

# CONCENTRATION AND VERTICAL DISTRIBUTION OF $^{137}\text{Cs}$ IN AGRICULTURAL AND UNDISTURBED SOILS FROM CHECHŁO and CZARNOCIN AREAS

GRZEGORZ POREBA<sup>1</sup>, ANDRZEJ BLUSZCZ<sup>1</sup> and ZBIGNIEW ŚNIESZKO<sup>2</sup>

<sup>1</sup>Department of Radioisotopes, Institute of Physics, Silesian University of Technology, Krzywoustego 2, 44-100 Gliwice, Poland

<sup>2</sup> Faculty of Earth Science, Silesian University, Będzińska 60, 41-200 Sosnowiec, Poland (e-mail: poreba@polsl.gliwice.pl)

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**Abstract:** This paper presents the results of measurements of  $^{137}\text{Cs}$  in soil profiles which were sampled in the Chechło and Zalesie areas, on the Proboszczowicka Highland, Poland, May 2002. Samples were collected from four soil profiles, three of them were taken on the cultivated field and one on the undisturbed field. The vertical distribution of  $^{137}\text{Cs}$  in collected profiles and the  $^{137}\text{Cs}$  inventory for all locations have been determined. The pH of soils (in distilled  $\text{H}_2\text{O}$  and KCl solution) and the organic matter content of soil samples were also measured. Caesium concentration ranged from 0.0 to 15.37 Bq/kg and from 0.0 to 101.61 Bq/kg in soils from cultivated and undisturbed sites, respectively. The measured  $^{137}\text{Cs}$  deposition has been compared with the nuclear weapon tests caesium fallout. The obtained results show that the Chernobyl  $^{137}\text{Cs}$  fallout had relatively large contribution to total  $^{137}\text{Cs}$  deposition in the study areas.

## 1. INTRODUCTION

The fallout of  $^{137}\text{Cs}$  has been used as a valuable sediment tracer in many environmental study cases, especially to provide information on rates of soil erosion (Elliot *et al.*, 1990; Lowrance *et al.*, 1988; Walling *et al.*, 1999), or rates of lacustrine sedimentation (Walling *et al.*, 1993). Caesium got to environment as a result of nuclear weapon tests and accidents in nuclear power stations. The main period of nuclear weapon caesium deposition was between 1958 and 1963; there was also a minor period of caesium deposition from 1971 to 1974 (Ritchie and McHenry, 1990).

Nuclear weapon caesium is being deposited globally on the surface of the Earth. The global caesium fallout depends on the latitude and precipitation (Ritchie and McHenry, 1990; Stach, 1996; Lee and Lee, 1997; Hien *et al.*, 2002). On the Earth's northern hemisphere the fallout is greater than on the southern one. The maximum value of fallout of  $^{137}\text{Cs}$  appears at about 45° latitude North and minimum at the equator (Davis, 1963).

In contrast, the deposition of caesium released during nuclear power plant accidents is local and strongly depends on meteorological conditions. The accident most relevant for our case took place in Chernobyl, in April 1986, and resulted in strongly spatially varying deposition

(e.g., Dubois and Bossew, 2003). Also in Poland values of Chernobyl caesium deposition are far from uniform and depended on the trajectories of main radioactivity clouds, and on the precipitation in the area of interest at that time (Stach, 1996).

Caesium is rapidly and strongly adsorbed by soil particles, especially by clay minerals (vermiculite in particular) (Tamura, 1964; Staunton *et al.*, 2002). The adsorption of  $^{137}\text{Cs}$  is mainly chemical and occurs through an ion exchange process (Staunton *et al.*, 2002). The adsorption capacity of the soil depends on the content of organic matter. When low (<40%), the abundance of clay minerals is enough for strong fixation of  $^{137}\text{Cs}$ , but if content of organic matter is higher, clay minerals are not sufficient, and also migration of caesium is higher. Generally, in soil with high organic matter content the adsorption of caesium is reversible and caesium is more available for uptake by plants (Valcke and Cremers, 1994). The adsorption of caesium is decreased by presence of competing ions of K or Na (Coleman *et al.*, 1963).

Caesium is carried to soil in several ways: direct deposition from atmosphere, wash-off from vegetation, turnover from vegetation, redeposition of eroded soil particles, and deposition from water on floodplains and coastal regions (Ritchie and Henry, 1990). Caesium is present in

atmosphere and precipitates to soil surface as a fallout which can be modified by over ground parts of plants. Even if fully adsorbed on soil particles, caesium may be mobilised within the soil profile due to pedoturbation processes (Southard and Graham, 1992). Caesium can also be taken by plants. The transfer of caesium from soil to plants is a more complex mechanism than simple ion exchange, which once was suggested. Recent studies point to the role of microorganisms in the retention and bioavailability of caesium in soils, for example the mycorrhiza fungi modify the effective area of plant roots (Drissner *et al.*, 1997). From an agricultural field caesium may be also removed with crops.

Generally, migration of caesium is a complicated process and is affected by a number of factors like type of soil, its chemical properties, or organic matter content. Additionally the mobility of caesium in soil depends on climatic conditions (such as rainfall, temperature, or humidity), and biological activity of microorganisms in soil.

There are only few places in the world where deposition of  $^{137}\text{Cs}$  have been measured since nuclear weapon tests started but the rate of fallout must be known (Ritchie and McHenry, 1990) for a purpose of using  $^{137}\text{Cs}$  method to study soil erosion. In Poland the deposition of  $^{137}\text{Cs}$  has been measured since 1970 (Stach, 1996). The initial caesium deposition can be determined by relation to environmental properties such as precipitation or by direct measurements in locations where neither erosion nor soil deposition has occurred.

## 2. MATERIAL

### Description of sites

Studied samples were taken from four locations on the Proboszczowicka Highland (Fig. 1). The soil type in all locations is loessial. Three of them were located on the agricultural field: Chechło 1 – base of the slope, old detrital fan (alluvial fan) cut by the drain; Chechło 2 – base

of the slope near the border of agricultural field; Chechło 3 – middle of the slope. The inclination of the slope is about 10-12°, the latitude is 50°24' N and longitude 18°24' E. Elevations of sites are in the range of 207 m to 221 m. The fourth sampling site (Czarnocin area) is located on the slope covered by forest (beech *Fagus sp.* and spruce *Picea*). This forest has existed for more than 50 years, i.e., from the time before the first deposition of  $^{137}\text{Cs}$ . The site co-ordinates are 50°26' N and 18°14' E, and the altitude is about 255 m.

All study areas are located in the region where larger  $^{137}\text{Cs}$  concentration in soil may be expected due to the Chernobyl accident. Concentration of Chernobyl  $^{137}\text{Cs}$  varies widely even within one field (Dubois and Bossew, 2003; Strzelecki *et al.*, 1994; Szewczyk, 1990; Szewczyk, 1994). Values of Chernobyl  $^{137}\text{Cs}$  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.*, 1994). The increased values of Chernobyl  $^{137}\text{Cs}$  concentration are found along the line Warsaw – Opole – the Kłodzko Valley. The largest values of Chernobyl  $^{137}\text{Cs}$  fallout were found in Voievodships of Opole and Silesia, and in the Kłodzko Valley in the Sudetes Mts. In these areas values of Chernobyl  $^{137}\text{Cs}$  concentration were between 15.6 kBq/m<sup>2</sup> and 19.3 kBq/m<sup>2</sup>, with the maximum value of 96 kBq/m<sup>2</sup> in the Nysa area (Stach, 1996; Strzelecki *et al.*, 1994). A strong spatial variability of  $^{137}\text{Cs}$  concentration from the Chernobyl accident makes the use of  $^{137}\text{Cs}$  in soil erosion studies more difficult.

### Sampling and laboratory procedures

The soil samples were collected in May 2002 by means of the 120 mm diameter borer. The samples were collected from the Chechło site 1 to the depth of 70 cm and from Chechło sites 2 and 3 to the depth of 50 cm. Each core was sectioned at 10 cm intervals. In a case of Czarnocin site soil samples were collected to the depth 50 cm and

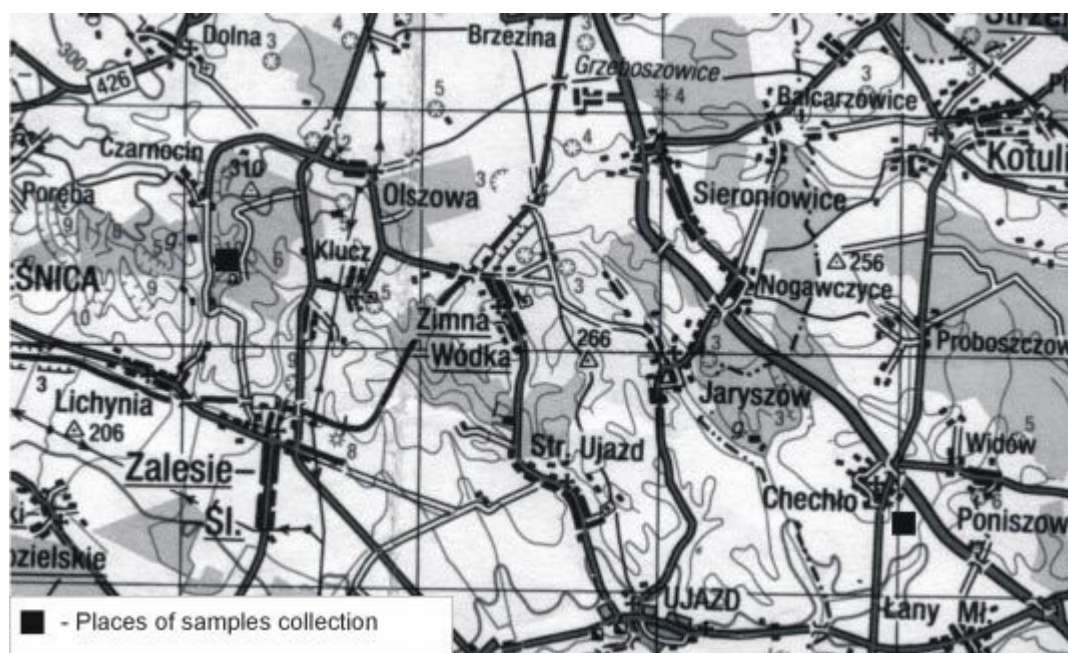


Fig. 1. The study area and the soil sampling places.

core was also sectioned at intervals of 10 cm (Czarnocin 1). Besides, an additional material was sampled from the uppermost 10 cm of this soil in 2 cm slides (Czarnocin 2).

All samples were first dried until their mass was constant. Afterwards samples were sieved to remove stones or visible organic parts and carefully mixed and placed in 0.65 dm<sup>3</sup> Marinelli beakers closed tightly (recommended for measurements of low-radioactivity samples). For smaller samples we used plastic boxes 3.5 cm high and 7.9 cm diameter (0.25 dm<sup>3</sup> volume). The <sup>137</sup>Cs concentration in soil samples were measured by means of a high-resolution gamma HPGe detector manufactured by CANBERRA (resolution 2.0keV at 1.33MeV; relative efficiency 30%). Detection limit for the Marinelli geometry is 0.5 Bq/kg while for the smaller box it is 1.0 Bq/kg. The detector is placed inside the low-background lead shield and cooled by a cryostat immersed in liquid nitrogen. Samples were counted for approximately 20 hours. The reference soil IAEA-375 (distributed by Laboratory of Seibersdorf IAEA, Vienna, Austria) was used as a standard of <sup>137</sup>Cs activity. We used 661.7 keV gamma peak and calculated <sup>137</sup>Cs activities with an aid of GENIE-PC software manufactured also by CANBERRA. Finally, specific activities of <sup>137</sup>Cs in soil were expressed in Bq/kg of dry mass of soil and corrected for the time elapsed since sample collection in the field.

Furthermore, total organic matter contents and pH values have been determined for all soil samples. Total organic matter contents were determined using a loss-on-ignition method. The values of pH were measured in water and in 1n water solution of KCl (soil suspended in liquid in proportion 1:2.5) and the results are shown in **Table 1**. For study areas Chechło and Czarnocin the organic matter content in soil samples are about 7.0-10.0% and about 2.5-5.5%, respectively.

### 3. RESULTS AND DISCUSSION

#### *Vertical distribution of <sup>137</sup>Cs in soil*

##### Chechło sites

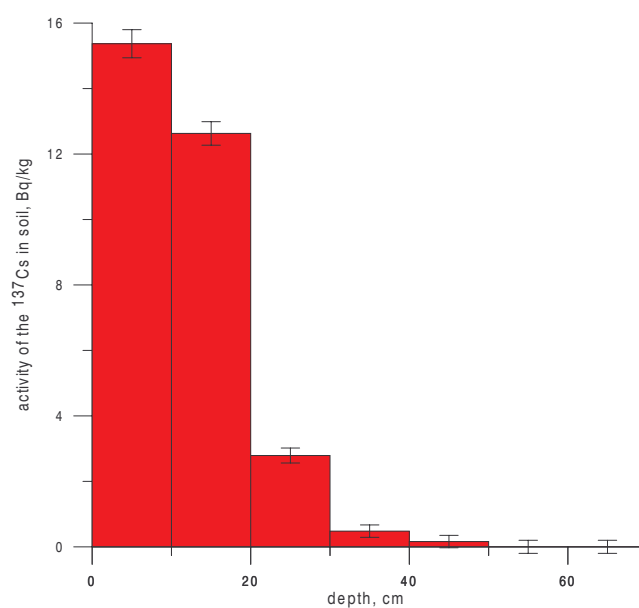
The vertical distributions of <sup>137</sup>Cs for soil profiles in Chechło areas are shown in **Figs. 2, 3** and **4**. The depth distributions of <sup>137</sup>Cs concentrations are similar to those reported for disturbed soils (Walling *et al.*, 1999; He and Walling, 1997). For all collected cores <sup>137</sup>Cs concentrations drastically decrease at the plough depth. The <sup>137</sup>Cs is almost uniformly distributed within the plough layer and then decreases sharply. Similar results were obtained for Odonów areas (Śnieszko *et al.*, in print). For core Chechło

**Table 1.** Results of measurement pH in soil samples from Chechło and Czarnocin.

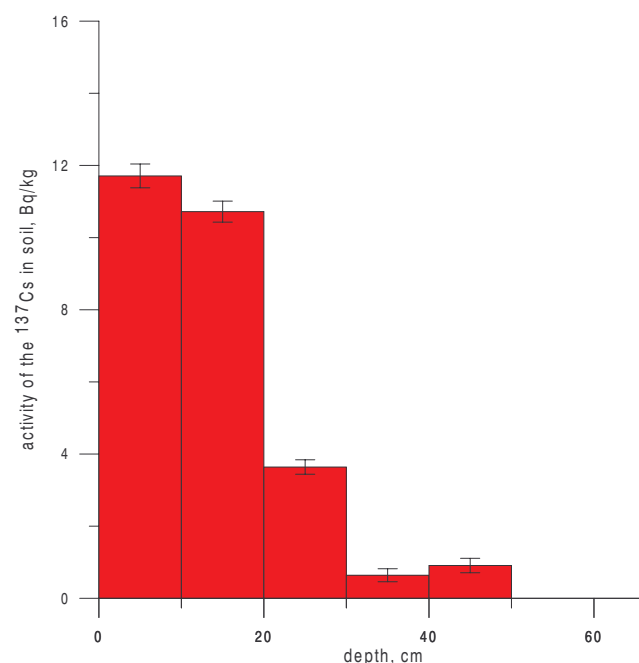
| Site      | The range of pH in water | The range of pH in KCl |
|-----------|--------------------------|------------------------|
| Chechło 1 | 5.50-6.55                | 4.65-5.79              |
| Chechło 2 | 5.66-6.06                | 4.65-4.83              |
| Chechło 3 | 4.95-6.27                | 4.21-5.26              |
| Czarnocin | 4.04-5.01                | 3.13-3.51              |

1 from the bottom of the slope, high concentrations of <sup>137</sup>Cs are observed in the first two 10 cm soil layers. Concentration of <sup>137</sup>Cs decreases gradually in the next soil layers. There is no <sup>137</sup>Cs below the depth of 50 cm in soil profile Chechło 1 which was sampled to a depth of 70 cm.

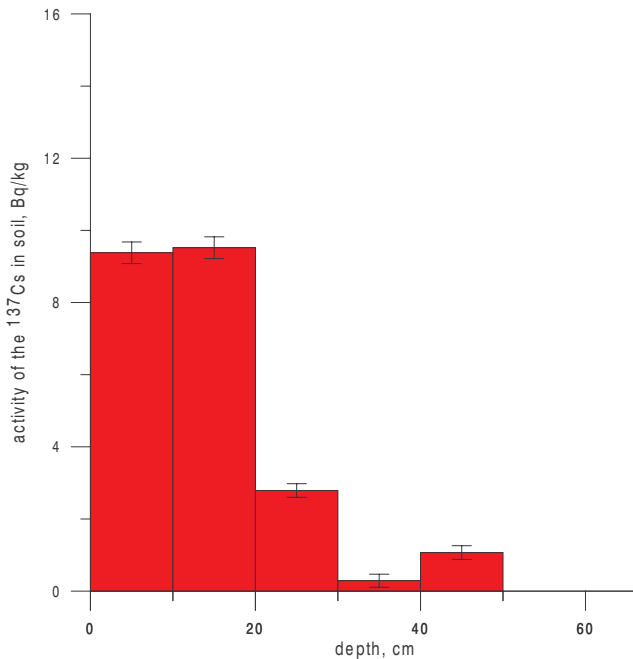
Cores Chechło 2 and 3 are located at the border between the cultivated field and meadow and on a slope on a cultivated field respectively. They have caesium profiles of similar shape. The <sup>137</sup>Cs concentrations are high within the plough range, similarly to what was found in Chechło 1. For Chechło 2 and 3 sites the highest values of the <sup>137</sup>Cs concentration are found in the first 20 cm, then it decreases to the value about 3 Bq/kg in the third one and to about the detection limit in the fourth one. <sup>137</sup>Cs concen-



**Fig. 2.** Vertical distribution of the <sup>137</sup>Cs in the soil profile at Chechło 1 site.



**Fig. 3.** Vertical distribution of the <sup>137</sup>Cs in the soil profile at Chechło 2 site.



**Fig. 4.** Vertical distribution of the  $^{137}\text{Cs}$  in the soil profile at Chechło 3 site.

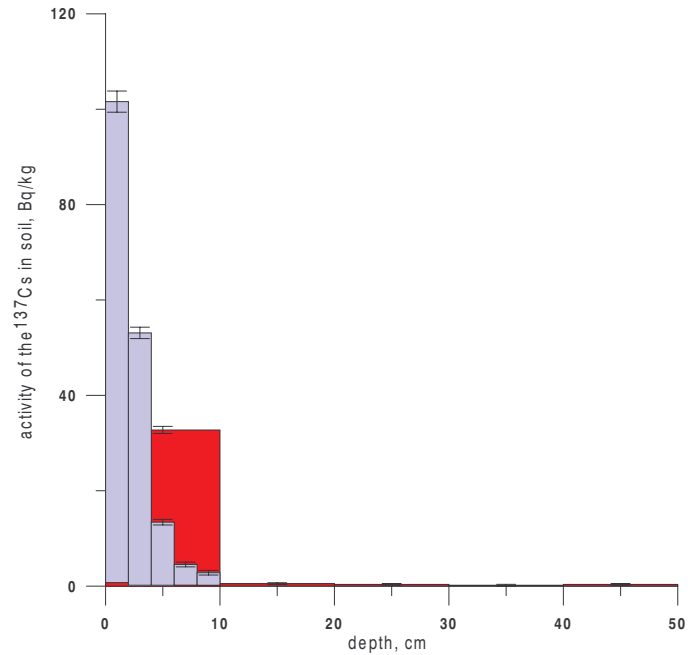
tration in the fifth soil layers (40-50 cm) increases to the value about 1 Bq/kg. Similar results was obtained for Kłodzko Valey (Bluszcz and Śnieszko, 2000). This differs these soil profiles from the soil profile Chechło 1. The behavior of  $^{137}\text{Cs}$  in soils Chechło 2 and 3 may be explained in two ways: first: the caesium was transported with water in solution and second, the caesium was transported in suspension adsorbed on the finest soil grains. It should be noted that for Chechło 2 and 3 the maximum depth of caesium occurrence in soil is unknown because the sampling depth was smaller than in case of Chechło 1.

For all soil profiles from Chechło site the caesium profiles have similar shape. Caesium is almost uniformly distributed within the plough layer. Then, for the next soil layer, the concentration of the  $^{137}\text{Cs}$  decreases to about a quarter of the value for the first layer. This pattern may be surely related to mechanical mixing of soil within the plough depth of ca. 22 to 28 cm.

#### Czarnocin site

The vertical distribution of  $^{137}\text{Cs}$  in undisturbed soil was investigated at Czarnocin site. Material was sampled as at the other sites in 10 cm depth steps down to 50 cm, and additionally the uppermost 10 cm was sampled again with higher resolution at 2 cm depth steps. Vertical distribution of caesium in the soil profile from Czarnocin site is presented in **Fig. 5**. Light bars lines show  $^{137}\text{Cs}$  concentrations measured in 2 cm slices and dark lines bars in 10 cm sections.

The first section (the topmost 10 cm layer) appeared to contain almost all of the  $^{137}\text{Cs}$  (32.75 Bq/kg). In the next soil layer (10-20 cm from surface) the concentration of  $^{137}\text{Cs}$  is about the detection limit (0.51 Bq/kg), and then, in the third layer,  $^{137}\text{Cs}$  concentration decreases below the detection limit (**Fig. 5**). The higher resolution data were obtained for the 10 cm soil core which was divided in 2 cm



**Fig. 5.** Vertical distribution of the  $^{137}\text{Cs}$  in the soil profiles at Czarnocin 1 and 2 sites. There are two independent cores on this figure. The soil profile at (Czarnocin 1) was collected to a depth 50 cm (with 10 cm resolution) and the soil profile at Czarnocin 2 was collected to a depth 10 cm (with 2 cm resolution).

layers. Vertical distribution of  $^{137}\text{Cs}$  in the additional soil profile from Czarnocin site is presented in **Fig. 5**. The concentration decreases apparently exponentially with depth and fitting the curve 1 (Isaksson and Erlandson, 1998):

$$C(d) = C(0)e^{-\beta d} \quad (3.1)$$

yields  $C(0) = 173.2(13)$  Bq/kg and  $\beta = 0.481(42)$   $\text{cm}^{-1}$ ; where  $C(d)$  is the concentration of the caesium at the depth  $d$ ;  $C(0)$  is the concentration of the caesium at the surface and  $\beta$  is the inverse of the relaxation length. The depth distribution of  $^{137}\text{Cs}$  shows that migration of  $^{137}\text{Cs}$  is relatively slow, similar to what was found in other studies (Isaksson *et al.*, 2001; Ivanov *et al.*, 1997; Schuller *et al.*, 2002).

To recapitulate, almost all of the  $^{137}\text{Cs}$  is contained within upper 10 cm of undisturbed soil (from the Eq. 3.1 may calculate the depth at which the concentration reaches the detection limit – it is about 11 cm in this case). The majority of  $^{137}\text{Cs}$  concentration is found in the top several centimeters in undisturbed soil from Czarnocin site.

#### Calculation of the $^{137}\text{Cs}$ deposition

The  $^{137}\text{Cs}$  inventory at a sampling point was calculated using the following equation (Sutherland, 1992):

$$Cs_{inv} = \sum_{i=1}^n C_i \cdot BD_i \cdot DI_i \quad (3.2)$$

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

**Table 2.** The measured  $^{137}\text{Cs}$  inventory for sampling points (with standard uncertainties  $u(\text{Cs}_{inv})$ ). All values of the  $^{137}\text{Cs}$  inventory in soil samples were corrected for the radioactive decay.

| Site      | $\text{Cs}_{inv}$<br>( $\text{Bq}/\text{m}^2$ ) | $u(\text{Cs}_{inv})$<br>( $\text{Bq}/\text{m}^2$ ) | Site description                  |
|-----------|---|--|-----------------------------------|
| Chechło 1 | 3980  | 230  | bottom of slope, old detrital fan |
| Chechło 2 | $\geq 5090^*$                                   | 220  | close to a border with meadow     |
| Chechło 3 | $\geq 3640^*$                                   | 180  | midslope                          |
| Czarnocin | 5430  | 240  | forest soil                       |

\*Caesium is probably present below the maximum sampling depth.

is the thickness of  $i$ -th sample (m). The  $^{137}\text{Cs}$  inventory values calculated in the above given way are shown in **Table 2**. For Chechło 2 and 3 locations the depth of existence of caesium may be more than the depth of sampling. Thus the calculated caesium inventory for those locations is underestimated.

The Estimated Weapons Caesium (EWCs) deposition for Poland is  $982 \text{ Bq}/\text{m}^2$  (Stach, 1996). The mean value of total (including Chernobyl)  $^{137}\text{Cs}$  inventory for Poland is about  $3770 \text{ Bq}/\text{m}^2$  (Stach, 1996). These values were decay corrected for the day of sample collection. There is no indication of erosion or accumulation processes on Czarnocin site, so a measured  $^{137}\text{Cs}$  inventory value higher than estimated EWCs may be attributed to Chernobyl fallout. The measured inventory of  $5430 \text{ Bq}/\text{m}^2$  is substantially higher than the mean total inventory for Poland. For Chechło locations the  $^{137}\text{Cs}$  inventories are smaller than for the Czarnocin location. This is probably a result of different deposition for agricultural field and forest area (Stach, 1996). Moreover caesium might be removed from a cultivated field by soil erosion and with crops. It has to be noted that only for location Chechło 1 the depth of sampling is enough. Although the soil accumulation is possible for two locations (Chechło 1 and 2) higher value of  $^{137}\text{Cs}$  inventory is observed only for the latter one. Chechło 2 site was located at the bottom of the slope and in this profile the  $^{137}\text{Cs}$  inventory is close to or higher than the inventory in Czarnocin location. Chechło 1 site was also located at the bottom of the slope, but the  $^{137}\text{Cs}$  inventory is much lower than the inventories for Chechło 2 or Czarnocin areas. The  $^{137}\text{Cs}$  inventory for Chechło 1 is similar to the value obtained for the sampling point which was located on the slope (Chechło 3).

To obtain reliable value of the contribution of the  $^{137}\text{Cs}$  from the Chernobyl accident to the total caesium inventory some authors used the ratio of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  activities (Chibowski and Mitura, 1995; Issakson *et al.*, 2001). At the present time, however, the concentration of  $^{134}\text{Cs}$  in soil is below the detection limit because of the relatively short half-life of this isotope ( $T_{1/2} = 2.06 \text{ y}$ ), which makes this method impossible to apply.

## CONCLUSIONS

The present work is the first study for the radiocaesium distribution in the soils on the Proboszczowicka Highland in Poland. The results obtained in this study show that for all soil sampling sites there is a large contribution of Chernobyl  $^{137}\text{Cs}$  fallout to total  $^{137}\text{Cs}$  deposition.

All values of calculated caesium deposition are higher than the estimated weapons caesium deposition for Poland and close to or higher the total mean.

For the non-eroded and non-mixed soil profile (Czarnocin site) the concentration of  $^{137}\text{Cs}$  rapidly decreases with depth (**Fig. 5**). The downward migration of  $^{137}\text{Cs}$  is slow, and many years after deposition caesium still remains in the top dozen centimetre layer. This suggests that caesium is strongly absorbed on soil particles, and allow to use caesium as a marker of sediments younger than about 50 years.

The experimental results for Chechło sites show similar thickness of the layer enriched in  $^{137}\text{Cs}$  (**Figs. 2 - 4**). The thickness of this layer corresponds to the plough depth what probably means that there was no significant accumulation of sediment there.

It is possible, however, that at Chechło 2 and 3 caesium is deposited below the maximum sampling depth. The problem could only be solved by additional and deeper sampling on the field.

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