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¹³⁷Cs AND ⁴⁰K CONCENTRATIONS IN FOREST SOILS AND WASTELANDS IN THE VICINITY OF SIEDLCE (EASTERN POLAND)

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Abstract: The activity of ¹³⁷Cs and ⁴⁰K in forest soils and wastelands in the vicinity of Siedlee (eastern Poland) were measured. The soil samples were collected on depths of 0–4 cm, 4–8 cm and 8–12 cm. The average specific radioactivity of ¹³⁷Cs in forest soils and wastelands were 57 Bq/kg and 15 Bq/kg, respectively. The highest specific radioactivity of ¹³⁷Cs was observed in superficial layers of forest soils with an arithmetic mean of 126 Bq/kq. The average specific radioactivity of ⁴⁰K in the soil samples was 200 Bq/kg independently of sampling depths. Positive correlations were found between ¹³⁷Cs and C_{org} concentrations. Both ¹³⁷Cs and ⁴⁰K were negatively correlated with the sand fraction and positively with silt and clay fractions.

INTRODUCTION

After the breakdown of the power reactor in Chernobyl in 1986, the highest concentration of radionuclides in the area of Poland was noted, for example, in eastern provinces [14]. A few places were registered as the ones where radioactive contamination was much higher than the national average. The neighborhood of Siedlee was one of those places [4, 16].

Out of all radionuclides released as a result of the breakdown of the power reactor in Chernobyl, ¹³⁷Cs isotope ($T_{1/2} = 30.1$ years) is the one which is still present in the environment. Cesium has only one stable isotope (¹³³Cs) and about 20 artificial radioactive isotopes; one of them is ¹³⁷Cs isotope [1]. ¹³⁷Cs isotope undergoes β - and γ -transformation. The content of ¹³⁷Cs depends on the level of contamination after the power reactor breakdown, and on the type of soil. In 2004, the mean radioactivity of cesium in the area of Poland was about 19 Bq/kg [3]. The literature data (e.g. Dołhańczuk-Śródka *et al.* [5]; Kubica [8]; Litorowicz *et al.* [9]; Pachocki *et al.* [12]; Zhiyanski *et al.* [17]) indicate that the surface layers of soil accumulate large amounts of ¹³⁷Cs.

Potassium has three natural isotopes (${}^{39}K$, ${}^{40}K$, ${}^{41}K$). ${}^{40}K$ nuclide is radioactive and it comprises 0.0119% of natural potassium. ${}^{40}K$ nuclei ($T_{1/2} = 1.3 \cdot 10^9$ years) undergo β -

and γ -transformation during which they change into stable ⁴⁰Ca [13]. Soil accumulates a considerable part of ⁴⁰K isotope. The ⁴⁰K radioisotope content depends on the content of stable isotopes of potassium (³⁹K, ⁴¹K) and the type of soil. It is greater in places where potassium fertilizers are used.

Describing the radioactivity of ¹³⁷Cs forest and wasteland soils in the neighbourhood of Siedlce 20 years after the breakdown of the power reactor in Chernobyl was the main aim of studies. The radioactivity of ⁴⁰K isotope was also measured during the studies, mainly because of similar chemical properties of cesium and potassium.

The researches aimed to:

- determine the specific radioactivity of ¹³⁷Cs and ⁴⁰K isotopes in forest and wasteland soils,
- assess the degree of migration of the studied isotopes into the soil profile,
- determine correlations between the specific radioactivity of ¹³⁷Cs and ⁴⁰K in soils and chosen soil properties (reaction, the content of C_{org}, granulometric composition).

THE AREA OF STUDIES

The town of Siedlee $(52^{\circ}10^{\circ} \text{ N}, 22^{\circ}17^{\circ} \text{ E})$ is situated at Siedlee Plateau, which is the part of a bigger geographical unit – the South Podlasie Lowland [7]. The Plateau lies in the area of frontal moraines of the Middle-Polish Glaciations (Warta stage). The terrain is flat; the uplifts reach 190–200 m a.s.l. Brown and podsolic soils predominate in the region.

Study sites were chosen at random. The soil was collected from the sites located northwest and southeast of the town of Siedlce. 9 sampling sites in forests (Mokobody, Kisielany, Opole, Siedlce, Mościbrody, Skórzec, Wiśniew settlement, Wiśniew, Gost-chorz) and 4 sampling sites in the area of wastelands (green strips along the roads running through the following places: Kiesielany, Opole, Mokobody, Wiśniew settlement) were appointed (Fig. 1).



Fig. 1. The location of sampling sites: 1 – Mokobody, 2 – Kisielany, 3 – Opole, 4 – Siedlee, 5 – Mościbrody, 6 – Skórzee, 7 – Wiśniew settlement, 8 – Wiśniew, 9 – Gostchorz

MATERIAL AND METHODS

The soil samples were collected in October and November 2005. The places from which the samples were collected were marked at the map (Fig. 1). The samples were taken from the surface layer of soil, from the depth up to about 12 cm. Soil samples were taken in the form of 12 cm high cores. Then the soil cores were cut into slices; each 4 cm high. Three layers of soil were obtained in this way: level A (0–4 cm), level B (4–8 cm), level C (8–12 cm). In forests, also litter was collected. Each of the soil and litter samples weighed 1 kg. The soil was dried aerially and then washed in a sieve of 2 mm mesh size which allowed to eliminate plant remains and small stones from the samples.

Altogether, 27 samples of forest soils, 9 samples of litter and 12 samples of wasteland soils were prepared for analysis. The following physical and chemical properties of soil were determined: reaction in 1 M KCl, the percentage concentration of organic carbon determined with the use of Tiurin method, granulometric composition determined with use of Bouyoucos method, modified by Casagrande and Prószyński [11].

The activity of ¹³⁷Cs and ⁴⁰K isotopes in the soil and litter samples was measured with use of γ-spectrometry method and semi-conductor spectrometer with coaxial germanium detector made by the Canberra Company. The spectrum analysis was carried out with use of Genie 2000 Applications Software (model S501C). Each sample was measured for 80000 seconds.

The results of the analysis of the radioactivity of ¹³⁷Cs and ⁴⁰K in soils, and of chosen physical and chemical parameters of soils (granulometric composition, the percentage concentration of organic carbon and the reaction of soils) were put to the statistical analysis (Statistica 5.0). Pearson correlation factors were calculated.

RESULTS

Soils chosen for studies were sandy, mainly represented by sand, weakly loamy sand and loamy sand. All the collected soils had similar granulometric composition, which results from the fact that the area of studies was relatively small.

The reaction of the studied soils was very acid and acid; it varied from 2.77 (forest in Gostchorz – A) to 4.75 (wastelands in Niwiski – level A). In forest soils, the reaction increased with every deeper level; it varied from 3.19 (level A) to 3.63 (level C). In wasteland soils, the highest reaction (pH - 4.15) was noted in level A, and the lowest – in level C (pH - 3.85).

The percentage concentration of organic carbon in the collected material varied between 0.4 and 8.83%. In forest soils, the content of C_{org} decreased with depth; the mean content of C_{org} in each level of forest soils was: level A – 3.63%, level B – 2.12% and level C – 0.93%. The mean content of C_{org} in each level of wasteland soils was about 1%.

The mean radioactivity of ¹³⁷Cs in forest soils, at the depth of 12 cm, was 57.5 Bq/kg. The greatest radioactivity of all sampling sites was measured in forest soils taken from Opole – 188.5 Bq/kg and Kisielany – 101.2 Bq/kg. The surface layer of soil (level A) was the richest in ¹³⁷Cs isotope. The radioactivity of cesium in that level was 126.2 Bq/kg. The highest radioactivity was noted in litter with an arithmetic mean of 205.9 Bq/kg (Fig. 2a).

104 ELŻBIETA KRÓLAK, ANETA MAJEWSKA, KATARZYNA SARNOWSKA, JADWIGA KARWOWSKA

The mean radioactivity of ¹³⁷Cs in wasteland soils was 15.6 Bq/kg. The greatest radioactivity (18.2 Bq/kg) was noted in samples taken from Mokobody. No significant differences in the radioactivity of ¹³⁷Cs in each level of wasteland soils were noted (Fig. 2b).



Fig. 2. Average ¹³⁷Cs and ⁴⁰K radioactivity versus sampling depth in (a, c) forest soils, (b, d) wastelands; uncertainties are quoted as one standard deviation (\pm SD), sampling depths: L – litter, A – 0–4 cm, B – 4–8 cm, C – 8–12 cm

The radioactivity measured for 40 K isotope was within the range: 103.6 Bq/kg (level B of Mościbrody forest soil) – 349.0 Bq/kg (level C of Opole forest soil). The mean content of 40 K was: 172.7 Bq/kg in forest soils, and 211.9 Bq/kg in wasteland soils. The values of 40 K radioactivity in each level of the studied soils were almost the same (Fig. 2c, d).

Calculated values of Pearson's (linear) correlation factors indicate that there are statistically significant correlations between the radioactivity of ¹³⁷Cs in soils and the reaction of soil (r = -0.5575, p < 0.001) (Fig. 3a), as well as the percentage concentration of organic carbon (r = 0.8639, p < 0.001) (Fig. 3b). Similar correlations were not noted for ⁴⁰K. The contents of the two radioactive nuclides were correlated with soil fractions: positively with silt and clay fraction, and negatively with the content of sand fraction (Tab. 1).



Fig. 3. Correlations between 137 Cs radioactivity in soil (a) soil reaction and (b) percentage concentration of C_{arr}

Table 1. Pearson correlation factors for ¹³⁷Cs and ⁴⁰K radioactivity and percentage soil fractions, (p - level of significance)

	Soil fractions (mm)		
	2.0-0.05	0.05-0.002	< 0.002
¹³⁷ Cs	-0.3573	0.3599	0.4161
	p = 0.026	p = 0.024	p = 0.008
⁴⁰ K	-0.6383	0.5986	0.4304
	p < 0.001	p < 0.001	p = 0.006

DISCUSSION

The analyses of soil conducted in a few places located in the vicinity of Siedlce showed that soils in the region are still contaminated with radioactive cesium.

The radioactivities of ¹³⁷Cs noted in the studied forest soils are three times higher than the mean contents of this isotope in soils of Poland (about 19 Bq/kg) calculated by Biernacka and Isajenko [3]. Also Chibowski *et al.* [4] and Szewczyk [16] confirm the existence of anomalous zones in the vicinity of Siedlee where, after the breakdown of the power reactor in Chernobyl, significantly higher radiation back-ground was noted as a result of ¹³⁷Cs nuclei disintegration. Our studies show that the anomalies could take place, for example, in the areas located northwest of Siedlee.

The distribution of cesium in forest soils is not the same in all the soil levels. The highest radioactivity of cesium was noted in the surface layers of the forest soil. The data are confirmed by Dołhańczuk-Śródka *et al.* [5], Kubica [8], Litorowicz *et al.* [9], Pachocki *et al.* [12] and Zhiyanski *et al.* [17]. No similar observations were made in wastelands.

105

The occurrence of ¹³⁷Cs is correlated with the soil reaction, the percentage concentration of organic carbon in soil, as well as with the content of silt and clay fractions in soils. The negative coefficient of the correlation between the radioactivity of ¹³⁷Cs and the soil reaction shows that the more acidified the environment the greater the content of cesium. The correlation between the radioactivity of cesium and the percentage concentration of organic matter in soil shows that the content of this element increases with the rise of carbon bound to organic compounds. It is confirmed by the literature data (e.g. Bergeijk *et al.* [2], Dołhańczuk-Śródka *et al.* [6], Zhiyanski *et al.* [17]) which indicate that the organic matter is a filter absorbing, for example, cesium.

The amount of finest fractions of soil has also an influence on the radioactivity of ¹³⁷Cs in the studied soils. Cesium is usually arrested in soils which are abundant in colloidal parts, while soils abundant in sand have little retention of that element [4, 10].

Isotope ⁴⁰K belongs to the group of natural radioactive isotopes which occur in the environment. The mean radioactivity of ⁴⁰K isotope in the soils of Poland is 400 Bq/kg [3]. The results of measurements of the content of ⁴⁰K in soils in the vicinity of Siedlce show that in relation to mean values calculated in the area of Poland, the soils in the neighborhood of Siedlce are poor in potassium. The type of parent rock has a great influence on that. In the studied area, the basement complex consists mainly of gravels, sands and boulder clay [15].

The level of soil has no significant influence on the radioactivity of 40 K in soils in the vicinity of Siedlce. Such observations are different from the data noted by Kubica [8] who indicates that the maximum radioactivity of 40 K is measured in the third level of the soil profile, at the depth of 7–10 cm.

It is worth-noting that the radioactivity of 40 K isotope is correlated with all soil fractions; positively with silt and clay fractions, and negatively with the sand fraction. The content of 40 K in the upper level of soil depends on the content of colloidal parts [4]. That isotope occurs in smaller amounts in sandy soils. Unlike in the case of 137 Cs, no significant correlations between the content of 40 K and the soil reaction, as well as the percentage concentration of organic carbon were noted.

The analysis of the obtained results allowed to follow the way in which two radioactive isotopes, which represent elements of similar chemical properties, accumulate in soil. The difference in the "behavior" of artificial ¹³⁷Cs and naturally occurring ⁴⁰K is distinct.

CONCLUSIONS

- 1. 20 years after the breakdown of a power reactor in Chernobyl, the contamination of soils by ¹³⁷Cs in the vicinity of Siedlce still holds. The highest accumulation of ¹³⁷Cs is noted in forest soils, the smallest – in wasteland soils.
- 2. The highest concentration of radioactive cesium is noted in the surface levels of forest soil; deep into the soil profile, the concentration of that nuclide decreases.
- 3. The occurrence of ¹³⁷Cs is strictly correlated with chosen properties of soil, such as: soil reaction, the percentage concentration of organic carbon and the content of silt and clay fractions.
- 4. In soils in the vicinity of Siedlee, the radioactivity of ¹³⁷Cs is considerably smaller in comparison with the radioactivity of ⁴⁰K.
- 5. The content of ⁴⁰K in soils in the vicinity of Siedlee is about two times smaller in comparison with the mean content of that isotope in the soils of Poland.

6. The ⁴⁰K content is correlated with all soil fractions: negatively with the sand fraction and positively with the silt and clay fractions.

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ZAWARTOŚĆ ¹³⁷Cs I ⁴⁰K W GLEBACH LEŚNYCH I NIEUŻYTKACH ROLNYCH OKOLIC SIEDLEC (WSCHODNIA POLSKA)

W glebach leśnych i nieużytkach rolnych okolic Siedlec (wschodnia Polska) mierzono aktywność radioizotopów ¹³⁷Cs i ⁴⁰K. Próbki gleby pobierano z glębokości: 0–4 cm, 4–8 cm i 8–12 cm. Średnia aktywność izotopu ¹³⁷Cs w glebach leśnych i nieużytkach wynosiła odpowiednio: 57 Bq/kg i 15 Bq/kg. Największą zawartość ¹³⁷Cs (126 Bq/kg) odnotowano w powierzchniowych warstwach gleb leśnych. Średnia aktywność radioizotopu ⁴⁰K w badanych próbkach gleby wynosiła około 200 Bq/kg i nie zależała od glębokości poboru prób. Odnotowano dodatnią korelację pomiędzy aktywnością ¹³⁷Cs i procentową zawartością węgla organicznego. Zawartości obu radioizotopów była ujemnie skorelowana z zawartością w glebach frakcji piasku i dodatnio z zawartością frakcji pyłu i iłu.