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Assessment of ^{137}Cs and ^{239,240}Pu Distribution in Forest Soils of the Opole Anomaly

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Abstract Distribution of ^{137}Cs and ^{239,240}Pu in the forest soils horizons of the Opole Anomaly was established. Gamma and alpha spectrometry was used for determination of these isotopes. It was found that the ^{137}Cs activity was approx. 1,000 times higher than that of ^{239,240}Pu. The highest activities of both radioisotopes were found close to the boundary region in soil profile where the organic horizon turns into the inorganic one. Cluster analysis did not clearly indicate the group's existence in data in respect to ^{137}Cs and ^{239,240}Pu activities and organic matter content. Distributions of ^{137}Cs and ^{239,240}Pu in soil horizons were non-normal but similar to each other. These distributions were substantially different from that one for organic matter content. The data were separated into two groups, for organic and inorganic soil horizons, respectively. Data transformation using Box–Cox formula was performed following by standardization. Mutual relationships between variables were investigated using ordinary and robust regression methods. Good correlation between ^{137}Cs and ^{239,240}Pu was found. No significant relationship between organic matter content and radioisotopes activity was asserted.

Keywords ^{137}Cs · ^{239,240}Pu · Forest soil · Robust linear regression

1 Introduction

Starting from the 1950s and 1960s of the twentieth century, radioactive dust, originating from nuclear weapon tested in the atmosphere, gave rise to the increased interest in radioactive isotope study in the environment. Radioactive fallout contained a number of radionuclides, among them ^{137}Cs and ^{239,240}Pu. Studies focused generally on the permissible dose for men and its influence on human health (UNSCEAR 2000a, 2006).

During the Chernobyl nuclear power plant breakdown in April 26th 1986, a large amount of radioactive matter was exhausted to the atmosphere, among them ^{137}Cs, ^{239}Pu, and ^{240}Pu, which are the most interesting ones as the long-life isotopes (UNSCEAR 2000b). Estimated initial ^{137}Cs activity in the damaged reactor core ranged from 2.6×10^{17} to 3.0×10^{17} Bq. About 33±10% of this amount was released to the atmosphere. Estimated activities of ^{239}Pu and ^{240}Pu in the reactor core were 8.5×10^{14}–

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9.5×10^{14}Bq ^{239}Pu and 1.2×10^{15}–1.8×10^{14}Bq ^{240}Pu. About 1.4±0.5% of plutonium isotopes were released, mainly to the reactor surroundings (Kashparov et al. 2003).

The isotopes introduced to the environment as radioactive fallout have started, depending on their chemical nature, circulation between various alive and abiotic components of natural environment. In forest, the intercepted radioactive dust was relocated to the forest floor by the processes of leaf fall or by rain leaching the material from the canopy (throughfall). Migration of radionuclides in soil depends on numerous and various parameters, among others, on soil type, physicochemical properties of soil horizons or chemical properties of soil solution (Odintsov et al. 2005). The distribution of deposited element in soil in a given area is influenced by amount of initial fallout, its physicochemical properties as well as horizontal and vertical migration rate in soil. Determination of radioisotope activity in vertical profile layers enable to calculate a migration rate. The factors that determine changes of radionuclide activity in soil are, among others: weather conditions, moisture content in soil, duration of radioactive fallout, soil structure, and water infiltration rate (Block and Pimpl 1990; Bunzl and Schimmack 1989; Nimis 1996).

The ^{137}Cs accumulation in soil is mainly caused by chemisorption processes and usually follows the general principles of metals sorption in natural or artificial ion exchangers (Berg and Shuman 1995) and depends mainly on the charge of an ion and its size, including hydration sphere (Janusz et al. 1997). Numerous experiments proved that the size of ^{137}Cs ion exchange depends upon the mineral particle size, structure, the ionic strength in the soil solution or the presence of the competing ions, especially K^+ and NH_4^+ (for example: Korobova et al. 2008). It was found that clay components of the organic horizons in forests soils caused decreasing in the available ^{137}Cs fraction (e.g., de Brouwer et al. 1994). Though organic soil horizons do not contain enough clay minerals to immobilize cesium, many authors (Beli et al. 1994; Takenaka et al. 1998; Nakamaru et al. 2007; El Samad et al. 2007; El-Reefy et al. 2006; Zhiyanski et al. 2008; Wacławek et al. 2004; Dołhańczuk-Śródka et al. 2006a, 2006b, 2006c) reported the highest ^{137}Cs activities just in upper horizons, rich in organic matter. A reason of Cs^+ retention in organic horizons could be explained by formation of certain complexes between cesium ions and humic acids (Macášek 1999). There is a hypothesis which suggested that organic matter modifies the adsorption properties of clay minerals in soil (El-Reefy 2006). It was found that low pH values improve cesium uptake from soil by plants (Kruyts and Delvaux 2002; Squire and Middleton 1966; Schuller et al. 1988) but decreases velocity of the Cs^+ cation transfer (Schuller et al. 1988). Type of clay and a sand content in mineral horizons may also influence the Cs transport parameters (Rogowski and Tamura 1970; Cremers et al. 1988; Giannakopoulou et al. 2007).

Plutonium behavior in environment is much more complicated than that for cesium. The most stable oxidation state for Pu ions in solution is +4, although some amounts of plutonium in its +3, +5, and +6 oxidation states can also exist simultaneously. The Pu chemistry in water is further complicated by the successive hydrolysis of Pu^{4+} compounds to form polymers of colloidal dimensions. It was found that the behavior of Pu cations in the soil–water system depends on the speciation and soil type (Skipperud 2000). In forest soils, both soluble and insoluble plutonium compounds remain in top layers (Bunzl 1998; Druteikienė 1999; Lujanienė 2002). The main reason of plutonium compounds retention in organic soil horizons are associations between humic substances and Pu ions (Fujikawa 1999; Macášek 1999; Sokolik 2003). It was found that besides organic matter Pu was also bound with Fe/Mn oxides (e.g., Komosa 2002). Transport of Pu in soil is influenced by physicochemical properties of soil and soil solution. The negative correlation between the exchangeable pH of soil and plutonium concentration in forest soils was found (Komosa 1999). It was also found that both oxidation and reduction mechanisms can play an important role in Pu transport through the vadose zone (Kaplan 2004).

One of the most important transport mechanisms of elements in the environment involves their uptake by living organisms. A considerably simplified description of this process includes transport of elements from abiotic components to autotrophic organisms and then to the heterotropic consumers within the food network system. Further transport occurs between successive higher trophic levels, e.g., from herbivores to carnivores. The presence of both ^{137}Cs and ^{239,240}Pu was confirmed in plants, for example in mushrooms (e.g., Kirchner 1998; Strandberg 2004;

Kuwahara 2005; Baeza 2006), lichen and mosses (e.g., Testa et al. 1998; Mietelski et al. 2000; Kłos et al. 2006), and green plants (Frissel et al. 2002; Dushenkov 2003; Sokolik et al. 2004). These radioisotopes were also found in animal organisms. Presence of ^{137}Cs and ^{239,240}Pu in different insects species was affirmed (Copplestone et al. 1999; Mietelski et al. 2004) as well as in small mammals (Copplestone et al. 1999; Mietelski 2003) and carnivores (Mietelski et al. 2006, 2008).

In Poland, some areas of increased radiocesium activity resulting from the Chernobyl fallout were observed. The highest activities were noted in southwestern 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 (Communique 1996; Jagielak 1996). In February and in March 1986, the mean beta activity in total fallout in this region was 13 Bq/m^2. On April 29th, the beta activity reached 100 Bq/m^2 and on May 1st the activity exceeded 10^5 Bq/m^2 (VSES 2008). In 2007, there were still areas of ^{137}Cs surface activities exceeding 70 kBq/m^2 in some forests on the region of the Opole Anomaly (Dołhańczuk-Śródka et al. 2007).

Determination of elements concentrations in particular constituents of environment is a crucial issue for description of their translocation mechanisms. However, information about other parameters of the system is indispensable. Among others, climate type, season of the year, local vegetation, soil type and its physicochemical properties as well as presence of certain bacteria and fungi species may affect elements circulation in ecosystems (Tamponnet et al. 2008). Unfortunately, it is usually not obvious which of the parameters play a key role in migration of element in the ecosystem considered.

A big number of factors affecting translocation of an element in woodland ecosystems significantly hampers modeling of this process. Transport of elements is influenced by a number of mutually independent or interrelated processes and depends on many parameters related to the current and former state of the environment. An insight in nature of these processes might be gained by analysis of the results received from statistical methods applied to the data obtained for different environmental components. It can be expected that proper statistical processing of the experimental data might reveal some of their specific features. These observations might be helpful in migration model designing creation.

In this, paper the results of ^{137}Cs and ^{239,240}Pu activities determination in forest soil horizons collected on the area of the Opole Anomaly were described. Because of supposed organic compounds importance in Cs and Pu ions retention, for each sample of horizon, the organic matter content OM was determined. The soil samples were collected in the sites located in the same forest complex. Vegetation at these sites was similar. All samples were collected within a few days in October. This approach enables reduction of variability in some parameters (e.g., soil humidity, or seasonal changes in vegetation) that might affect ^{137}Cs and ^{239,240}Pu migration in forest ecosystem.

Chemometric methods were used for examination of the results obtained.

2 Materials and Methods

2.1 Sampling Locations

Samples were collected in an area of the Opole Anomaly in October 2006. They were taken from Bory Stobrawskie forest, located in the north-east direction from Opole (PL). The sampling sites were located close to Zagwiździe (50°52′25″ N, 17°58′31″ E), Rzędów (50°44′46″ N, 18°08′38″ E), Staniszcze W. (50°38′55″ N, 18°20′56″ E), Olesno (50°52′31″ N, 18°24′54″ E), and Szumirad (50°50′20″ N, 18°14′36″ E). The soil samples were taken in the vicinity of at least 20-year-old trees, not less than 100 m from the roads. The ground surrounding the trees was in large part covered by fallen leaves and few forest bed plants were growing.

2.2 Sampling Methods

The consecutive soil layers were carefully taken starting from forest litter and finishing at approx. 30–35 cm depth to obtain sample of soil horizons. In our study, the samples of the horizons: Ol, Of, Oh, A, Ees, Bbr, and C were collected. Not all of the horizons appeared in each soil sample, usually a single profile, were composed of four to five horizons. The horizon sample was dried in 105°C until constant mass of the sample were recorded. Dry samples were manually cleaned to remove pieces of wood, cones, stones, and other intrusion bodies. This material was crumbled until no bodies on the 5-mm mesh sieve remained. The material obtained was kept in a tightly closed container.

2.3 ^{137}Cs and ^{239,240}Pu Activity Measurements

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 (Canberra) of high resolution: 1.29 keV (FWHM) at 662 keV and 1.70 keV (FWHM) at 1332 keV. Relative efficiency was 21.7%. Energy and efficiency calibration of the gamma spectrometer was performed with the standard solutions MBSS 2 (Czech Metrological Institute, Praha), which covers an energy range from 59.54 keV to 1836.06 keV. Geometry of calibration source was a Marinelli beaker (volume of 447.7± 4.48 cm^3), with a density of 0.985±0.01 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 the 450 cm^3 Marinelli beaker. The measuring process and analysis of spectra were computer controlled with use of the software GENIE 2000 (Canberra). The spectra were recorded for 24 h. Counting uncertainty did not exceed 4%.

Determination of plutonium in environmental samples requires separation of these isotopes in a pure form and fixing them on a steel plate, which enables alpha spectrometric measurements. This is realized by a radiochemical multi-stage procedure. In the first stage of this procedure, a dried and ashed (450–500°C) sample was leached with 6 M HCl. From the obtained solution, trace elements were co-precipitated with iron (III) hydroxide by 25% NH_4OH addition. The precipitate was dissolved in 6 M HCl and then separated from Fe by co-precipitation with calcium oxalate at pH=3. Next, the precipitate was dried, burned in an oven, dissolved in 12 M HCl and then a second co-precipitation with iron (III) hydroxide was performed. In the next stage, the precipitate was dissolved in 8 M HNO_3, boiled on a sand bath with $NaNO_2$ until nitric oxides disappeared. This stage allows changing the oxidation state of plutonium to the Pu^{4+} form. The solution was introduced on an ion-exchange column filled with Dowex 1×8 (50–100 mesh). Before sample introduction, the column was washed with 8 M HNO_3 in order to exchange chloride ions on nitrate ions. After the sample was passed through the column, the next solutions were introduced as follows: 8 M HNO_3, 6 M HCl, and concentrated HCl in order to eliminate trace elements (Th, Am) which disturb Pu determination. In the final stage of anion exchange, Pu was eluted from the column using concentrated HCl with 0.1 M NH_4I, which changed the plutonium oxidation stage to +3. The obtained solution was evaporated to dryness with aqua regia to eliminate iodine and ammonium salts. Finally, Pu was electrodeposited onto stainless steel disks from 0.4 M ammonium oxalate with 0.3 M HCl and measured by alpha spectrometry (Komosa 1996).

Measurements were performed using the 7401 Canberra Alpha Spectrometer with a 1520 mixer/router and S-100 multichannel analyzer. The PIPS detector of 17 keV FWHM resolution was used. For quantitative analysis, the Canberra Genie-2000 software was used. Chemical yield was monitored by addition of a standard ^{242}Pu solution to the sample. Measurements were performed usually during 600,000 s and radiochemical yield varied from 60 to 100%.

3 Computations

The area on which the samples were collected was not contaminated uniformly. It was obvious that location of the place from which the samples were taken affects its total ^{137}Cs or ^{239,240}Pu activity. To omit or, at least, to diminish the effect of unequal initial soil contamination, the relative activities a_r were calculated. The activities of isotope considered 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 mutual relationships between isotope concentrations in soil horizons than the activities on their own.

For statistical computations, the R language (R Development Core Team 2008) 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 used. This method allows to assign objects to different groups, so that the data in each subset share some common traits. For our computation, the functions provided by the “cluster” library of R were used. Functions available in this library were described by Kaufman and Rousseeuw

(2005). For each variable, the 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.

The overviews of cluster existence in data were shown on dendrograms—tree diagrams illustrating the arrangement of the clusters. Diagrams were constructed using "agnes" and "diana" functions from "cluster" library. Function "agnes" uses agglomerative nesting procedure, while "diana" realizes divisive hierarchical clustering.

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:

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

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 $\bar{s}$ parameter values. The silhouette coefficient SC is defined as the maximal value of all $\bar{s}$ computed for k=2, 3 ... $n-1$, where n is 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 0.70 and a strong structures for SC>0.71 (Kaufman et al. 2005).

It was found that distributions of our data considerably differ from each other. This may cause problems in interpretation of significance test results. To avoid this problem, the data were separated into two groups and variables were transformed to make their distributions similar to the normal one. For this purpose the Box–Cox transformation was used. If the original random variable is x and the transformed one is x_t, the Box–Cox formula can be expressed by the following relationships (Eq. 2a–b):

$$x_t = (x^m - 1)/m \text{ for m} \neq 0 \tag{2a}$$

and

$$x_t = \log(x) \text{ for m} = 0 \tag{2b}$$

where m is the dimensionless exponent.

The exponents found for each column of data frame were utilized for x' computations.

Normality of variable distribution was verified using one sample Kolmogorov–Smirnov test.

For computations the transformed and standardized values of variables were taken. The data were standardized using the following formula:

$$x_{\text{st}} = \frac{x - \overline{x}}{\sigma} \tag{3}$$

The existence of interrelations between variables was investigated. For this purpose, ordinary linear regression (OLS) and robust regression methods were chosen. Preliminary analysis of our results revealed possible problems due to data dispersion. In our computations, the robust regression methods were used, which are forms of weighted least squares regression. In robust regression, the linear equation $y=x\beta+\varepsilon$ is considered. To find estimators b of β, the following approach can be applied. Assuming a scaled probability distribution function $f(e/s)/s$ of ε and setting $\rho=-\log f$, the maximum likelihood estimator (MLE) minimizes

$$\sum_{i=1}^{n} \rho\left(\frac{y_i - x_i b}{s}\right) + n \log s \tag{4}$$

The MLE b of β solves

$$\sum_{i=1}^{n} x_i \psi\left(\frac{y_i - x_i b}{s}\right) = 0 \tag{5}$$

where Ψ is derivative of ρ with respect to b.

Equation 5 can be solved by iterated re-weighted least squares. For s value estimation, the median absolute deviation (MAD) was used. The Hampel, Huber, and Tukey (bisquare) M-estimators (MLE-like) were used in data processing. Each of these estimators is additionally described by its own parameters, but in our computations, we used the default values from function "rlm" (from "MASS" library in R language).

The M-estimation methods are robust to outliers in the response variable but are not resistant to outliers in the explanatory ones. The S-estimation was proposed to solve this problem, the method highly resistant to leverage points. But this method was also found to be inefficient. Advantages of M and S estimations were joined in MM-estimation.

Each estimator assigns a weight to residual e. For OLS, the weights w_{OLS} of each residual are the same ($w_{\mathrm{OLS}}=1$). Huber weights w_{H} are described by the following relationships (Eq. 6a–b):

$$w_{\mathrm{H}} = 1 \text{ for } |e| \leq k \tag{6a}$$

and

$$w_{\mathrm{H}} = \frac{k}{|e|} \text{ for } |e| > k \tag{6b}$$

In Huber weighting, observations with small residuals get a weight of 1. To the residuals bigger than k, decreasing with e weights are assigned.

The Tukey weights w_{T} are described by the relationships (Eq. 7a–b):

$$w_{\mathrm{T}} = \left[1 - \left(\frac{e}{k}\right)^2\right]^2 \text{ for } |e| \leq k \tag{7a}$$

and

$$w_{\mathrm{T}} = 0 \text{ for } |e| > k \tag{7b}$$

In biweighting, all observations with a non-zero and smaller than k residuals are down-weighted. Cases with e bigger than k are removed (Rousseeuw 1987; Venables and Ripley 2002; Fox 2002).

4 Results

In Table 1 statistical parameters of ^{137}Cs activities a_{Cs}, ^{239,240}Pu activities a_{Pu} and organic matter contents OM are listed. Activities a_{Cs} and a_{Pu} are expressed in kBq/kg and Bq/kg, respectively. OM is expressed in weight %. The following parameters of our data are shown: minimal value (min), lower quartile (q_1), median (ME), upper quartile (q_3), maximal value (max), mean value ($\bar{x}$), standard deviation (SD), skewness (g_1), kurtosis (g_2), and relative variability coefficient (ratio of difference between maximal and minimal values to mean value, RVC).

The ^{239,240}Pu activities were approx. 1,000 lower than ^{137}Cs activities. For the isotopes investigated, the differences between mean and median as well as g_1 and g_2 values were similar, supposing similar and non-normal type of variables distribution. Values of the same parameters for OM also indicate non-normal variable distribution, but its type is dissimilar to the isotopes distributions. The relative variability coefficients of a_{Cs} and a_{Pu} were similar too and they were approx. 2.5 times bigger than RVC for OM.

In Fig. 1, the box plots showing ^{137}Cs relative activities distribution in soil horizons are drawn. In this plots lower base of the rectangle is a lower quartile, upper base is an upper quartile and a horizontal 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).

Figure 1 shows that the highest ^{137}Cs relative activities were found in the Oh and A horizons.

Figure 2 shows the box plots of ^{239,240}Pu distribution in soil horizons.

The highest ^{239,240}Pu relative activities were found in the A horizon.

High activities of the investigated isotopes were observed in the lower organic horizons and in the upper inorganic. The plots in Figs. 1 and 2 indicate

Table 1 Parameters of ^{137}Cs activities a_{Cs} [kBq/kg], ^{239,240}Pu activities a_{Pu} [Bq/kg] and organic matter content OM [%]

	min	q_1	ME	q_3	max	$\bar{x}$	SD	g_1	g_2	RVC
a_{Cs}	0.003	0.056	0.187	0.779	2.867	0.578	0.836	1.71	1.83	4.95
a_{Pu}	0.009	0.028	0.127	0.579	2.059	0.453	0.613	1.52	1.14	4.53
OM	1.36	3.62	57.05	84.58	97.31	50.34	38.89	−0.19	−1.79	1.91

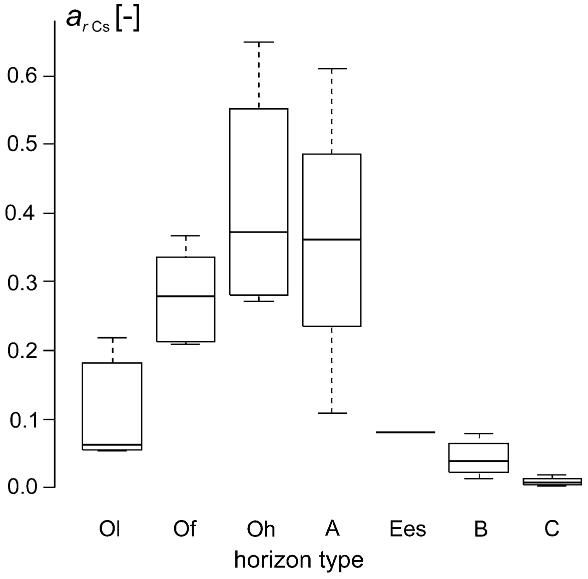

Fig. 1 Boxplots of a_{rCs} in soil horizons and subhorizons

quite well maximal a_r in the soil region where organic horizon turns into inorganic one.

Figure 3 shows the organic matter distribution in soil horizons.

As it could be expected, organic matter content continuously decreases downward the soil profile.

Investigation of clustering in data was performed for activities of radionuclides and organic matter content. Positions of points were determined by a_{rCs}, a_{rPu}, and OM variables, so that cluster formation was investigated in 3D space of parameters. Application of both agglomerative and divisive methods leads to similar structure of dendrogram. Figure 4 shows the dendrogram constructed using divisive algorithm. Additionally the horizon or subhorizon type was marked at each point.

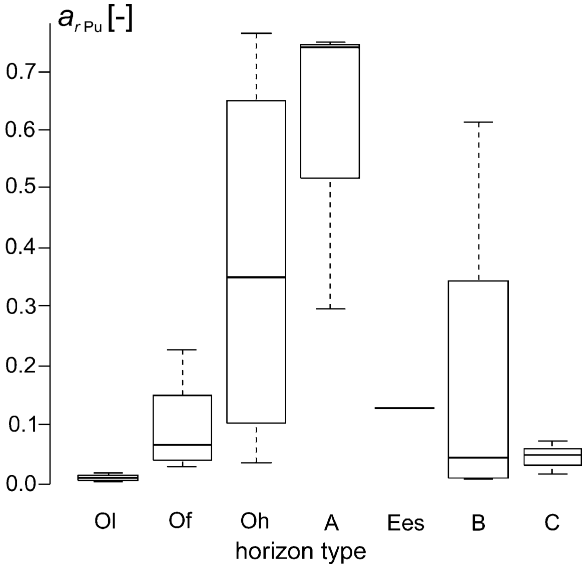

Fig. 2 Boxplots of a_{rPu} in soil horizons and subhorizons

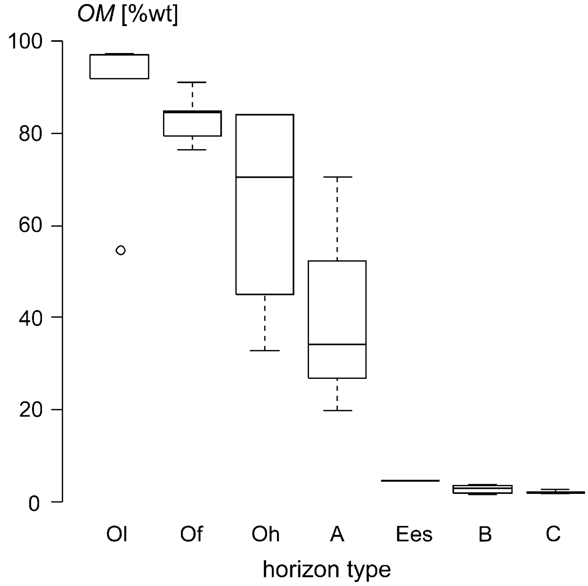

Fig. 3 Boxplots of organic matter content in soil horizons and subhorizons

A tendency to group formation based on subhorizon or horizon type can be observed. Most of the points representing organic subhorizons are located in

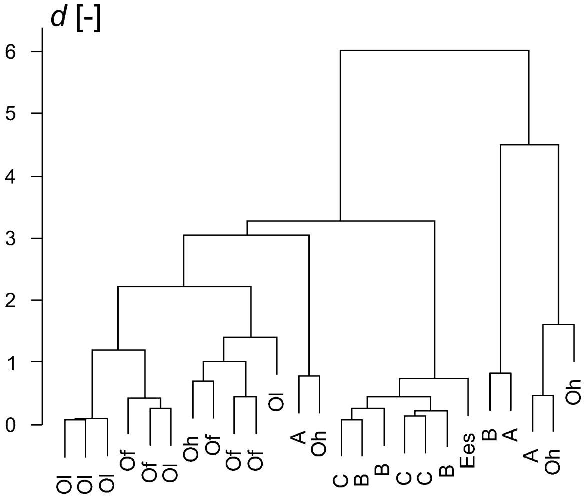

Fig. 4 Dendrogram of structures appearing in data comprising a_{rCs}, a_{rPu}, and OM

structures on the left side of dendrogram whereas the inorganic ones are on the right side.

To examine whether clustering in data is apparent or true, the SC coefficient was determined. Figure 5 shows the $\bar{s}$ parameter dependence on number of clusters k.

The SC value of 0.639 was found for seven clusters. These results supposes existence of reasonable (but not strong) structure in the data. But for $k=4$, 6 to 9 the $\bar{s}$ values are comprised in the range from 0.600 to 0.628. These values are only a bit smaller than SC. Taking this data into account, it is rather difficult to conclude that the certain number of clusters indicates well the structures existence in the data. It might be supposed that points with a_{rCs}, a_{rPu}, and OM coordinates tend to form some structures in respect to soil horizon or subhorizon type. But these structures are rather fuzzy and they may partly overlap with the nearest neighbors. For analysis of such kind of data, the fuzzy clustering methods might be utilized. These methods are used in many areas of environmental sciences. Among others, they are helpful in e.g., water, air, and soil pollution assessment (e.g., Lehn and Temme 1996; Chang et al. 2001; Argiriou et al. 2004; Grande et al. 2005; Ocampo-Duque et al. 2007) or assessment of toxic substances influence on the environment (e.g., Melcher and Matthies 1996; Friedrichs et al. 1996; Sârbu and Pop 2000; Salski 2007).

In Fig. 6, density of a_{rCs} distribution is shown. Position of individual points is marked with vertical dashes above the horizontal axis. In the upper part of the plot description of soil horizon or subhorizon, the type is shown for each point. No clear relationship between horizon type and point location on histogram can be observed. The distribution is positively skewed.

In Fig. 7, density of a_{rPu} distribution is shown. Similar to a_{rCs} distribution, no clear relationship

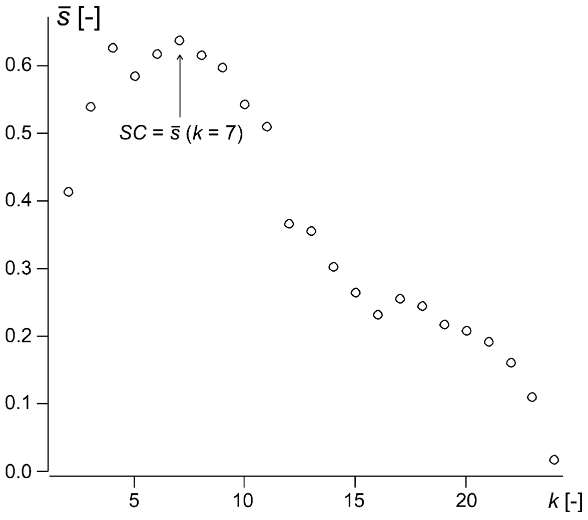

Fig. 5 Mean s dependence on cluster number in data comprising a_{rCs}, a_{rPu}, and OM

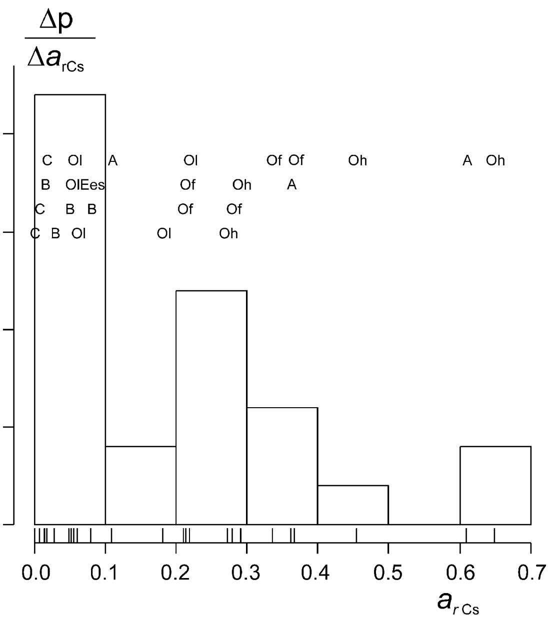

Fig. 6 Density of a_{rCs} distribution in soil horizons and subhorizons

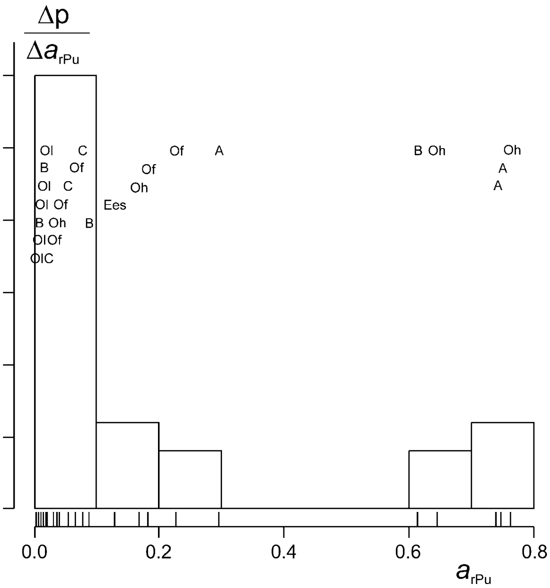

Fig. 7 Density of a_{rPu} distribution in soil horizons and subhorizons

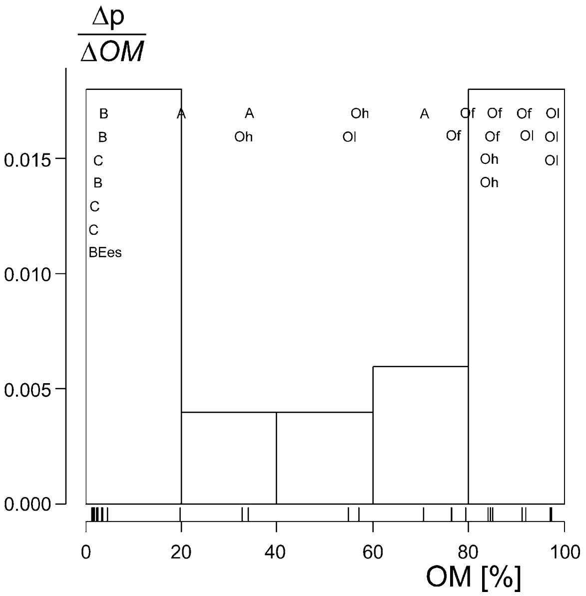

Fig. 8 Density of OM distribution in soil horizons and subhorizons

between horizon type and point location in the histogram can be observed. The distribution seems to be positively skewed, but density of points in the right side of the histogram is bigger and in the middle part of histogram is smaller than the ones for a_{rCs} distribution.

The OM distribution shown in Fig. 8 is antimodal. The left side of the histogram is formed by points representing inorganic soil horizons while the right side contains points representing organic soil horizon. It can be concluded that OM distributions in inorganic and organic soil horizons are substantially different.

We would like to examine possible co-variabilities that may occur between variables. Linear relationship is the simplest one and its credibility can be assessed (considering some assumptions) by correlation coefficient *r*. Linear transformation of one variable into the second implies similar distribution of both variables. Though distributions of a_{rCs} and a_{rPu} are somewhat similar, both of them are essentially different from OM distribution. The shape of the histogram in Fig. 8 indicates necessity of OM data separation into two groups. The first group contains data from organic horizon and the second comprises data from inorganic horizon. Before further computations, the data were separated in two data frames containing a_{rCs}, a_{rPu}, and OM values for organic and inorganic horizons. To avoid problems in interpretation of significant test results, we have decided to transform the variables to make their distributions similar to the normal one. For this purpose, the Box–Cox transformation was used (Eq. 2a–b). In Table 2, the computed exponent *m* values and their standard errors, as well as mean values and standard deviations of transformed variables are shown.

The values of transformed variables were standardized using Eq. 3. After standardization, the data concerning the relative activities and organic matter content in organic and inorganic horizons were joined together. Transformed and standardized ^{137}Cs relative activities from the O horizon were connected with those from inorganic horizons. The same operations were carried out for ^{239,240}Pu relative activities and for organic matter content. All operations performed did not change data arrangement in rows of data frame. In this way, the parameters investigated became continuous again through organic and inorganic soil horizons. The new a_{rtCs}, a_{rtPu}, and OM_t variables have mean values close to 0 and standard deviations close to 1. Normality of data distribution was examined with Shapiro–Wilk test. The lowest confidence level $\alpha_{SW}=0.11$ was computed for a_{rtPu}. The computations results show that normality of a_{rtCs}, a_{rtPu}, and OM_t distributions cannot be rejected regarding usually accepted critical $\alpha_{SW}=0.05$.

Existence of groups in the data may cause false evidence of relationship existence, e.g., results of

Table 2 The values of *m* exponent and their standard errors SE_m, mean values and standard deviations SD_{ar} calculated for transformed a_{rCs}, a_{rPu}, and OM in organic and inorganic horizons

	a_{rCs}				a_{rPu}				OM			
	m	SE_m	$\bar{x}$	SD_{ar}	m	SE_m	$\bar{x}$	SD_{ar}	m	SE_m	$\bar{x}$	SD_{OM}
Organic horizon O (Ol, Of, Oh)	0.48	0.35	−1.05	0.34	−0.04	0.17	−3.3	1.9	3.2	1.2	4.4×10^5	2.3×10^5
Inorganic horizons (A, B, C, Ees)	0.08	0.16	−2.7	1.3	0.13	0.23	−1.07	0.52	−0.58	0.31	0.90	0.43

Table 3 Parameters of linear equation computed using robust regression methods

b_0	SD_{b0}	b_1	SD_{b1}	Estimation type	M-estimator
0.02	0.13	0.76	0.14	M	Tukey
0.02	0.14	0.76	0.15	M	Huber
0.00	0.13	0.77	0.13	M	Hampel
0.03	0.13	0.75	0.14	MM	–
0.00	0.13	0.77	0.13	OLS	–

computations using data in which two separated clusters of points can be recognized, might indicate false linear relationship between variables. But the results from CA showed rather poor grouping tendency in our data. It might be supposed that regression methods can be applied with a low risk of false interpretation of the computation results.

Linear correlation coefficients r between a_{rtCs}, a_{rtPu}, and $\mathrm{OM_t}$ and their confidence levels α_r were computed. Coefficient of correlation between a_{rtCs} and a_{rtPu} was 0.774 (α_r=0.00), and this was the highest value among the computed ones. Correlations between a_{rtCs}, a_{rtPu}, and $\mathrm{OM_t}$ were negligible. For a_{rtCs} and $\mathrm{OM_t}$ the r value was 0.038 (α_r=0.86). For a_{rtPu} and $\mathrm{OM_t}$, the r value was 0.053 (α_r=0.80).

Parameters of linear equation $a_{\mathrm{rtCs}} = b_0 + b_1 \times a_{\mathrm{rtPu}}$ describing relationship between ^{137}Cs and ^{239,240}Pu activities were computed using robust regression and OLS methods. In Table 3, values of the b_0 and b_1 parameters, their standard deviations SD, scaling factor s, estimation method, and M-estimator types are shown.

In Fig. 9, the plots of the relationship between a_{rtCs} and a_{rtPu} as well as lines representing fitted models are shown.

The lines presenting results from Huber and Tukey M-estimators nearly overlap with each other. For this reason, in Fig. 9, a single line is drawn to show results obtained from this methods. Utilization of the Hampel estimator and OLS method deliver the same b_0 and b_1 values, so, in Fig. 9, a single line represents both approaches.

Regardless of the estimation method or M-estimator type, the values of b_0 and b_1 parameters were similar. Comparison of b_0 and SD_{b0} values suppose that the intercept value is 0 in population. Linear relationship between variables can be simplified to ratio of transformed ^{137}Cs and ^{239,240}Pu relative activities.

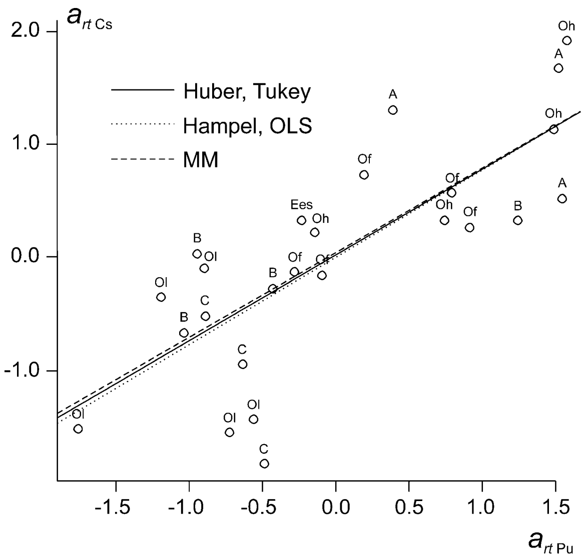

Fig. 9 Relationship between a_{rtCs} and a_{rtPu}

Figure 10 show weights assigned to consecutive points from the plot in Fig. 9. For the Hampel estimator, all weights were 1, like in OLS. That is why the results computed in this regression models were the same for both methods.

Organic matter content influence on ^{137}Cs and ^{239,240}Pu accumulation was investigated. While very low values of correlation coefficient between OM and ^{137}Cs or ^{239,240}Pu were computed, one could suppose

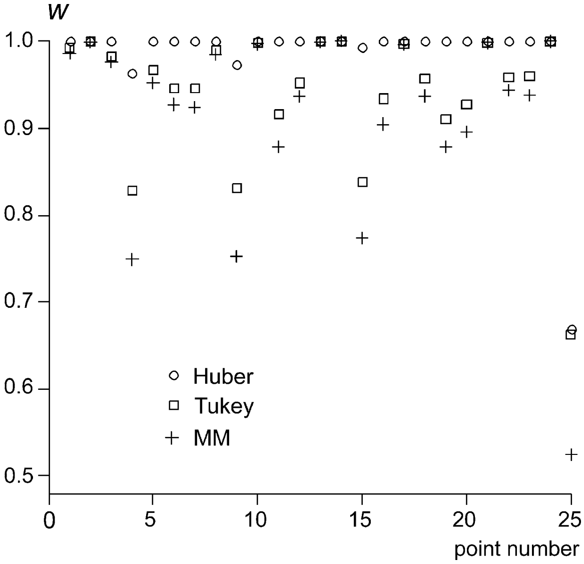

Fig. 10 The weights w assigned to consecutive points from the plot in Fig. 9

that it might be a result of outstanding OM observations. To verify this presumption, the robust regression methods were used to find linear relationships between OM_t and a_{rtCs} as well as OM_t and a_{rtPu}. But the computed slope values and their standard deviations did not allow to accept significancy of organic matter content influence on the investigated isotopes activity. The computed t statistics values for slopes did not exceed 0.32. The hypothesis that in population values of the slopes were 0 cannot be rejected. This result does not confirm conclusions of some authors mentioned in the "Introduction" section in this paper. However, absorption of Cs and Pu cations is determined not by organic matter generally but rather by some of its components. Concentration of certain compounds, originated from decaying of organic matter and affecting Cs and Pu cations retention, may depend on climate, plant species growing on the area, fungi, and microorganism activities and many others factors. Earlier investigation carried out on the area of the Opole Anomaly showed that ^{137}Cs activity only in the Ees horizon depends on organic matter content (Ziembik et al. 2009). It was also shown that common influence of two or more soil parameters is needed to affect significantly the ^{137}Cs activity in soil horizon. It is possible that Pu ions transport in soil also depends on more than only OM parameter. This conclusion can be a starting point for further investigations.

5 Conclusions

In our, samples the ^{137}Cs activity was about one thousand higher than ^{239,240}Pu activity. For both isotope activities, similar relative variability coefficients were computed. The RVC of organic matter content was approx. 2.5 times lower than the ones for ^{137}Cs and ^{239,240}Pu activities.

The CA method applied to points with a_{Cs}, a_{Pu}, and OM coordinates show a tendency to group formation based on subhorizon and horizon type. However, the clusters were not well separated, presumably partly overlapping with nearest neighbors. Poor tendency to cluster formation in the data allowed us to utilize regression methods in further computations.

The distributions of a_{rCs}, a_{rPu}, and OM were investigated. It was found that distributions of a_{rCs} and a_{rPu} were similar, whereas distribution of OM was substantially different. For further computations the data were divided into two groups. The first group comprised the data from O horizon and the second contains data from inorganic soil horizons. The data were transformed using the Box–Cox formula then standardized, and data from organic and inorganic horizons were joined together.

The co-variability of transformed variables was investigated using OLS and robust regression methods. No significant differences between results obtained from these methods were found. The linear dependence between a_{rtCs} and a_{rtPu} was affirmed. But due to variables transformation, this relationship was not essentially linear. No relationships between a_{rtCs} and OM_t as well as a_{rtPu} and OM_t were found.

References

Argiriou, A. A., Kassomenos, P. A., & Lykoudis, S. P. (2004). On the methods for the delimitation of seasons. *Water Air and Soil Pollution Focus, 4*, 65–74. doi:10.1023/B:WAFO.0000044787.71076.38.

Baeza, A., Guillén, F. J., Salas, A., & Manjón, J. L. (2006). Distribution of radionuclides in different parts of a mushroom: Influence of the degree of maturity. *Science of the Total Environment, 359*, 255–266. doi:10.1016/j.scitotenv.2005.05.015.

Beli, M., Sansone, U., & Menegon, S. (1994). Behaviour of radiocaesium in a forest in the easter Italian Alps. *Science of the Total Environment, 157*, 257–260. doi:10.1016/0048-9697(94)90587-8.

Berg, M. T., & Shuman, L. J. (1995). A three-dimensional stochastic model of the behavior of radionuclides in forest. *Ecological Modelling, 83*, 359–372. doi:10.1016/0304-3800(94)00104-3.

Block, J., & Pimpl, M. (1990). *Cycling of radiocesium in two forest ecosystems in the state of Rhineland-Palatinate* (pp. 450–458). London: Elsevier.

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. *Radiation and Environmental Biophysics, 27*, 165–176. doi:10.1007/BF01214606.

Bunzl, K., Kracke, W., Schimmack, W., & Zelles, L. (1998). Forms of fallout of 137Cs and 239, 240Pu in successive horizons of a forest soil. *Journal of Environmental Radioactivity, 39*, 55–68. doi:10.1016/S0265-931X(97)00042-8.

Chang, N.-B., Chen, H. W., & Ning, S. K. (2001). Identification of river water quality using the fuzzy synthetic evaluation approach. *Journal of Environmental Management, 63*, 293–305. doi:10.1006/jema.2001.0483.

Communique II.411.K.S.96 (1996). *Soil contamination in communes of Opole Voivodship*. Opole: WIOŚ/PIOŚ (in Polish).

Copplestone, D., Johnson, M. S., Jones, S. R., Toal, M. E., & Jackson, D. (1999). Radionuclide behaviour and transport in a coniferous woodland ecosystem: vegetation, invertebrates and wood mice, *Apodemus sylvaticus*. *Science of the Total Environment, 239*, 95–109. doi:10.1016/S0048-9697(99)00294-6.

Cremers, A., Elsen, A., De Preter, P., & Maes, A. (1988). Quantitative analysis of radiocaesium retention in soil. *Nature, 335*, 247–249. doi:10.1038/335247a0.

De Brouwer, S., Thiry, Y., & Myttenaere, C. (1994). Availability and fixation of radiocaesium in a forest brown acid soil. *Science of the Total Environment, 143*, 183–191. doi:10.1016/0048-9697(94)90456-1.

Dołhańczuk-Śródka, A., Majcherczyk, T., Ziembik, Z., Smuda, M., & Wacławek, M. (2006a). Spatial ^{137}Cs distribution in forest soil. *Nukleonika, 51*(Suppl. 2), 69–79.

Dołhańczuk-Śródka, A., Ziembik, Z., Majcherczyk, T., Smuda, M., Wacławek, M., & Wacławek, W. (2006b). Factors influencing vertical and horizontal translocation of ^{137}Cs in fores environment. In: K. Pachocki (red.), *Chernobyl – 20 Years After: Contamination of Environment and Food, Health Effects. Nuclear Energy in Poland: pro and con* (pp. 427-435). Published by Polish Radiation Research Society, Zakopane 2006 (in Polish). ISBN 83-89379-66-X.

Dołhańczuk-Śródka, A., Ziembik, Z., Wacławek, M., & Wacławek, W. (2006c). Research of radiocaesium activity in the Opole Anomaly area and in the Czech Republic. *Environmental Engineering Science, 23*, 642–649. doi:10.1089/ees.2006.23.642.

Dołhańczuk-Śródka, A., Ziembik, Z., Wacławek, M., & Hyšplerova, L.(2007). *Radiocesium Activity in the Polish-Czech Border Region.* Published by Society of Ecological Chemistry and Engineering. ISBN 978-83-917511-5-2.

Druteikienė, R., Lukšienė, B., & Holm, E. (1999). Migration of ^{239}Pu in soluble and insoluble forms in soil. *Journal of Radioanalytical and Nuclear Chemistry, 242*, 731–737. doi:10.1007/BF02347387.

Dushenkov, S. (2003). Trends in phytoremediation of radionuclides. *Plant and Soil, 249*, 167–175. doi:10.1023/A:1022527207359.

El Samad, O., Zahraman, K., Baydoun, R., & Nasreddine, M. (2007). Analysis of radiocaesium in the Lebanese soil one decade after the Chernobyl accident. *Journal of Environmental Radioactivity, 92*, 72–79. doi:10.1016/j.jenvrad.2006.09.008.

El-Reefy, H. I., Sharshar, T., Zaghloul, R., & Badran, H. M. (2006). Distribution of gamma-ray emitting radionuclides in the environment of Burullus Lake: I. Soils and vegetations. *Journal of Environmental Radioactivity, 87*, 148–169. doi:10.1016/j.jenvrad.2005.11.006.

Friedrichs, M., Fränzle, O., & Salski, A. (1996). Application of fuzzy clustering to data dealing with phytotoxicity. *Ecological Modelling, 85*, 27–40. doi:10.1016/0304-3800(95)00009-7.

Frissel, M. J., Deb, D. L., Fathony, M., Lin, Y. M., Mollah, A. S., & Ngo, N. T. (2002). Generic values for soil-to-plant transfer factors of radiocesium. *Environmental Radioactivity, 58*, 113–128. doi:10.1016/S0265-931X(01)00061-3.

Fox, J. (2002). http://cran.r-project.org/doc/contrib/Fox-Companion/appendix-robust-regression.pdf. Accessed 15 January 2009.

Fujikawa, Y., Zheng, J., Cayer, I., Sugahara, M., Takigami, H., & Kudo, A. (1999). Strong association of fallout plutonium with humic and fulvic acid as compared to uranium and ^{137}Cs in Nishiyama soils from Nagasaki, Japan. *Journal of Radioanalytical and Nuclear Chemistry, 240*, 69–74. doi:10.1007/BF02349138.

Giannakopoulou, F., Haidouti, C., Chronopoulou, A., & Gasparatos, D. (2007). Sorption behavior of cesium on various soils under different pH levels. *Journal of Hazardous Materials, 149*, 553–556. doi:10.1016/j.jhazmat.2007.06.109.

Grande, J. A., Andújar, J. M., Aroba, J., de la Torrea, M. L., & Beltrán, R. (2005). Precipitation, pH and metal load in AMD river basins: an application of fuzzy clustering algorithms to the process characterization. *Journal of Environmental Monitoring, 7*, 325–334. doi:10.1039/b410795k.

Jagielak, J., Biernacka, M., Grabowski, D., & Henschke, J. (1996). *Changes in environmental radiological situation in Poland during 10 years period after Chernobyl breakdown*. Warsaw: PIOŚ. (in Polish).

Janusz, W., Kobal, I., Sworska, A., & Szczypa, J. (1997). Investigation of the electrical double layer in a metal oxide/monovalent electrolyte solution system. *Journal of Colloid and Interface Science, 187*, 381–387. doi:10.1006/jcis.1996.4690.

Kaplan, D. I., Powell, D. A., Demirkanli, D. I., Fjeld, R. A., Molz, F. J., & Serkiz, S. M. (2004). Influence of oxidation states on plutonium mobility during long-term transport through an unsaturated subsurface environment. *Environmental Science & Technology, 38*, 5053–5058. doi:10.1021/es049406s.

Kashparov, V. A., Lundin, S. M., Zvarich, S. I., Ioshchenko, V. I., Levchuk, S. E., & Khomutinin, Y. V. (2003). Soil contamination with fuel component of Chernobyl radioactive fallout. *Radiochemistry, 45*, 189–200. doi:10.1023/A:1023897612740.

Kaufman, L., & Rousseeuw, P. J. (2005). *Finding groups in data. An introduction to cluster analysis*. New York: Wiley.

Kirchner, G., & Daillant, O. (1998). Accumulation of ^{210}Pb, ^{226}Ra and radioactive cesium by fungi. *Science of the Total Environment, 222*, 63–70. doi:10.1016/S0048-9697(98)00288-5.

Kłos, A., Rajfur, M., Wacławek, M., & Wacławek, W. (2006). Use of lichens to assess local soil aerosol pollution with radiocesium-137. *Ecological Chemistry and Engineering, 13*, 833–838.

Komosa, A. (1996). Study on plutonium isotopes determination in soils from the region of Lublin (Poland). *Science of the Total Environment, 188*, 59–62. doi:10.1016/0048-9697(96)05160-1.

Komosa, A. (1999). Migration of plutonium isotopes in forest soil profiles in Lublin region (Eastern Poland). *Journal of Radioanalytical and Nuclear Chemistry, 240*, 19–24. doi:10.1007/BF02349131.

Komosa, A. (2002). Study on geochemical association of plutonium in soil using sequential extraction procedure. *Journal of Radioanalytical and Nuclear Chemistry, 252*, 121–128. doi:10.1023/A:1015252207934.

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). *Journal of Geochemical Exploration, 96*, 236–255. doi:10.1016/j.gexplo.2007.04.014.

Kruyts, N., & Delvaux, B. (2002). Soil organic horizons as a major source for radiocesium biorecycling in forest ecosystems. *Journal of Environmental Radioactivity, 58*, 175–190. doi:10.1016/S0265-931X(01)00065-0.

Kuwahara, C., Fukumoto, A., Ohsone, A., Furuya, N., Shibata, H., & Sugiyama, H. (2005). Accumulation of radiocesium in wild mushrooms collected from a Japanese forest and cesium uptake by microorganisms isolated from the mushroom-growing soils. *Science of the Total Environment, 345*, 165–173. doi:10.1016/j.scitotenv.2004.10.022.

Lehn, K., & Temme, K.-H. (1996). Fuzzy classification of sites suspected of being contaminated. *Ecological Modelling, 85*, 51–58. doi:10.1016/0304-3800(95)00014-3.

Lujanienė, G., Plukis, A., Kimtys, E., Remeikis, V., Jankünaitė, D., & Ogorodnikov, B. I. (2002). Study of ^{137}Cs, ^{90}Sr, ^{239,240}Pu, ^{238}Pu and ^{241}Am behavior in the Chernobyl soil. *Journal of Radioanalytical and Nuclear Chemistry, 251*, 59–68. doi:10.1023/A:1015185011201.

Macášek, F., Shaban, I. S., & Mátel, L. (1999). Cesium, strontium, europium(Ill) and plutonium(IV) complexes with humic acid in solution and on montmorillonite surface. *Journal of Radioanalytical and Nuclear Chemistry, 241*, 627–636. doi:10.1007/BF02347223.

Melcher, D., & Matthies, M. (1996). Application of fuzzy clustering to data dealing with phytotoxicity. *Ecological Modelling, 85*, 41–49. doi:10.1016/0304-3800(95)00010-0.

Mietelski, J. W., Gaca, P., & Olech, M. A. (2000). Radioactive contamination of lichen and mosses collected in South Shetlands and Antarctic Peninsula. *Journal of Radioanalytical and Nuclear Chemistry, 245*, 527–537. doi:10.1023/A:1006748924639.

Mietelski, J. W., Kitowski, I., Gaca, P., & Tomankiewicz, E. (2003). Elevated plutonium and americium content in skulls of small mammals. *Journal of Radioanalytical and Nuclear Chemistry, 256*, 593–594. doi:10.1023/A:1024584707374.

Mietelski, J. W., Szwałko, P., Tomankiewicz, E., Gaca, P., Małek, S., & Barszcz, J. (2004). ^{137}Cs, ^{40}K, ^{90}Sr, $^{238,\ 239+240}$Pu, ^{241}Am and $^{243+244}$Cm in forest litter and their transfer to some species of insects and plants in boreal forests: Three case studies. *Journal of Radioanalytical and Nuclear Chemistry, 262*, 645–660. doi:10.1007/s10967-004-0488-5.

Mietelski, J. W., Kitowski, I., Gaca, P., Grabowska, S., Tomankiewicz, E., & Szwałko, P. (2006). Gamma-emitters ^{90}Sr, $^{238,\ 239+240}$Pu and ^{241}Am in bones and liver of eagles from Poland. *Journal of Radioanalytical and Nuclear Chemistry, 270*, 131–135. doi:10.1007/s10967-006-0319-y.

Mietelski, J. W., Kitowski, I., Tomankiewicz, E., Gaca, P., & Błażej, S. (2008). Plutonium, americium, ^{90}Sr and ^{137}Cs in bones of red fox (*Vulpes vulpes*) from Eastern Poland. *Journal of Radioanalytical and Nuclear Chemistry, 275*, 571–577. doi:10.1007/s10967-007-7062-x.

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 and Surfaces A: Physicochemical and Engineering Aspects, 306*, 111–117. doi:10.1016/j.colsurfa.2007.01.014.

Nimis, P. L. (1996). Radiocesium in plants forest ecosystems. *Studia Geobotanica, 15*, 3–49.

Ocampo-Duque, W., Schuhmacher, M., & Domingo, J. L. (2007). A neural-fuzzy approach to classify the ecological status in surface waters. *Environmental Pollution, 148*, 634–641. doi:10.1016/j.envpol.2006.11.027.

Odintsov, A. A., Pazukhin, E. M., & Sazhenyuk, A. D. (2005). Distribution of ^{137}Cs, ^{90}Sr, $^{239+\ 240}$Pu, ^{241}Am, and ^{244}Cm among components of organic matter of soils in near exclusion zone of the chernobyl NPP. *Radiochemistry, 47*, 96–101. doi:10.1007/s11137-005-0056-z.

R Development Core Team. (2008). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. ISBN 3-900051-07-0, URL http://www.R-project.org.

Rogowski, A., & Tamura, T. (1970). Erosional behavior of cesium-137. *Health Physics, 18*, 446–477. doi:10.1097/00004032-197005000-00002.

Rousseeuw, P. J., & Leroy, A. M. (1987). *Robust regression and outlier detection*. USA: Wiley.

Salski, A. (2007). Fuzzy clustering of fuzzy ecological data. *Ecological Informatics, 2*, 262–269. doi:10.1016/j.ecoinf.2007.07.002.

Sârbu, C., & Pop, H. F. (2000). Fuzzy clustering analysis of the first 10 MEIC chemicals. *Chemosphere, 40*, 513–520. doi:10.1016/S0045-6535(99)00285-4.

Schuller, P., Handl, J., & Trumper, R. E. (1988). Dependence of the ^{137}Cs soil-to-plant transfer factor on soil parameters. *Health Physics, 5*, 3–15.

Skipperud, L., Oughton, D., & Salbu, B. (2000). The impact of Pu speciation on distribution coefficients in Mayak soil. *Science of the Total Environment, 257*, 81–93. doi:10.1016/S0048-9697(00)00443-5.

Sokolik, G. A., Ovsyannikova, S. V., & Kimlenko, I. M. (2003). Effect of humic substances on plutonium and americium speciation in soils and soil solutions. *Radiochemistry, 45*, 176–181. doi:10.1023/A:1023893511831.

Sokolik, G. A., Ovsiannikova, S. V., Ivanova, T. G., & Leinova, S. L. (2004). Soil–plant transfer of plutonium and americium in contaminated regions of Belarus after the Chernobyl catastrophe. *Environment International*, *30*, 939–947.

Squire, H. M., & Middleton, L. J. (1966). Behaviour of ^{137}Cs in soils and pastures: a long-term experiment. *Radiation Botany, 6*, 413–423. doi:10.1016/S0033-7560(66)80074-1.

Strandberg, M. (2004). Long-term trends in the uptake of radiocesium in *Rozites caperatus*. *Science of the Total Environment, 327*, 315–321. doi:10.1016/j.scitotenv.2004.01.022.

Takenaka, C., Ondaa, Y., & Hamajima, Y. (1998). Distribution of cesium-137 in Japanese forest soils: correlation with the contents of organic carbon. *Science of the Total Environment, 222*, 193–199. doi:10.1016/S0048-9697(98)00305-2.

Tamponnet, C., Martin-Garin, A., Gonze, M.-A., Parekh, N., Vallejo, R., & Sauras-Yera, T. (2008). An overview of BORIS: bioavailability of radionuclides in Soils. *Journal* *of Environmental Radioactivity, 99*, 820–830. doi:10.1016/j.jenvrad.2007.10.011.

Testa, C., Degetto, S., Jia, G., Gerdol, R., Desideri, D., & Meli, M. A. (1998). Plutonium and americium concentrations and vertical profiles in some Italian mosses used as bioindicators. *Journal of Radioanalytical and Nuclear Chemistry, 234*, 273–276. doi:10.1007/BF02389784.

UNSCEAR. (2000). Report, Sources and effects of ionizing radiation, vol. 2. http://www.unscear.org/unscear/en/publications/2000_2.html. Accessed 10 August 2008.

UNSCEAR. (2000a). Report, Annex J: Exposures and effects of the Chernobyl accident, http://www.unscear.org/docs/reports/annexj.pdf. Accessed 10 August 2008.

UNSCEAR. (2006). Report, Effects of ionizing radiation, vol. 1, http://www.unscear.org/unscear/en/publications/2006_1.html. Accessed 12 August 2008.

Venables, W. N., & Ripley, B. D. (2002). *Modern applied statistics with S*. New York: Springer.

VSES (2008). Data provided by Voivodship Sanitary and Epidemiological Station in Opole, PL.

Wacławek, M., Dołhańczuk-Śródka, A., & Wacławek, W. (2004). Radioisotopes in environment. In *Pathways of pollutants and mitigation strategies of their impact on the ecosystems* (pp. 245-256). Monographs of Environmental Engineering Committee, Polish Academy of Science, vol.27, Lublin.

Zhiyanski, M., Bech, J., Sokolovska, M., Lucot, E., Bech, J., & Badot, P. M. (2008). Cs-137 distribution in forest floor and surface soil layers from two mountainous regions in Bulgaria. *Journal of Geochemical Exploration, 96*, 256–266. doi:10.1016/j.gexplo.2007.04.010.

Ziembik, Z., Dołhańczuk-Śródka, A., & Wacławek, M. (2009). Multiple regression model application for assessment of soil properties influence on ^{137}Cs Accumulation in forest soils. *Water, Air, and Soil Pollution, 198*, 219–232. doi:10.1007/s11270-008-9840-7.