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VERTICAL PROFILE OF ^{137}Cs IN SOIL FIFTEEN YEARS AFTER CHERNOBYL NUCLEAR ACCIDENT

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Abstract

The subject of research in this paper is the vertical distribution of the ^{137}Cs in undisturbed soil. The presence of the ^{137}Cs in the soil is largely a result of the Chernobyl's accident, which occurred in 1986. Soil samples were taken in central Serbia, in the surroundings of the city of Kragujevac during year 2001. Specific activities of the ^{137}Cs in soil samples were measured by gamma spectrometric method using coaxial germanium detector HPGe. Fifteen years after the Chernobyl accident the largest activity of the ^{137}Cs is within 10 cm of the upper layer of the soil. The obtained result shows that the migration of Cs in soil is a very slow process.

1. INTRODUCTION

As a consequence of nuclear probes and nuclear accidents (the largest one was Chernobyl at 1986) a certain amount of the ^{137}Cs was ejected into the atmosphere, from where it was deposited on the soil. After the accident in Chernobyl in 1986, large scale contamination occurred in Europe and some other parts of the northern hemisphere. The main mechanism of the ^{137}Cs deposition was wet deposition (fallout), although there was some dry deposition. Chernobyl's contamination was inhomogeneous in space and time and was dependent on local weather and other conditions. Being biologically important, the fission product ^{137}Cs , gamma emitter with energy of 661.6 keV and with halflife of 30.17 years, was extensively investigated in the past [1]. From the soil, ^{137}Cs reach food chains and humans, where it irradiates sensitive tissues.

The ^{137}Cs concentration in surface soil decreases under the influence of various processes like decay, mechanical removing with the rain water, vertical migration and diffusion into deeper layers of soil. Several models have been developed to describe the ^{137}Cs migration in soils and to explain its vertical distribution [2-7].

Szerbin et al. [6] applied the diffusion-convection model, and a homogeneous Green function as a solution of partial differential equation for infinite medium. Likar et al. [7] used a "proper" Green function, which satisfies boundary conditions at the soil-air interface, to describe the ^{137}Cs vertical profile in soil. It was claimed by Likar et al. (2001) that proper Green function fits experimental results better than function used by Szerbin et al. (1999). This was tested for two groups of experimental data. The first group was obtained at the location "Krsko Nuclear Power Plant", and the second was taken from literature (Baranya region

in Hungary). In the shallower layer up to 8 cm, there was no important difference between homogeneous and “proper” Green function. Deeper in the soil, the “proper” Green function is closer to the experimental data, but the fit is still below the experimental data and out of the error bars.

The objective of this paper is experimental investigation of the ^{137}Cs vertical profile in undisturbed (not cultivated) soil and determination of its diffusion coefficient, and convective velocity in different types of soil. Our experimental results obtained for ten locations were fitted according to two different diffusion equations, those reported by [6,7].

2. THEORETICAL TREATMENT OF THE ^{137}CS MIGRATION IN SOIL

It is generally accepted that migration of the ^{137}Cs in soil was governed by diffusion and convection this processes were described by the equation, usually known as Fokker-Planck equation [8], which takes into account convection and diffusion:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} - \lambda C \quad (1)$$

where: C is ^{137}Cs concentration in soil, λ is its decay constant, D is effective diffusion coefficient of the ^{137}Cs in soil, v is convective velocity, x is soil depth in respect to the surface and t is time from the deposition. Term $-\lambda C$ which represents the radioactive decay is included in Eq. 1.

The convection–diffusion equation describes physical phenomena where particles are transferred inside a physical system due to two processes: diffusion and convection. The basic processes controlling mobility of anthropogenic nuclides in soil include convective transport by water and diffusive movement within the soil matrix. Convection is mass transfer due to the bulk motion of a fluid, where convection velocity is fluid velocity. Diffusion coefficient is proportionality constant between the flux due to diffusion and the gradient in the concentration of the species.

The solution of Eq. 1. is given by Eq. 2, taken from Szerbin et al. [6]. Since C_0 is the initial surface contamination in $\text{Bq}\cdot\text{m}^{-2}$, $C(x,t)$ is obtained in $\text{Bq}\cdot\text{m}^{-3}$.

$$C(x,t) = C_0 e^{-\lambda t} \frac{1}{2\sqrt{\pi Dt}} \cdot e^{-\frac{(x-vt)^2}{4Dt}} \quad (2)$$

The ^{137}Cs concentration in soil is usually given in $\text{Bq}\cdot\text{kg}^{-1}$; density of soil, ρ in $\text{kg}\cdot\text{m}^{-3}$ could be used to recalculate the ^{137}Cs concentration in $\text{Bq}\cdot\text{kg}^{-1}$ of soil. Total content, C_1 , of the ^{137}Cs in soil is the sum of two components, namely contribution from nuclear probes and Chernobyl accident;

$$C_1 = C_{Ch} + C_{Np} \quad (3)$$

$$C_1(x,t) = C_{0Ch} e^{-\lambda t} \frac{1}{2\sqrt{\pi Dt}} \cdot e^{-\frac{(x-vt)^2}{4Dt}} + C_{0Np} e^{-\lambda(t+t')} \frac{1}{2\sqrt{\pi D(t+t')}} \cdot e^{-\frac{(x-v(t+t'))^2}{4D(t+t')}} \quad (4)$$

where C_{0Ch} and C_{0Np} are initial surface contamination from Chernobyl and nuclear probes, respectively; t' is time between nuclear probes and Chernobyl accident (we took it as 21 years). In order to take into account the "reflection at the air soil interface" [6] considered the term $C(-x,t)$ and finally used:

$$C_{tot} = C_1(x,t) + C_1(-x,t) \quad (5)$$

The expression

$$G_0(x,t) = \frac{1}{2\sqrt{\pi Dt}} \cdot e^{-\frac{(x-vt)^2}{4Dt}} \quad (6)$$

which appeared in Eqs. 2 and 4 is the homogeneous Green function.

Recently, Likar et al. [7] used the following form of the Green function

$$G(x,t) = \frac{e^{-\frac{v\left(\frac{x-vt}{2}\right)}{2D}}}{\sqrt{\pi Dt}} \left[e^{-\frac{x^2}{4Dt}} - \frac{v}{2} \sqrt{\frac{\pi t}{D}} \cdot e^{-\frac{v\left(\frac{x+vt}{2}\right)}{2D}} \left(1 - \Phi\left(\frac{x+vt}{\sqrt{4Dt}}\right) \right) \right] \quad (7)$$

which satisfies the condition of conservation of the activity deposited on the soil in the case of very long life radionuclides, where Φ is the error function in the form

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt \quad (8)$$

The depth profile of the ^{137}Cs is given again as the sum of two contributions

$$C(x,t) = C_{0Ch} G(x,t) e^{-\lambda t} + C_{0Np} G(x,t+t') e^{-\lambda(t+t')} \quad (9)$$

We compared our experimental results to Eqs. (4,5) and Eqs. (7-10). To do the fitting, we used SigmaPlot 4.0 commercial software that is intended for graph creating, statistical calculations and fitting. In order to avoid numbers of numerical integration of the error function in Eq. (7) an Eq. (8) when the fitting was running, we used the polynomial approximation of the error function given as

$$\Phi(x) = 1 - (a_1 t + a_2 t^2 + a_3 t^3) e^{-x^2} \quad (10)$$

where $t = \frac{1}{1+px}$

with constants

$$p=0.47047; a_1=0.3480242; a_2=-0.0958798; a_3=0.7478556.$$

The error of this approximation is less than $2 \cdot 10^{-5}$.

3. EXPERIMENTS

The soil samples were taken during the spring-summer of 2001, fifteen years after Chernobyl accident. Totally ten locations were chosen surrounding Kragujevac city in central Serbia, characterized with different soil types. The soils were uncultivated for a long time and we considered them as not disturbed. It might not be true particularly for the vertical soil where some of "normal" mixing naturally exists even without cultivation. The list of locations and soil characteristics and types is given in Table 1. A specially tool that was used earlier in military applications was applied for soil sampling, so that disturbance and mixing was avoided. The samples were taken in rectangular blocks (10 x 10) cm² up to the depth of 20 cm, and cut in horizontal layers of 2 cm thick, and "depth sampling" was realized. Stones and plant roots were removed before treatment. Soil samples were dried at 105 C⁰ during 24 hours, minced, sifted through the sieve and prepared for gamma spectrometry. The activity of the ¹³⁷Cs in samples was measured by HpGe EG&G ORTEC detector with resolution 1.7 keV on 1.33 MeV energy and relative efficiency of 22.2%, and a multi channel analyzer with 8192 channels. Maestro II software was used to analyze obtained spectrum. The samples were measured between 72 ks and 100 ks, depending on the ¹³⁷Cs activity in the samples. Calibration was performed with calibration sources of ¹⁵²Eu obtained from Laboratoire de Metrologie des Rayonnements Ionisants, Buureau National de Metrologie, France. Since ¹⁵²Eu emits gammas with many different energies, the calibration curve was established for our spectrometer. Calibration source was in the same cylindrical geometry as the samples, and correction on geometry was not needed. Activity of the calibration source at the moment of calibration measurement was 3.44 kBq with a relative error of 3%.

4. RESULTS

Physical and chemical characteristics of the soil samples are given in Table 1. Contents of K, CaCO₃ and organic matter were determined by standard chemical methods. pH values are in the range between 4.8 and 7, which means soils are mostly acid. CaCO₃ was presented only in samples from locations 6 and 10 and the values of 2.1 and 2.2% were found, respectively. Contents of organic mater were from 1 to 8.8%. Samples from location 1 to 4, 8 and 10 had high contents of organic matter, while it was low in location 5, 6, 7 and 9.

The vertical distribution of the ¹³⁷Cs in soil in Bq·kg⁻¹ in different locations is shown on Figs. 1-10. We separate locations in two groups depending on the vertical profile character; first group are locations 1 to 6 and second group is 7 to 10.

Table 1. Location, soil types and its physical and chemical characteristics

Location number	Soil type	pH (H ₂ O)	CaCO ₃ %	K ₂ O (mg/kg)	Organic matter %
1	Vertisol	6.00	0	1000	5.99
2	Brown forest	5.50	0	300	7.02
3	Gray brown	5.88	0	310	5.79
4	Gray brown	5.81	0	700	7.86
5	Vertisol	5.72	0	350	4.43
6	Vertisol	7.5	2.10	100	3.87
7	Alluvial	4.84	0	210	1.04
8	Brown forest	5.22	0	300	8.79
9	Skeletoid soil	5.18	0	130	3.32
10	Alluvial	6.99	2.20	160	7.35

Results for locations 1, 2, 3, 4, 5 and 6 are presented in Figs. 1-6. Experimental results are given as a scatter graph with error bars. The concentration of the ¹³⁷Cs at these locations decreases monotonically with the depth and there is no peak. Such character of vertical profile can be explained assuming pure diffusion; convective migration of the ¹³⁷Cs is weak or did not occur in these soils.

Curves in Figs. 1-6 are results of fitting according to Eqs. 4,5 and Eqs. 7-10. Two curves are mostly overlapped in the subsurface layer of the soil up to 12 cm. In all locations deeper in the soil between 12 and 20 cm, curves are below experimental points and out of experimental error bars. The difference between functions (4, 5) and (7-10) could be seen in the deeper layers of soil where the homogeneous Green function is a little above the Eq. 7-10. The difference is very small and we do not consider it as important. At location 5 there is no difference between two sets of equations and the curves are completely overlapping.

In Figs 7-10, the results for locations 7-10 are given. The peak in the ¹³⁷Cs concentration appears few cm below the surface. At location 7, the peak is very weak and it is really uncertain if there any peak here or not. Peaks are very clearly pronounced at locations 8-10. The peak appearance may be explained by convective transport of the ¹³⁷Cs in these soils. Migration velocity in these locations is an order of magnitude larger than that in location 1-6. Some authors recommended neglecting the convective migration of the ¹³⁷Cs in soil [4]. That is in the case where the concentration of the ¹³⁷Cs decreases monotonically with the depth. In our work, we have found monotonically decreasing of concentration in some cases, but in another cases the peak is clearly seen.

Fitting parameters C_{Och} , C_{ONp} , D and v obtained by two sets of equations (4,5) and (7-10) are given in Table 2.

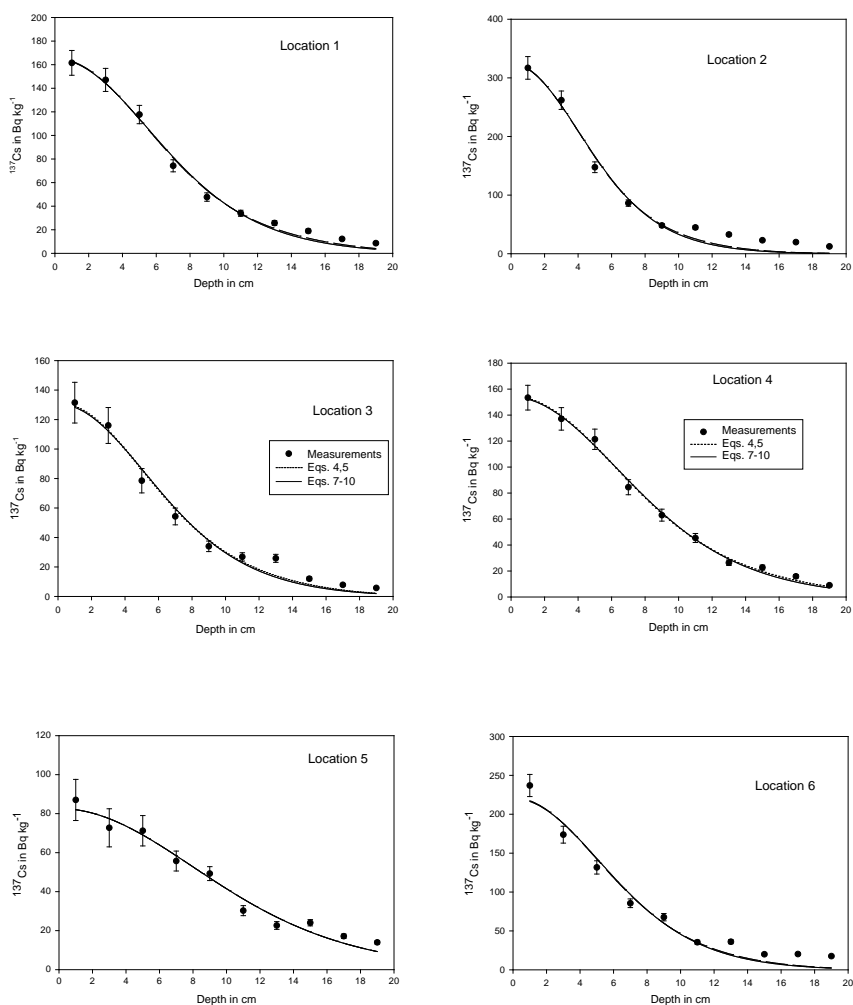


Fig. 1-6. Depth distribution of the ^{137}Cs in locations 1 to 6. Solid line, Eqs. 7-10. Long dashed lines, Eqs. 4,5

Chernobyl contamination is in range from $6 \text{ kBq}\cdot\text{m}^{-3}$ up to $17 \text{ kBq}\cdot\text{m}^{-3}$. The concentration of the ^{137}Cs in the rain water that fell in our terrain at 1 and 2 May of 1986 was 250 Bq/l [9] (the measured rain sample was taken from location that was not included in location 1-10 in this paper). The total amount of water was between 11 and $20 \text{ liters}\cdot\text{m}^{-2}$ which gave surface contamination between $3 \text{ kBq}\cdot\text{m}^{-3}$ and $6 \text{ kBq}\cdot\text{m}^{-3}$. That is on the lower level of surface contamination found by the fitting described above. Considering that the measured locations are different, (data for water precipitation was taken from meteorology station in the city, while the sampled locations are around the city within 25 km) and that Chernobyl contamination was very inhomogeneous, the agreement is not too bad.

Contamination from nuclear probes is in the range from $4 \text{ kBq}\cdot\text{m}^{-3}$ up to $25 \text{ kBq}\cdot\text{m}^{-3}$. The diffusion coefficient D , also given in Table 2, is in the range from 0.24 up to $1.47 \text{ cm}^2/\text{year}$. The values found here for D are in the range obtained for Hungarian soil [6]. We have examined the correlation between soil characteristics and the diffusion coefficients obtained here. The correlation coefficient between diffusion coefficient and contents of organic mater was about 0.5, and is significant. The correlation coefficients between the diffusion coefficient and pH value and contents of K_2O were below 0.3, which is very a weak correlation.

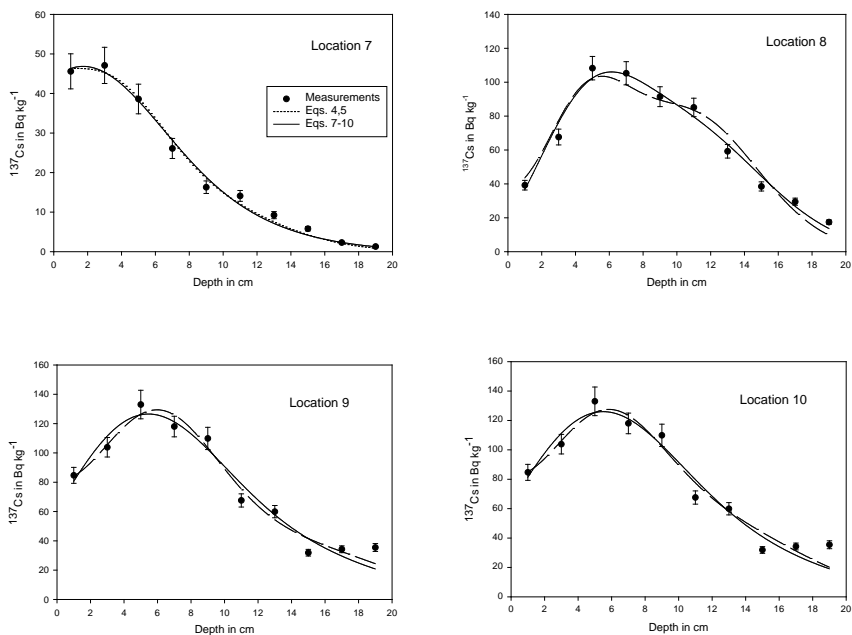


Fig. 7-10. Depth distribution of the ^{137}Cs in locations 7 to 10. Solid line, Eqs. 7-10. Long dashed lines, Eqs. 4,5

Correlation between convective velocity and soil characteristics is weak for all parameters measured here. The set of functions Eqs.7-10 gave $v=0$ on the locations without peak; this seems to us reasonable while the homogeneous Green function (set Eqs. 4,5) gave some small values for v at these locations. Convective velocity may be determined from the position x_m of the peak in the graph as $v=x_m/t$ (time between Chernobyl accident and sampling moment). For example, at the location 8, $v\approx 0.3 \text{ cm/year}$. If we multiply this value by 36 years (time from nuclear probes) we may expect some peak between 11 and 12 cm that originates from nuclear probes. This peak is not seen being weak because longer time was passed, but in Fig 8 some irregularities in vertical profile about 12 cm exists.

Table 2. Fitted initial surface contamination, convection velocities and diffusion coefficients

Location	C_{0ch} (Bq/m ²)		C_{0Np} (Bq/m ²)		D (cm ² year ⁻¹)		v (cm year ⁻¹)	
	Eqs. 7-10	Eqs. 4,5	Eqs. 7-10	Eqs. 4,5	Eqs. 7-10	Eqs. 4,5	Eqs. 7-10	Eqs. 4,5
1	9300	11000	16700	14200	0.83	0.59	0	0.15
2	12900	14200	19900	17700	0.46	0.35	0	0.10
3	6200	6900	12700	11700	0.72	0.53	0	0.13
4	9700	11500	19500	16700	1.07	0.76	0	0.16
5	6900	6700	16500	16800	1.47	1.45	0	0
6	12700	13500	25100	23800	0.62	0.49	0	0.10
7	3400	3700	5000	4400	0.73	0.34	0.06	0.20
8	6000	6500	18500	17500	0.34	0.24	0.26	0.30
9	10400	11100	9500	8600	0.56	0.31	0.15	0.26
10	16900	16600	19800	19900	0.75	0.39	0.21	0.31

5. CONCLUSION

In this paper, the diffusion of the ¹³⁷Cs in soil was investigated experimentally and theoretically. The concentration of the ¹³⁷Cs in some locations (1-6) decreased monotonically in soil. A peak in the vertical profile appeared in 4 of the examined locations. The experimental data were fitted according to homogeneous Green function and "correct" Green function, and no important difference was found between them. Both functions do not fit experimental results very well for deeper layers of the soil. Experimental results, given in Figs 1-10, show that the ¹³⁷Cs activity is highest in the subsurface layer up to the depth of 8 cm. With the depth, the activity decreases, and at about 20 cm it is almost negligible. The shape of the curves depends on the physical and chemical characteristics of the soil. Correlation between diffusion coefficient and parameters of the soil were weak, except with contents of organic matter, where the correlation coefficient of 0.5±0.5 was found.

The diffusion coefficient was determined by fitting the experimental data using two versions of the Green function. The values are between 0.2 and 1.47 cm²/year. Some authors found two peaks in vertical profile of the ¹³⁷Cs in soil; the first shallower and stronger is attributed to Chernobyl accident, and the deeper and weaker peak originated from earlier nuclear probes. In our graphs we do not see

two peaks in the vertical profile of the ^{137}Cs in soil. Some irregularities are found in vertical profiles which are attributed to the nuclear probes.

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VERTIKALNI PROFIL ^{137}CS U TLU PETNAEST GODINA POSLE ČERNOBILJSKOG AKCIDENTA

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Predmet istraživanja u ovom radu je vertikalna raspodela koncentracije ^{137}Cs u tlu. Prisustvo ^{137}Cs u tlu je u najvećoj meri posledica akcidenta u Černobilju, koji se dogodio 1986. godine. Uzorci zemlje su uzimani na teritoriji centralne Srbije, u okolini Kragujevca u toku 2011. godine. Specifična aktivnost ^{137}Cs u uzorcima tla je merena gama-spektrometrijskom metodom pomoću poluprovodničkog germanijumskog detektora. Nađeno je da je najveća aktivnost ^{137}Cs u površinskom sloju tla od 10 cm. Dobijeni rezultat pokazuje da je migracija ^{137}Cs veoma spor proces.

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