseeuw PJ (2005) Finding groups in data. An introduction to cluster analysis. John Wiley & Sons, Inc., Hoboken, New Jersey
- Korobova E, Linnik V, Chizhikova N (2008) The history of the Chernobyl ^{137}Cs contamination of the flood plain soils and its relation to physical and chemical properties of the soil horizons (a case study). *J Geochem Explor* 96:236–255
- Kruyt N, Delvaux B (2002) Soil organic horizons as a major source for radiocesium biorecycling in forest ecosystems. *J Environ Radioact* 58:175–190
- Nakamaru Y, Ishikawa N, Tagami K, Uchida S (2007) Role of soil organic matter in the mobility of radiocesium in agricultural soils common in Japan. *Colloids Surf A* 306:111–117
- Nimis PL (1996) Radiocesium in plants forest ecosystems. *Stud Geobot* 15:3–49
- Ostrowska A, Gawliński S, Szczubiałka Z (1999) Methods of analysis and assessment of soil and plants properties. The catalog. Institute of Environmental Protection, Warsaw (in Polish)
- R Development Core Team (2008) R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna. <http://www.R-project.org>
- Rogowski A, Tamura T (1970) Erosional behavior of cesium-137. *Health Phys* 18:446–477
- Schuller P, Handl J, Trumper RE (1988) Dependence of the ^{137}Cs soil-to-plant transfer factor on soil parameters. *Health Phys* 5:3–15
- Squire HM, Middleton LJ (1966) Behaviour of ^{137}Cs in soils and pastures: a long-term experiment. *Radiat Bot* 6:413–423
- Takenaka Ch, Ondaa Y, Hamajima Y (1998) Distribution of cesium-137 in Japanese forest soils: correlation with the contents of organic carbon. *Sci Total Environ* 222:193–199
- Waclawek M, Dołhańczuk-Śródka A, Waclawek W (2004) Radioisotopes in environment. In: Pathways of pollutants and mitigation strategies of their impact on the ecosystems. Monografie Komitetu Inżynierii Środowiska. Vol. 27. Państwowe Wydawnictwo Naukowe, Lublin, pp 245–256
- Zhiyanski M, Bech J, Sokolovska M, Lucot E, Bech J, Badot PM (2008) Cs-137 distribution in forest floor and surface soil layers from two mountainous regions in Bulgaria. *J Geochem Explor* 96:256–266
- Ziembik Z, Dołhańczuk-Śródka A, Waclawek M (2009) Multiple regression model application for assessment of soil properties influence on ^{137}Cs accumulation in forest soils. *Water, Air Soil Pollut* 198:219–232