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DETERMINATION OF THE INITIAL ^{137}CS FALLOUT ON THE AREAS CONTAMINATED BY CHERNOBYL FALLOUT

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Abstract: The fallout radioisotope ^{137}Cs is widely used to study rates and patterns of soil redistribution. This method requires the knowledge about the initial fallout of cesium in the study area. This paper describes the method of establishing the initial fallout of cesium for a study area which is contaminated by Chernobyl fallout. The study was carried out on the loess area near the Ujazd village (South-West Poland). The ^{137}Cs activities for reference soil cores varied from 4.41(24) kBq/m^2 to 5.97(26) kBq/m^2. The average value of the reference inventory of ^{137}Cs for the study area is 5.23(15) kBq/m^2. The calculated contribution of the Chernobyl ^{137}Cs fallout in the total cesium is equal 69 %. Moreover the annual values of the ^{137}Cs fallout based on the precipitation data were calculated and presented. This study provides the method of calculating the ^{137}Cs fallout connected with the nuclear weapon testing based on the precipitation data. Moreover, this study also indicated that the spatial variability on the highly contaminated by Chernobyl cesium study area is small (RSD about 10%) and thus it is possible to use the cesium method to study soil redistribution.

Keywords: ^{137}Cs fallout, soil erosion

1. INTRODUCTION

Cesium-137 is a valuable soil redistribution tracer for many environments (Ritchie and McHenry, 1990). ^{137}Cs was introduced into the atmosphere as a result of nuclear weapon tests and also as a result of the accident of the nuclear power plant in Chernobyl. The main period of cesium from nuclear weapon was in the 1950s and 1960s with the maximum cesium deposition in 1963. ^{137}Cs deposition depends on the latitude and amount of precipitation (Ritchie and McHenry, 1990). After deposition on the ground surface ^{137}Cs is strongly absorbed by the clay minerals (especially by the colloidal fraction) and organic matter in soil. The uptake of cesium by plants is limited. The cesium technique involves measuring of the total inputs of ^{137}Cs on the reference areas with neither erosion nor accumulation visible. The comparison between the value of the ^{137}Cs inventory for the reference site with the ^{137}Cs inventory measured for the disturbed location allows to estimate the intensity of soil redistribution processes. To obtain the qualitative results of soil erosion from the ^{137}Cs data one of the model should be used for calculations. Moreover, the mass balance models to calculate soil erosion based on the cesium data require the annual values of cesium deposition. Nowadays, there is a serious problem to obtain the contribution of the Chernobyl cesium in the total cesium deposition. In the past, it was possible to use the cesium isotope ^{134}Cs for this purpose, which was emitted to atmosphere together with the ^{137}Cs during the nuclear power plant accident in Chernobyl and the ratio between ^{134}Cs and ^{137}Cs is well known. Unfortunately, the half-live time of ^{134}Cs is about 2 years and nowadays the activity of the ^{134}Cs in soil is below the detection limit.

The main assumption of the cesium technique is that cesium deposition was uniformly distributed in the study area. Although for the global cesium deposition this assumption is usually fulfilled, for the areas with high Chernobyl cesium contamination could be a problem.

In this study, the ^{137}Cs concentration in soil from reference sites was measured to establish the initial value of the ^{137}Cs fallout for the study area. Moreover, the calculation of the global cesium fallout based on precipitation

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record was carried out allowing calculating the contribution of the Chernobyl ^{137}Cs fallout in the total ^{137}Cs fallout. This knowledge is necessary to use mathematical models to calculate soil erosion from the cesium activity data.

2. THE STUDY AREA AND METHODS

The study area is located an a loess area on the Proboszczowicki tableland, near Ujazd village, SW Poland (50°24´ N, 18°24´ E). The study area is a part of the Raciborska valley which is a south part of the Śląsk lowland, the altitude ranging between 200 and 220 m above sea level. The thickness of the loess layer for the study area is about 10 meters. The study area is used mainly as an agricultural field. This fact in connection with the presence of slopes up to 12° makes soil erosion a serious problem for the study area. The average annual rainfall is 675 mm, with a range from 277 in 1953 to 933 mm in 1981. For the study area the highest rainfall intensity is in July.

The study area is located in the region where larger ^{137}Cs concentration in soil may be expected due to the Chernobyl accident. Concentration of Chernobyl ^{137}Cs varies widely even within one field (Dubois *et al.*, 2003; Strzelecki *et al.*, 1994; Strzelecki *et al.*, 1992). Quantities of Chernobyl ^{137}Cs depend on the trajectories of main radioactivity clouds, and on the precipitation in the area of interest at that time (Stach, 1996; Strzelecki et al., 1992; Strzelecki *et al.*, 1994).

All soil samples were collected during the fall 2003 from uncultivated places with neither soil erosion nor deposition visible. In total, 12 soil cores by means of a 80 mm diameter corer were collected. The samples were collected to the depth of 60 cm and each soil core was sectioned in 10 cm intervals.

All soil samples were first dried in a dryer at 60°C until the weight was not changing any more in 24 hours. Samples were then sieved through a 2 mm mesh, to remove stones and visible organics parts, and carefully mixed to homogenize. Measurements of ^{137}Cs activities in soil samples were carried out by means of gamma-ray spectrometry, using high resolution high purity germanium detector. The counting time of each sample was usually 24 hours. The resolution of the germanium detector was 2 keV at 1333 keV. The detection limit was equal to 0.5 Bq/kg. As a reference, IAEA-375 standards were used. Count rates were determined by peak fitting using commercially available software GENIE PC manufactured by CANBERRA and the ^{137}Cs activities in the soil samples were decay corrected to the date of sampling.

The ^{137}Cs inventory, i.e. the total ^{137}Cs activity per unit surface area, for a given sampling point was calculated as follows (Sutherland, 1992; Poręba *et al.*, 2003):

$$Cs_{inv} = \sum_{i=1}^{n} C_i \cdot BD_i \cdot DI_i \qquad (2.1)$$

where:
Cs_{inv} is the ^{137}Cs inventory (Bq/m^2), i is the sample index, n is the number of the deepest sample with detectable ^{137}Cs,
C_i is the activity of ^{137}Cs in i-th soil sample (Bq/kg),
BD_i is the air-dry bulk density of the soil (kg/m^3),
DI_i is the thickness of i-th sample (m).

3. RESULTS

Results of the ^{137}Cs in reference soil cores

The values and uncertainties of ^{137}Cs inventories measured in the soil cores collected from the study area are presented in **Table 1**. The ^{137}Cs activities for reference soil cores varied from 4.41(24) kBq/m^2 to 5.97(26) kBq/m^2. The mean value of ^{137}Cs inventory for the study area is 5.23 kBq/m^2. The standard deviation is 0.53 kBq/m^2 and the standard deviation for the mean value is 0.15 kBq/m^2. The relative standard deviation of the ^{137}Cs inventories in reference sampling points is 10.1 % which is in good agreement with the results published by other authors (Golosov *et al.*, 1999; Golosov, 2003; Sutherland, 1994; Bacchi *et al.*, 2003). The obtained mean value of ^{137}Cs inventory for the Ujazd area is higher than the mean value for Poland (3,65 kBq/m^2 for 1996 year), but it is in good agreement with the mean values of ^{137}Cs inventory for the province of Opole and the former province of Katowice. For those provinces, the mean values of ^{137}Cs inventories are equal to 11.24 kBq/m^2 and 6.80 kBq/m^2 respectively (Stach, 1996). Moreover, for those provinces the contribution of Chernobyl ^{137}Cs fallout in the total ^{137}Cs fallout is about 80 % or more.

The results of calculation of the global cesium deposition based on the precipitation

To obtain the part of the ^{137}Cs fallout connected with the nuclear weapon testing for the study area the formula described by Sarmiento and Gwinn was used (Sarmeinto and Gwinn, 1986). Originally this formula was provided to calculate the ^{90}Sr fallout per unit area as a function of precipitation rate. Fortunately, the relation between the ^{137}Cs fallout and ^{90}Sr fallout is known and thus the model delivered for ^{90}Sr fallout prediction could be used to cal-

Table 1. *The results of ^{137}Cs deposition measurements in Ujazd area.*

Number of the reference core	Activity of ^{137}Cs (Bq/m^2)	Uncertainty (Bq/m^2)
R_1	5850	280
R_2	5040	230
R_3	5550	250
R_4	4870	300
R_5	5970	260
R_6	5400	240
R_7	4820	190
R_8	4530	200
R_9	5650	190
R_{10}	4410	240
R_{11}	5750	200
R_{12}	4890	240
Mean value of ^{137}Cs deposition (Bq/m^2)	5230	
Standard deviation (Bq/m^2)	530	
Standard deviation of mean value (Bq/m^2)	150	

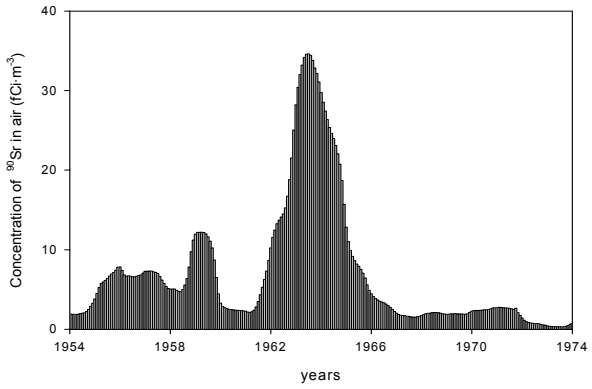

Fig. 1. *The value* $\overline{C}_{ref}(t)$ *for the years 1954-1974.*

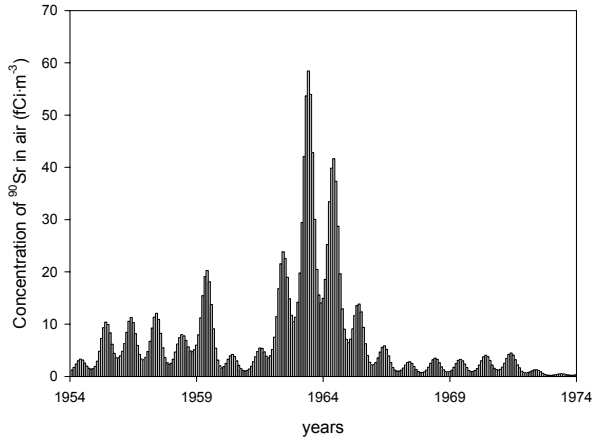

Fig. 2. *The calculated values of* $C(\phi, t)$ *for the Ujazd for the years 1954-1974.*

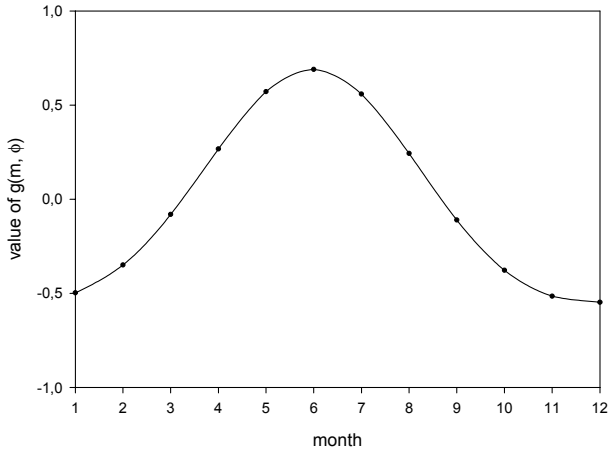

Fig. 3. *The calculated values of* $g(m, \phi)$ *for the Ujazd area.*

culate the ^{137}Cs fallout. Moreover, by this method the values of annual deposition of ^{137}Cs could be calculated (Lu and Higgitt, 2000). This knowledge is necessary for using mass balance models to calculate soil erosion (Walling and He, 1999).

The ^{90}Sr fallout is described by the equation (Sarmiento and Gwinn, 1986):

$$F(\phi,t) = C(\phi,t)\left[v_d(\phi) + v_w(\phi,t)\right] \tag{3.1}$$

where:
$F(\phi, t)$ – monthly ^{90}Sr deposition; fCi/cm^2/s;
$C(\phi, t)$ – ^{90}Sr concentration in air near the ground surface for given month; fCi/cm^3;
$v_d(\phi)$ – dry deposition rate; cm/s;
$v_w(\phi, t)$ – wet deposition rate; cm/s.
ϕ – latitude;
t – time

The wet deposition rate could be calculated from equation:

$$v_w(\phi,t) = a(\phi)\left[P(\phi,t)/P_0\right]^{b(\phi)} \tag{3.2}$$

where:
$a(\phi)$ and $b(\phi)$ – parameters for wet deposition rate for a given latitude,
$P(\phi, t)$ – mean monthly precipitation rate; cm/month,
P_0 = 1 cm/month.

Concentration of ^{90}Sr in air near the ground surface $C(\phi, t)$ could be described by the equation:

$$C(\phi,t) = R(\phi)\overline{C}_{ref}(t)\left[1 + g(m,\phi)\right] \tag{3.3}$$

where:
$R(\phi)$ – the relation between mean annual concentration of ^{90}Sr in air for a given latitude to mean annual concentration of ^{90}Sr in air for reference latitude,
$\overline{C}_{ref}(t)$ – mean annual concentration of ^{90}Sr in air for the reference latitude; fCi/m^3;
$g(m, \phi)$ – inter annual relative deviations of ^{90}Sr concentration in air for a given latitude ϕ and for given month m.

The $\overline{C}_{ref}(t)$ values are tabulated by Sarmiento and Gwinn (1986) whereas the values $R(\phi)$ and $g(m, \phi)$ as well as parameters $a(\phi)$ and $b(\phi)$ could be calculated for a given localization based on Tables provided by the authors (Sarmiento and Gwinn, 1986). The values of $\overline{C}_{ref}(t)$ for 1954 to 1974 are presented in **Fig. 1**, whereas the calculated values of $C(\phi, t)$ and $g(m,\phi)$ for the study area were presented in **Fig. 2** and **3**, respectively.

The calculated value of ^{137}Cs deposition connected with the nuclear weapon testing based on the precipitation data and Sarmiento-Gwinn model for the study area is 1.61 kBq/m^2. Moreover, by this model were calculated the annual values of ^{137}Cs deposition, which is necessary for mass balance model to calculate soil erosion. The annual values of ^{137}Cs are presented in **Fig. 4**, whereas the cumulated annual values of ^{137}Cs deposition are presented on **Fig. 5**. It is clearly visible, that for the years 1962-1964 occurred more than 48% of the cesium deposition due to nuclear weapon tests and the highest intensity of the global cesium deposition was in 1963. The cesium deposition connected with the Chernobyl accident could be calculated as a difference between the total cesium deposition measured in reference sites and the estimated value of global cesium deposition based on the precipitation record. For the study area the about 69% of

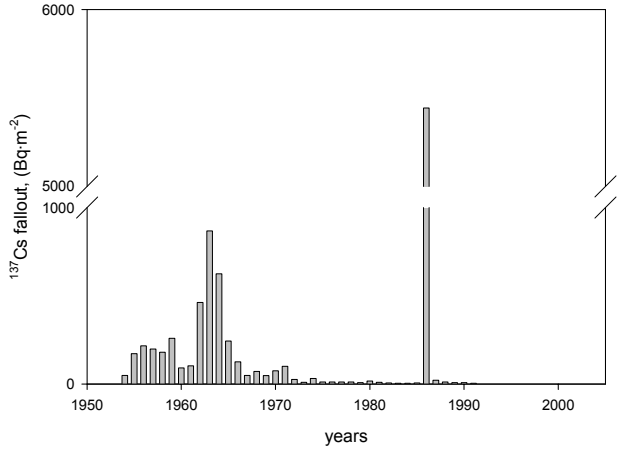

Fig. 4. *Calculated values of annual ^{137}Cs deposition based on the precipitation records according to Sarmiento-Gwinn model for Ujazd area.*

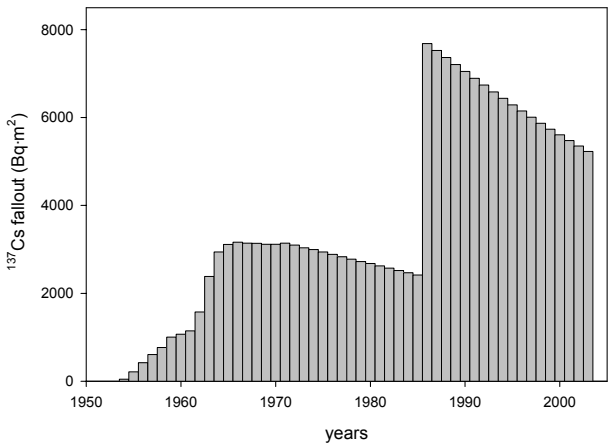

Fig. 5. *The cumulated values of ^{137}Cs deposition for Ujazd area.*

the total cesium deposition is connected with the Chernobyl accident.

5. CONCLUSION

The results presented in **Table 1** clearly confirm the potential for using ^{137}Cs measurements to investigate the soil redistribution in the study area which was affected by contamination with cesium from the Chernobyl fallout. The spatial variability of the ^{137}Cs inventory for the investigated location is quite low and is similar for an area uncontaminated by Chernobyl cesium. The contribution of the Chernobyl cesium in investigated soil profiles for the location under study constitutes about 69 % of the total. It was found, that the presence of Chernobyl cesium does not influence the potential to use cesium as a soil erosion tracer in the investigated area.

The information about both total cesium deposition and annual values of cesium deposition is essential for using cesium as a soil erosion tracer. Here, using measurements for the reference site only the total cesium deposition could be obtained; in fact, there are only few places in the world where cesium deposition has been measured for the whole nuclear weapon test period. The method used in the present work to calculate annual values of global cesium deposition based on the precipitation data and the Sarmiento-Gwinn model seems to be a valuable tool in the case of lack of other data.

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