¹³⁷Cs CONTAMINATION AND ITS VERTICAL DISTRIBUTION IN THE CURONIAN SPIT SOIL

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Abstract. In 2003–2004 soil depthprofile sampling was carried out at Juodkrantė in the Curonian Spit. The activity concentrations of anthropogenic radionuclide ¹³⁷Cs in soil samples from the terrestrial and coastal environment of the Curonian Spit were determined and a study of their distribution was performed. The range of activity concentrations of accumulated deposits of ¹³⁷Cs in a 0–30 cm soil layer is 0,2–370,9 Bq·kg⁻¹ and 2,2–11,2 Bq·kg⁻¹ in samples of the forest soil and beach sand, respectively. Soil depth profiles display higher activity concentration levels in their upper layer of the forest soil and insignificant variations of lower activity concentrations in the surf zone–the dune ecosystem.

Keywords: the Curonian Spit, soil, radionuclide, ¹³⁷Cs.

1. Introduction

The presence of artificial radionuclides in the terrestrial environment may be conditioned by nuclear weapon tests, nuclear accidents and unauthorized releases.

The Lithuanian territory was contaminated substantially by artificial radionuclides due to nuclear weapon tests in the atmosphere in the period 1945-1980 and the Chernobyl Nuclear Power Plant (ChNPP) accident in 1986. One of the most important radionuclides in the releases was ¹³⁷Cs. Owing to the global shedding of radioactive substances ¹³⁷Cs surface activity density in the soil of the Lithuanian region is 1,1–2,2 kBq·m⁻² [1]. After the ChNPP accident values from 7,4·10² to 3,0·10⁴ Bq·m⁻² were obtained [2]. It was established that the territory of Lithuania was contaminated non-uniformly, and thus far the Southern, South-Eastern and Western regions are the most contaminated by ¹³⁷Cs [3]. Major part of ¹³⁷Cs is still accumulated in the upper soil layer [3].

The Curonian Spit is one of the regions where the highest radionuclide concentrations after the ChNPP accident were detected. ¹³⁷Cs surface activity concentration in a 0–5 cm deep undisturbed soil layer in the Curonian Spit was studied at Institute of Physics in 1992–1995 and 1999–2000 [4]. It was found that an average ¹³⁷Cs surface activity density changed from 2760 to 2130 Bq·m⁻² in the period of time between investigations. Few amount of information on ¹³⁷Cs distribution according to the depth in the mentioned

region was obtained. The activity concentrations determined only for two points in the Curonian Spit in 1999–2000 show non-uniform distribution of 137 Cs in the soil with two peaks at 0–2 cm and 8–10 cm depth [4].

The assessment of the radiocaesium inventory and distribution in the soil layers down to 20 or 25 cm depth is of great interest due to its relevance to the research of caesium uptake by plants [5] and its importance for the calculations of caesium contribution to the total absorbed gamma dose rate in the air [6], which strongly depends on radionuclide concentration in the upper 25 cm soil layer [7].

The Curonian Spit is a significant recreational area. The objective of our study was to assess spatial variations in the radionuclide activity concentrations in the soil of dune and forest ecosystem for further investigations of ¹³⁷Cs migration on the Lithuanian seaside.

2. Sampling and measuring methods

In 2003–2004 soil sampling at Juodkrantė in the Curonian Spit was carried out. Soil samples were collected from 19 locations (Fig 1). The sampling area was plotted out in 2 transects (a and b) and 10 longitudinal sections. The transects were chosen 30–50 m apart on the beach and in the pine wood the distance between them reached 200–300 m.

The soil samples were collected by a corer $12 \text{ cm} \times 12 \text{ cm}$ and 30 cm depth, segmented in 0–5, 5–10, 10–15, 15–20, 20–25 and 25–30 cm layers. Then they were weighed and transported to the laboratory in plastic



Fig 1. The Curonian Spit and location of soil sampling points

bags, dried in an oven at 100–110 $^{\circ}$ C for about 24 h, weighed again, and placed in to 100 ml volume plastic containers.

The ¹³⁷Cs activity concentration of the soil samples was determined with the help of a high-resolution hyper pure germanium (HPGe) gamma ray spectrometer system (CANBERRA) (resolution 2 keV, efficiency 15 %). The spectrometer was calibrated by means of the solid ¹⁵²Eu + ¹³⁷Cs standard of various densities.

Each sample was counted from 24 to 168 h. ¹³⁷Cs activity was determined according to the peak at 662 keV. The background radiation spectrum was

evaluated for 168 h counting time. The maximum fractional measurement error didn't exceed 12 %.

3. Results and discussion

The soil sample analysis results are presented in Table 1. As can be seen, the widest range of ¹³⁷Cs activity concentrations (2,5-370,9 Bq·kg⁻¹) was observed in the upper 0–5 cm soil layer. In the surf zone and dunes (points 1–4) they varied in the limits 2,5–11,2 Bq·kg⁻¹, while in a pine forest (points 5–8) they were 10,1 to 370,9 Bq·kg⁻¹. The concentration ranges in deeper forest soil layers were getting narrower. The lowest levels of ¹³⁷Cs activity concentration (0,2–10,4 Bq·kg⁻¹) were determined in the forest soil in 25–30 cm layer. In the surf zone and the sand dunes ¹³⁷Cs activity concentrations variations were insignificant, except the point behind the dune, where the activity concentration increased up to 11,2 Bq·kg⁻¹.

Current studies show that the distribution of ¹³⁷Cs in the surface soil may be attributed to the topographical situation. A characteristic property of radiocaesium distribution was observed – an increment in ¹³⁷Cs activity concentration before the dune (point 2) and its decrement on the top (point 3) and behind the dune (point 4) (Fig 2). As for the forest, it was observed that ¹³⁷Cs activity concentration was getting lower when passing from the sea upward a hill. The lowest activity concentrations were observed on the top of hills (point 7 (transect *a*)) and the highest ones – in forest lowlands (points 8 (transect *a*) and 7 (transect *b*)). Radionuclide concentration on lowlands probably occurried under the impact of contaminated water runoff from hills. On the other hand, the influence of organic matter could take place as well.

The obtained ¹³⁷Cs surface activity density results in a 0–5 cm undisturbed soil layer in the Curonian Spit with analogical results of other authors [4] were compared (Table 2). We can see from the data that an average surface activity density of ¹³⁷Cs decreased approximately by the factor of two in comparison with the data of measurements in 1999–2000. Significant decrease in maximum values are observed as well. It could be probably related to inadequacy of sampling points.

¹³⁷Cs activity concentration distribution according to depth was investigated, and it was established that ¹³⁷Cs activity concentrations varied in a wide interval of values (Fig 3).

Table 1. Statistical data of ¹³⁷Cs specific activity (Bq·kg⁻¹) in Juodkrantė environment soil in 2003–2004

	¹³⁷ Cs					
	0–5 cm	5–10 cm	10-15 cm	15–20 cm	20–25 cm	25–30 cm
Surf zone and sand dunes:						
Range	2,5–11,2	2,8–4,8	2,4–4,5	2,7–4,5	2,4–4,0	2,2-6,8
Arithmetical mean (AM)	4,5	3,8	3,7	3,7	3,4	3,9
Forest soil:						
Range	10,1-370,9	1,6–173,7	0,4–67,6	0,3–29,2	0,2–19,6	0,2–10,4
Arithmetical mean (AM)	153,8	67,5	24,0	12,1	7,4	4,7



Fig 2. Distribution of ¹³⁷Cs activity concentration ($Bq\cdot kg^{-1}$) in surface soil (0–5 cm) of the Curonian Spit at sampling points of transects *a* and *b* (Fig 1)

Table 2. Comparison of 137 Cs surface activity density (Bq·m⁻²) in 0–5 cm depth soil layer in the Curonian Spit at different time of periods

Year	Parameter $(^{137}$ Cs, Bq·m ⁻²)			References	
	Average	Maximum	Minimum	-	
1992–1995	2760	19700	130	[4]	
1999-2000	2130	15800	90	[4]	
2003-2004	1102	4769	197	Current study	

Profiles 1–4 present the vertical distribution of 137 Cs activity concentration in the surf zone and dune ecosystem. It is found that the distribution of mentioned radionuclide is non-uniform, the activity concentration increases and diminishes at different depths but the variations observed are not significant. A maximum 137 Cs activity concentration value of 6,8 Bq·kg⁻¹ was found in the surf zone in a 25–30 cm layer, and a minimum value of 2,2 Bq·kg⁻¹ was on the top of the dune at the same depth. The observed 137 Cs depth profiles in the surf zone can be formed by the sea water wash. Radiocaesium distribution in sand dunes may be influenced by the wind and rain in the case of the absence of vegetation.

Profiles 5, 7, 8 show the distribution of radiocaesium in the pine forest soil. It is found that in the hills (profile 5) and on the top of the hills (transect *a* (profile 7)) the maximum values of 137 Cs activity concentration are located in the uppermost part of the soil (0 to 5 cm in depth). In the deeper 5–10 cm layer the specific activities decrease significantly and keep the same level up till 25–30 cm. The different situation has been observed in the lowland (transect *b* (profile 8)), where the highest activity concentration exists in the uppermost part of the soil and then it decreases

exponentially with depth. Here the changes in caesium activity concentration with depth can be described by an exponential law which in general form is written as

$$A = A_0 \cdot e^{-kz},\tag{1}$$

here A_0 is the caesium activity concentration in the soil on the zero level, A is the activity concentration at depth z (Bq·kg⁻¹), k is the parameter.

The vertical distribution of ¹³⁷Cs activity concentration in some profiles of the soil in the Curonian Spit (Fig 3) can be expressed as

$A = 570 \cdot e^{-0.69z}$,	for transect <i>b</i> profile 7;	(2)

- $A = 584 \cdot e^{-0.91z}$, for transect *a* profile 8; (3)
- $A = 526 \cdot e^{-0.71z}, \qquad \text{for transect } b \text{ profile 8.} \qquad (4)$

The various depth distributions of ¹³⁷Cs content in the forest soil on the top of the hills and in the lowland may possibly be due to the different amount of litter and influence of precipitation in these topographically so different places.



Fig 3. Depth distribution of ¹³⁷Cs activity concentration ($Bq \cdot kg^{-1}$) in soils of the Curonian Spit in profiles 1, 2, 3, 5, 7, 8 of transects *a* and *b*

4. Conclusions

1. 137 Cs activity concentration values in the upper 0–5 cm soil layer of the Curonian Spit varied at an interval of 2,5-370,9 Bq kg⁻¹. In a pine forest the arithmetical mean (AM) of 137 Cs activity concentration values was by the factor of 30 greater than that in beach sand.

2. The present studies show that the distribution of 137 Cs in the surface soil may be possibly attributed to a topographical situation.

3. Insignificant variations of 137 Cs activity concentrations by depth in beach sand was observed, while in the forest soil their maximum values were located in the uppermost part of the soil (0 to 5 cm in depth).

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TARŠA¹³⁷Cs IR JO VERTIKALUSIS PASISKIRSTYMAS KURŠIŲ NERIJOS DIRVOŽEMYJE

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Santrauka

Nustatytas dirbtinės kilmės radionuklido ¹³⁷Cs savitasis aktyvumas kranto smėlyje bei miško dirvožemyje, tam tikslui atlikta 2003–2004 m. Kuršių nerijoje ties Juodkrante paimtų grunto bandinių analizė. ¹³⁷Cs savitasis aktyvumas miško dirvožemio 0–30 cm sluoksnyje kito nuo 0,2 iki 370,9 Bq·kg⁻¹, o kranto ir kopų smėlyje – nuo 2,2 iki 11,2 Bq·kg⁻¹. Didžiausias radionuklido savitasis aktyvumas nustatytas paviršiniame 0–5 cm miško dirvožemio sluoksnyje, gilesniuose kranto bei kopų smėlio sluoksniuose jis pakito neženkliai.

Raktažodžiai: Kuršių nerija, dirvožemis, radionuklidas, ¹³⁷Cs.

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