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^{137}Cs transfer from local particulate matter to lichens and mosses

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Abstract. Data on ^{137}Cs radioactivity in the lichen *Hypogymnia physodes* and the moss *Pleurozium schreberi* were compared with the ^{137}Cs content in soil for a Chernobyl ^{137}Cs contaminated forest area of Bory Stobrawskie, southern Poland. The study area partly overlaps the so-called Opole Anomaly, where ^{137}Cs radioactivity of forest upper soil layer in some places is 100 times greater than the average for Poland. A clear correlation between ^{137}Cs radioactivity in the upper soil layers and in lichens and mosses was found, the relationship of which can be expressed as follows: $A_{\text{lichen(moss)}} = a + b\cdot\ln(\Delta A_s)$, where (ΔA_s) is the radioactivity of mobile ^{137}Cs cations, leached from soil to an HCl solution of pH = 3.9. Lichen transplantation method was used to demonstrate the transfer of ^{137}Cs from soil via local soil particulate matter to transplanted epiphytic lichens. During 220 days of exposure, ^{137}Cs radioactivity in the transplanted lichens increased by 145 Bq·kg^{-1} (d.m.). Atmospheric activity of ^{137}Cs during this time was low (on average it was 5.4 μBq·m^{-3}). The lichens were transplanted from regions with low ^{137}Cs soil radioactivity (ca. 30 Bq·kg^{-1} (d.m.)) to areas where it exceeded 1400 Bq·kg^{-1} (d.m.).

Key words: radioactivity • ^{137}Cs transfer to biomonitors • lichens • mosses • lichens transplantation • Chernobyl fallout

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Introduction

Biomonitors (organisms which react to changes of the chemical composition of their environment [29]) are frequently used to assess environmental pollution. Lichens and mosses are perceived as two of the best bioindicators of various air pollutants [7].

Analysis of trace elements accumulation in lichens and mosses is a useful tool for evaluation both levels and sources of pollution. The main problem associated with quantitative assessment of environmental pollution based on trace element accumulation in lichens and mosses is the multifactorial impact of the environment on the bioindicators themselves [25]. Mutual relations between accumulation of trace elements in soil, lichens and mosses have been the subject of numerous publications (e.g. [5, 26]).

Radionuclide accumulation in biological material has been addressed in many previous studies. Early researches on radionuclide accumulation in lichens focussed mainly on radioactive isotopes resulting from nuclear tests (in the middle of the 20th century) such as caesium (^{137}Cs, $t_{1/2}$ = 30.2 y) and strontium (^{90}Sr, $t_{1/2}$ = 28 y). Following the Chernobyl accident in April 1986, several studies focused on the concentration of isotopes with relatively short half-lives, including iodine (^{131}I, $t_{1/2}$ = 8 d) [11] and caesium (^{134}Cs, $t_{1/2}$ = 2.06 y) [6]. However, ^{137}Cs released due to the accident, whose total radioactivity is estimated to 3.77 × 10^{16} Bq is still monitored [13]. The data concerning the results of

Chernobyl NPP fallout were published in the report of United Nations Scientific Committee on the Effects of Atomic Radiation [28].

Ionic exchange is regarded to be the main mechanism of radionuclide sorption [24, 27], as it is indicated, among others by the results of our investigations on the mechanism of heavy metal accumulation [14–16, 18, 19] and by research on radionuclide accumulation in lichens [21, 22], and in mosses by [3]. Caesium ions, which are chemically similar to potassium (plant nutrient metal) ions, are easily incorporated into the biological structure of lichens and mosses. This paper is concerned with ^{137}Cs released to the atmosphere due to the Chernobyl nuclear power plant accident (1986), which is still present in forest soil throughout Europe. Therefore, it seems to be interesting to check possibile correlation between ^{137}Cs radioactive in the lichen *Hypogymnia physodes* (and the moss *Pleurozium schreberi*) and in soil of Chernobyl fallout terrains. The objective of this research was a study of ^{137}Cs radioactivity concentration in lichens (or mosses) and that in forest soil in the area of the so-called Opole Anomaly, southern Poland, where ^{137}Cs radioactivity concentration in forest soil upper layer in some sites is 100 times greater than the average for Poland [12]. The nutrients are uptaken by lichens and mosses (in gaseous or particulate forms) from air. So one should expect that if any dependence of ^{137}Cs content exists in the plants growing there and the ^{137}Cs content in the upper soil layer of the sites, then one could presume that a source of the radioisotope contaminant are the local soil particulate matter containing the pollutant. In addition, one should expect that the dependence obtained (^{137}Cs content in the plants vs. that in soil) can also be useful for describing the transfer of other metallic ions, including anthropogenic contaminants, from soil via local soil particulate matter to lichens and mosses.

In Poland, ^{137}Cs has not been found from global fallout for a number of years [28]. In 2006 its radioactivity in air was 0.1–10.7 (mean: 1.4 $\mu Bq \cdot m^{-3}$) in the 2nd quarter and 0.1–6.7 (mean 1.0 $\mu Bq \cdot m^{-3}$) in the 3rd quarter [1, 2]. From quarterly data published in the Report of President of the National Atomic Energy Agency, it was calculated that in time period when the samples were collected the ^{137}Cs activity in the air changed in the range from 0.1 to 11.4 $\mu Bq \cdot m^{-3}$ (mean: 1.3 $\mu Bq \cdot m^{-3}$).

Therefore, one might claim that at present the main source of ^{137}Cs accumulated in lichens and mosses in some places contaminated due to the Chernobyl accident is the soil contamination which can reach a level of activity 1000 $Bq \cdot kg^{-1}$ (d.m.) [17]. Radioactivity in these plants is caused by soil particulate matter (derived from the surface layer of local soil), which is moved by the wind and deposited on the surface of lichens and mosses. Our earlier research on the equilibrium processes of metal ion sorption by lichens from aqueous solutions showed that the lichens, as the result of substituting lichen hydrogen ions by the metallic ones, may acidify the solution wetting the thallus surface up to ca. 3.9 pH with no damage to the lichen biological structure [14, 15, 19].

Materials and methods

Hypogymnia physodes and *Pleurozium schreberi* as well as soil were collected from Bory Stobrawskie (partly belonging to the so-called Opole Anomaly), a forest area of ca. 3000 km^2 situated 20–50 km to the north-east of the provincial city of Opole, Poland. Lichens, mosses and soil sampling was carried out from March 2005 to the end of November 2006 and from April to June 2007. In some places of this region ^{137}Cs radioactivity concentration in soils measured in 1994 was up to a hundred times higher than the average for Poland [12]. Twenty years after the Chernobyl accident, ^{137}Cs is still present in the forests and wastelands. The map of Bory Stobrawskie (Fig. 1) shows the locations of the 24 sampling sites where in the years 2005–2006 the

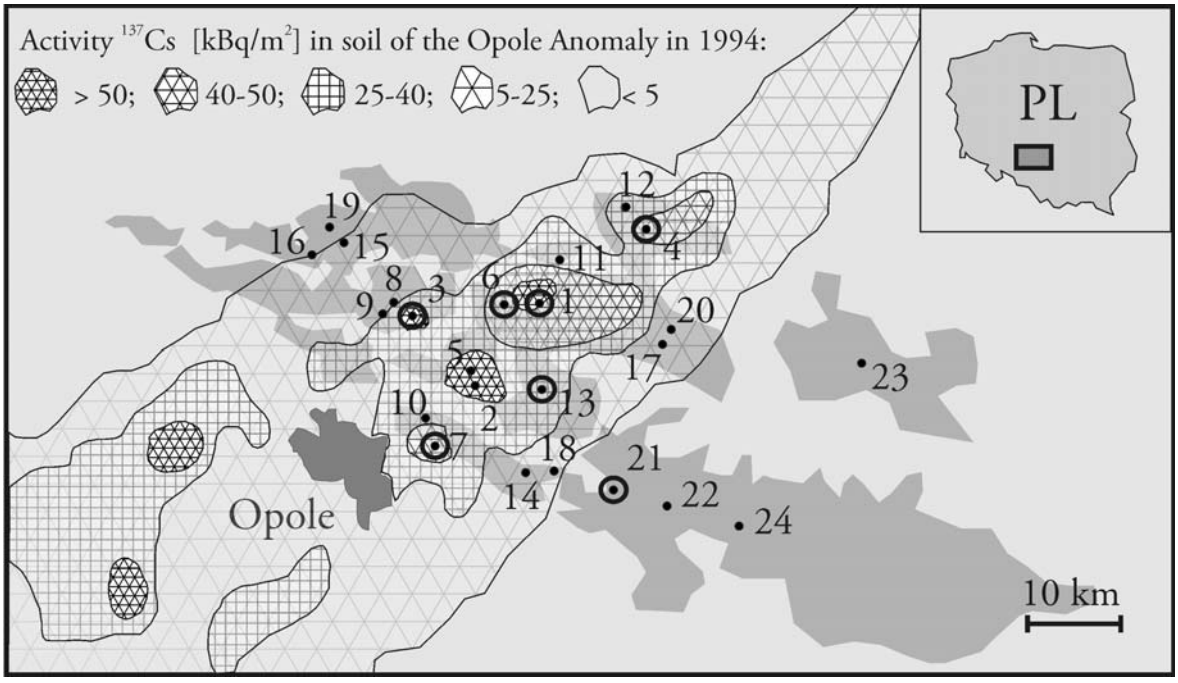

Fig. 1. Sampling sites in 2005–2006 (1–24) and in 2007 (1, 3, 4, 6, 7, 13 and 21).

samples were collected as well as contour lines indicating the ^{137}Cs radioactivity concentration levels in soils, as measured in 1994. On the sites 1, 3, 4, 6, 7, 13 and 21 the samples were collected and concentration of ^{137}Cs measured again in 2007.

The samples of lichens growing on spruce (*Picea abies*) or larch (*Larix decidua*) branches were placed horizontally 1.0 to 1.5 m above the ground level.

Samples of lichens, mosses and soil taken at six sampling points on the sampling site (of about 50 m^2 in the area) were homogenized. The lichen, moss and soil sampling has been repeated on 4 consecutive days (Table 2) at each case at new sampling points within the same sampling site, in order to test the results reproducibility. The data concern 5 independent measurements. In the case of soil ^{137}Cs mobile forms the data describe differences in the individual sample activity before and after leaching. The statistical significance of the results has been confirmed by Student's test approx. 100 g of mosses, 100 g of lichens and 500 g of soil were collected on each sampling site. For the soil examination samples from sub-horizon Oh (located at 2 cm to 7 cm depth from soil surface) were collected. Mechanical impurities (bark, sand) were removed from moss and lichen samples by hand and subsequently air stream was passed through samples in order to remove finer dust particles upon which they were dried at a temperature not exceeding 303 K. The soil was sieved (mesh diameter was 2 mm) and subsequently dried at a temperature of 373 K. After homogenization of composition the samples of lichens, mosses and soil, the following mass 30, 40 and 150 g, respectively were used for the ^{137}Cs radioactivity measurements. To study the rate of transfer (transfer factor) of ^{137}Cs from soil (via soil particulate matter) to *Hypogymnia physodes*, lichen samples were transplanted on their natural substrate (phorophytes) from an area of low ^{137}Cs radioactivity (site 21, Fig. 1) and they were exposed in the area, where ^{137}Cs radioactivity in naturally growing lichens was in excess of 1100 Bq·kg^{-1} (d.m.), and in soil 1400 Bq·kg^{-1} (d.m.) (site no. 2, Fig. 1). Branches with lichens were placed horizontally 1.0 to 1.5 m above the ground in places not overshadowed by crowns of larger trees in order to eliminate the impact of canopy through-fall. The branches with lichens growing on them were tied with a nylon cord to dead dry spruce or larch trees ca. 2 m in height. Blueberries were the basic undergrowth in the places from which the lichens originated. A reference sample was placed in the same way in location 19 (Fig. 1), at which low ^{137}Cs activities in lichen and soil were observed. The lichens were exposed over the period from February to August, which coincides with the season of moderate and low precipitation in the area.

The ^{137}Cs radioactivity concentration measurement of samples 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: 21.7%. Measurements and analysis of spectra was computer controlled with the use of software GENIE 2000. Energy and efficiency calibration of the gamma spectrometer was performed with standard solutions, type MBSS 2 (Czech Metrological Institute, Prague, Czech Republic), which covers an energy range from

Table 1. The ^{137}Cs radioactivity concentration in lichen, moss and soil samples [Bq·kg^{-1} (d.m.)], recalculated for the 01.03.2005. Minimum detectable activity, MDA < 2.0 Bq·kg^{-1}, u_A – mean activity uncertainty

No.	Soil before leaching (A_{soil})		Soil after leaching ($A_{soil,leach.}$)		Moss (A_{moss})		Lichen (A_{lichen})	
	$A \pm u_A$ [Bq·kg^{-1} (d.m.)]							
	2005/06	2007	2005/06	2007	2005/06	2007	2005/06	2007
1	1765±40	1740±42	1718±41	1689±40	748±22	785±28	769±34	752±31
2	1425±36	–	1238±32	–	–	–	1147±23	–
3	1357±29	1313±32	1329±34	1284±28	593±10	622±11	575±11	579±14
4	1327±31	1419±33	1279±33	1372±31	731±11	731±17	696±16	718±12
5	1010±20	–	890±21	–	1075±47	–	1010±41	–
6	801±21	850±18	777±20	830±24	502±16	483±13	420±18	458±10
7	668±14	711±12	660±15	702±17	17.42±0.56	34.7±1.2	116.1±5.4	91.3±2.9
8	627±21	–	612±14	–	404±19	–	221±11	–
9	361.7±8.3	–	350.0±6.5	–	–	–	312.9±7.9	–
10	318.1±6.0	–	307.9±5.9	–	–	–	116.3±4.1	–
11	276.2±5.2	–	249.1±5.5	–	–	–	681±10	–
12	190.0±7.0	–	171±6.3	–	412±14	–	455±16	–
13	167.6±3.3	170.2±4.1	156.2±3.2	156.1±2.9	211.2±7.2	203.2±6.4	166.1±5.3	174.9±4.4
14	155.2±4.6	–	147.8±4.2	–	133.3±3.0	–	13.2±1.1	–
15	105.8± 3.5	–	101.0±3.6	–	62.5±3.1	–	35.3±1.3	–
16	99.0± 2.7	–	94.8± 2.2	–	39.6±1.7	–	52.4± 2.4	–
17	87.3±4.3	–	80.1±3.2	–	55.0±2.4	–	34.8±1.9	–
18	80.1±3.6	–	74.7±3.0	–	104.8±2.8	–	54.7±2.6	–
19	72.2±1.9	–	68.4±2.2	–	–	–	34.5±1.6	–
20	52.6±1.5	–	44.8±1.0	–	–	–	35.1±1.6	–
21	28.9±1.1	27.8±1.1	24.9±1.0	23.4±1.1	53.0±2.3	19.7±1.1	17.3±0.8	16.3±0.9
22	13.95±0.35	–	6.95±0.30	–	112.1±4.1	–	37.2±1.5	–
23	12.95±0.34	–	3.64±0.14	–	105.9±.6	–	13.9±0.57	–
24	8.74±0.30	–	5.11±0.24	–	60.3±1.6	–	25.5±1.1	–

59.54 keV (^{241}Am) to 1836.06 keV (^{88}Y). Geometry of calibration source and samples: Marinelli 450 cm^3. The ^{137}Cs radioactivity concentration of soil samples was reanalysed (after 30 min) once they had been leached with hydrochloric acid solution of pH = 3.9 (1 dm^3 of acid for 100 g of soil). The soil samples were leached by shaking at room temperature. After leaching the samples were washed with demineralized water and then dried at 373 K.

Results

Distribution of ^{137}Cs radioactivity concentration in lichens, mosses and soil samples

Table 1 lists data on ^{137}Cs radioactivity concentration in lichens, mosses and soil collected on 24 sampling sites (Fig. 1). Some data concerned with the ^{137}Cs radioactivity concentration in lichens and soil have been already published in a recent paper [17].

Table 2 contains a summary of the measurements of ^{137}Cs, mean and standard deviation s determined for five samples of lichens, mosses and soil collected at each sampling place 5, 12 and 21 (Fig. 1). The sites were selected taking into account different ^{137}Cs activity level and big quantity of lichens growing there.

From Table 2, one can conclude that the standard deviation s [±%] increases with decreasing activity of ^{137}Cs of the examined samples.

Results of measurements presented in Table 1 do not show clear correlations between the radioactivity of ^{137}Cs in lichens A_{lichen} (and mosses A_{moss}) and its total radioactivity in soil; the correlation coefficient $R^2 < 0.7$. One could guess that the poor correlation is due to the fact that instead of the radioactivity of total ^{137}Cs content in soil one would take into considerations only part of it, namely that available for lichens and mosses. The content of mobile form of ^{137}Cs in soil can be determined by leaching the soil samples with HCl solution of pH = 3.9.

Figure 2 compares the results of ^{137}Cs radioactivity concentration in lichens and mosses with that of

Table 2. Measurements of ^{137}Cs, mean and standard deviation s determine for five samples of lichens, mosses and soil collected on the sampling sites 5, 12 and 21; $\text{Mean}_{(1-5)}$ – mean value for 5 independent measurement results; $s_{(1-5)}$ – standard deviation of measurements of 5 independent samples at the site; u_A – weighted mean activity uncertainty; $u_{\Delta A_{s.}}$ – standard uncertainty of the difference: $\Delta_{A_{s.}} : u_{\Delta A_{s.}} = [(u_{\Delta A,\text{soil}}/3^{1/2})^2 + (u_{\Delta A,\text{soil leach}}/3^{1/2})^2]^{1/2}$

No.	1	2	3	4	5	$\text{Mean}_{(1-5)}$	$s_{(1-5)}$ [±%]
	[Bq·kg^{-1} (d.m.)]						
	Sampling site 5						
Lichen	1024	918	968	1050	1092	1010	6.1
u_A	±43	±38	±36	±40	±46		
Moss	1075	962	1154	992	1191	1075	8.3
u_A	±51	±43	±48	±45	±49		
Soil before extraction (A_{soil})	918	1064	1042	944	1082	1010	6.6
u_A	±21	±23	±19	±20	±24		
Soil after extraction ($A_{\text{soil, leach.}}$)	808	936	930	826	951	890	6.8
u_A	±21	±20	±22	±19	±20		
$\Delta A_{s.} = A_{\text{soil}} - A_{\text{soil, leach.}}$	110	128	112	118	131	120	7.0
$u_{\Delta A_{S.}}$	±17	±18	±17	±16	±18		
	Sampling site 12						
Lichen	412	498	420	482	465	455	7.4
u_A	±16	±15	±19	±16	±14		
Moss	380	374	444	412	449	412	7.6
u_A	±12	±15	±16	±14	±15		
Soil before extraction (A_{soil})	212.0	201.9	194.3	166.1	177.8	190	8.7
u_A	±8.1	±7.3	±7.2	±6.0	±6.5		
Soil after extraction ($A_{\text{soil, leach.}}$)	189.9	181.0	175.2	149.8	159.3	171	8.5
u_A	±7.4	±7.5	±6.1	±5.6	±5.1		
$\Delta A_{s.} = A_{\text{soil}} - A_{\text{soil, leach.}}$	22.1	20.9	19.1	16.3	18.5	19	10.8
$u_{\Delta A_{S.}}$	±6.3	±6.0	±5.4	±4.7	±4.8		
	Sampling site 21						
Lichen	18.00	14.09	20.6	19.5	14.12	17.3	17
u_A	±0.85	±0.68	±1.0	±1.0	±0.69		
Moss	43.8	46.0	61.7	62.4	51.1	53.0	15
u_A	±2.1	±2.0	±2.8	±2.6	±2.2		
Soil before extraction (A_{soil})	28.6	22.9	36.1	24.8	31.9	28.9	16
u_A	±1.0	±0.95	±1.1	±1.1	±1.2		
Soil after extraction ($A_{\text{soil, leach.}}$)	24.86	20.2	30.8	22.34	26.1	24.9	15
u_A	±0.83	±1.0	±1.2	±0.95	±1.2		
$\Delta A_{s.} = A_{\text{soil}} - A_{\text{soil, leach.}}$	3.7	2.7	5.3	2.5	5.8	4.0	28
$u_{\Delta A_{S.}}$	±0.75	±0.80	±0.94	±0.84	±0.98		

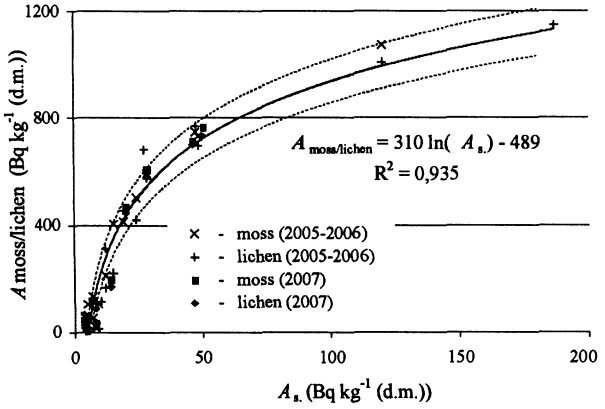

Fig. 2. Comparison of ^{137}Cs radioactivity concentration in lichens and mosses with that of radiocaesium $\Delta A_{s.}$ leached from soil to HCl solution (pH = 3.9).

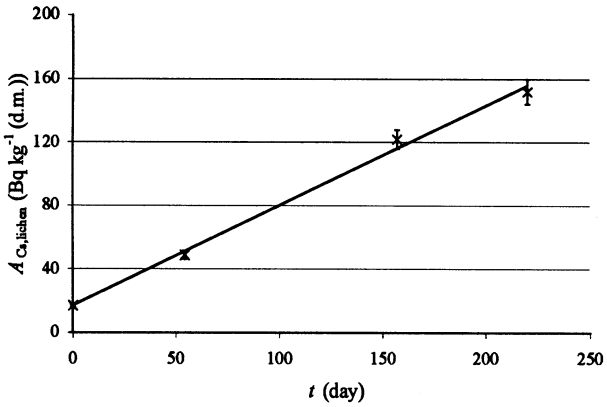

Fig. 3. Changes in ^{137}Cs radioactivity of transplanted lichen exposed in the area, where ^{137}Cs radioactivity exceeding 1000 Bq·kg^{-1} (d.m.) in naturally-occurring lichens.

Table 3. The a and b parameters and their standard deviations s_a and s_b of equation: $y = a + b \cdot \ln x$ and correlation coefficient R^2 describing the relationship between ^{137}Cs radioactivity concentration in lichens, mosses and in the fraction of soil extracted using HCl solution (pH = 3.9)

Materials	a	s_a	b	s_b	R^2
Lichen	–504	43	311	15	0.934
Moss	–473	45	309	16	0.946
Moss/lichen	–489	31	310	11	0.935

^{137}Cs released from soil by hydrochloric acid solution (pH = 3.9) $\Delta A_{s.} = A_{soil} - A_{soil,leach}$.

In the diagram is shown the function: $A_{moss/lichen} = a + b \cdot \ln(\Delta A_{s.})$. The parameters s_a (±31) and s_b (±11) (shown also in Table 3) were taken into account.

Table 3 shows the parameters of logarithmic function describing the relationships observed: $A_{lichen} = a + b \cdot \ln(\Delta A_{s.})$, $A_{moss} = a + b \cdot \ln(\Delta A_{s.})$ and $A_{moss/lichen} = a + b \cdot \ln(\Delta A_{s.})$. As one can see, the ^{137}Cs radioactivity concentration in lichens and mosses correlate very well with that of the leachate solution. The data also show that the ^{137}Cs sorption capacity of the lichen and moss is very similar.

Rate of ^{137}Cs transfer from soil particulate matter to lichens

Lichens transplanted on their phorophytes from site no. 21 (Fig. 1), where ^{137}Cs radioactivity concentration in the lichen was only 17 Bq/kg (d.m.), were exposed during February and August 2006 at site no. 2 (Fig. 1) where ^{137}Cs radioactivity concentration measured for lichen naturally growing there exceeded 1100 Bq·kg^{-1} (d.m.), and for soil was above 1400 Bq·kg^{-1} (d.m.). During the above-mentioned period, three lichen samples were analysed (Fig. 3). Lichens collected for analysis were not exposed again.

Figure 3 shows an increase of ^{137}Cs radioactivity concentration [Bq·kg^{-1}] in transplanted lichen from an initial of 17.4 (t = 0) to 49.1 (±1.9) after 54 d, to 122.3 (±4.1) after 157 d and reaching 151.9 (±5.6) after 220 d. In reference samples (at location no. 19) ^{137}Cs activity in exposed lichens did not change significantly: 20.8 (54 days), 16.2 (157 days) and 19.1 (220 days) Bq·kg^{-1} (d.m.). Atmospheric radioactivity in Poland during the time of the experiment was in the range (0.1–10.7) μBq·m^{-3} [1, 2]. Therefore, its effect is negligible. It seems that the relationship can be only explained by transfer of ^{137}Cs from the local soil particulate matter to the lichens. The slope of straight-line shown in Fig. 3 is a measure of the rate of ^{137}Cs transfer from soil via soil particulate matter to transplanted epiphytic lichens. In time period when experiment was carried out the mean increase in ^{137}Cs activity was 0.63 Bq in 24 h. It might be expected that this increase will diminish in time.

Discussion

The presented data on radiocaesium content in the forest environment of Bory Stobrawskie can be rationalized in terms of circulation of this radionuclide in the soil-plant-soil system. Dołhańczuk-Śródka [8] determined the greatest concentration of radiocaesium in the upper (10 cm) renewable organic layer (O horizon) of soil, and other researches have also shown deeper but insignificant translocation of radiocaesium into soil profiles (e.g. [9, 10]) of the forest environment, ^{137}Cs uptake by plants depends on the type of soil (mainly content of clay and organic matter), soil pH value and humidity (e.g. [4]) as well as the type of undergrowth.

Data presented in Fig. 2 show that the ^{137}Cs radioactivity concentration in mosses and lichens depends only on the soil radionuclide mobile forms. This relationship was described by the equation $A_{lichen\ (moss)} = a + b \cdot \ln(\Delta A_{s.})$. It should be remembered that the concentration of the mobile forms was set as the ^{137}Cs radionuclide concentration released from soil into HCl solution (pH = 3.9). Our earlier investigations showed [14, 17, 19] that water of pH value more than 3.9 might wet lichens in their natural environment without damaging the lichen biological structure. Radiocaesium cations released from the particulates covering lichen (from secondary deposition) were transported into thallus inside in the way of ion exchange.

A linear character of the dependence $A_{mosses} = (A_{lichens})$ (the straight line slope a = 1.05; and the correlation coefficient R^2 = 0.948) indicates that the plants have very similar sorption properties as regards ^{137}Cs.

Such proportional relationships for epigeic mosses and epiphytic lichens were also mentioned by Chiarenzelli *et al*. [5]. Proportional relationships have also been found for sorption from solutions of various chemical species. The research concerned, among others, the kinetics equilibrium constant and the impact of cadmium ions and pH value on heavy metal and radionuclide sorption by mosses [23] and lichens [19, 21].

It seems that changes of ^{137}Cs radioactivity concentration in lichens (Fig. 3) exposed in the area with a high content of this radionuclide in soil (over 1400 Bq·kg^{-1}) can be rationalize by assuming a soil particulates deposition on the lichen surface. Insignificant changes of ^{137}Cs radioactivity concentration were measured for the reference sample exposed on site no. 19 with a low ^{137}Cs soil radioactivity [72 Bq·kg^{-1} (d.m.)]. No significant changes in the radioactivity of lichen samples collected in comparing with those collected earlier were observed (Table 1). The investigation results show a large ^{137}Cs content in the upper 2–7 cm organic soil layer (sub-horizon Oh). Similar results were described in [8, 9, 20].

Conclusions

The research was carried out in the area with high (compared with other regions of Poland) radioactivity of ^{137}Cs which is currently found mainly in woodlands and wastelands, e.g. in some areas of the Opole Anomaly. It was shown that the ^{137}Cs radioactivity concentration accumulated in lichens and mosses depends on the content of mobile forms of this radionuclide in soil. Moreover, epigeic mosses and epiphytic lichens growing close to each others exhibit similar sorption properties in respect of this radionuclide. It was shown that at present the main source of ^{137}Cs accumulated in the two plants is dust originating from the surface of the organic horizon of forest soil. The temporal dependence found for ^{137}Cs transfer from soil to epiphytic lichens and epigeic mosses may be also useful for explaining translocation of other ionic substances, e.g. anthropogenic metallic ionic contaminants (heavy metals), from soil surface layers via 'local soil particulate matter' to lichens and mosses.

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