MEASUREMENTS AND MODELLING OF ^{137}Cs MIGRATION INTO VARIOUS TYPES OF SOIL

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INTRODUCTION

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. Therefore, the investigation of radiocaesium migration in soil is a focus of several research all over the world (1,2).

The level of soil contamination in Central and Eastern Europe after the nuclear accident at Chernobyl has been high enough to study the vertical migration of radicaesium under natural conditions. In Hungary, ^{137}Cs measurements have been carried out regularly since the Chernobyl accident at several location to determine the long-term migration of radiocaesium in soil system (3).

In the project presented here, the distribution of ^{137}Cs activity concentration has been measured and analyzed over the period of 1987-1996. On the experimental data collected during these ten years a model has been developed which can be used for long-term prediction of radioisotope penetration.

SAMPLING AND SAMPLE PROCESSING

Approximately 30 * 30 cm^2 areas on plane, grassy, uncultivated fields are sampled at each of five sites. Seven layers extending from the surface down to a depth of 20 cm are excavated at each of 11 sampling times (12, 14, 17, 24, 29, 36, 41, 48, 53, 83 and 118 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 °C, then homogenized and the grains exceeding 1.25 mm diameter are removed. After this processing activities of 400 cm^3 volumes are measured by HpGe semiconductor gamma spectrometry in Marinelli geometry.

SOIL CHARACTERISTICS

Five sites have been selected to represent five different soil types, characteristic in Hungary. Several measured soil parameters are given in the Table below.

Identification	black rendzina	humic sandy soil	meadow soil	typical Ramann brown forest soil	brown forest soil with clay illuviation.
Site	Ady-liget	Szada	Domony	Galgamácsa	Dobogókô
pH (KCl)	5.5	7.5	7.3	7.2	4.2
$CaCO_3$ (%)	-	0.5	7.2	4.2	-
org. matter (%)	3.9	1.3	4.6	3.1	5.1
cation exhange capacity (me/100g)	43.0	9.8	26.8	31.0	23.1
changeable K (me/100g)	0.6	0.04	1.12	1.65	2.72
density (g/cm^3)	1.14	1.70	1.45	1.24	1.13

ERROR SOURCES

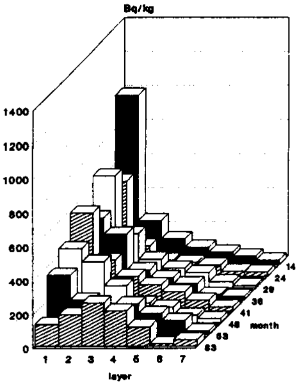
There are many sources of errors and uncertainties in the final results: short-range soil inhomogenities, the roughness of soil surface, the uncertainties in soil layer thicknesses, and errors of the spectrometric measurements.

To improve the error estimation at the last but one sampling three samples were taken and evaluated parallelly at each site. The standard deviations of the three sample sets are in the range of 25-40 %

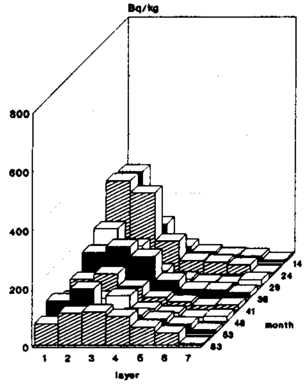
The effect of the sampling uncertainties are decreased by determining the actual weights of the layers, in g/cm^2, and using these values in the evaluation, rather than the nominal thicknesses in cm.

MEASURED CONCENTRATIONS

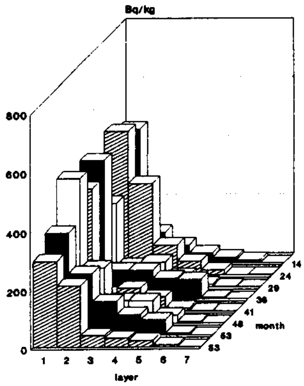
As an illustration, the measured activity concentrations are shown for three types of soil



brown forest soil with clay illuviation (Dobogókõ)



meadow soil (Domony)



humic sandy soil (Szada)

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

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

where
- $a(z,t)$ is the activity concentration (Bq/cm^3) at time t and depth z,
- a_{Ch} is the Chernobyl fall-out on the soil surface (Bq/cm^2),
- a_{nt} is the fall-out of the nuclear weapon tests on the soil surface (Bq/cm^2),
- D is the diffusion coefficient (cm^2/month),
- u is the convection velocity (cm/month),
- λ is the decay constant of ^{137}Cs (1/month),
- τ is the average time between the nuclear tests and the Chernobyl accident (set to 300 months).

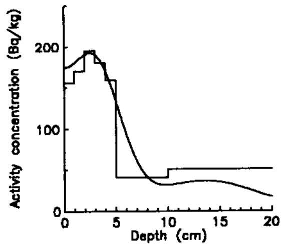
Furthermore, a total reflection at the air-ground interface is assumed.

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 the next table.

Identification	black rendzina	humic sandy soil	meadow soil	typical Ramann brown forest soil	brown forest soil with clay illuviation
Site	Ady-liget	Szada	Domony	Galgamácsa	Dobogókô
a_{Ch} (Bq/cm^2)	0.76	0.61	0.53	0.70	1.43
a_{nt} (Bq/cm^2)	0.61	0.84	0.76	0.68	1.14
u (cm/y)	0.082	0.042	0.220	0.106	0.424
D (cm^2/y)	0.343	0.053	0.400	0.041	0.433

According to the best fits the ratio of the maximum and minimum convection velocities is about 10, and the diffusion coefficients vary also within a factor of about 10.

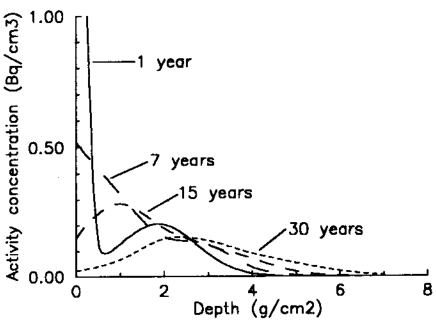
As an illustration, the measured activity concentrations (hystogram) and the fitted depth profile (continuous curve) for brown forest soil with clay illuvation (Dobogókô) are given in the figure for 83 months after the accident.



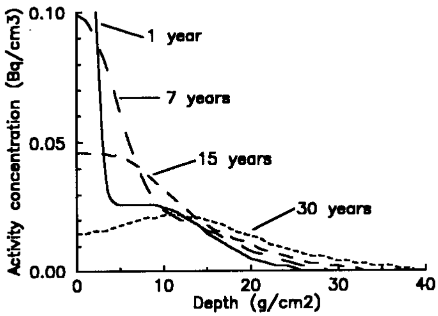
The goodness of fit is somehow characterized by the fact that the initial surface activities are in the range of 0.6-0.8 Bq/cm^2, both for Chernobyl and the nuclear tests, as expected form earlier measurements at these sites around Budapest, i.e. at one of the most contaminated areas of Hungary. Both values are slightly but significantly higher at Dobogókô, where samples were taken at a site with higher precipitation.

MODEL PREDICTIONS

Assuming the correctness of the model for longer time periods, predictions can be made for future times. Activity concentration curves computed for 15 and 30 years are given in the next figures for two types of soil.



Humic sandy soil (Szada)



Meadow soil (Domony)

CONCLUSIONS

The penetration of caesium into the soil is a very slow process, the majority of the activities is still in the top 1-5 cm layer.

According to the measurements carried out in the first ten years following the Chernobyl accident the migration can well be modelled by the diffusion-convection model. Since the uncertainties are relatively large, further measurement series are required to confirm the validity of the model and to improve the parameter estimations.

REFERENCES

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