

INFLUENCE OF PHYSICAL PROPERTIES OF SOIL ON ^{137}Cs MOBILITY

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A model to account for the mobility of radiocesium in soil is presented. The model requires a minimal set of coefficients that describe radiocesium migration and fixation rates, which can be related to physical soil properties. The peculiarities of experimental radiocesium profiles in soil are explained by the composition of soil, which affects the radiocesium fixation rate. It is shown that the migration of radiocesium in soil is governed by vertical convection of a mobile form, whereas diffusion is a slower process due to strong fixation. The results show that the velocity of vertical migration downward of mobile radiocesium can be set constant, because the overall migration rate depends on fixation. Modelling of experimental radiocesium soil profiles suggests that organic (humic) layers with reduced mineral content and humidity have a high radiocesium fixation rate. Soil structure that maintains high soil humidity and mineral content has an increased cesium exchangeability and, consequently, a higher radiocesium mobility.

Keywords: radiocesium, soil, migration, compartment modelling

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1. Introduction

Significant amounts of radionuclides were deposited on soil of Europe after the Chernobyl accident (1986) by fallout. Radiocesium is considered as one of key artificial radionuclides in the environment due to external irradiation exposure and internal radiation of man through the food chain. Therefore, the study of the vertical migration process of radiocesium in soil is important in order to evaluate the external irradiation from gamma-emitters and to estimate the plant root uptake.

There is a lot of theoretical and experimental works devoted to the study of the radiocesium transfer through the soil matrix. Models, which have been proposed to describe the migration process and the profile of radiocesium concentration in soil, may roughly be divided into three groups. The first group is compartment models [1], where migration is described by rate equations. The rate parameters are transfer coefficients, which characterize the rate of migration of radionuclides between soil layers. When transfer coefficients are equal for all layers the rate equations can be solved analytically and the answer drives towards the second group of models with Gaussian distribution of radioce-

sium [2, 3], where the main parameters are the diffusion coefficient and vertical migration rate. In the third group of models the radiocesium concentration profile in undisturbed soil is approximated by a function, for example, a polynomial [4] or exponential [5] one.

Although it is often accepted that transport of radiocesium is governed by diffusion and convection (Gaussian model), the variety of models reflects the fact that there is no general model, which could be applied for the analysis of the experimental radiocesium profiles in soil without significant assumptions or corrections. These difficulties are comprehensible because experimental results depend on sample conditions such as soil type, structure, amount of precipitation, etc. The knowledge of the dependence of analysed radiocesium profiles on any common soil feature could provide additional useful information for evaluation of the external irradiation from gamma-emitters in soil and estimation of the plant root uptake. There are successful attempts to relate the migration process to soil features. At least two approaches could be selected: the first one relies on soil and radionuclide chemistry [6, 7]; the second one is based on physical soil properties [8] such as structure and composition.

The aim of this work is to analyse how radiocesium migration depends on the structure of undisturbed soil. Soil is considered as a structure of several layers. The difference of layer composition affects the migration process and this influence can be elucidated by the investigation of the parameters that describe migration. This investigation is carried out using both Gaussian and compartment models. Experimental results were taken from [10]: four samples of cesium soil profiles.

2. Modelling approach

Since the mobility of radionuclides in soil depends on complex physical, chemical, and biological processes, a black-box approach based on a multicompartment model is frequently employed for describing radionuclide migration in soils [1]. By using this model the residence times of radionuclides in soil layers and/or migration rates between adjacent layers are deduced from measured depth distributions.

The soil is split into a series of N horizontal layers (compartments), which are connected by downward transport rates of the certain radionuclide. The time dependences (for $t > 0$) of radionuclide concentrations in the compartments are written then as

$$\begin{cases} \frac{dC_1(t)}{dt} = -(\lambda + k_{12})C_1(t), \\ \frac{dC_i(t)}{dt} = -(\lambda + k_{i,i+1})C_i(t) + k_{i-1,i}C_{i-1}(t), \\ i = 2, \dots, N, \end{cases} \quad (1)$$

where C_i is the concentration of the radionuclide in soil layer i , $k_{i,j}$ is its transport rate from soil layer i to j , and λ is the radioactive decay constant. Usually, the deposition of radionuclides from the atmosphere is modelled as pulse-like inputs into the uppermost compartment. For these initial and boundary conditions analytical solutions of Eqs. (1) are available, but numerical methods of solution that can be used with defined deposition histories and initial radionuclide concentrations in the compartments have also been developed. Usually, the solution of Eqs. (1) is fitted to measured concentrations in soil layers at a field site in order to estimate the unknown $k_{i,j}$. From these one can deduce a residence half-life in layer i as $\tau_i = \ln 2 / k_{i,i+1}$ and a migration rate from layer i of thickness Δx_i to the adjacent layer as $\Delta x_i / \tau_i$. The conditions of applicability of the multicompartment model are discussed elsewhere [1].

It is generally accepted that transport of a trace solute in the liquid phase is governed by diffusion and

convection. For one-dimensional transport (along the x -axis) the flux densities are

$$J_d(x, t) = -D_d \frac{\partial C(x, t)}{\partial x} \quad (2)$$

for diffusion, where D_d is the diffusion coefficient of the solute and $C(x, t)$ is its concentration in the liquid phase, and

$$J_c(x, t) = vC(x, t) \quad (3)$$

for convection, where v is the convection velocity of water. Combining these equations results in the convection–dispersion equation, which for the transport of a radionuclide in a porous medium of a constant water content is written as

$$\frac{\partial C(x, t)}{\partial x} = D \frac{\partial^2 C(x, t)}{\partial^2 x} - v_w \frac{\partial C(x, t)}{\partial x} - \lambda C(x, t), \quad (4)$$

where v_w is the mean pore water velocity, and $D = D_d + D_c$ takes into account both molecular diffusion and longitudinal dispersion, which is determined by the velocity profile of laminar flow and by the irregular geometry of streamlines in porous media.

The diffusion-determined flux density of a radionuclide at a depth x ($i = 1$ for the surface layer) depends on its concentration gradient, i. e.

$$\begin{cases} \frac{dC_1(t)}{dt} = -(\lambda + k_{12})C_1(t) + k_{21}C_2(t), \\ \frac{dC_i(t)}{dt} = -(\lambda + k_{i,i+1} + k_{i,i-1})C_i(t) \\ \quad + k_{i-1,i}C_{i-1}(t) + k_{i+1,i}C_{i+1}(t), \\ i = 2, \dots, N, \end{cases} \quad (5)$$

$k_{i,j+i}$ and $k_{i,j-i}$ represent downward and upward transfer rates, respectively. Since the system of Eqs. (5) corresponds to convection–diffusion Eq. (4), it can be noted that downward transfer rates represent a convective and diffusive transport process, while the upward transfer rates are due to diffusion only.

As a consequence, the system of Eqs. (1) can be used if water and solute transport in soil is dominated by a convective process. Only if radionuclide transport in soil is dominated by convection, Eqs. (5) can be approximated by the system (1), i. e. if

$$k_{i,i+1} \gg k_{i+1,i}, \quad i = 1, \dots, N - 1. \quad (6)$$

Often radionuclide concentrations decline monotonically with depth even a long time after deposition, and

Gaussian model approach cannot explain experimental radiocesium profiles; therefore, polynomial [4–6] or exponential [2, 5, 6] fit is used. The fitted functions can describe the changes of radionuclide distribution in soil with time; however, the processes that govern transport are difficult to elucidate [2–6]. In this case a compartmental model with backflow (Eqs. (5)) has to be used, but it requires a large number of transfer rates to be known. Because of these difficulties new approaches are being developed for the description of radiocesium transport in soil.

Recently, hard efforts have been made to explain radionuclide migration dependence on the chemical properties of their carriers [7, 8]. Particularly, radiocesium is effectively fixed by clay minerals. The presence of organic substance affects adsorption of cesium in soil as well. In [9] the following chemical forms of ^{137}Cs were identified: (1) dissolved cations, (2) exchangeable forms including radionuclides absorbed onto soil by ion-exchange mechanism, (3) nonexchangeable forms including radionuclides in hot particles and fixed radionuclides on mineral or organic components of soil. Of the four forms identified, only species in exchangeable forms move into the dissolved state, the other forms move only with the particles on which they are sorbed. But because of the filtering in the porous soil these forms do not move with the infiltrating water considerably.

Therefore, it is reasonable for the analysis of radionuclide migration in soil to choose two components of radiocesium: mobile (m) and fixed (f). The characteristic length of a soil sample by radionuclide concentration determination during measurements is usually equal to 1 cm. Therefore, let us assume that the thickness of the compartment in vertical direction is equal to 1 cm. In each soil compartment (horizontal layer i) the total radiocesium concentration C_i [$\text{Bq}\cdot\text{m}^{-2}$] is formed by the mobile fraction $C_{i,m}$ and the fixed one $C_{i,f}$ (Fig. 1).

C_a [$\text{Bq}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$] denotes concentration of radiocesium fallout. One can assume Chernobyl fallout as one-time (peak) deposition of all radiocesium activity whereas the old (“bomb”) fallout should be distributed over some time; we use a three year period, i. e. years 1961–1963. C_h [$\text{Bq}\cdot\text{m}^{-2}$] denotes radiocesium concentration in hot particles on the soil surface. According to [9], let us assume that at the initial stage after fallout all radiocesium is located in hot particles.

Hot particles infiltrate into soil usually to a depth of 1 cm, but the layer thickness of modelled soil is assumed to be equal to 1 cm, so we do not need any spe-

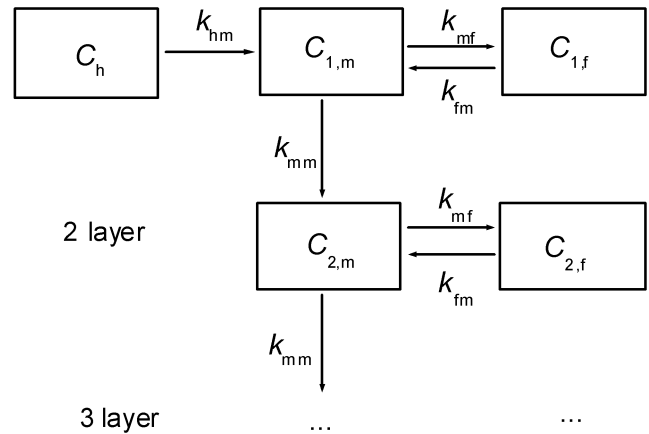


Fig. 1. Schematic representation of the model with mobile and fixed cesium fractions.

cial model condition to take infiltration into account. Due to erosion or washout radiocesium dissolves into the first (surface) layer ($i = 1$) of soil to a mobile fraction of radiocesium ($C_{1,m}$). Then the radionuclide may be fixed on the top level ($C_{1,f}$) or it moves downward to the second layer of soil ($C_{2,m}$). The rate equations that describe the process can be written in the following way:

$$\left\{ \begin{array}{l} \frac{dC_h}{dt} = C_a - k_{hm}C_h - \lambda C_h, \\ \frac{dC_{1,m}}{dt} = k_{hm}C_h + k_{fm}C_{1,f} - (k_{mm} + k_{mf} + \lambda)C_{1,m}, \\ \frac{dC_{1,f}}{dt} = k_{mf}C_{1,m} - (k_{fm} + \lambda)C_{1,f}, \\ \frac{dC_{i,m}}{dt} = k_{mm}C_{i-1,m} + k_{fm}C_{i,f} - (k_{mm} + k_{mf} + \lambda)C_{i,m}, \\ \frac{dC_{i,f}}{dt} = k_{mf}C_{i,m} - (k_{fm} + \lambda)C_{i,f}, \\ i = 2, \dots, N, \end{array} \right. \quad (7)$$

where k_{hm} [day^{-1}] is a constant representing the rate of formation of mobile radiocesium from hot particles, k_{mm} [day^{-1}] is a constant representing the rate of movement of the mobile fraction downward, λ is the radioactive decay rate constant [day^{-1}], k_{mf} and k_{fm} are rate constants representing the rates of formation of the fixed fraction from the mobile one and of the mobile fraction from the fixed one, respectively. These con-

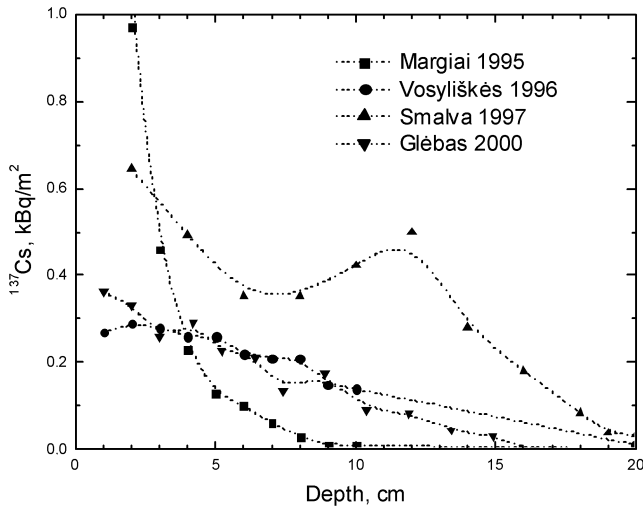


Fig. 2. Vertical distributions of ^{137}Cs in soil [10].

stants may depend on soil structure and composition (clay, organic substance, major ions, and the oxidation–reduction state of soil solution, etc). The initial values of k_{mf} , k_{fm} , and k_{hm} for the simulation were taken from [9], where artificial rain was used to simulate the formation of natural surface runoff, to estimate the vertical distribution of all forms of ^{137}Cs (also ^{90}Sr), and then the rate constants were evaluated.

3. Experimental results

All experimental results for the simulation were taken from [10]. Several areas with natural noneroded or little-eroded soils in local flat places were selected for the investigation of radionuclide activities in vertical profiles of soil: near Ignalina NPP (Table 1, rows 1, 2), in southeastern Lithuania near Margiai bog and Lake Glėbas (Table 1, rows 3, 4). Short general description of soil profiles used for simulation is presented in Table 1 as well. Additional information about soil (density, activity concentration of ^{40}K , ^{226}Ra , ^{232}Th) and measurement procedure may be found in [10]. We present radiocesium profiles of studied soil only in the graphical form (Fig. 2).

The sources of ^{137}Cs inventory in soil may be evaluated using the known activity ratio $r_{1986} = {}^{134}\text{Cs}/{}^{137}\text{Cs} = 0.59$ for fallout following the Chernobyl NPP accident, and its change r_t , described by the ratio of two exponents. Then ${}^{137}\text{Cs}_{\text{ChNPP}} = {}^{134}\text{Cs}_{\text{year}}/r_t$, where ${}^{134}\text{Cs}_{\text{year}}$ is the measured ${}^{134}\text{Cs}$ activity concentration in soil t years after the accident. The equality ${}^{137}\text{Cs}_{\text{nt}} = {}^{137}\text{Cs}_{\text{tot}} - {}^{134}\text{Cs}_{\text{year}}/r_t$ is also valid, where ${}^{137}\text{Cs}_{\text{tot}}$ is the total measured ^{137}Cs activity concentration in soil, ${}^{137}\text{Cs}_{\text{nt}}$ is the ^{137}Cs activity

concentration in soil caused by fallout of nuclear tests in the atmosphere. Using these relationships the contributions of the Chernobyl NPP and nuclear tests at each measured site are established (Table 2).

For model validation, additional information about soil structure was taken from Smalva and Margiai sites (2003 autumn–winter). Determined soil horizons with general description are presented in Table 3.

4. Model application and discussion

In our simulation model the vertical structure of soil contains several horizontal layers of different thickness that are distinguished by soil formation peculiarities. For satisfactory model validation the results of radiocesium concentration profiles in soil should be accompanied by the information about soil structure and composition. In most cases, however, experimental results on soil characterization lack required knowledge. Nevertheless, the presented model allows one to simulate radiocesium concentration profiles in good agreement with measurement results; moreover, by varying the rate constants k_{mf} and k_{fm} with depth it is possible to elucidate some soil properties governing the radionuclide migration. It should be noted that the model uses a small set of coefficients (rate constants); these coefficients can be related to some soil properties.

The model described by Eqs. (7) was applied to simulate the radiocesium migration process in soils with radiocesium profiles presented in Fig. 2. The initial values of rate constants for chemical speciation were taken from [8]: $k_{hm} = (4.0–12.0) \cdot 10^{-4} \text{ day}^{-1}$, $k_{mf} = (2.0–6.0) \cdot 10^{-2} \text{ day}^{-1}$, $k_{fm} = (2.0–6.0) \cdot 10^{-4} \text{ day}^{-1}$. The values obtained in the present work show that these rate constants have not exceeded the range estimated from the results of the artificial experiment.

In Fig. 3 simulated and experimental distribution of radiocesium in soil near Margiai bog (Varėna district) are presented. The ratio ${}^{134}\text{Cs}/{}^{137}\text{Cs}$ shows that the nuclear test load of ^{137}Cs is approximately equal to fallout after the accident in Chernobyl NPP. However, the experimental profile does not indicate that migration of radiocesium is governed by a Gaussian diffusion process. It shows that migration of radiocesium in soil is slow due to its strong fixation in the top layer. Most likely the fraction of mobile radiocesium depends on the amount of precipitation and moisture distribution in the soil profile (ratio of percolation and lateral flow) [5]. Dissolved radiocesium moves downward at a constant rate (described by k_{mm}) until it is fixed in soil. It should be noted that the obtained vertical migration

Table 1. Brief description of ^{137}Cs profiles used for simulation [10].

Profile	Soil description	Soil typological units (STU)
1. Vosyliškės	Sandy loam (basal till) (stagni-eutric alboluvisol) with organic remains in the 5–7 cm layer, large gravel grains in fissures in whole profile available	ABse
2. Smalva	Various sand (glaciofluvial) with very high and changeling content of organics in the top 8 cm layer (haplic podzol)	PZh
3. Margiai	Fine sand (lacustrine) with high content of organics in the top 11 cm layer (gleytic arenosol), 30–31 cm old humus layer, ironized spots of organic remains in the whole profile below available	ARg
4. Glėbas	Fine sand (lacustrine) with high content of organics in the top 11 cm layer (fluvisol)	FL

Table 2. Sources of ^{137}Cs in topsoil (0–5 cm) according to the $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio [10].

Profile	Measurement time	Measured activity concentration, Bq/kg		Load of ^{134}Cs in 1986, Bq/m ²	Load of ^{137}Cs in 1986, Bq/m ²		
		^{137}Cs	^{134}Cs		total	nucl. test, %	ChNPP, %
1. Vosyliškės	1996	21	<dl*	very low	2300	~100	very low
2. Smalva	1997	70	0.4	475	3870	80	20
3. Margiai	1995	374	1.4	3918	12900	51	49
4. Glėbas	2000	119	0.3	1711	9000	68	32

* below detection limit

Table 3. Concentration of ^{137}Cs in soil horizons of Smalva and Margiai sites.

Horizon	Depth, cm	^{137}Cs , Bq/kg	Short horizon description
Smalva (04 December 2003)			
O(L)	0–8	111.0 ± 6.5	Organic litter, mor, significant water percolation
E	8–14	8.0 ± 0.4	Sand leached, 1% fine coal and Fe–Mn concretions, wavy bottom bound
Bs	14–19	2.6 ± 0.2	Cemented and mottled sand, 1% fine Fe–Mn concretions, wavy bottom bound (bottom bound more wavy than top and synchronizes with large roots), clay minerals
B	19–53	<dl	Fine sand, 1% fine coal and Fe–Mn concretions, 5% mottled
Margiai (13 November 2003)			
O(H)	0–11	50.0 ± 2.5	Peat (low water percolation, lateral flow is possible) with eolic sand and dust admixtures ($C_{\text{org}} \sim 90\%$)
AE	11–19	2.0 ± 0.2	Sand, organic substances, pockets of 15 cm width, bottom bound is wavy with 1 cm thick hard ochre sublayer (shell), 3.7% C_{org} , 95.8% mineral matter, 0.5% CaCO_3
1B	19–40	<dl	Medium sand, 7% brown spots, and <1% black concretions
2B	40–59	<dl	Fine sand, 2% ochre spots

rate in this profile (Fig. 3) is the smallest ($k_{\text{mm}} = 0.02$) in comparison with other radiocesium profiles.

As follows from Table 3, radiocesium is fixed in the first soil horizon (O(H), 0–11 cm), which is characterized as an organic layer with mineral content (dust particles). It has a rather small fraction of minerals and is usually not flooded with water due to active surface runoff. These conditions may be the reason that reduces radiocesium exchange with soil. Because of fixation the radiocesium does not move with the infiltrating water.

Figure 4 represents the simulated and experimental distribution of ^{137}Cs in soil near Smalva (Zarasai district). Because of deeper penetration the downward mi-

gration rate is higher, but at the depth of 10–14 cm soil should have a layer with increased cesium fixation ($k_{\text{mf}} \sim 0.09$). Additional study of soil structure has shown that at this depth a transition from soil horizon E to soil horizon Bs is formed. From the general soil description it follows that this sand leached layer E lacks in clay minerals and iron, which are accumulated at a greater depth, in Bs horizon. From both analysed cases it follows that organic (humic) layers with reduced mineral content have an increased effect on radiocesium fixation.

The next simulated profile is presented in Fig. 5. Comparison with the profile analysed earlier shows that radiocesium penetrates the soil at a higher rate

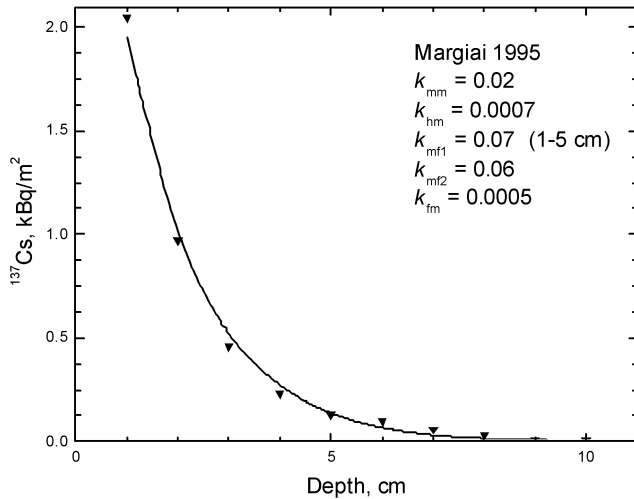


Fig. 3. Simulated and experimental distribution of ^{137}Cs in soil near Margiai bog (Varėna district).

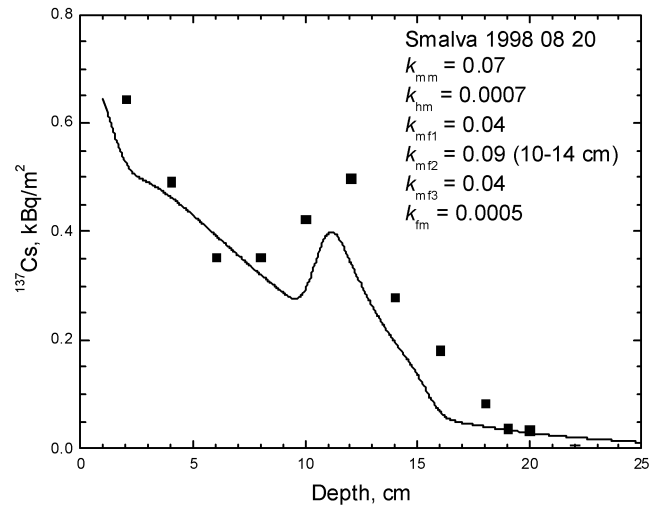


Fig. 5. Simulated and experimental distribution of ^{137}Cs in soil in southeast of Lithuania near Lake Glėbas (Varėna district).

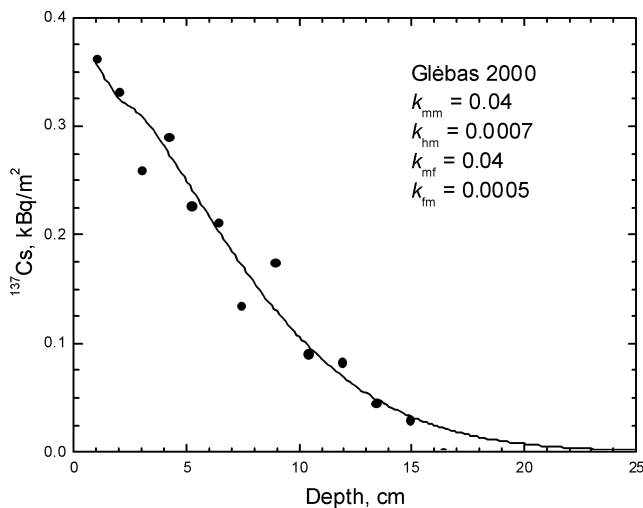


Fig. 4. Simulated and experimental distribution of ^{137}Cs in soil in background areas of the Ignalina NPP region (Smalva, Zarasai district).

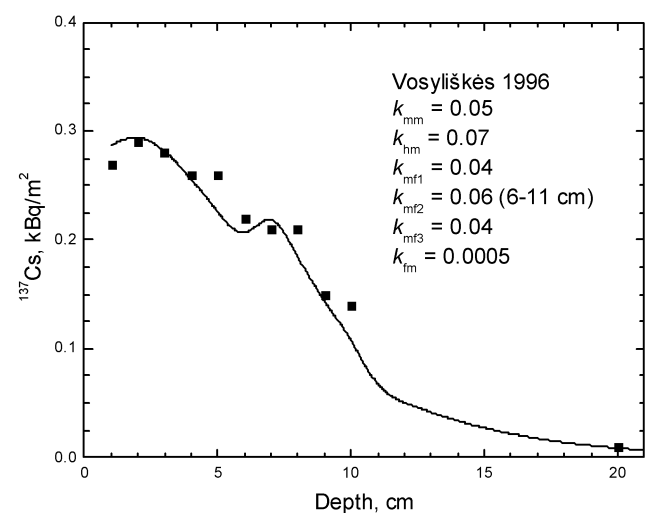


Fig. 6. Simulated and experimental distribution of ^{137}Cs in soil in background areas of the Ignalina NPP region (Vosyliškės, Ignalina district).

($k_{mm} = 0.04$) and lower fixation rate. This can be explained by the fact that in this case soil has less organic content and higher water content or even is flooded due to lake water level changes. This case is contrary to the Margiai one, where the upper organic soil horizon is dry or affected by lateral water flow. In the Glėbas profile water penetrates soil easily and minerals are washed downward to a greater depth, so possibilities for cesium exchange are limited. Stagnic (flooded) properties of soil at Glėbas site imply that the upper soil layer is saturated with water for a long time during a year. Therefore, permanent soil humidity should be attributed to increased exchangeability of radiocesium between the mobile and fixed

form and, consequently, to increased migration downward.

The upper layer of soil at Vosyliškės site (Fig. 6) is sandy loam (with clay minerals, stagni-eutric podzolvisol). Some soil properties are similar to Glėbas site: water-saturated upper soil horizon, minerals are not washed downward because of impermeable deeper soil horizon, which is formed from clay particles. As a consequence, all rate coefficients are similar in both cases. Additional information is required about soil structure in these cases, but it seems that at a greater depth (>12 cm) radiocesium penetrates very slowly. Probably the main mechanism of cesium migration in loam-podzol fertile soils is biotransportation.

Table 4. Characteristics of ^{137}Cs migration in soil.

Profile number	Characteristics of medium of radiocesium migration	k_{mm} , day^{-1}	k_{mf} , day^{-1}
1. Margiai	O(H) horizon: organic (peat), fine sand, washed downward minerals, relatively dry	0.02	0.07; 0.06
2. Smalva	E horizon (8–14 cm): leached sand, lack of minerals, iron; high concentration of sand and dust particles	0.07	0.04; 0.09; 0.04
3. Glėbas	Fluvisol: periodically flooded, fine sand with organics; clay minerals at deeper depth	0.04	0.04
4. Vosyliškės	Stagni-eutric alboluvisol: sandy loam with gravel, organics; clay minerals at deeper depth	0.05	0.04; 0.06; 0.04

The main results are summarized in Table 4. It follows that organic (humic) layers with reduced mineral content and humidity have a high radiocesium fixation rate. Minerals are washed out and are located at deeper levels (usually B horizon). Soil structure that maintains high soil humidity (stagni-eutric podzolovisol) has an increased cesium exchangeability and, consequently, higher radiocesium mobility. It should be noted that these soils are fertile and wet. Therefore, biotransportation must be taken into account.

5. Conclusions

A model to account for the mobility of radiocesium in soil is presented. The model requires a minimal set of coefficients that describe radiocesium migration and fixation rates, which can be related to soil properties. Diversity of soil properties (mainly, structure, and composition) form rather different radiocesium distribution in soil profiles. It is shown that organic (humic) layers with reduced mineral content and humidity have a high radiocesium fixation rate. Soil structure that maintains high soil humidity and mineral content has an increased cesium exchangeability and, consequently, higher radiocesium mobility. The migration of radiocesium in soil is governed by vertical convection of the mobile form, whereas molecular diffusion is a slower process due to strong fixation. The results show that the velocity of vertical migration of mobile radiocesium downward can be set constant, because the overall migration rate depends on fixation.

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^{137}Cs MOBILUMO PRIKLAUSOMYBĖ NUO DIRVOŽEMIO FIZINIŲ SAVYBIŲA. Kanapickas ^a, J. Mažeika ^b, I. Paulaitienė ^a, I. Baužienė ^b^a Vytauto Didžiojo universitetas, Kaunas, Lietuva^b Geologijos ir geografijos institutas, Vilnius, Lietuva**Santrauka**

Išnagrinėti radiocezio pasiskirstymo dirvožemyje profiliai ir, remiantis gautaisiais rezultatais, pasiūlytas būdas, kaip atsižvelgti į radiocezio mobilumą dirvožemyje. Pateiktu modeliu rastais rezultatais parodyta, kad cezio migraciją dirvožemyje lemia stačioji mo-

bilios cezio fazės konvekcija, tuo tarpu molekulinė difuzija pasireiškia silpnai. Stačiosios mobilios cezio fazės pernešimo koeficientą galima laikyti pastoviu, nes didžiausią įtaką turi radiocezio surišimas dirvožemyje. Parodyta, kad radiocezio surišimas stipriausiai pasireiškia organika praturtintuose sausuose dirvožemiuose su maža mineralinių medžiagų koncentracija.