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The activity concentration of post-Chernobyl ^{137}Cs in the area of the Opole Anomaly (southern Poland)

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Abstract During the years 2007 and 2010, the activity concentration of ^{137}Cs accumulated in soil, mosses *Pleurozium schreberi* and lichens *Hypogymnia physodes* was measured. The studies covered the areas of the so-called Opole Anomaly. In consequence of the Chernobyl nuclear power plant breakdown in 1986, relatively large amounts of this radionuclide were deposited in this area. In some areas of the Anomaly, over 100 times higher surface activity of ^{137}Cs was detected, compared to the lowest values registered in Poland. Currently, ^{137}Cs is still present in woodlands and wastelands. As at 2 April 2013, the surface activity concentration of ^{137}Cs in soil on the tested area was from 0.34 to 67.5 kBq m^{-2}. In comparison, the surface activity concentration of ^{137}Cs as at 1 June 1986, soon after deposition, was from 2.08 to over 125 kBq m^{-2}. The maximum specific activity concentrations of ^{137}Cs in mosses and lichens sampled for testing in 2010 were respectively 1234 and 959 Bq kg^{-1}. It was also proven that the changes in activity concentration of ^{137}Cs in the area of the Anomaly are mainly the consequence of the radioactive decay of this radionuclide.

Keywords Mosses · Soil · Lichens · Radionuclides

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Introduction

The Chernobyl reactor breakdown disturbed global gamma-ray radiation for a long time and by varying intensities. The breakdown also changed the radiation environment in Poland. Radioactive air pollution in Poland at the end of April through May of 1986 exceeded the values registered before the breakdown many times over. The main components of the pollution were inert gases, iodine and caesium isotopes. Pollutants were transferred to the soil by atmospheric deposition, and local variation in rainfall resulted in uneven soil pollution.

After the Chernobyl breakdown, most countries introduced monitoring of post-Chernobyl ^{137}Cs concentrations in soil. Studies were carried out in countries including Belarus (Knatko et al. 1996), Chile (Schuller et al. 2000), Israel (Lavi et al. 2006), Japan (Takenaka et al. 1998), Jordan (Hamarneh et al. 2003), Canada (Blagoeva and Zikovsky 1995), Mexico (Gaso et al. 1998), Germany (Schimmack et al. 1997), Poland (Jagielak et al. 1997), Russia (Strand et al. 1999), Serbia and Montenegro (Bikit et al. 2005), Switzerland (Riesen et al. 1999), Sweden (Melin et al. 1994) and Italy (Giovani et al. 1994).

From 1980 to 1990 in Poland, the geographical distribution of radiocaesium pollution (^{137}Cs and ^{134}Cs) was studied by three independent teams from the Central Laboratory for Radiological Protection (Biernacka et al. 1991), the Institute of Nuclear Physics of the Polish Academy of Science PAN (Mietelski et al. 1996) and the National Geological Institute (Strzelecki et al. 1993).

Maps that were produced from these studies agreed with the geographical pollution distribution, with most ^{137}Cs deposition (more than 50 kBq m^{-2}) in the Opole–Nysa–Kłodzko region in southwest Poland, known as the Opole Anomaly.

The objective of this study was to assess historical changes in the surface ^{137}Cs concentrations in woodland and wasteland soils in the Opole Anomaly and to assess the potential influence of ^{137}Cs translocation mechanisms on these changes. To allow comparison, ^{137}Cs activity in epigeic mosses (*Pleurozium schreberi)* and epiphytic lichens (*Hypogymnia physodes*) was tested.

Materials and methods

The locations of the woodland and wasteland study sites were in the area within the co-ordinates 50°–51° N and 15.5°–18.5° E and are shown in Fig. 1.

The surface ^{137}Cs concentrations were determined for randomly selected locations, distributed evenly throughout the study area. We sampled 87 measurement points in 2007 and 35 in 2010. We collected samples of *P. schreberi* mosses and *H. physodes* lichens in 2010 at 31 randomly selected points in woodlands in the study area. We sampled from locations only when both mosses and lichens were present. Approximately 20 g of mosses and 20 g of lichens were collected from each sampling site. Lichens and mosses were cleaned and dried at a temperature of up to 303 K.

Equipment and reagents

The surface ^{137}Cs activities were detected using a Canberra portable scintillation spectrometer InSpector1000. This is a portable, digital, multichannel analyser and is suitable for monitoring in situ. It can be used to determine radiation dose and power, identify the source location, for nuclide identification with activity measurements in the field, and for spectrum acquisition and analysis. Genie 2000 software (Gamma Analysis Option model S501C) was used to analyse the spectrum.

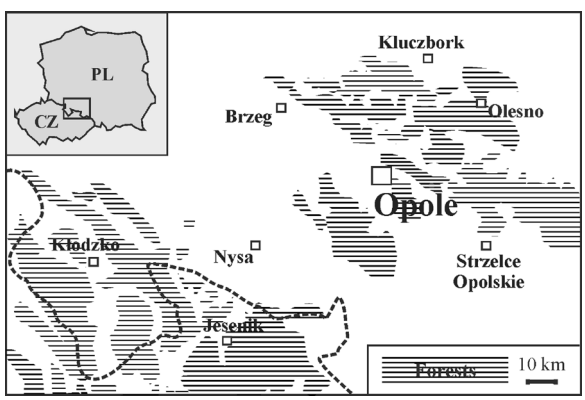

Fig. 1 Location of the study area

The ^{137}Cs activity measurements were determined using a gamma spectrometer with a high-resolution germanium detector HPGe (Canberra) as follows: 1.29 keV (FWHM) at 662 keV and 1.70 keV (FWHM) at 1332 keV. The relative efficiency was 21.7 %. Energy and efficiency calibration of the gamma spectrometer energy and efficiency was done using a MBSS 2 standard solution (Czech Metrological Institute, Prague, Czech Republic), which covers an energy range from 59.54 to 1836.06 keV. The calibration source geometry was Marinelli (447.7±4.5 cm^3), had a density of 0.99± 0.01 g/cm^3, and contained ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y and ^{203} Hg. The Marinelli sample container had a volume of 450 cm^3. GENIE 2000 software was used to measure and process the spectra 24 h a day. The measurement uncertainty for ^{137}Cs, estimated by the GENIE 2000 software, was approximately 1–2 %.

An ordinary kriging technique was used to produce isoline maps of ^{137}Cs surface activity distribution and the specific activity of ^{137}Cs in mosses and lichens. An exponential model was used to describe the experimental semi-variance. The geostatistical software Surfer (Golden Software Ltd.) was used to prepare the maps based on the WGS 1984 reference co-ordinate system.

Results

Table 1 presents information on the surface activity of ^{137}Cs in the study area for 1994, 2007 and 2010, and the specific activity of ^{137}Cs accumulated in the sampled mosses and lichens in 2010. The values of the surface activity of ^{137}Cs registered in 1994 were read from the map (Jagielak et al. 1997), which displays surface activity ^{137}Cs isolines in steps of 5 kBq m^{-2}. The data from this period do not distinguish values of surface activities greater than 65 kBq m^{-2}.

There was a decrease in the surface ^{137}Cs activity in soil over time. To assess the influence of translocation on ^{137}Cs activity, we compared our results for 2007–

Table 1 The activity factors of ^{137}Cs accumulated in soil during the years 1994, 2007 and 2010 and in mosses and lichens in 2010

Parameter	Soil 1994 kBq m^{-2}	Soil 2007	Soil 2010	Mosses Bq kg^{-1}	Lichens
Minimum	5.0	1.29	0.36	10.9	6.94
Lower quartile	7.5	4.84	3.25	43.8	21.0
Median	12.5	12.0	7.31	81.5	53.0
Mean	17.0	18.0	11.6	190	155
Upper quartile	22.5	23.0	16.3	223	161
Maximum	>65	77.6	41.6	1234	959
The standard deviation	11.7	18.5	11.0	248	239

2010 with those reported by Jagielak et al. (1997). The measured ^{137}Cs activities at 2 April 2013 were calculated using formula 1:

$$A(t) = A_0 \cdot \exp\frac{-\ln 2 \cdot t}{t_{1/2}} \quad (1)$$

where A_0=initial activity of ^{137}Cs, $A(t)$=activity after time t and $t_{1/2}$=half-life ($t_{1/2Cs}$ =30.08 years (National Nuclear Data Center)).

The distribution of the surface activity of ^{137}Cs in 1994 and for 2007–2010 is presented in Figs. 2 and 3. The letters A–D mark the parts of the study area that had the highest surface ^{137}Cs activity.

It should be noted that the ^{137}Cs activity distribution shown in the map for 1994 (Fig. 2) does not cover the whole area that was studied in 2007 and 2010, but that it mainly covers the area bounded by the co-ordinates: 50°–51° N and 15.5°–18.5° E.

Figure 4 presents a semi-variogram of how the surface ^{137}Cs activity changed in the study area in the years 2007–2010.

The theoretical model of the semi-variogram accurately describes the dependence defined by the experimental semi-variogram and suggests that the assumed model of the spatial distribution of surface ^{137}Cs activities that was used to prepare the map shown in Fig. 3 was accurate.

The distribution of the specific ^{137}Cs activity accumulated in mosses and lichens is shown in Figs. 5 and 6. As mentioned before, mosses and lichens are good biosorbents of heavy metals and ^{137}Cs.

The maps (Figs. 5 and 6) were based on theoretical models of semi-variograms, which showed good agreement with experimental semi-variograms as shown in Fig. 4.

Compared with the surface ^{137}Cs activity of soil, the highest specific activities in mosses and lichens were recorded at locations A and B.

Discussion

To meet the objectives of the study, we analysed soils to help us understand historical changes in the surface

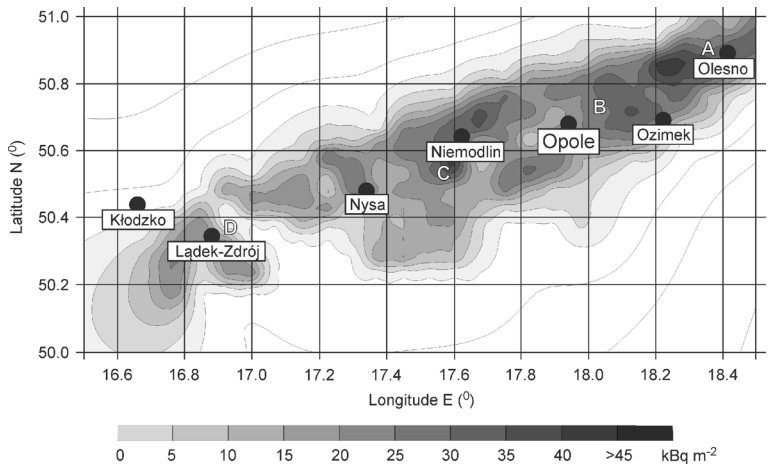

Fig. 2 Distribution of surface activity of ^{137}Cs in the area of the Opole Anomaly in 1994 (Jagielak et al. 1997), calculated for 2 April 2013

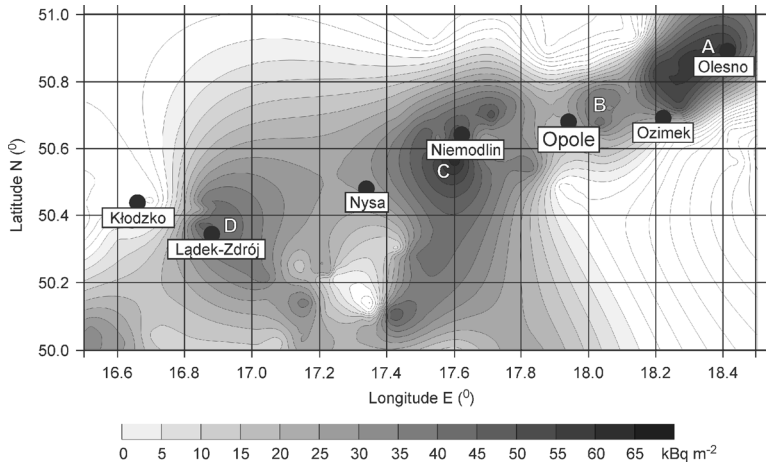

Fig. 3 Distribution of surface ^{137}Cs activity in the Opole Anomaly for 2007–2010, calculated for 2 April 2013

^{137}Cs activity accumulated in soil. The statistical parameters of surface ^{137}Cs activities in the study area, calculated using formula 1 as at 2 April 2013, based on data from 1994, 2007 and 2010, are shown in Table 2. For comparison, data from 2007 and 2010 were back-calculated for 1 June 1986.

There was little variation in the values of statistical parameters for surface ^{137}Cs activity calculated for 2 April 2013, based on data from 1994, 2007 and 2010. It has been shown that changes in the surface activity of ^{137}Cs in soil are mainly due to radioactive disintegration of the radionuclide. This can be confirmed by comparing the surface activity concentrations of ^{137}Cs, measured in 2007 and 2010. If we assume that the reduction in the amount of the radionuclide is due only to radioactive decay, then, based on formula 1, ^{137}Cs activity concentrations have decreased such that its activity ratio a_{2007}/a_{2010} in those two measurement periods lies within the range 1.09–1.12. This activity ratio was calculated for five measurement points located within 2 km of each other. The mean of the calculated ratios was 1.13, which is slightly higher than that estimated by the model that assumes that change in radionuclide concentrations at a given location is due only to radioactive disintegration.

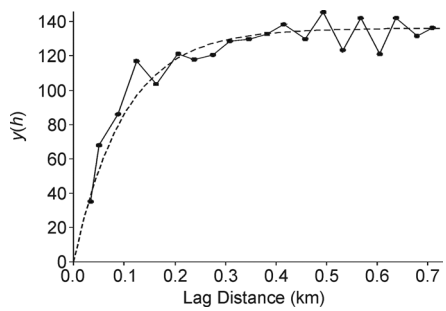

Fig. 4 Variability in surface ^{137}Cs activity in the study area for 2007–2010

The results show limited ^{137}Cs migration into the soil profiles. It has been demonstrated that approximately 90 % of ^{137}Cs is stored in the top 10 cm of soil (e.g. Takenaka et al. 1998; Beli et al. 1994; Ziembik et al. 2010).

The values of surface ^{137}Cs activity in soil, back-calculated for 1 June 1986 (Table 2), show that the amount of ^{137}Cs deposited after the Chernobyl nuclear power plant breakdown in the Opole Anomaly area (mean 26.4, max. 125 kBq m^{-2}) is comparable to that deposited in areas of Southern Belarus (Knatko 1996).

We compared the specific ^{137}Cs activity accumulated in mosses and lichens using values of their respective specific ^{137}Cs activities from the maps (Figs. 5 and 6) for locations where the geographical network coincided with the map. This comparison showed a linear correlation as follows: $a_{moss}=0.952 \cdot a_{lichen}-0.03$. This linear correlation method has been used in other studies to estimate recent deposition of various analytes (Kłos et al. 2010, 2011). Numerous examples show that recent deposition of analytes can be defined by the comparison factor (CF) >0.62 ($CF=2\times(c_{lichen}-c_{moss})/(c_{lichen}+c_{moss})$, where c_{lichen} and c_{moss}=concentration of an analyte in lichens and mosses, respectively). The mean CF value (0.15) calculated from measurements of the specific ^{137}Cs activity in mosses and lichens shows that it did not drift in 2010, which agrees with the results published by Message (2010).

Surface ^{137}Cs activity in soil (Fig. 3) and radionuclide activity in mosses and lichens were not

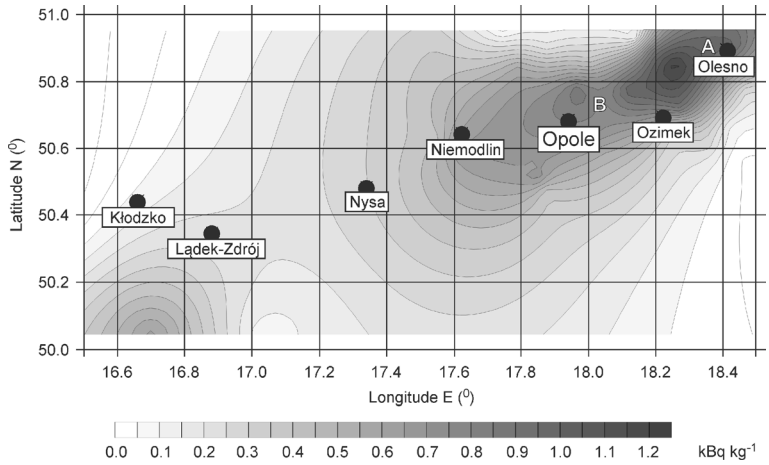

Fig. 5 Distribution of the specific ^{137}Cs activity in mosses collected in 2010

significantly correlated (Figs. 5 and 6), nearly 25 years after the ^{137}Cs deposition. As stated before, the maximum activities overlap only at points A and B marked on the map. Therefore, the specific ^{137}Cs activity in mosses and lichens cannot be used as an indicator of the surface activity of soil. Studies have suggested that metal cations may translocate from soil to epigeic mosses and epiphytic lichens via dust from soil and, in the case of mosses, via water (Kłos et al. 2012). However, the bioavailability of ^{137}Cs differs, depending on the physical and chemical characteristics of soil and of dust that originates from the soil. Statistically significant correlations between the specific ^{137}Cs activity in mosses and lichens and the specific ^{137}Cs activity of mobile forms in the surface layer of soil released in cation form to a solution with a pH of 3.9 were reported by Kłos et al. (2009). Various studies of the specific ^{137}Cs activity in mosses, carried out in the Yamal Peninsula (Nifontova 1995), large areas of the Ural and Siberia (Nifontova 2006) and near Ekaterinburg, where the radionuclide was deposited in large quantities after the Chernobyl disaster (Nifontova 2006), have shown that the level of ^{137}Cs accumulation is influenced by biological half-life time ($t_{B1/2}$). The completed studies clearly suggest that $t_{B1/2}<t_{1/2}$. In summary, accumulation of radiocaesium and other pollutants in mosses and lichens depends on the biological half-life and climate conditions, and pollutants in soil reflect polluting atmospheric aerosols. This study, similar to other studies that have documented translocation of ^{137}Cs from soil to higher plants (Tagami et al. 2012; Lukšienė et al. 2013), demonstrates that there may be local dispersion of ^{137}Cs accumulated in soil. ^{137}Cs in the environment is tied up mainly in topsoil, and its circulation is limited to genetic levels O–A, from which it is integrated into the plant structure via the root system, creating a new sub-level Ol

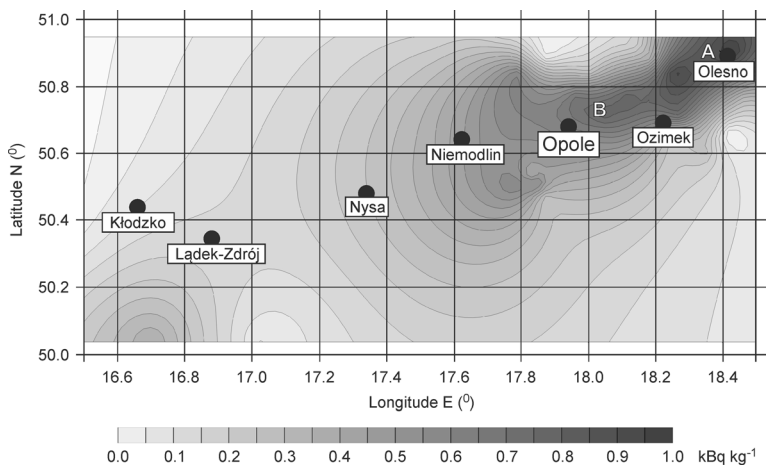

Fig. 6 Distribution of the specific ^{137}Cs activity in lichens collected in 2010

Table 2 Statistical parameters regarding the surface activity concentration of ^{137}Cs in the studied area (kBq m^{-2}), calculated as at 2 April 2013 and 1 June 1986, on the basis of the data from 1994 and the years 2007 and 2010

Parameter	1994 calculated for 2013	2007 and 2010 calculated for 2013	2007 and 2010 calculated for 1986
Minimum	3.2	0.34	0.63
Lower quartile	4.9	3.9	7.3
Median	8.1	9.5	17.6
Mean	11.5	14.3	26.4
Upper quartile	14.6	19.1	35.5
Maximum	>48.6	67.5	125
The standard deviation	8.6	14.8	27.5

over time (Ziembik et al. 2010; Dołhańczuk-Śródka et al. 2006; Dołhańczuk-Śródka and Wacławek 2007). Such local dispersion can be illustrated by wider zones, limited by isolines, created by comparing the results from studies carried out in 2007 and 2010 (Fig. 3) with those from 1994 (Fig. 2). Other studies have shown that radioactive disintegration and migration are the main causes of decreased ^{137}Cs activity in woodlands (Isajenko et al. 2010, 2012).

The lack of correlation between the ^{137}Cs activity in soil, mosses or lichens does not diminish the importance of using these organisms as biomonitoring indicators of radionuclide environmental pollution. Indeed, biomonitoring studies, initiated as early as the 1960s, are still being continued (Yoshida et al. 2004; Adelinė et al. 2006; Celik et al. 2009).

Conclusions

Results from this study show that the decreasing activity of ^{137}Cs deposited in woodlands and wastelands after the breakdown of the Chernobyl nuclear power plant is mainly due to radioactive disintegration of ^{137}Cs. These changes are also, to a lesser degree, caused by the migration of ^{137}Cs with dust from soil and translocation to plants.

Relative to other parts of Poland, high ^{137}Cs activities are still detected in the Opole Anomaly area. The average surface activity of ^{137}Cs calculated for 2 April 2013 was 14.3 kBq m^{-2}, and the maximum surface activity concentration was 125 kBq m^{-2}. ^{137}Cs concentrations detected in the mosses sampled in 2010 were also high, with mean specific concentrations in lichens and mosses of 190 and 155 Bq kg^{-1}, respectively.

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