

Depth and Areal Distribution of Cs-137 in the Soil of a Small Water Catchment in the Sopron Mountains

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Abstract – The study presents the depth and areal distribution of Cs-137 activity concentration in the forest soils of Farkas Trench, a small water catchment in the Sopron Mountains, in 2001 and 2010, moreover the possible reason of the alteration in activity concentration. The Cs-137 activity values were measured in 30 plots in 2001, and in 5 in 2010. In 2001, the depth distribution of Cs-137 activity concentration in the measurement plots was shaped in accordance with a decreasing exponential function. It appeared in the 2010 data that the highest Cs-137 activity concentration had shifted lower from the top layers, and the depth distribution changed along an increasing or a stagnating function until a depth of 4 to 6 cm (in function of slope inclination), then along a decreasing function. In 2001, activity concentration in the surface soil layers (0 – 2 cm) altered between 15 and 609 Bq/kg, whereas in 2010 between 26 and 72 Bq/kg. A correlation was found between activity concentration in the surface soil layer and slope inclination. It was concluded that one of the main reasons for differences in distribution of Cs-137 activity concentration was the differing extent of soil movement.

Cs-137 distribution / Cs-137 activity concentration / forest soil

Kivonat – Cs-137 mélységi és területi eloszlása a Soproni hegyvidék egy kis vízgyűjtőjének talajában. A tanulmány bemutatja a Farkas-árok, a Soproni-hegység egy kis vízgyűjtője, erdei talajában a Cs-137 aktivitáskoncentráció mélységi és területi eloszlását 2001 és 2010-ben, valamint az aktivitáskoncentráció változás lehetséges okát. A Cs-137 aktivitás értékek 2001-ben 30, 2010-ben 5 helyen lettek mérve. 2001-ben a mérési helyek Cs-137 aktivitáskoncentrációjának mélységi eloszlása a talajban csökkenő exponenciális függvény szerint alakult. A 2010-es mérések adataiban látható volt, hogy a legmagasabb Cs-137 aktivitáskoncentráció a felső rétegekből lejjebb tolódott, és a mélységi eloszlás 4–6 cm mélységig egy növekvő, vagy stagnáló függvény (lejtőszög függvényében), majd egy csökkenő függvény szerint változott. 2001-ben a felszíni talajrétegek (0–2 cm) aktivitáskoncentrációja a mintavételi pontokban 15 és 609 Bq/kg között, míg 2010-ben 26 és 72 Bq/kg között változott. A felszíni talajréteg aktivitáskoncentrációja és a lejtőszög között összefüggést lehetett kimutatni. A területi eloszlás lejtőszög függéséből azt a következtetést vontuk le, hogy a Cs-137 aktivitáskoncentráció területi eltéréseinek egyik fő oka az eltérő mértékű talajvándorlás volt.

Cs-137 eloszlás / Cs-137 aktivitáskoncentráció / erdei talaj

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1 INTRODUCTION

Fallout of caesium-137 (Cs-137) is associated worldwide with the nuclear weapon tests performed in the 1950s and 1960s (Du et al. 1998). In addition, the 1986 Chernobyl reactor accident has also made its effect felt in our home country, as a result of which the Cs-137 activity concentration which had originated from earlier atmospheric nuclear weapon tests increased in the soil. Cs-137 arrived at the surface by dry and wet deposition, then, due to natural processes in the environment, a significant portion of it migrated into the soil. Its depth and areal distribution was set by how it was bound in the soil. Since Cs-137 is strongly bound to clay minerals and to the organic matter in the soil, this radioisotope can even be used to follow soil movement (Du et al. 1998).

In general, the depth distribution of Cs-137 in undisturbed soils shows an exponential decrement in function of soil depth. Even though the Cs-137 concentrations of undisturbed sites show great spatial variation, still the depth distribution will chiefly depend on soil type and on soil structure, on the differing distribution of clay minerals and of organic matter (Du et al. 1998). Walling and Quine (1992) surveyed the depth distribution of Cs-137 in several soil species in the United Kingdom. They concluded that activity concentration strongly decreases by depth, more than 75 per cent of the total activity is to be found in the uppermost 15 cm layer, and the total Cs-137 accumulation of the soil strictly correlates with the total fallout amount. Berg and Shuman (1995) carried out studies in coniferous forests, and the results of their 3D model created on the basis of measurements showed that in any equilibrium state, up to 85 per cent of the active radiocaesium is found in the soil. Furthermore, Cs-137 migration, according to the predictions, is rather slow, its most essential part taking place within the uppermost 10 cm of the soil profile.

The objective set in this study was to study the depth and areal distribution of Cs-137 activity concentrations in undisturbed forest soils in a small water catchment in the Sopron Mountains, and to serve with data about Cs-137 accumulation to serve as a basis for surveys to come, for estimating soil movement.

2 MATERIAL AND METHODS

2.1 Study area

The examination was carried out in one of the small water catchments of the Sopron Mountain Range, in the Farkas Trench (*Figure 1*).

The Farkas Trench (0.63 sq. km) is a southwest-northeast oriented trench along a branch of Rák Brook. The altitude of the outlet is 401 m above the sea level and the highest point of the catchment reaches 545 m. Terrestrial clastic rocks deposited at different siltation conditions (conglomerate, gravel, sand, silt) on crystalline shale bedrock. Average slope of Farkas Creek exceeds 21%, valley-bottom has 7.7% inclination (Csáfordi et al. 2010 p. 4). On the basis of fluvial sediment, podzolic brown forest soils, highly acidic non-podzolic brown forest soils and lessivated brown forest soils have evolved (Gribovszki et al. 2006 p. 85). At the lowermost third of the trench, even landslides can be traced. Mediterranean, continental and also oceanic effects can be sensed in the local climate. Long-time average precipitation of the area is 917 mm, but as an example, in the 2001 examination year there was only 607 mm (Gribovszki et al. 2006). Farkas Trench is mostly covered by forests, with deciduous and coniferous vegetation present alike (Csáfordi 2010). The main conifer species is Norway spruce (*Picea abies*), the main deciduous beech (*Fagus sylvatica*). From 2001 on there have been clear cuttings performed at several sites along the trench, which have probably affected precipitation intake and runoff (Gribovszki et al. 2006).

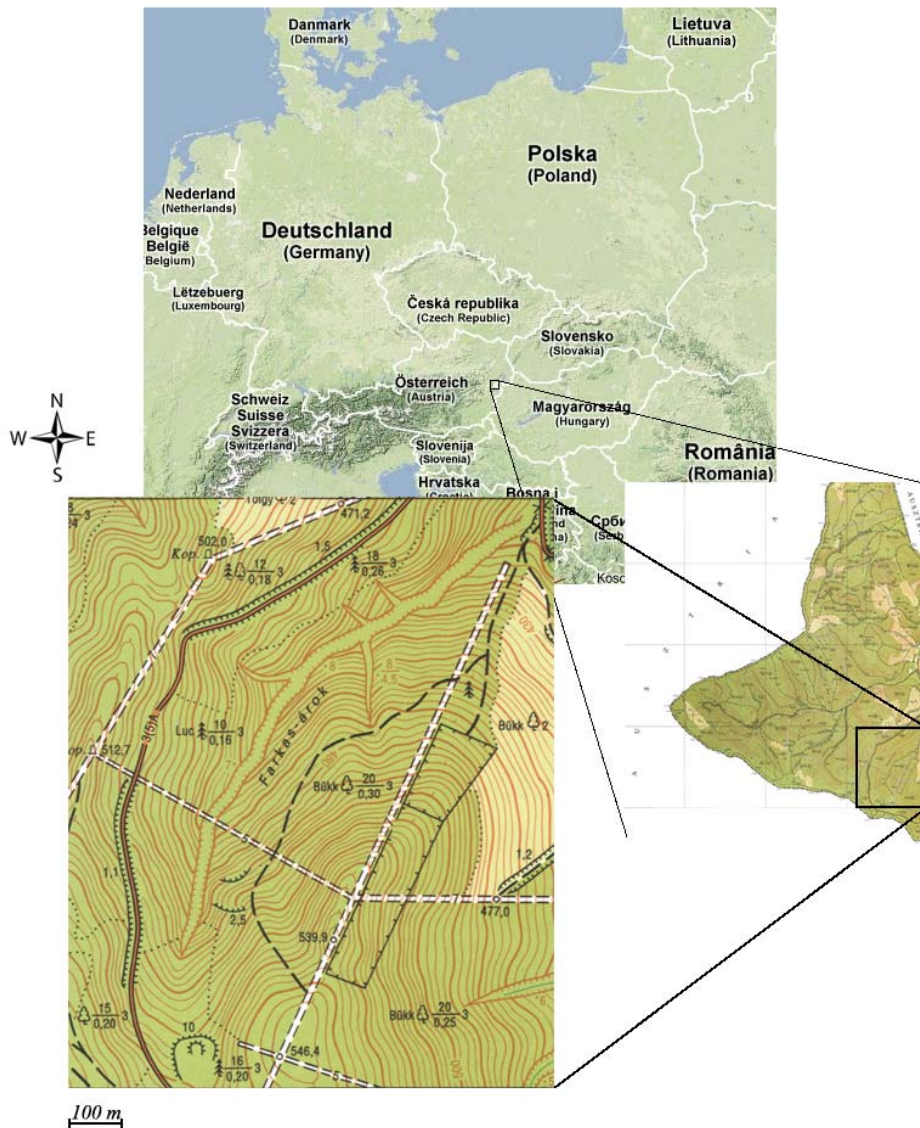


Figure 1. Location of the studied water catchment

2.2 Cs-137 deposition values

Cs-137 accumulated in the examined area originates from two sources. One of them is the fallout originating from nuclear weapon tests, the other one being the Cs-137 amount deposited during the 1986 Chernobyl reactor accident. According to the data in the literature (Pellet 2006), in the examined area the extent of deposition originating from Chernobyl (Figure 2) was nearly one magnitude level higher than the activity concentration of nuclear weapon test origin (Figure 3) present in the soil at the time of the accident. Thus, the activity values determined during the examination give information primarily about the period which has passed since 1986.

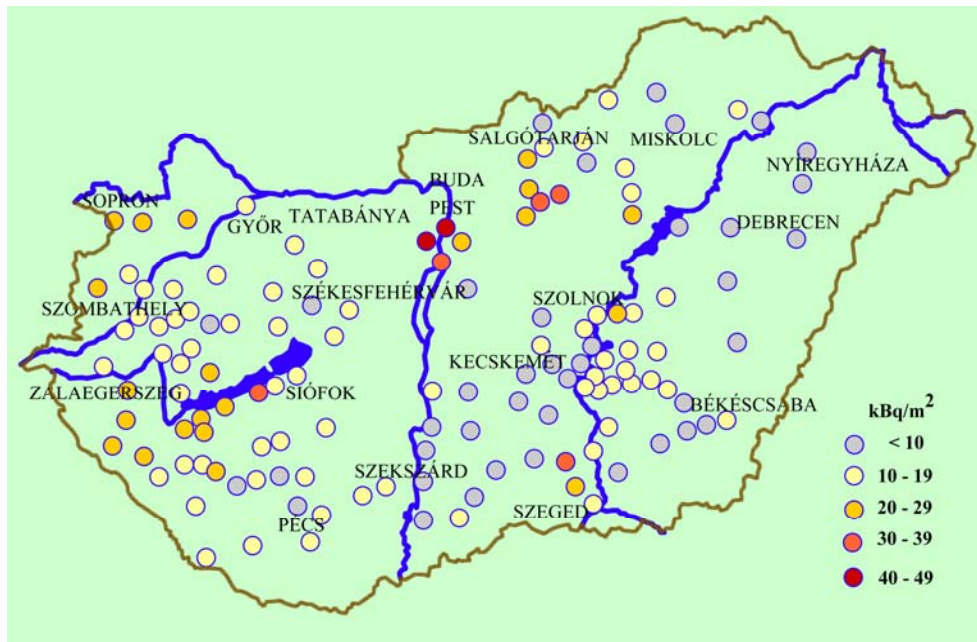


Figure 2. Soil surface Cs-137 pollution in Hungary in 1986, after the Chernobyl accident (Pellet 2006)

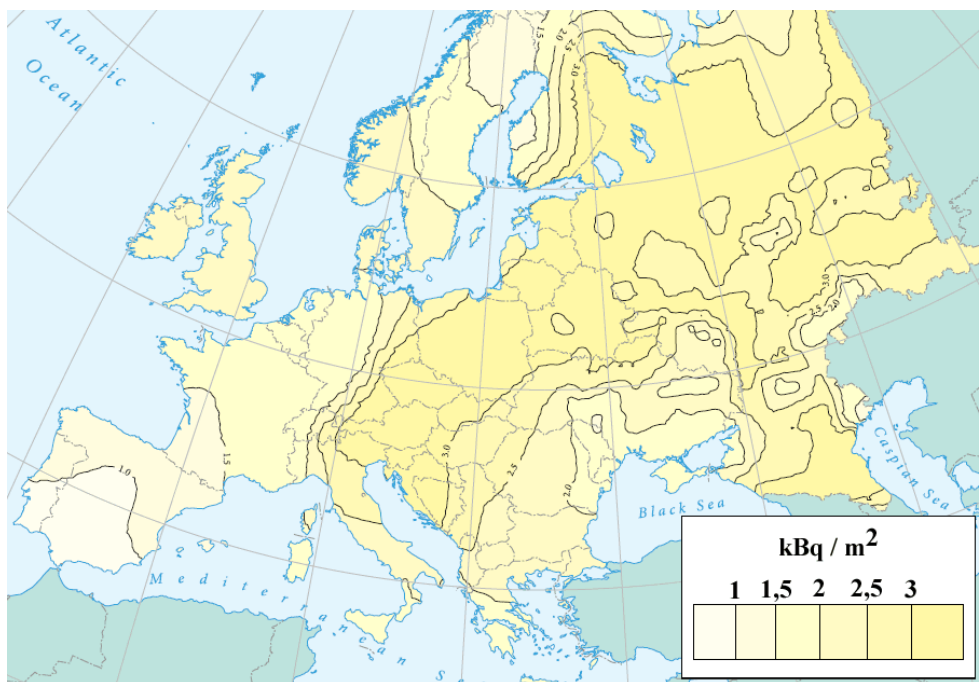


Figure 3. Europe's Cs background pollution before the Chernobyl accident (Pellet 2006)

2.3 Measurement method

When sampling an undisturbed area, acquiring the soil samples without significantly disturbing the area is an important consideration. Sampling device and method have to be such that they either do not disturb or mix up the different layers of the soil, or only to a minimal extent.

Sampling took place within the examined area at 30 plots (numbered points from 1 to 3, and L1/1 to L9/3) in 2001, then at 5 plots (numbered points from L1-2010/0 to L1-2010/4) in 2010 (Table 1) (Figure 4).

Table 1. EOY coordinates of the sampling plots, and their measured slope inclination

Site	EOV(Y) (m)	EOV(X) (m)	Slope (deg.)	Site	EOV(Y) (m)	EOV(X) (m)	Slope (deg.)
1	455493	260122	<4	L5/1	455470	260536	38,8
2	455657	260371	<4	L5/2	455534	260495	17,7
3	455951	260909	<4	L6/1	455428	260415	18,8
L1/1	455899	260926	4,6	L6/2	455472	260398	16,1
L1/2	455905	260910	9,3	L7/1	455371	260333	25,1
L1/3	455911	260899	8,4	L7/2	455312	260336	26,3
L2/1	455891	260950	29,3	L8/1	455392	260329	22,7
L2/2	455890	260977	9,4	L8/2	455429	260318	16,7
L3/1	455874	260903	8,1	L8/3	455469	260311	15,6
L3/2	455880	260888	6,3	L8/4	455521	260295	19,1
L3/3	455886	260862	11,9	L9/1	455375	260267	32,8
L4/1	455727	260796	28,2	L9/2	455417	260230	17,3
L4/2	455736	260791	16,5	L9/3	455435	260207	26,2
L4/3	455745	260743	11,8	L1-2010/1	455932	260943	21,6
L4/4	455762	260720	16,9	L1-2010/2	455939	260948	7,8
L4/5	455765	260680	13	L1-2010/3	455932	260940	13,3
L4/6	455778	260643	15,3	L1-2010/4	455923	260922	9,1
				L1-2010/0	455919	260911	< 4

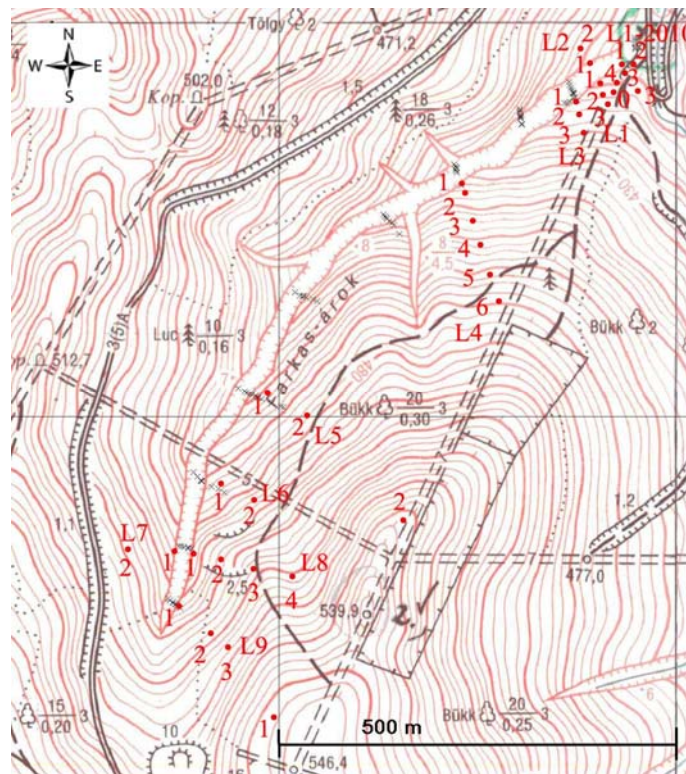


Figure 4. Location of the sampling plots

Sampling was performed using a sampler made of steel plates, down to a depth of 10 to 12 cm, by 2-cm depth intervals. The sampler consisted of a one lateral side opened metal frame (size: 20*25 cm, high: 20 cm) and a horizontal metal plate (frame fitting). In some plots trowel was used to the layers separation instead of the metal plate (due to the roots). Preliminary sampling demonstrated that a significant portion of the Cs-137 amount is found in the uppermost 10-cm layer, hence the sampling depths were adjusted accordingly. In 2001,

at the plots with minor (< 4 degrees) slope inclination at a depth of 10 cm, measured Cs-137 activity concentrations were at least one magnitude level lower than in the surface layer. Samples collected in the area were measured after drying, pulverizing, sieving (< 2 mm). By measuring, 0.5 or 1-liter Marinelli sample dishes were used, the weight of the samples was determined on a certified Precisa 3100D scale. Cs-137 activity of the samples was determined by gamma spectrometry in the OSJER (Hungarian Radiation-Monitoring Detection and Verification System) laboratory at the University of West Hungary. Cs-137 activity was measured using a high-resolution ORTEC HPGe detector (Gem-10185), moreover by a PC-attached multi-channel (8K) ORTEC analyser card. For evaluating the measurements, the ORTEC Maestro software which belongs to the card was used. Measurement duration was 1800 or 3600 s. Channel-energy calibration of the software programme was completed by using Co-60, Cs-137 and Am-241 certified standard sources of known activity.

Activity referring to a given isotope of the sample can be calculated from the area under the net gamma peak (661.62 keV in case of Cs-137) determined using the Maestro programme using the following formula (Eq. 1) (Bódizs 1997):

$$A = \frac{N}{\eta_{E_i} \cdot K\gamma_{E_i} \cdot t_m} \quad (1)$$

Where:

- A – is activity referring to a given isotope of the sample (Bq).
- N – net area of the gamma peak generated by that given isotope (impulse).
- $K\gamma_{E_i}$ – the E_i -energy gamma radiation yield by that given isotope, expressing the number of gamma quanta at a certain energy referring to 100 radioactive decaying atomic nuclei.
- η_{E_i} – the efficiency value read from the energy-efficiency curve referring to a given geometry of E_i -energy radiation. It expresses how many of all the gamma photons emitted by the radiating source at a certain energy are registered in the full energy peak.
- t_m – measurement duration (live time) (s).

Knowing the activity values thus derived one can calculate the activity concentration, A_K , (Bq/kg), of the sample referring to one mass unit (Eq. 2), (A – being the activity referring to a certain isotope in the sample (Bq), m – the mass of the sample (kg)):

$$A_K = A / m \quad (2)$$

The activity concentrations can be converted to the inventory (Bq/m²) according to the following Eq. (3) (FANG et al. 2006):

$$CPI = \sum_{i=1}^n C_i \times B_i \times D_i \times 10^3 \quad (3)$$

where:

- CPI – is Cs-137 point inventory (Bq/m²),
- i – is the No. of soil horizon,
- C_i – is Cs-137 activity concentration of the i th soil horizon (Bq/kg),
- B_i – is bulk density of the i th soil horizon (g/cm³), and
- D_i – is thickness of the i th soil horizon (m).

Even with properly set measurement devices, random statistical errors may occur. Equation 4, which describes the time-dependence of radioactive decay, only gives a probability.

$$N_t = N_{t0}e^{-\lambda t} \tag{4}$$

(λ = decay constant, t = time, N_{t0} = number of initial radioactive nuclei)

The simple unweighted average of measured impulse number (N_i) and the empirical standard deviations (s) of measurement points calculated from an average are described by the following formulas (Pátzay 2008):

$$\bar{N} = \frac{\sum_{i=1}^n N_i}{n} \tag{5}$$

$$s = \pm \sqrt{\bar{N}} \approx \sqrt{N_i} \tag{6}$$

$$s = \pm \sqrt{\frac{\sum_{i=1}^n (N_i - \bar{N})^2}{n-1}} \tag{7}$$

With the first standard deviation formula (Eq. 6) only the errors arising from the decay, while with the second formula (Eq. 7) the errors arising from the decay as well as those arising from measurement device fluctuation, can be estimated. If the standard deviations calculated in these two ways nearly match (difference < 10 %), then the measurement device error is negligible compared to the error arising from decay fluctuation.

3 RESULTS

Vertical distribution of Cs-137 in undisturbed soil (Blagoeva – Zikovsky 1995, Perrin et al. 2006, Porto et al. 2001, Zhiyanski et al. 2008) has been examined in several studies. For this purpose, evenly decreasing two-parameter functions were fitted to the measured values of the profile, by computer (Blagoeva – Zikovsky 1995):

$$C(X) = A \times X^B \tag{8}$$

$$C(X) = A \times \exp(-B \times X) \tag{9}$$

$$C(X) = A / (1 + B \times X) \tag{10}$$

where the x is the depth in cm and C the activity concentration in Bq/kg. The best fitting was displayed by the second function (Eq. 9).

Based on worldwide published data, Cs-137 concentration distribution by depth in undisturbed soil profiles was classified into three groups (Du et al. 1998). The three types of regression functions are the following:

$$Cs = a \times e^{-bz} \tag{a>0, b>0}$$

$$Cs = a \times (1-(k-z/H)^b) \times (k-z/H)^{b-1} \tag{a>0, b>1 and 0<k<=1}$$

$$Cs = a \times (1-z/H)^b \tag{a>0, b>=0}$$

where Cs is the concentration of Cs-137 at a given depth (Bq/kg), z represents the given depth in the soil profile (m), a , b and k are coefficients of the used function and H is the thickness in which Cs-137 can be detected.

3.1 Depth distribution of Cs-137 at the sampling plots in 2001

The Cs-137 activity concentration and inventory values of the soil samples acquired at the sampling plots as determined by measurement appear in the following table (Table 2).

Table 2. Cs-137 activity concentration by each soil layer at the sampling plots and Cs-137 inventories of the 1-3 sampling plots

Site	Cs-137 concentration (Bq/kg)					
	Soil depth					
	0 – 2 cm	2 – 4 cm	4 – 6 cm	6 – 8 cm	8 – 10 cm	10 – 12 cm
1	608.8	360.5	150.4	46.8	36.2	
2	449.0	344.6	108.0	27.0	7.1	
3	489.5	250.2	88.8	25.2	15.5	
L1/1	572.1	326.6	199.8	169.2	88.3	
L1/2	438.1	165.8	58.9	34.3	25.1	
L1/3	493.0	208.8	106.8	36.2	19.6	
L2/1	112.4	91.4	39.6	13.7	12.0	
L2/2	255.7	157.2	98.2	43.5	17.5	
L3/1	233.2	248.3	121.1	53.5	23.7	
L3/2	408.9	229.0	103.0	48.5	26.2	
L3/3	368.4	146.5	60.7	11.2	7.0	
L4/1	70.2	62.2	27.9	15.5	10.4	
L4/2	169.2	117.4	38.2	12.5	9.7	
L4/3	156.9	129.4	100.8	42.5	19.4	
L4/4	310.7	135.9	35.4	18.1	10.4	
L4/5	331.0	141.2	27.0	20.2	11.9	
L4/6	310.2	125.8	48.3	20.7	12.4	
L5/1	104.0	107.1	67.5	38.4	18.1	
L5/2	390.6	242.4	161.2	36.4	21.5	
L6/1	155.1	93.6	43.3	26.9	14.2	
L6/2	235.1	253.1	125.1	51.4	16.4	
L7/1	15.5	8.4	3.3	3.6	2.6	
L7/2	158.2	130.6	83.6	38.5	20.1	
L8/1	165.0	131.2	92.2	41.2	21.7	
L8/2	328.9	120.4	48.7	32.0	14.5	
L8/3	397.4	227.4	53.6	19.7	10.1	
L8/4	255.5	160.2	46.4	32.6	13.6	7.5
L9/1	38.8	68.4	35.2	12.4	8.2	
L9/2	221.7	73.2	29.6	7.5	6.0	
L9/3	202.1	103.6	7.9	0.9	2.0	

Site	Cs-137 inventory (Bq/m ²)
1	10569.9
2	9137.0
3	8909.8

The values for some soil profiles from among the values derived are shown in function of soil depth in the following figures as examples. First, 3 sample plots with lesser slope steepness (Figure 5), then, a slope-directed sampling series (Figure 6). On fitting regression functions onto the sampled values it was realised that by the plots with lesser steepness (< 4 degrees) exponential functions provide the best match. As the slope grade grows, this Cs-137 profile alters, too. At the plots which have greater slope inclination, a decrease in Cs-137 activity concentration of the top layers appeared. With the consideration that Cs-137 is strongly bound to soil, this decrease in soil Cs-137 activity suggests soil movement.

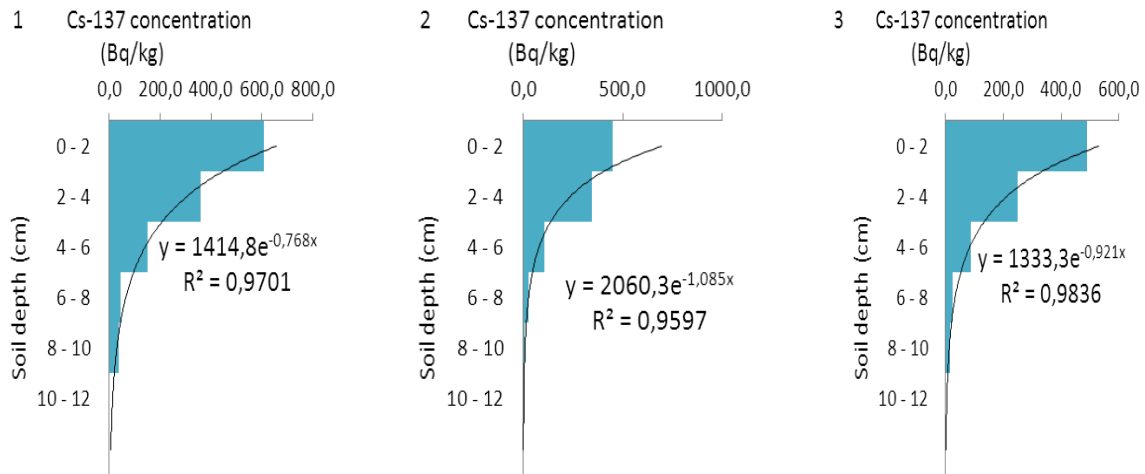


Figure 5. Cs-137 activity concentration distribution by depth in the soil of sampling plots 1 to 3 (slope < 4 degrees)

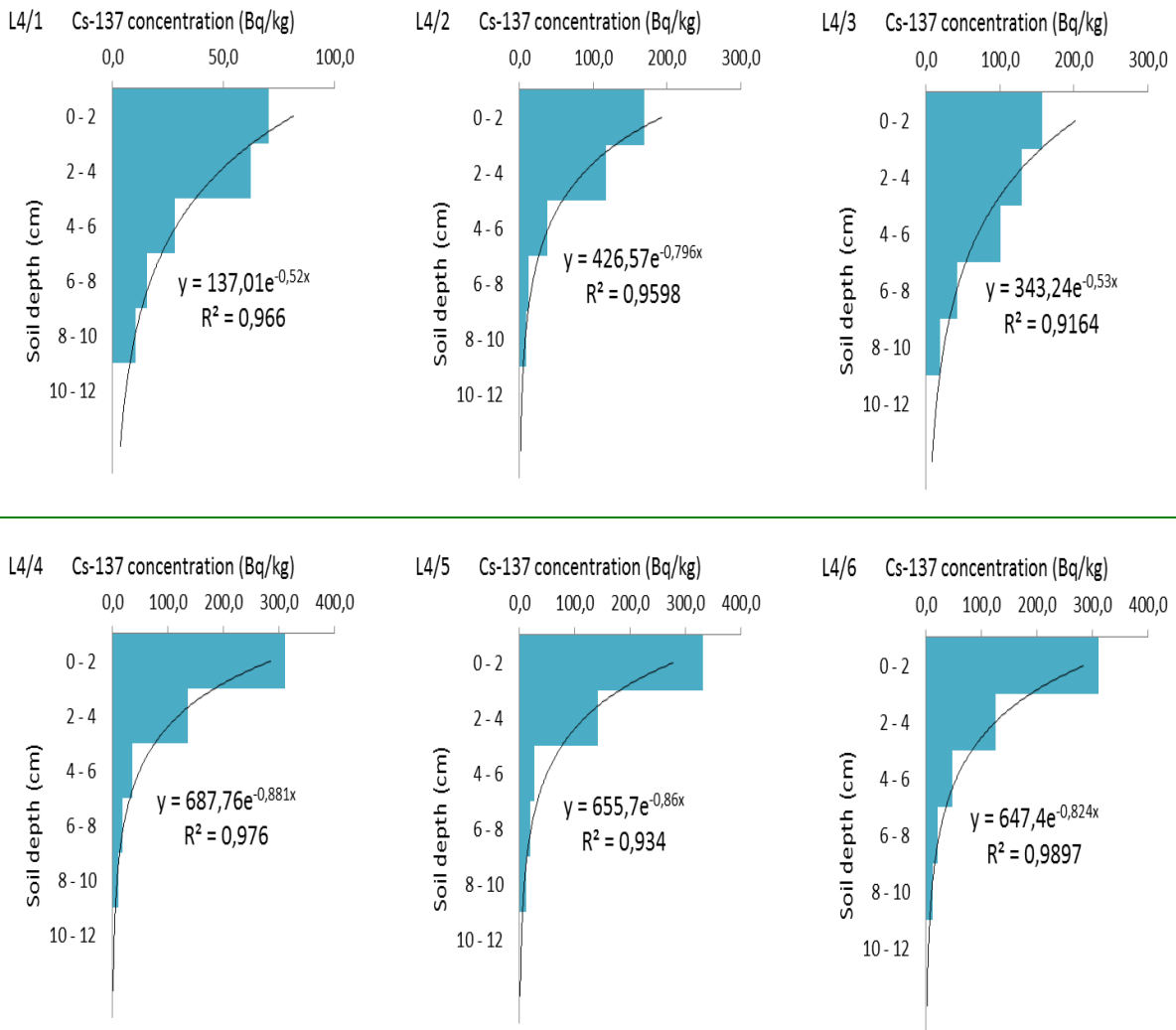


Figure 6. Along a slope (at the L4/1 – L4/6 measurement points) the Cs-137 activity concentration distribution by depth in the soil (slope > 4 degrees)

3.2 Depth distribution of Cs-137 at the sampling plots in 2010

The 2010 repetition of sampling was performed by a smaller number of samples in the lower part of the area. The Cs-137 activity concentration and inventory values determined by measurements of the soil samples taken at the sampling plots appear in *Table 3*.

Table 3. The Cs-137 activity concentrations of the sampling plots in each soil layer and Cs-137 inventory of the L1-2010/0 sampling plot

Site	Cs-137 concentrations (Bq/kg)					
	Soil depth					
	0 – 2 cm	2 – 4 cm	4 – 6 cm	6 – 8 cm	8 – 10 cm	10 – 12 cm
L1-2010/1	26.0	42.0	41.3	29.4	19.8	
L1-2010/2	45.6	69.4	76.0	58.0	29.8	
L1-2010/3	32.6	42.9	59.4	52.8	23.7	
L1-2010/4	29.8	58.9	80.4	47.2	38.2	11.1
L1-2010/0	72.1	62.7	79.4	47.5	33.4	8.9

Site	Cs-137 inventory (Bq/m ²)
L1-2010/0	3887.6

The Cs-137 profile of the slightly sloped sampling plot (< 4 degrees) altered showing stagnating values to a depth of 4 to 6 cm, then exponentially decreasing ones (*Figure 7*).

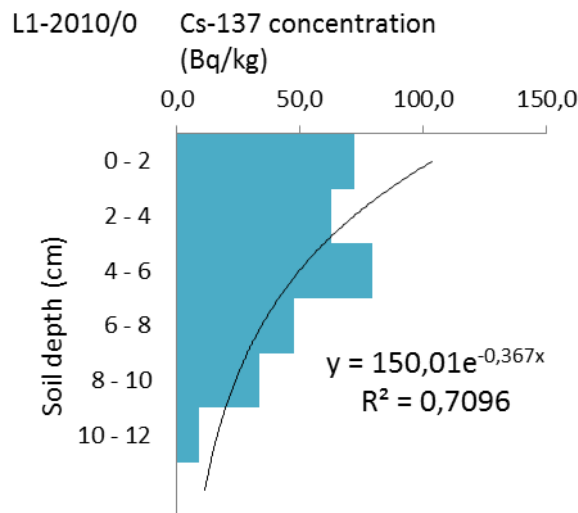


Figure 7. Depth distribution of Cs-137 in the soil at the L1-2010/0 measurement point (slope < 4 degrees)

During the recent period since 2001, at the sampling plot with small slope grade (< 4 degrees) one of the factors shaping the profile besides a slight extent of soil displacement and the natural radioactive decay must have been the vertical migration of Cs-137, as a consequence of which the highest Cs-137 activity concentration shifted to the 4 to 6 cm deep zone. A similar phenomenon was experienced at the sampling plots with greater slope grades, but there, in addition to the aforementioned, also soil movement did shape the surface-close Cs-137 distribution (*Figure 8*) to a great extent.

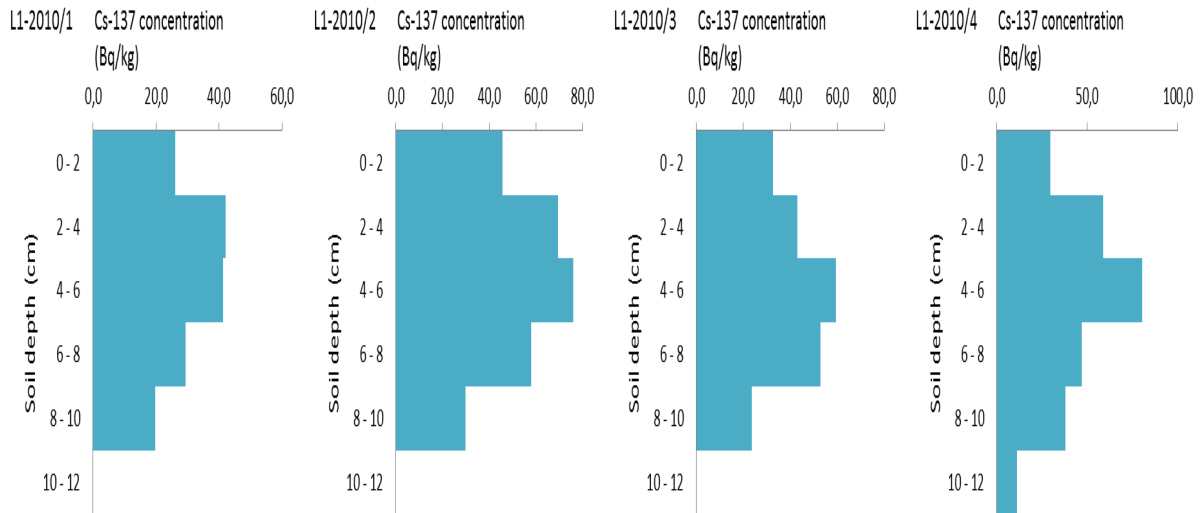


Figure 8. Cs-137 activity concentration depth distribution in the soil at the L1-2010/1 - L1-2010/4 measurement points (slope > 4 degrees)

From this soil profile alteration, one can deduce the vertical speed of movement, which can be estimated based on the samples to be 4 to 6 cm in 9 years (0.44 to 0.66 cm/yr) in the soil of the area. Arapis et al. (1997) estimate this value to be between 0.4 and 1.2 cm/yr. In order to gain better understanding of the presumed phenomenon, further samplings will be needed in the future, through which the presumed Cs-137 relocation can be traced.

3.3 The dependence of Cs-137 activity concentration on slope inclination

On creating a graphic representation of the Cs-137 activity concentration of the surface-close (0 to 2 cm) layer of sampling plots in function of slope inclination it became apparent that there is a connection between Cs-137 activity concentration at the surface and slope inclination. The regression line fitting to the values derived is shown in Figure 9. Based on the connection between the data it was concluded that one of the main reasons of differing surface activity values was soil movement (considering that Cs-137 is tightly bound to the organic matter and the clay minerals of soils).

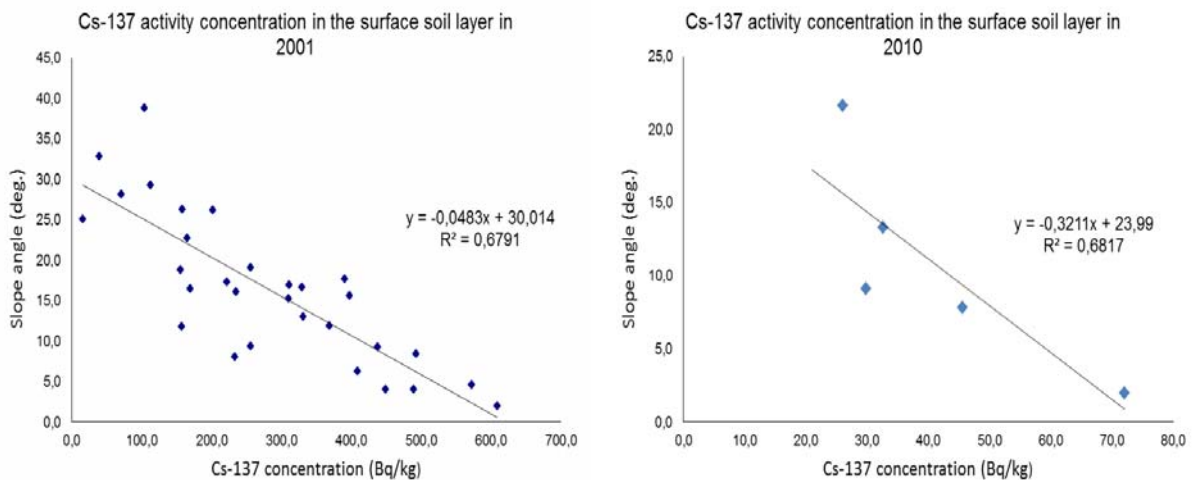


Figure 9. Slope inclination dependence of surface-close (0 to 2 cm) Cs-137 activity concentration of the measurement points in 2001 and 2010

4 SUMMARY AND CONCLUSIONS

Measurements on soil samples of a small water catchment (Farkas Trench) of the Sopron Mountains showed that there is Cs-137 isotope present in the area in well measurable quantities. In 2001, the depth distribution of Cs-137 activity concentration in the soil of the tested area took the shape of an exponential function. When fitting regression functions to the sampled values it was found that at the plots with lesser slope steepness (< 4 degrees) exponential functions provided the best match. As slope inclination increased, so did this Cs-137 profile. At sampling plots owning greater inclination a decrease in Cs-137 activity concentration of the top layers appeared. In 2010, at the sampling points, the activity concentration of the surface layers (0 to 2 cm) of the soil varied between 15 and 609 Bq/kg, whereas in 2001 between 26 and 72 Bq/kg. An interconnection between activity concentration in the surface soil layer and slope inclination could be indicated. From the slope inclination dependence of areal distribution it was concluded that one of the main reasons of areal alteration of Cs-137 activity concentration was differing extent of soil movement.

It appeared in the 2010 measurement data that the highest Cs-137 activity concentration had shifted lower from the top layers, and the depth distribution was altering along an increasing or a stagnating function to a depth of 4 to 6 cm (in function of slope inclination), then along a decreasing function. From this soil profile alteration the vertical migration speed could be deduced, which can be estimated to be 0.44 to 0.66 cm/year in the soil in the area.

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