

Measurements and Modelling of ^{137}Cs Migration into Various Types of Soil

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ABSTRACT

The behaviour and migration of long-lived radioisotopes is an important part of information in the prediction of the consequences of radioactive contamination in agricultural areas.

The soil contamination in Central and Eastern Europe after the Chernobyl accident has been high enough to study the vertical migration of radiocaesium under natural conditions. In the project presented here, the distribution of ^{137}Cs activity concentration has been measured and analysed over the period of 1987-1999.

Four sites have been selected to represent four different soil types. Samples are taken regularly from plane, grassy, uncultivated fields. After physical processing, activities are measured by gamma spectrometry.

The physical and chemical processes of the migration are assumed to follow the diffusion-convection model, taking into account contaminations from both the atmospheric nuclear weapon tests and the Chernobyl accident. Fits of the initial surface activities, the effective convection velocities and the effective diffusion coefficients are obtained by Monte Carlo technique. The convection velocities vary from about 0.06 to 0.4 cm/y, whereas the range of the diffusion coefficients is about 0.02 to 0.8 cm²/y.

The goodness of fit is somehow characterised by the fact that the initial surface activities are in the range of 0.5-1.0 Bq/cm², for both the Chernobyl and the nuclear tests, as expected from earlier measurements.

The penetration of caesium into the soil is a very slow process, the majority of the activities is still in the top 10 cm layer. Assuming the correctness of the model for longer time periods, predictions can be made for future times.

INTRODUCTION

During the recent years extensive studies have been initiated to gain more precise knowledge on the soil behaviour and biological availability of fission and activation products released by the accident at the Chernobyl nuclear power plant. The fallout originated from the radioactive plume was composed of condensed fraction with the attached volatile radionuclides, such as caesium isotopes, and fuel particles. In the territory of Hungary the condensed fraction dominated in the fallout.

From the whole range of radionuclides the most important ones are ^{90}Sr and ^{137}Cs due to their relatively long half-lives ($T_{1/2} = 28$ and 30 years, respectively), abundance in the fallout, and radioecological importance. In Hungary the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio varied between 6 and 21% [1], and the ^{137}Cs ground contamination ranged from about 0.1 to 1.0 Bq/cm² [1, 2]. As a consequence of the low amount of fallout, ^{90}Sr can hardly be measured in Hungarian soils.

The mobility of ^{137}Cs in the soil-pore/water system and the biological availability of a radionuclide are determined by the physico-chemical properties of the soil, as well as by the physico-chemical properties of the radionuclide. Caesium is likely immobilised rapidly in soils which have high proportion of clay minerals, contain free carbonates, have pH-s between 4 and 7, and contain small amounts of organic matter [3, 4].

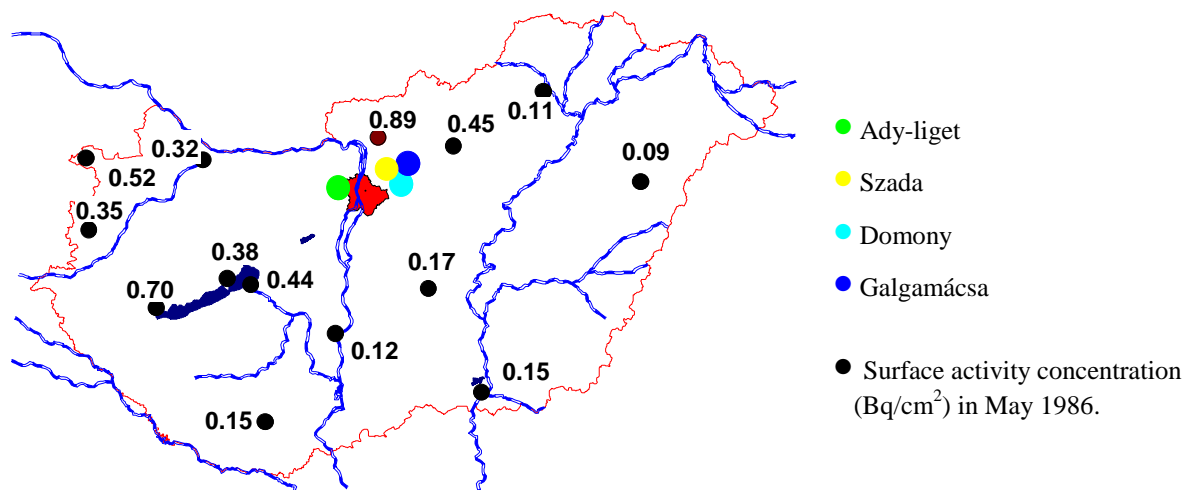
Even very low concentrations of clay minerals are sufficient for caesium fixation, and most of the soils contain some minerals. The only exclusion are the soils with very high organic matter content. In these soils the caesium fixation, as a consequence of the low clay content, is very low and due to the presence of regular ion exchange complexes the caesium remains available for plant uptake. Therefore, caesium contamination of soils with high levels of organic matter (e.g. peat soils) may present a serious hazard in terms of the soil-to-plant transfer processes [5].

Several attempts have been made to describe the transport of radionuclides in soils [6]. The most widespread for vertical migration description is the diffusion-convection model [7, 8, 9, 10, 11]. The model parameters are usually fitted to the available experimental data and then the model can be used to calculate the radionuclide distribution profile for any given time [12].

The main goals of the present study are: (i) determination of site-specific vertical distribution profiles of ^{137}Cs originated from the global fallout and from the Chernobyl fallout, with particular emphasis on the possible correlation between the downward migration and the type of the soil; (ii) mathematical modelling of the ^{137}Cs migration in order to give predictions of radionuclide distribution in the root layer.

SAMPLING SITES

One of the mostly contaminated areas in Hungary following the Chernobyl accident has been the vicinity of Budapest (see ^{137}Cs surface concentration (Bq/cm^2) values in the map), therefore, the four sampling sites were selected in that region.



SAMPLING AND SAMPLE PROCESSING

Approximately $30 * 30 \text{ cm}^2$ areas on plane, grassy, uncultivated fields are sampled at each of the four sites. Ten layers extending from the surface down to a depth of 20 cm are excavated at each of 12 sampling times (12, 14, 17, 24, 29, 36, 41, 48, 53, 83, 118 and 160 months) after the accident. The distances between the sampling spots of the same site never exceeded 10 m.

The samples are first dried at $105 \text{ }^\circ\text{C}$, then homogenised, and the grains exceeding 1.25 mm in diameter are removed. After this processing activities of 400 cm^3 volumes are measured by HpGe semiconductor gamma spectrometry in Marinelli geometry.

SOIL CHARACTERISTICS

The four sites have been selected to represent four different soil types, characteristic in Hungary. Several measured soil parameters are given in Table 1.

ERROR SOURCES

There are many sources of errors and uncertainties in the final results: short-range soil inhomogeneities, the roughness of the surface, temporal changes in humidity, the uncertainties in layer thicknesses, and errors of the spectrometric measurements.

The effects of the sampling uncertainties are decreased by determining the actual weights of the layers, in g/cm^2 , and using these values in the evaluation, rather than the nominal thicknesses in cm.

Table 1. Soil characteristics

soil type	black rendzina	humic sandy soil	meadow soil	typical Ramann brown forest soil
soil texture	clay-loam	sandy	loam	loam
Site	Ady-liget	Szada	Domony	Galgamácsa
pH (KCl)	5.5	7.5	7.3	7.2
CaCO ₃ (%)	-	0.5	7.2	4.2
org. matter (%)	3.9	1.3	4.6	3.1
cation exchange capacity (me/100g)	43.0	9.8	26.8	31.0
changeable Ca (me/100g)	40.1	8.8	21	26.2
changeable Mg (me/100g)	2.56	0.99	5.24	4.18
changeable K (me/100g)	0.26	0.04	0.3	0.51
changeable (Ca+Mg)/K	163	250	87	60

MEASURED CONCENTRATIONS

The measured activity concentrations are shown in the next figures for the four types of soil, respectively. The layer numbers given in the figures are related to depths in the following manner: the first six layers are 1 cm thick each, layers 7 to 10 extend from 6 to 8, 8 to 10, 10 to 15 and 15 to 20 cm, respectively.

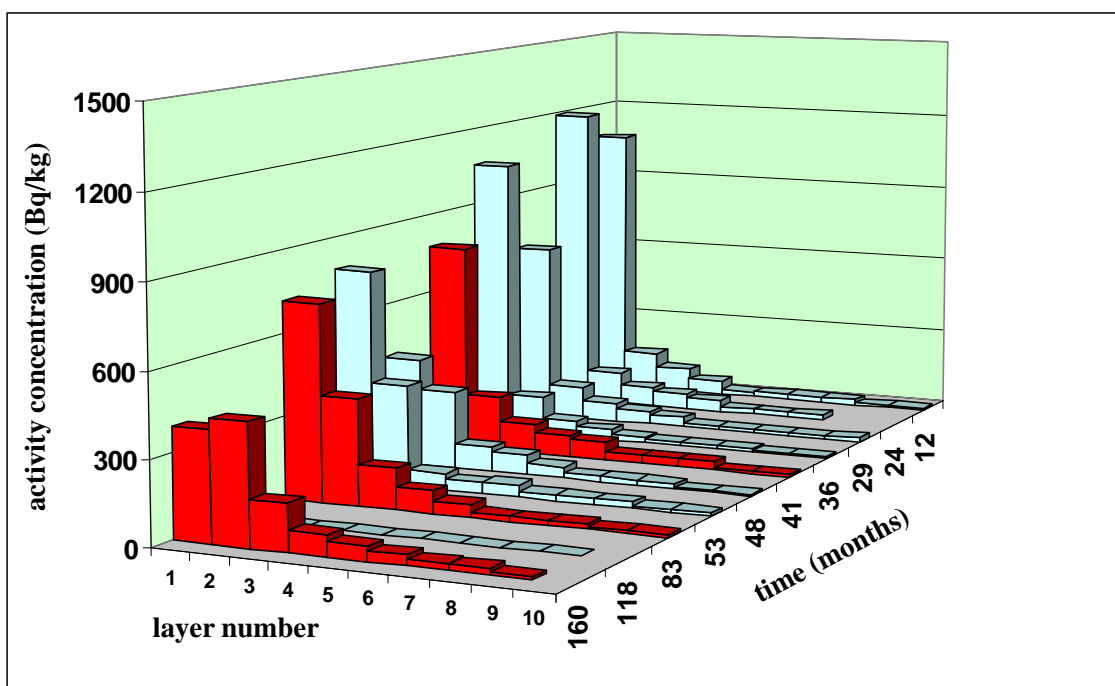


Figure 1. Activity concentrations in Ady-liget (black rendzina)

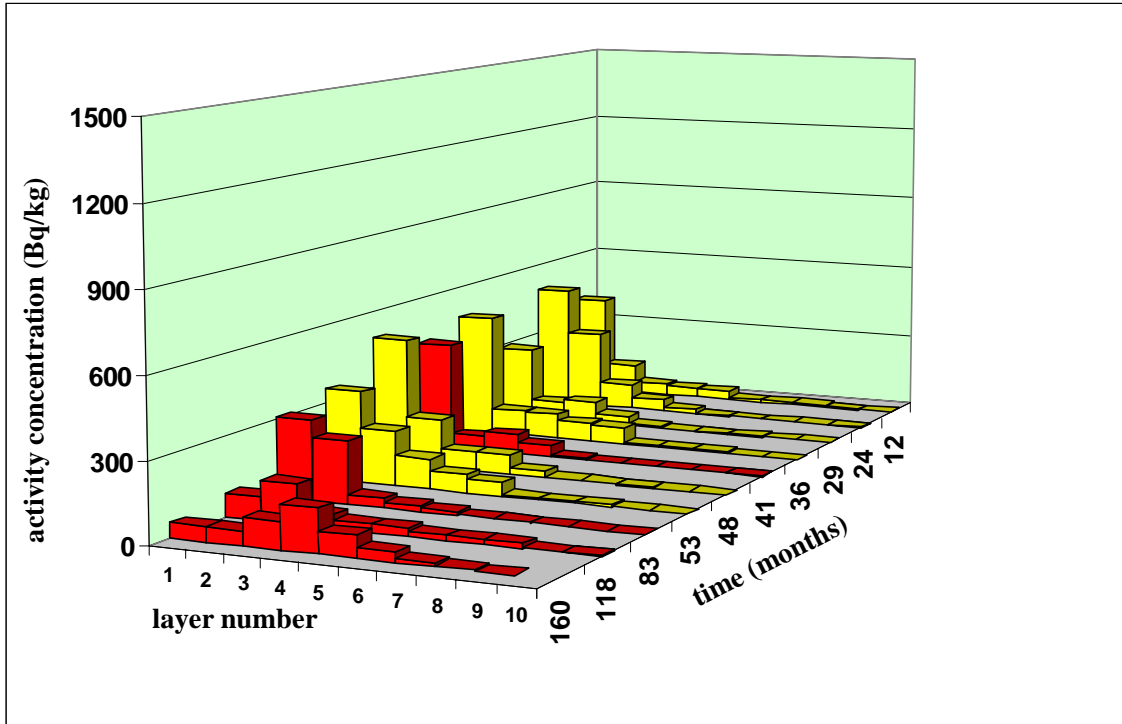


Figure 2. Activity concentrations in Szada (humic sandy soil)

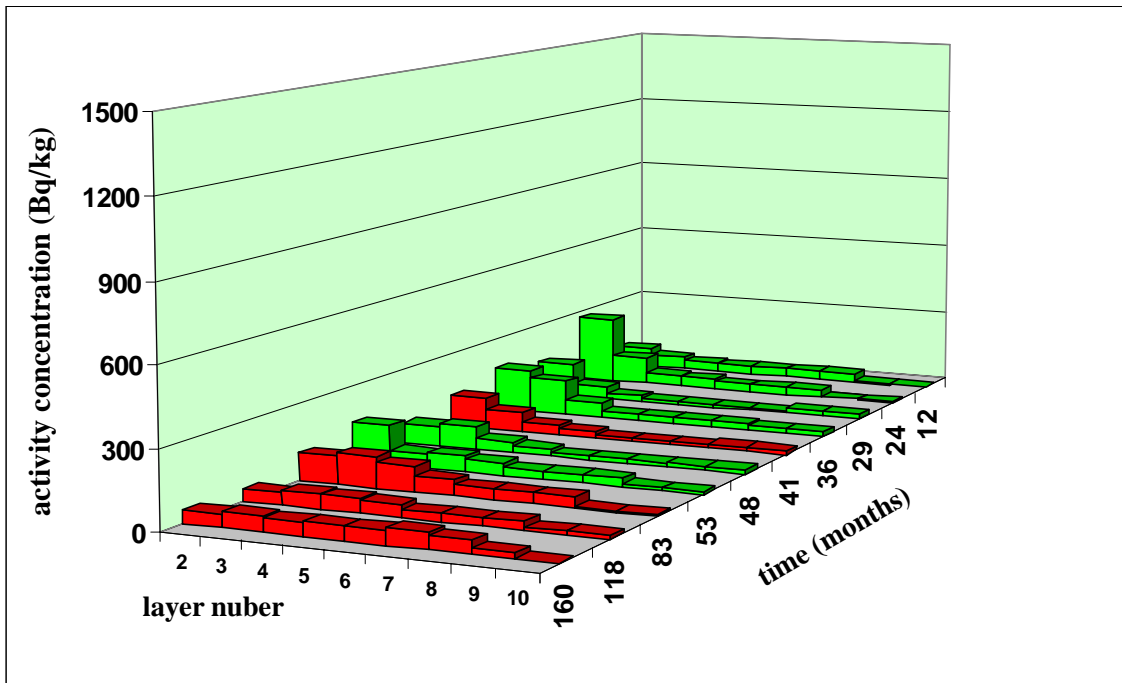


Figure 3. Activity concentrations at Domony (meadow soil)

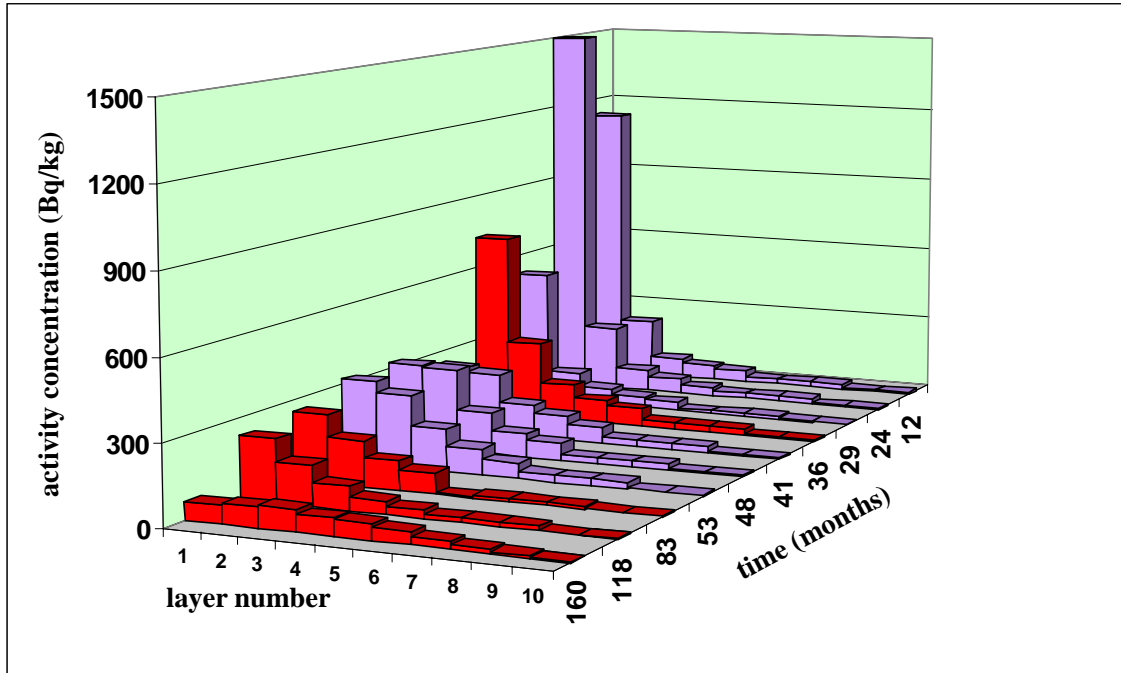


Figure 4. Activity concentrations at Galgamácsa (typical Ramann brown forest soil)

MIGRATION MODEL

The physical and chemical processes of the migration are assumed to follow the diffusion-convection model. Let the initial surface concentrations from the Chernobyl and nuclear test fall-outs be a_{Ch} and a_{nt} , respectively, then the concentration a at depth z , at time t after the Chernobyl accident is

$$a(z, t) = a_{Ch} \exp[-\lambda t] \frac{1}{2[\pi Dt]^{1/2}} \exp\left\{-\frac{[z-ut]^2}{4Dt}\right\} + a_{nt} \exp[-\lambda(t+\tau)] \frac{1}{2[\pi D(t+\tau)]^{1/2}} \exp\left\{-\frac{[z-u(t+\tau)]^2}{4D(t+\tau)}\right\},$$

- where
- $a(z,t)$ is the activity concentration (Bq/cm³) at time t and depth z ,
 - a_{Ch} is the Chernobyl fall-out on the soil surface (Bq/cm²),
 - a_{nt} is the fall-out of the nuclear weapon tests on the soil surface (Bq/cm²),
 - D is the diffusion coefficient (cm²/y),
 - u is the convection velocity (cm/y),
 - λ is the decay constant of ¹³⁷Cs (1/y),
 - τ is the average time between the nuclear tests and the Chernobyl accident (set to 25 y).

Further, we assume that there is a total reflection at the air-ground interface, thus the activity $A_{i,k}$ in layer i at sampling time t_k reads

$$A_{i,k} = \phi \int_{z_{i,2}}^{z_{i,1}} [a(z, t_k) + a(-z, t_k)] dz,$$

where $z_{i,1}$ and $z_{i,2}$ are the depth limits of layer i , and ϕ is the area of the samples.

Four parameters are to be fitted according to the measurements: D , u , a_{ch} and a_{nt} . If $B_{i,k}$ denotes the activity measured in layer i at time t_k , then the quantity to be minimised in a least square fit is:

$$\sum_i \sum_k (A_{i,k} - B_{i,k})^2.$$

At the actual fitting procedure four sampling times, approximately equally distributed over the whole period, 41, 83, 118 and 160 months after the accident are selected. The activity concentrations measured at these times are marked in red in figures 1-4. The inclusion of the more frequent early results, e.g. from 12, 14 etc months measurements, would have led to an over-weighting of the first period.

There was a fatal error in the evaluation of the samples taken at Ady-liget at 118 months, therefore, for that site only three sets of data are considered.

The best fits of the initial surface activities, the convection velocities and the diffusion coefficients obtained by applying a Monte Carlo technique are given in Table 2.

Table 2. Fitted model parameters

Identification	black rendzina	humic sandy soil	meadow soil	typical Ramann brown forest soil
Site	Ady-liget	Szada	Domony	Galgamácsa
a_{ch} (Bq/cm ²)	0.52	0.81	0.69	0.53
a_{nt} (Bq/cm ²)	0.57	0.43	0.57	1.02
u (cm/y)	0.062	0.130	0.43	0.083
D (cm ² /y)	0.023	0.031	0.82	0.24

According to the best fits the ratio of the maximum and minimum convection velocities is about 7, and the diffusion coefficients vary within a factor of about 35.

If one compares these coefficients with the data published after the evaluation of the measurements carried out in the first 118 months [14], there is a significant increase in the diffusion coefficient in the cases of Domony (from 0.39 to 0.82 cm²/y) and Galgamácsa (from 0.04 to 0.24 cm²/y). This increase can be explained by the unusually high precipitation in the last years and by the fact that these areas used to be covered by flood water for several weeks.

The goodness of fit is somehow characterised by the fact that the initial surface activities are in the range of 0.5-1.0 Bq/cm², both for Chernobyl and the nuclear tests, as expected from earlier measurements at these sites around Budapest, i.e. at one of the most contaminated areas of Hungary.

For illustration the measured activity concentrations and the fitted theoretical profiles for typical Ramann brown forest soil (Galgamácsa) are given in figure 5.

MODEL PREDICTIONS

Assuming the correctness of the model for longer time periods, predictions can be made for future times. Activity concentration curves computed for 20, 30 and 50 years are given in figure 6.

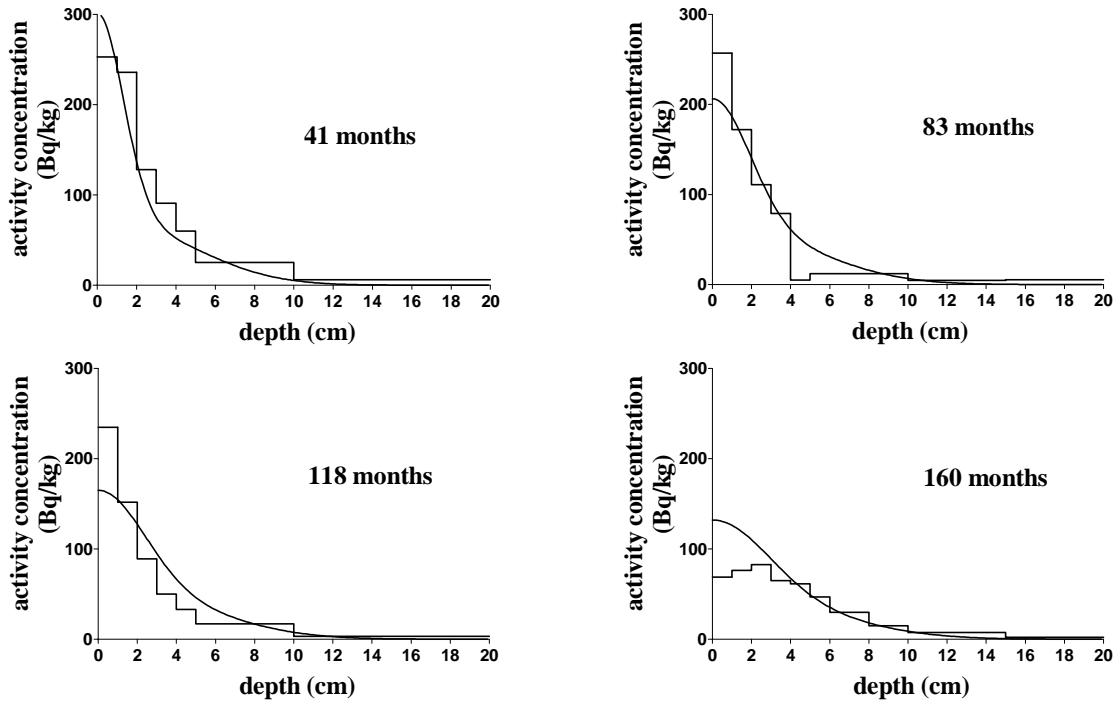


Figure 5. Measured activity concentrations (hystograms) and fitted activity profiles (continuous curves) for the typical Ramann brown forest soil at Galgamácsa

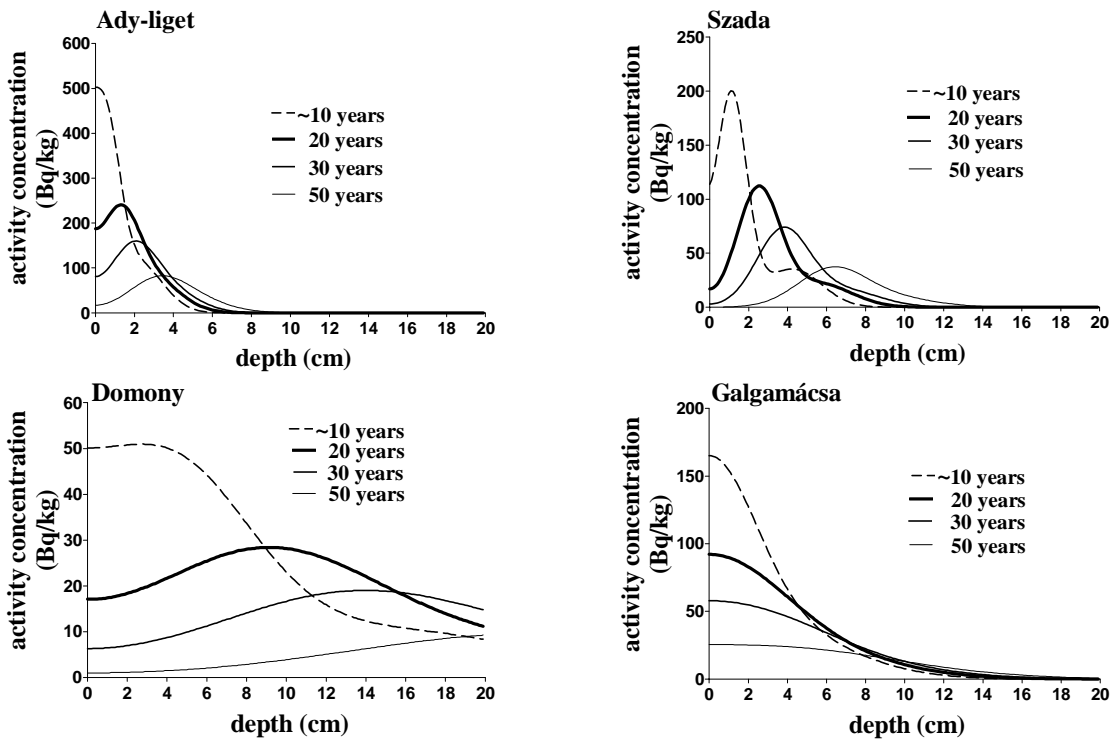


Figure 6. Predicted activity concentration profiles

DISCUSSION

It is known from the literature, that soils with low or moderate (<40 %) organic matter content have sufficient clay mineral content for strong caesium fixation [5]. In soils selected here the organic matter content was always less than 10 %. In good agreement with the expectations, figures 1-4 show a strong ^{137}Cs fixation.

In a laboratory study it was demonstrated that the (Ca+Mg)/K ratio may play a key role in accelerating the radiocaesium fixation process [13]. Comparing the changeable Mg and Ca contents given in Table 1 and the migration parameters shown in Table 2, a weak correlation is found between the (Ca+Mg)/K ratio and the calculated convection velocity (u) and the diffusion coefficient (D). In general, the higher the (Ca+Mg)/K ratio, the lower the migration parameters are.

According to earlier studies [4] caesium fixation is enhanced in soils having higher clay content. Our investigation confirms this finding: the lowest migration parameters are found at Ady-liget, where the clay content is the highest.

In another study [12] convection velocities and diffusion coefficients were derived for many other locations in Hungary, however, only at one sampling time. The coefficients obtained are similar to those presented here.

CONCLUSIONS

- The results presented in this work indicate that penetration of caesium into the soil is a very slow process, at more than 13 years after the accident, most of the activity is still in the top 10 cm layer.
- The amount of caesium deposited on the ground surface in Hungary from the global fallout and from the Chernobyl fallout are in the same order.
- A slight correlation is found between the migration parameters and certain physico-chemical characteristics of the soil: (Ca+Mg)/K and clay content.
- The model is adequate to reproduce the time variation of the ^{137}Cs concentrations profile in the soil.

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