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### INVESTIGATION OF COMMITTED RADIATION DOSE RATE AND RELATIONSHIPS BETWEEN ALKALINE METALS CONCENTRATIONS IN MUSHROOM *Xerocomus badius*

### BADANIE WCHŁONIĘTEJ, SKUTECZNEJ DAWKI PROMIENIOWANIA I ZALEŻNOŚCI POMIĘDZY STĘŻENIAMI METALI ALKALICZNYCH W OWOCNIKACH Xerocomus badius

**Abstract:** The fruiting bodies of fungi sprout from mycelium are capable of accumulating significant amounts of trace elements, both metals and metalloids. Content of these elements in fruiting bodies may exceed their concentration in the substrate where fungi develop. Among the elements the radioactive nuclides are also present. In this work health risk caused by increased radioactivity dose absorbed with *Xerocomus badius* bay bolete consumption was estimated. In analysis concentrations of radioactive isotopes <sup>137</sup>Cs and <sup>40</sup>K were taken into consideration. It was found that moderate ingestion of bay bolete does not create health risk due to increased radioactive substances intake. The amount of consumed mushrooms that could deliver the dose exceeding the safe one, is rather improbable in real life. Possible relationships between radioactive isotopes concentrations and concentrations of common alkali metals were investigated using methods designed for compositional data analysis. No clear relationships between <sup>137</sup>Cs, Ca, K and Mg concentrations in samples of bay bolete were found and significant influence of outlying data points on statistical inference was noticed.

Keywords: fungi, isotopes, alkali metals

### Introduction

Fungi are organisms which form a separate kingdom of living organisms, different from plants and animals [1]. Their fruiting bodies sprout from mycelium, which colonizes and penetrates various types of substrates and absorbs numerous elements and organic substances contained in these grounds [2, 3]. They are capable of accumulating significant amounts of trace elements, both metals and metalloids, including the elements necessary for

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the organism: iron, copper, manganese, cobalt, selenium, molybdenum, chromium, as well as toxic heavy metals: cadmium, lead and mercury [4]. In many cases, the content of these elements in fungi exceeds their concentration in the substrate where fungi develop [5]. Fungi have the ability to accumulate trace elements thanks to the specific structure of mycelium, *ie* the exposed surface of vegetative cells, as well as the large surface of hyphae [4]. Fungi can absorb trace amounts of elements from soil and store them. The species of fungus, type of soil, fruiting body growth rate and its level of ripeness are of crucial importance for accumulation of heavy metals [6].

The heavy metal ion content levels in fruiting bodies are essentially different in stem and cap. The concentrations of metals are higher in the cap, which is probably the result of a higher density of hyphae [7-8]. While higher metal concentration is observed in younger fruiting bodies, it decreases with the growing weight of the fruiting body [9].

The content of heavy metals in fruiting bodies depends on their level in the environment. The fruiting bodies collected in the areas contaminated with copper, lead and mercury (former coal mining sites and metal steelworks) are characterized by a significantly higher metal content in comparison with the fruiting bodies from the uncontaminated areas [10-12].

The metal content in fungi has been a subject of research for scientists such as Grzybek, Witusik, Lasota, Szefer and Falandysz [13-18]. The results of research carried out by both Polish and foreign authors indicate that metals are cumulated in fungi by bonding with proteins (metallothioneins) [8, 18, 19].

Numerous tests have shown that fungi are also good biomonitors of radioactive contaminations. Radionuclides present in the environment are absorbed both from soil through mycelium, and directly through the whole fruiting body surface [20]. The examination of <sup>137</sup>Cs content in fungi demonstrated significant selectivity in absorbing this radionuclide from soil. The following species demonstrated the highest level of caesium absorption: poisonous (though sometimes consumed) brown roll-rim (*Paxillus involutus*), as well as the tasty and popular bay bolete (*Xerocomus badius*) [21-25]. The mechanism of absorbing caesium by bay bolete was explained by the presence in its fruiting body cap of phenyl dye (the so-called bad ion A), which complexes potassium ions and alternatively caesium ions [26].

The relations between <sup>137</sup>Cs in fungi and alkali metal concentrations were also investigated.

The research performed in laboratory conditions suggests that there are two pathways of passive transport of caesium in mycelium [27]:

- through the potassium channel, and then by means of diffusion within hyphae,

extracellular transport between mycelium hyphae to the interior of fruiting bodies.

The papers [28, 29] demonstrated a lack of connection between <sup>137</sup>Cs activity and K concentration in fungi, which is suggested by different <sup>137</sup>Cs and K absorption mechanisms. In opposite to this result the statistical analysis of the influence of alkali metals on the content of Cs-137 in fungi demonstrated a significant positive correlation between the activity of this radionuclide with Cs (r = 0.940) and Rb (r = 0.901) concentrations [29].

Apart from <sup>137</sup>Cs there are other non-radioactive isotopes of this element in the forest ecosystem, and in fact the <sup>137</sup>Cs isotope transported from soil constitutes only a small part of caesium migrating in this way. The absorption of <sup>137</sup>Cs to the fruiting body together with

dust is also possible and may cause an increase of the osmotic potential of potassium penetrating mycelium [27].

The purpose of our research is to determine the relationship between the  $^{137}$ Cs activity and the concentration of alkali metals (K, Ca and Mg), as well as to evaluate potential effective doses being a result of absorbing  $^{137}$ Cs and  $^{40}$ K together with the consumed fungus. The assessment of the relationship between the content of metal radioactive isotopes and the content of other, non-radioactive isotopes will provide information allowing for a better assessment of radioactive risk related to the consumption of fungi. The results of this research may also be helpful in modelling the circulation of metals in the forest environment.

### Materials and methods

The samples of bay bolete were collected in three areas:

- A forests in the Hradce Kralove area,
- B forests near Trebechovice pod Orebem,
- C forests of Kotlina Klodzka, the border zone between Poland and Czech Republic. Mushrooms samples were collected in four consecutive years, starting from 2002.

Samples of mushrooms were collected from the places located at least 100 m from forest edges and 200 m from roads and buildings. Distance from the tree trunk was always at least 2 m. Where it was possible, samples were taken from six sites situated close to each other, so that the total weight of the samples was about 200 g. They were cut using plastic knife to obtain slices which were dried at 298 K to constant weight. Then the material was homogenized in an agate mortar and kept in a closed plastic containers (PP, PE).

The dried and homogenized sample was mineralized in mixture of  $HNO_3$  and  $H_2O_2$ . For mineralization the microwave digestion system MLS-1200 MEGA was used, controlled by appropriate program for the decomposition of biological material. All operations in the processing of the samples were carried out in such a way that they prevent sample contamination from the surrounding.

The Ca, K and Mg concentrations in mushroom samples were determined using *Atomic Absorption Spectrometers* AAAnalyst 400 from PerkinElmer and Solaar M5, AVANTA SUM, from UNICAM. Both instruments used two methods of atomization, *Flame Atomization* AAS (FA AAS) and *Graphite Furnace* AAS (GF AAS). The level of determined concentrations of elements is mg/dm<sup>3</sup> for FA AAS and  $\mu$ g/dm<sup>3</sup> for GF AAS.

In potassium determination the standard calibration solution (ASTASOL M 001)  $1.000 \pm 0.002 \text{ g/dm}^3$  (analyst) was used. To suppress ionization of samples and standards, CsCl (MERCK) was added to the analyzed solution. In calcium and magnesium determination the standard calibration solution (ASTASOL M 002)  $1.000 \pm 0.002 \text{ g/dm}^3$  (analyst) was used. To suppress ionization of samples and standards, LaCl<sub>3</sub> (MERCK) was added to the analyzed solution.

The measurement of <sup>137</sup>Cs activity in mushrooms were 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: 21.7%. Energy and efficiency calibration of the gamma spectrometer was performed with the standard solutions type MBSS 2 (Czech Metrological Institute, Prague, CZ) which covers an energy range from 59.54 to 1836.06 keV. Geometry of calibration source was

Marinelli (447.7 ±4.48 cm<sup>3</sup>) with density 0.985 ±0.01 g/cm<sup>3</sup>, containing <sup>241</sup>Am, <sup>109</sup>Cd, <sup>139</sup>Ce, <sup>57</sup>Co, <sup>60</sup>Co, <sup>137</sup>Cs, <sup>113</sup>Sn, <sup>85</sup>Sr, <sup>88</sup>Y and <sup>203</sup> Hg. Geometry of samples container was Marinelli, 450 cm<sup>3</sup>. Measuring process and analysis of spectra were computer controlled with use of the software GENIE 2000. The radiation spectrum was recorded day and night. Concentration of <sup>40</sup>K activity was calculated from K concentration in sample. Potassium determined with AAS contains constant amount of <sup>40</sup>K, equal to 0.012%. From this concentration and half-life time (1.25 · 10<sup>9</sup> years) of <sup>40</sup>K isotope, its activity concentration was calculated.

The measurement uncertainties of Ca, K and Mg concentrations were 10%. The measurement uncertainty of  $^{137}$ Cs activity concentration was 10%, too.

## Estimation of radioactivity dose rate absorbed with ingested mushrooms and related health risk

The obtained measurement results, as well as the information published in relevant legal acts binding in Poland, were used for the assessment of risk related to the consumption of bay bolete. Pursuant to the Regulation of the Council of Ministers of 18 January 2005 on Ionizing Radiation Dose Limits (Journal of Laws No. 20, item 168) [30], the dose limit of ionizing radiation from artificial sources, expressed as the effective dose, amounts to 1 mSv in a calendar year for the total population, excluding staff professionally exposed to ionizing radiation. Such dose may be exceeded in a calendar year on the condition that its total value within the following five-year period will not exceed 5 mSv above the natural background level.

The method of calculating internal exposure doses presented in the regulation is based on the following international regulations:

- basic international standards of protection from ionizing radiation and security of radiation sources (developed under the auspices of UNO, IAEA and International Labour Organization),
- basic security standards of health protection from ionizing radiation for employees and total population (EU Council Directive 96/29/EURATOM) [31].

The indicators included in the Regulation [30] were applied in order to calculate the annual dose of internal exposure, caused by the consumption of bay bolete collected in the examined area. The loading effective dose E, being a result of a nuclide penetrating through the digestive system, was calculated from the formula:

$$E = e(g)A \tag{1}$$

where e(g) is convertion factor for persons in the age group g expressed in Sv/Bq, and A is the activity of a radionuclide, which penetrated into the organism through the digestive and respiratory systems, expressed in Bq.

Together with the consumed fungus, our organism is penetrated not only by the natural radioactive isotopes (especially  $^{40}$ K), but also by the radioactive  $^{137}$ Cs, which is accumulated in all soft tissues, and consequently removed from the organism.

The committed effective doses of  $^{137}$ Cs and  $^{40}$ K, included in Table 4 of the Regulation for the total population, related to the penetration of a nuclide with the activity concentration of 1 Bq, are shown in Table 1.

It can be noticed that with the same level of nuclide activity penetrating the organisms of a child and adult, the child will receive a bigger dose. Due to the smaller total weight, the absorbed energy for one body weight unit will be higher for a child than for an adult.

Table 1

<i>e</i> ( <i>g</i> ) [Sv/Bq]	$g \leq 1$ year	g > 1 year	g = 2-7 years	g = 7-12 years	g = 12-17 years	<i>g</i> > 17 years
<sup>137</sup> Cs	$2.1 \cdot 10^{-8}$	$1.2 \cdot 10^{-8}$	9.6·10 <sup>-9</sup>	1.0.10-8	1.3.10-8	$1.3 \cdot 10^{-8}$
<sup>40</sup> K	$6.2 \cdot 10^{-8}$	$4.2 \cdot 10^{-8}$	$2.1 \cdot 10^{-8}$	$1.3 \cdot 10^{-8}$	7.6·10 <sup>-9</sup>	$6.2 \cdot 10^{-9}$

Values of the committed effective doses of  $^{137}$ Cs and  $^{40}$ K in different age groups *g*, related to penetration of a nuclide with the activity concentration of 1 Bq [30]

Considering the fact that fungi are not an obligatory component of children's diet and are introduced only for older children, there were two age groups - 12-17 and above 17 - taken into account.

In order to calculate the committed effective dose E, resulting from the penetration of a nuclide through the digestive system (*ie* by the consumption of the examined fungi), it was assumed on the basis of EFSA data [32, 33] that the maximum fungus consumption in a longer period may reach the level of 100 g per week. This amount corresponds to the consumption of approximately 5 kg of fresh fungi per year and a higher level of consumption should only take place sporadically. Taking into consideration the fact that fresh fungi contain 90% of water, while dry fungi contain 15% of water, the adopted annual consumption level of dry fungi for further calculations of loading effective dose was determined at the level of 0.5 kg.

Table 2 presents data characterizing the distribution of committed effective dose as a result of the absorption of <sup>137</sup>Cs and <sup>40</sup>K together with the consumed fungus, for persons in a relevant age group. Minimum (*Min*) and maximum (*Max*) values, lower quartiles ( $Q_1$ ), medians ( $Q_2$ ), upper quartiles ( $Q_3$ ), arithmetic mean values (*mean*) and standard deviation (*SD*) were presented.

Table 2

E [mSv/year]	Min	Max	$Q_1$	$Q_2$	$Q_3$	mean	SD
	g = 12-17 years						
<sup>137</sup> Cs	0.003	0.016	0.005	0.006	0.009	0.008	0.005
<sup>40</sup> K	0.002	0.004	0.002	0.003	0.004	0.003	0.001
	$g \ge 17$ years						
<sup>137</sup> Cs	0.003	0.016	0.005	0.006	0.009	0.008	0.005
<sup>40</sup> K	0.001	0.004	0.002	0.002	0.003	0.002	0.001

Distribution of committed effective dose as a result of <sup>137</sup>Cs and <sup>40</sup>K absorption together with the consumed fungus, for persons in a relevant age group

The data included in the table indicate that the effective dose received together with the consumed fungus from  $^{137}$ Cs is significantly higher than the dose from  $^{40}$ K. Therefore, it can be ascertained that a diet rich in fungi may cause an increase of the received annual effective dose.

The analysis of data presented in the table allows to notice that the calculated maximum value of effective dose originating from  $^{137}$ Cs amounts to 0.016 mSv/year, which is only 1.6% of the limit dose referred to in Regulations [30].

Figure 1 presents the relationship between the consumption rate of bay boletes contaminated with <sup>137</sup>Cs and the level of this contamination, assuming that the dose of 1 mSv will not be exceeded (for conversion factor  $1.3 \cdot 10^{-8}$ ).



Fig. 1. The relationship between the consumption rate of bay boletes contaminated with  $^{137}$ Cs and the level of this contamination, assuming that the dose of 1 mSv will not be exceeded (conversion factor  $1.3 \cdot 10^{-8}$ )

Assuming that a human being does not consume any other contaminated fungi or stay in a contaminated environment, the dose of 1 mSv will only be exceeded after consuming the amount of 0.25 kg of dried bay bolete with the activity of 300 kBq/kg. Also the consumption of 5 kg of dried fungi with the activity of approximately 15 kBq/kg will cause the dose of 1 mSv to be exceeded. Due to a small presence of fungi in the annual diet, as well as the maximum <sup>137</sup>Cs activity in dried bay bolete, amounting to approximately 1.2 kBq/kg, it can be acknowledged that the health risk related to the consumption of bay bolete collected in the examined area is insignificant.

# Relationships between Ca, <sup>137</sup>Cs, K and Mg concentrations in mushrooms

The measurement results show composition of mushrooms expressed as ratio of element weight in the sample to total weight of the sample. If mass of an element is expressed in mg and total sample mass is expressed in kg then the element's concentration changes are limited in the range from 0 to  $10^6$  mg/kg.

Concentrations of each element can be regarded as parts (components) of the vector representing compositional variable. Composition  $\mathbf{x}$  of a system composed of D components can be calculated by the closure operation C defined as follows:

$$C(\mathbf{m}) = \left[\frac{km_{1}}{\sum_{i=1}^{D} m_{i}}, \frac{km_{2}}{\sum_{i=1}^{D} m_{i}}, \dots, \frac{km_{D}}{\sum_{i=1}^{D} m_{i}}\right] = \left[x_{1,}x_{2,}\dots, x_{D}\right] = \mathbf{x}$$
(2)

where **m** is the vector of component amounts, *k* depends on the units of measurement and **x** is vector of compositions. Usual *k* values are 1 for proportions, 100 for %,  $10^6$  for mg/kg and  $10^9$  for µg/kg. Subcomposition can be obtained by applying the closure operation to a vector of *S* parts taken from D (S < D) [34-37].

Within composition, concentrations are not independent on each other. If content of one of components increases the others have to decrease. It is the reason for which results of standard statistical analysis of the relationships between raw components or parts are spoiled by spurious effects [35, 38].

The main principles of compositional data analysis are scale invariance and subcompositional coherence. Scale invariance originates from the idea that a composition provides information only about relative values and not about the absolute ones. The principle of subcompositional coherence requires the same results of statistical inherence of common parts regardless of whether full composition or subcomposition was used in computations. These principles are obeyed when variables ratios (or logratios) are used in statistical computations. It is so because ratios within a subcomposition are equal to the corresponding ratios within the full composition [39].

The constrained and intrinsically related compositional data need application of special statistical approach. First of all, to avoid false interpretation of results and misleading conclusions, appropriate sample space has to be selected for data analysis.

Compositional data occupy a restricted space where variables can vary only from 0 to a given constant. The unit simplex  $S^{D}$  was proposed for the geometrical representation of sample space. The simplex can be defined in the following way [34]:

$$S^{D} = [x_{1,}x_{2,}\dots,x_{D}]: x_{i} > 0(i = 1,\dots,D), x_{1} + x_{2} + \dots + x_{D} = 1$$
(3)

In this space two operations can be defined. The first one is the perturbation operation denoted by  $\oplus$  which takes two vectors **x** and **p** then calculates vector **y** of pairwise products and applies the closure operation:

$$\mathbf{y} = \mathbf{x} \oplus \mathbf{p} = C[x_1 p_1, \dots, x_D p_D]$$
(4)

Perturbation operation is analogous to addition in real space.

The second operation defined in simplex is power operation. For any real number a and any composition vector **x** the result **y** of power transformation  $\otimes$  is defined as follows:

$$\mathbf{y} = \mathbf{a} \otimes \mathbf{b} = C \Big[ x_1^a, \dots, x_D^a \Big]$$
(5)

This operation is analogous to multiplication by a scalar in real space.

The perturbation and power transformation are used for the compositional straight line description. The following equation describe the line:

$$\mathbf{x}(t) = \mathbf{x}_{\mathbf{0}} \oplus (t \otimes \mathbf{v}) \tag{6}$$

where  $\mathbf{x}_0$  is a starting point and  $\mathbf{v}$  is direction of the line and *t* is a real parameter [38].

To describe difference between two compositions the scalar measure of distance was introduced. Aitchison distance  $d_A$  between two compositions  $\mathbf{x}[x_1, x_2, ..., x_D]$  and  $\mathbf{X}[X_1, X_2, ..., X_D]$  is described by the formula [35]:

$$d_A(\mathbf{x}, \mathbf{X}) = \sqrt{\frac{1}{D} \sum_{j < l} \left( \ln \frac{x_j}{x_l} - \ln \frac{X_j}{X_l} \right)^2} = \sqrt{\sum_{j=1}^{D} \left( \ln \frac{x_j}{g(\mathbf{x})} - \ln \frac{X_j}{g(\mathbf{X})} \right)^2}$$
(7)

where  $g(\mathbf{x})$  denotes the geometric mean of x.

This measure reflects relative changes in points positions in restricted sample space.

Aitchison [34] introduced methods which should be used in computations of statistical parameters characterizing data from the constrained sample space. Certain approaches to constrained data were developed also by other authors [35]. These methods include, among others, transformations of the constrained data to unconstrained values [40, 41].

One of transformations is centered logratio (*clr*). For a vector  $\mathbf{x}$  of non-zero compositions the *clr* transformation is defined by the relationship:

$$clr(\mathbf{x}) = \left[\frac{x_1}{g(\mathbf{x})}, \dots, \frac{x_D}{g(\mathbf{x})}\right]$$
 (8)

This transformation enables computation of distances between compositions (Aitchison distances) in the same way like distances in Euclidean space of clr coordinates. The limitation of this transformation is constant, equal to 0, sum of all transformed parts of compositional vector. The points representing transformed compositions are located on a plane in D dimensional space.

Covariability of two compositional vector variables  $\mathbf{x}$  and  $\mathbf{X}$  can be estimated using variance of logratio *VR* [34]:

$$VR = var \left( \ln \frac{x_A}{x_B} \right) \tag{9}$$

If a linear relationship between A and B compositions exists then  $x_A/x_B$  ratio is constant and its variance is 0. If there is a weak or no relationship between  $x_A$  and  $x_B$  then the VR value increases.

In investigations of possible structures in data the hierarchical clustering methods were used. These methods allow to assign objects to different groups, so that the data in each subset share some common trait.

All of pairwise dissimilarities between *clr* transformed compositions in the data set were computed. The agglomerative nesting procedure and divisive hierarchical clustering were used in cluster structure determination. The overviews of cluster existence in data can be shown on dendrogram - a tree diagram illustrating the arrangement of the clusters.

For statistical computations the R language [42] was utilized. R is a free software environment for statistical computing and graphics. Besides standard R libraries, functions from package "compositions" were used [43-45]. For investigations of data clustering the functions provided by the "cluster" library of R were used [46]. Functions available in this library were described by Kaufman et al [47].

Concentration  $c_{\rm Cs}$  of <sup>137</sup>Cs was calculated from its specific activity concentration *a* from the formula:

$$m = \frac{t_{1/2}M}{\ln(2)N_A}a$$
 (10)

where  $t_{1/2}$  is the half lifetime of <sup>137</sup>Cs equal to 30.08 years, *M* is its molar mass equal to 137 g/mol and  $N_A$  is Avogadro constant. Because the radioisotope activity *a* was related to 1 kg of sample mass, the calculated *m* value corresponds to mass concentration of <sup>137</sup>Cs expressed in g/kg units.



Fig. 2. Distributions of K, Mg, Ca and <sup>137</sup>Cs concentrations in the places, where the mushroom samples were collected

Distributions of the investigated metals concentrations in mushrooms are shown in boxplots. In these plots the lower base of the rectangle is a lower quartile, the upper base is an upper quartile and a horizontal line dividing the rectangle represents 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). If the points marked by a circle appear in the plot, they are located more than 1.5 times of interquartile range from the median and can be regarded as outliers.

Boxplots in Figure 2 show distributions of K, Mg, Ca and <sup>137</sup>Cs concentrations in the places, where the mushroom samples were collected. In this graph and the subsequent ones the description of <sup>137</sup>Cs is simplified to "Cs". Among the elements and nuclides investigated in mushrooms, the biggest concentrations of potassium were determined. The changes in K concentrations are limited in the range from approx. 4 to 40 g/kg. There are some differences in medians but comparison of interquartile ranges as well as maximal and minimal values do not indicate significant differences between distributions of potassium concentration in mushrooms collected in various places. None of measurement results was supposed to be outlier.



Fig. 3. Distributions of K, Mg, Ca and <sup>137</sup>Cs concentrations in mushrooms samples, collected in the years 2002-2005

Concentrations of magnesium in mushrooms were at least one order of magnitude lower than potassium concentrations. Similarly like for K, taking into account medians, interquartile ranges and ranges of data changes, no influence of collection place on Mg distribution could be supposed. The same conclusion could be drawn for calcium, which concentrations are approx. 2.5 times lower than these for Mg.

Concentrations of <sup>137</sup>Cs in mushroom samples did not exceed 1 ng/kg. Median values, interquartile ranges and ranges of data changes are nearly uniform for all places. It could be supposed that the regions investigated were contaminated by <sup>137</sup>Cs very uniformly. Comparing to the formerly discussed elements, concentrations of <sup>137</sup>Cs seem to depend on other factors than the ones affecting K, Mg and Ca concentrations.

In Figure 3 the boxplots of elements distributions in samples collected in the years 2002-2005 are shown.

For K and Ca no relationship between element concentration and time of samples collection could be supposed. A trend of concentration decrease in time could be noticed for Mg. Such direction of concentration changes is clearly shown in the boxplots showing <sup>137</sup>Cs concentration in relation to the year of sample collection.

Possible data clustering in sample space was investigated using appropriate methods. Distances  $d_A$  between pairs of compositions were calculated in simplex. The distances were used in dendrogram construction. Results of hierarchical clustering were similar, both for agglomerative nesting procedure and divisive hierarchical clustering. In Figure 4 the plot of dendrogram with structure determined by agglomerative procedure is shown.



agglomerative procedure

In the dendrogram a separate branch composed of 2 points can be recognized. These points represent samples collected in 2005 from locations B and C. Currently it is rather not possible to state, why this effect occurs.

To estimate covariability of elements concentrations the VR coefficients of all pairs of variables were calculated. In calculations the complete data set was used and the one with 2 records excluded, which are represented by separate branch in dendrogram. The calculation results are shown in Table 3. Because of different number of records in both groups of data, the VR values can not be compared between groups. But within a group of variables comparison leads to valid conclusions.

Table 3

The VR values calculated for pair of variables representing concentration of elements. The results below diagonal, composed of 0.0, were calculated for all records in data set. Above diagonal parameters were calculated excluding 2 records, represented by clearly separate branch in dendrogram

VR	K	Ca	Mg	<sup>137</sup> Cs
K	0.0	0.33	0.54	0.33
Ca	2.1	0.0	1.0	0.49
Mg	1.7	2.1	0.0	0.51
<sup>137</sup> Cs	0.34	1.6	1.4	0.0

The least *VR* value in the complete data was found for  $^{137}$ Cs and K. It supposes linear covariability between their concentrations. But significance of this conclusion is diminished by observation, that in the reduced data set the corresponding *VR* value is less different from the others.

In Figure 5 summary of information about relationships between concentrations are shown. In the diagonal, density plots of distributions of *clr* transformed concentrations are shown.



Fig. 5. Distributions of *clr* transformed concentrations of metals (in diagonal), relationships between concentrations of metal pairs in simplex space (below diagonal) and relationships between *clr* transformed concentrations of metal pairs in Euclidean space

Below diagonal the relationships between metals concentration in simplex space are shown. The third component, marked by asterisk, represents geometric mean of concentrations of all remaining components. Solid line shows the linear estimation of relationship between components, defined by expression 6. In calculations of line parameters all data points were utilized. The parameters of dashed line were calculated from the reduced data set, 2 points represented by separate branch in Figure 4 were omitted. The data points in the plot were marked with circles, besides the two ones from separate branch in dendrogram which were marked by triangles. The plots above diagonal show relationships between pairs of *clr* transformed concentrations. Similar like parameters of lines in simplex space the ones in Euclidean space were calculated from full (solid line) and reduced data set (dashed line). To facilitate localization of possibly outlying points marked by triangles their positions in graphs were connected with dotted line.

Distributions of *clr* transformed data are unimodal. Only distribution of  $clr(c_{Mg})$  seem to be symmetric. Distributions of  $clr(c_K)$  and  $clr(c_{Cs})$  are positively skewed while distribution of  $clr(c_{Ca})$  is negatively skewed. The distributions are different but rather in moderate manner. Results of investigation of relationships between these variables should not be seriously burdened by the bias caused by different distributions.

In data analysis both the plots of relationships between variables in simplex and Euclidean space were considered. Some outlines of relationships between variables can be noticed. But analysis of graphs shown in Figure 5 reveals an influence of outlying points on relationship between concentrations. This influence is better visible in simplex space than in the Euclidean one. However some plots show that among two outlying points only one of them might be influential. It is clearly shown in the plot of  $c_{Mg}$  and  $c_{Ca}$  relationship in simplex space.

The main source of committed effective dose in mushroom samples is <sup>137</sup>Cs. A constant ratio  $c_{Cs}/c_{K}$  was supposed by low *VR* value in Table 3, and appropriate graphs in Figure 5, when all data points were used in plots construction and in computations. But this relationship is much weaker when the reduced data set was used and its character changes. After two points removal the proportionality between these metals disappears. Instead of that the constant ratio between <sup>137</sup>Cs and geometrical mean of the remaining metals concentrations could be supposed. The conclusion is that concentrations of potassium and cesium in mushroom are not essentially related with each other.

No reliable relationships between concentrations of <sup>137</sup>Cs and the remaining metals could be observed. The same conclusion could be drawn for K. Besides Ca and Mg which sum of concentrations is constant, no clear relationship between concentrations of the investigated metals could be found.

### Conclusions

Moderate ingestion of bay bolete collected in the investigated area does not create health risk due to increased radioactive substances intake. The amount of consumed mushrooms that could deliver the dose exceeding the safe one, is rather improbable in real life. The risk is also lowered because of obvious decrease of <sup>137</sup>Cs content in bay bolete.

In studies of relationships between metals concentration the methods designed for compositional data analysis were used. Besides constant sum of magnesium and calcium concentrations, no clear relationships between <sup>137</sup>Cs, Ca, K and Mg concentrations in

samples of bay bolete were found. Significant influence of outlying data points on statistical inference was noticed.

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### BADANIE WCHŁONIĘTEJ, SKUTECZNEJ DAWKI PROMIENIOWANIA I ZALEŻNOŚCI POMIĘDZY STĘŻENIAMI METALI ALKALICZNYCH W OWOCNIKACH Xerocomus badius

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**Abstrakt:** Owocniki grzybów wyrastające z grzybni są zdolne do gromadzenia znacznych ilości pierwiastków śladowych zarówno metali, jak i niemetali. Zawartość tych pierwiastków w owocnikach może wielokrotnie przekroczyć ich stężenia w podłożu. Wśród pochłoniętych pierwiastków są również nuklidy promieniotwórcze. Dokonano oceny potencjalnych dawek skutecznych promieniowania gamma w wyniku wchłonięcia <sup>137</sup>Cs i <sup>40</sup>K wraz ze spożywanym grzybem. Na podstawie wyników pomiarów stwierdzono, że umiarkowane spożycie podgrzybka brunatnego nie stwarza zagrożenia dla zdrowia ze względu na zwiększone spożycie substancji radioaktywnych. Badano również relacje między aktywnością izotopów promieniotwórczych a stężeniem metali alkalicznych, wykorzystując metody przeznaczone do analizy składu danych.

Słowa kluczowe: grzyby, radionuklidy, metale alkaliczne