^{90}Sr profile in soil samples from the East Urals Radioactive Trail (EURT): a quantitative approach

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Abstract

An analysis both of the physico-chemical properties and the radionuclide concentrations in some soil samples obtained in the vicinity of Lake Tygish (Sverdlovsk Region, Middle Urals, Russia) are reported. By taking into account the meteorological parameters of the region, a model has been developed and applied to the experimental data. A good agreement with the experimental data for ^{90}Sr concentration was obtained by assuming, respectively, an effective transport velocity (v_{eff}) varying between 0.1 and 0.8 cm yr^{-1} and an effective diffusion coefficient (D_{eff}) varying between 0.4 and 3.4 cm^2 yr^{-1} from the surface to a depth of 40 cm. These results may be useful for the estimation of the propagation of ^{90}Sr in such kinds of soil. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Many papers have been published on the East Urals Radioactive Trail (EURT) on account of the relevant amount of radionuclides released into the environment by the

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accident. The first study ever published on this special topic is due to Agre and Korogodin (1960). Their report after the Kyshtym accident, which happened on 27 September 1957, was presented for publication on 11 October 1958 and was published only 2 years later without any indication of the place where the research was carried out because anything concerning the accident was classified.

On June 21, 1995, during an Advanced Course on Radioecology in Zarechny (Russia) organized by the International Union of Radioecology and financed by NATO as an Advanced Study Institute, an excursion in the region around Kamenks-Ural'skiy was carried out. Soil samples were collected at two points. The first one was at some tens of metres from an abandoned graveyard; the soil was sampled from the surface to a depth of 80 cm; the second one was at 100 m from the shore of Lake Tygish.

This area is included within the EURT, originated by the Kyshtym accident when about 740 PBq of radionuclides were released into the atmosphere (Romanov, Nikipelov & Drozhko, 1991) after a chemical explosion.

One of the aims of the paper is a quantitative analysis of the migration process to determine the average values of the effective migration diffusion coefficients and transport velocities of ^{90}Sr in the soils examined. Such a quantitative analysis has been done by developing and calibrating a migration model based on the transport–diffusion equation.

Previous studies (see, e.g. Bachhuber, Bunzl, Schimmack & Gans, 1982) suggested that the values of the effective transport and diffusion coefficients depend on the soil horizon. The calibration of the transport-diffusion model gives the opportunity of evaluating the migration parameters, as a function of depth, in soils collected in an area characterised by a severe nuclear accident. Data relevant to the main characteristics of the soils are reported in Table 1 in order to supply any relevant information for any future application of this model to other situations.

The composition of the radioactive mixture has been estimated with some uncertainty by different authors, and the contribution of ^{90}Sr + ^{90}Y ranges between 5.4 and 7.0% (Trabalka & Auerbach, 1991). At present, i.e. 40 years after the accident, the short-lived radionuclides have disappeared and only ^{90}Sr + ^{90}Y can be measured since ^{137}Cs contribution from Kyshtym was too small to be detected. As a consequence of the Chernobyl accident and weapons fallout some ^{137}Cs was also found.

2. Sampling description

A trench 80 cm deep was dug in undisturbed soil in the vicinity of an abandoned graveyard. Layers 10 cm thick were collected but, in the upper one, the first 5 cm with the grass root mat was discarded.

About 100 m north of the lake Tygish shore other samples were collected. For practical reasons only the upper layer of 10 cm was considered and divided into two sublayers: 0–5 and 5–10 cm; in order to ascertain the variability of the deposition, two samples of the sublayer 5–10 cm were taken. Underneath a depth of 45 cm the soil texture changed abruptly and a gley (sticky clay) horizon was found; a sample of the gley was also collected.

Table 1
Physical characteristics and chemical composition of the soil samples (uncertainties = 2 sigma)

Sampling point	Layer (cm)	Density (g cm⁻³)	Porosity (cm³ g⁻¹)	Organic matter %	Pb (ppm) (±5%)	Cr (ppm) (±5%)	Ca (%) (±2%)	Fe (%) (±2%)	Mn (%) (±0.01)
Graveyard	0–5	—	—	—	—	—	—	—	—
	5–10	1.53 ± 0.30	0.06 ± 0.02	8.4 ± 1.8	17	90	1.22	2.17	0.16
	10–20	1.52 ± 0.30	0.04 ± 0.01	5.5 ± 1.9	18	95	1.07	2.24	0.12
	20–30	1.74 ± 0.35	0.05 ± 0.02	3.8 ± 1.9	14	90	1.07	3.29	0.08
	30–40	1.88 ± 0.32	0.10 ± 0.04	5.8 ± 1.9	14	100	1.07	4.06	0.06
	40–50	1.92 ± 0.38	0.12 ± 0.05	5.6 ± 1.9	15	115	1.00	4.20	0.07
	50–60	1.93 ± 0.39	0.14 ± 0.06	4.9 ± 1.9	16	100	1.07	3.92	0.07
	60–70	1.97 ± 0.40	0.14 ± 0.06	5.3 ± 1.9	18	100	1.07	3.99	0.09
	70–80	1.93 ± 0.39	0.16 ± 0.07	4.3 ± 1.9	17	95	1.07	3.92	0.08
Lake shore	0–5	1.89 ± 0.22	0.75 ± 0.18	46.7 ± 1.4	45	91	7.58	2.73	0.29
	5–10	1.65 ± 0.19	0.75 ± 0.17	49.3 ± 1.4	73	120	6.79	3.01	0.20
	10–45	1.71 ± 0.23	0.72 ± 0.19	41.3 ± 1.4	50	100	4.72	2.80	0.12
	45–50	2.08 ± 0.42	0.07 ± 0.03	3.5 ± 1.9	12	178	1.22	2.52	0.03

3. Sample treatment and measurement

Samples were contained in hermetically sealed plastic bags and safely transported to CIEMAT for later processing and analysis. On reaching the laboratory, samples were dried at 60°C until constant weight (wet and dry weight recorded) and a part of the sample of each layer was taken in order to determine bulk density, porosity and organic matter. The rest of the sample was further dried at 200°C for 24 h, and after homogenizing, different aliquots were taken for radionuclide and chemical analyses; ^{90}Sr analysis was carried out after gamma-ray spectrometry.

4. Determination of soil parameters

The physical parameters characterizing the soil samples were determined according to the procedures described below.

4.1. Bulk density (ρ)

An aliquot of the dried sample (60°C) was compacted into a known volume (V), and its weight recorded (R_w); a duplicate was assayed for each sample layer. The density was calculated from

$$\rho(g/cm^3) = R_w/V.$$

4.2. Porosity (ϕ)

This was calculated from the following formula where D_w is the dry weight (sample at 60–70°C for 24 h) and W_w is the wet weight (sample as it arrived in the laboratory)

$$\phi(cm^3/g) = \frac{1}{D_w/\rho(W_w - D_w) + 1}.$$

4.3. Organic matter (Ω)

The remainder of the sample was ashed at 600°C for 48 h and the weight recorded (A_w). The concentration of organic matter was calculated from the following formula:

$$\Omega(\%) = \frac{D_w - A_w}{D_w} \times 100.$$

5. Chemical analyses

The composition of soil samples (Ca, and heavy metals) was determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES), except for Pb,

which was analyzed by atomic absorption spectroscopy (AAS). 0.5 g sample was dissolved by the use of a mixture of 4 ml of concentrated HF and 2 ml of aqua regia, in a PTFE (Polytetrafluorethylene) closed bomb, and standing overnight at 100°C. After cooling, the content was transferred to a PTFE evaporating dish and 2 ml of concentrated $HClO_4$ were added and evaporated until fumes using infrared lamps. Finally, the content of the dish was dissolved with 1 M HNO_3 and transferred to a 50 ml volumetric flask.

Major elements were determined in a 1 : 5 diluted solution, after addition of 10 mg l^{-1} of Y, used as internal standard. Trace elements were determined directly from the sample solution.

6. Gamma spectrometry

Each sample was measured by gamma spectrometry in a coaxial HP-Ge (n-type) detector. The necessary amount of each soil sample was transferred to the 115.45 cm^3 volume petri dish-geometry used in calibration. After recording gamma-emitting radionuclide concentrations in each sample, different aliquots of soil were prepared for radiochemical analyses of ^{90}Sr.

7. ^{90}Sr Analyses

The classical precipitation method has been used for Sr determination in samples. 10–20 g of sample was leached with HNO_3 and Ca and other interferents were separated by NO_3^- precipitation. Y and Ba were eliminated by precipitation as $Fe(OH)_3$ and $BaCrO_4$, respectively, and radiostrontium was separated as $SrCO_3$. The precipitate was purified and then counted twice in a low background alpha/beta gas flow detector with a time interval between the counts of about two weeks in order to allow the ingrowth of ^{90}Y to approach secular equilibrium with the ^{90}Sr. Chemical recovery was obtained by the addition of stable Sr carrier (Alvarez, 1993).

8. Results

The results of the measurements described in the preceding paragraph are summarized in Table 2. Concerning the sampling point close to an abandoned graveyard, it is evident that ^{90}Sr migrated rather slowly into the soil; in fact most of the ^{90}Sr is still in the upper layer of 20 cm. ^{137}Cs has a similar distribution but its origin is quite different because most of it originated from the Karachay and the Chernobyl accident (Aarkrog et al., 1992,1997).

The soil sampled in the vicinity of the lake shore has a definitely higher concentration of both radionuclides which can be explained by the contribution of run-off since the lake shore is obviously at a lower altitude than the first sampling point. The layer 5–10 cm from the surface was sampled twice and the concentrations measured in these

Table 2
Concentrations of ^{90}Sr and ^{137}Cs in the soil samples (uncertainties = 2 sigma)

Sampling point	Layer (cm)	^{90}Sr (Bq kg$_{dw}^{-1}$)	Ca (g kg$_{dw}^{-1}$)	^{137}Cs (Bq kg$_{dw}^{-1}$)	K (g kg$_{dw}^{-1}$)
Graveyard	0–5	—	—	—	—
	5–10	406 ± 17	12.2 ± 0.2	47 ± 3.3	17 ± 1
	10–20	130 ± 9.2	10.7 ± 0.2	12 ± 1	15 ± 1
	20–30	17 ± 7.7	10.7 ± 0.2	< 1.2	18 ± 1
	30–40	7.3 ± 3.0	10.7 ± 0.2	< 1.2	19 ± 1
	40–50	3.7 ± 1.5	10.0 ± 0.2	< 0.98	16 ± 1
	50–60	0.96 ± 0.94	10.7 ± 0.2	< 1.1	18 ± 1
	60–70	< 1.0	10.7 ± 0.2	< 1.3	17 ± 1
	70–80	< 3.8	10.7 ± 0.2	< 1.2	17 ± 1
Lake shore	0–5	74 ± 17	75.8 ± 1.5	497 ± 34	15 ± 2
	5–10	736 ± 50	67.9 ± 1.3	828 ± 57	17 ± 2
		514 ± 37	47.2 ± 0.9	539 ± 37	18 ± 2
	10–45	—	—	—	—
	45–50	1.8 ± 1.4	12.2 ± 0.2	< 1.9	11 ± 1

samples are different outside the range due to the error of measurement. Since the sample with a higher concentration of ^{90}Sr has also a higher concentration of ^{137}Cs, such a difference should be due to variability of the soil characteristics and not to a "hot particle" because ^{90}Sr and ^{137}Cs were released by totally different events.

In fact, it must be pointed out that a gamma spectrum of the most active sample performed at the ENEA Saluggia Research Centre a few months after the sampling, gave a ratio ^{137}Cs/^{134}Cs = 1.8 ± 0.5 (corrected back to 26 April 1986); therefore the radiocaesium in the soil sample originated from the Chernobyl accident only.

The results obtained for this sampling point confirm the hypothesis that radionuclides had a rather low migration into the soil, because no ^{137}Cs above the detection limit was found in the layer from 45 to 50 cm which is formed of gley, a sticky clay as reported before, which would have captured any ^{137}Cs which came into contact with it.

By assuming a soil density of 1.5 kg dm^{-3} and an average concentration of 600 Bq kg^{-1} of ^{90}Sr in the upper layers of soil, a mean value of 6.8 Ci km^{-2} was obtained for the deposition in 1957 in good agreement with the data given by Romanov, Nikipelov and Drozhko (1991) and Yáchmenyov and Isageva (1996), who reported a range of 5–20 Ci km^{-2} in the area where the samples were collected.

9. Meteorological data

In order to evaluate the influence of precipitation on the penetration of ^{90}Sr in the soil, the amounts of yearly precipitation divided between the cold period (November–March) and the warm one (April–October) since 1957 until the sampling have

Table 3
Atmospheric precipitation (mm) from November 1957 to March 1995

Year	Amount of precipitation		Year	Amount of precipitation	
	Cold period[a] Nov.–March	Warm period April–October		Cold period[a] Nov.–March	Warm period April–October
1958	148	286	1977	90	250
1959	98	371	1978	93	450
1960	62	373	1979	145	364
1961	116	375	1980	71	355
1962	177	338	1981	115	260
1963	139	293	1982	85	370
1964	103	419	1983	119	375
1965	119	238	1984	95	449
1966	176	347	1985	105	279
1967	58	353	1986	166	424
1968	113	325	1987	123	524
1969	106	432	1988	77	264
1970	102	436	1989	153	368
1971	125	444	1990	140	460
1972	115	337	1991	127	396
1973	105	314	1992	110	428
1974	104	250	1993	110	513
1975	105	232	1994	83	483
1976	109	261	1995	98	178

[a]The cold period starts in November of the previous year and finishes in March of the current year.

been obtained from the Urals Territorial Management on Hydrometeorology and Environment Monitoring for the Meteorological Station of Kamenks-Ural'skiy which is about 20 km from the sampling point (Table 3).

10. Modelling the migration of ^{90}Sr in soil

The migration of radionuclides in soils is controlled by the following processes:
(a) sorption of radionuclides by the solid soil particles;
(b) diffusion of radionuclides in the aqueous phase;
(c) transport of radionuclides due to the flow of interstitial water in soil;
(d) radioactive decay.

The sorption of a radionuclide by soil particles may be modelled by means of the partition coefficient concept:

$$\frac{C_s}{C_w} = k_d, \tag{1}$$

where C_s is the radionuclide concentration in soil (Bq kg^{-1}), C_w is the radionuclide concentration in the soil interstitial water (Bq m^{-3}) and k_d is the partition coefficient (m^3 kg^{-1}). Eq. (1) is based on the hypothesis that both the dissolved and the attached phases of the radionuclide reach a quick, reversible equilibrium. More generally, it is necessary to account for non-reversible processes controlling the interactions of the radionuclide in dissolved form with the soil particles. Such processes are of importance for some radionuclides such as ^{137}Cs (see, for instance, Comans & Hockley, 1992).

The non-reversible process may be modelled by the following general equations:

$$F = k_{12}C_s, \quad \frac{\delta C_s^1}{\delta t} = k_{12}C_s, \tag{2}$$

where F is the non-reversible fixation rate of the radionuclide (Bq kg^{-1} s^{-1}), k_{12} is the non-reversible fixation constant (s^{-1}) and C_s^1 is the concentration (Bq kg^{-1}) in soil of the radionuclide in non-reversible form.

The solute flux $\mathbf{J}$ (Bq m^{-2} s^{-1}) is

$$\mathbf{J} = (-D\mathbf{grad}C_w + \mathbf{v}C_w)\theta, \tag{3}$$

where D is the diffusion coefficient (m^2 s^{-1}), $\mathbf{grad}$ the gradient operator, $\mathbf{v}$ (m s^{-1}) the mean pore water velocity and θ the volumetric water content of the soil (m^3 m^{-3}).

Combining formula (3) with the equation of the mass balance we get

$$\frac{\delta}{\delta t}(\theta C_w + \rho C_s + \rho C_s^1) + \lambda(\theta C_w + \rho C_s + \rho C_s^1)\text{div }\mathbf{J} = 0, \tag{4}$$

where ρ is the soil density (kg m^{-3}), λ the radioactive decay constant (s^{-1}) and div the divergence operator. Eqs. (1)–(4), together with the suitable initial conditions allow one to predict the time behaviour of a radionuclide in soil. Obviously, it is quite impossible to obtain all the time-dependent site specific values of the model parameters in Eq. (1)–(4). Indeed the parameters in the above formulae, such as the soil moisture content and the infiltration velocity of the water, are functions of time and of depth. Therefore, even when copious experimental data of radionuclide concentrations at different depths are available, the "degrees-of-freedom" of the migration model give ample room for more complex and refined theoretical speculations. These may be used to explain any kind of behaviour of the radionuclide vertical profiles in any soil and in any circumstance by claiming presumptive, approximate knowledge of the values of the non-measured parameters.

It seems more convenient to simplify the above model and to evaluate, by means of a calibration, the values of specific parameters that may be useful for approximate predictions of radionuclide migration in general cases.

The radionuclide concentrations in the interstitial water of each soil horizon characterized by approximately constant values of the parameters in Eq. (4) may be evaluated, after simple (but boring) calculations, by the following equation:

$$\frac{\delta C_w}{\delta t} D_{\text{eff}} \frac{\delta^2 C_w}{\delta z^2} - v_{\text{eff}} \frac{\delta C_w}{\delta z} - \lambda C_w, \tag{5}$$

where D_{eff} and v_{eff} are, respectively, the effective diffusion coefficient and the effective transport velocity which are related to D and v according to the following equations:

$$D_{eff} = \frac{D}{R}, \quad v_{eff} = \frac{v}{R}, \tag{6}$$

where R is the, so-called, retardation factor

$$1 + k_d \frac{\rho}{\theta} = R. \tag{7}$$

Eq. (5) has been obtained on the hypothesis of a completely reversible equilibrium of the two phases and such a hypothesis is reasonable for ^{90}Sr.

It is interesting to note that using constant values for the effective diffusion and the effective velocity in Eq. (5), the predicted concentration profiles in soil do not agree with the experimental data. Indeed, following a single deposition pulse of a radionuclide on the ground (when the effective transport velocity is equal to 0) the diffusion Eq. (5) has the following analytical solution:

$$C_w = \frac{L}{(\theta + k_d \rho)\sqrt{\pi D_{eff} t}} e^{-x^2/4D_{eff}t - \lambda t}, \tag{8}$$

where I is the initial inventory of the radionuclide. As its concentration in soil is proportional to the concentration in interstitial water, the profile, on a logarithmic scale, of radiostrontium in soil is proportional to x^2, i.e. is a concave parabola whereas the experimental profile (Figs. 1 and 2) is convex.
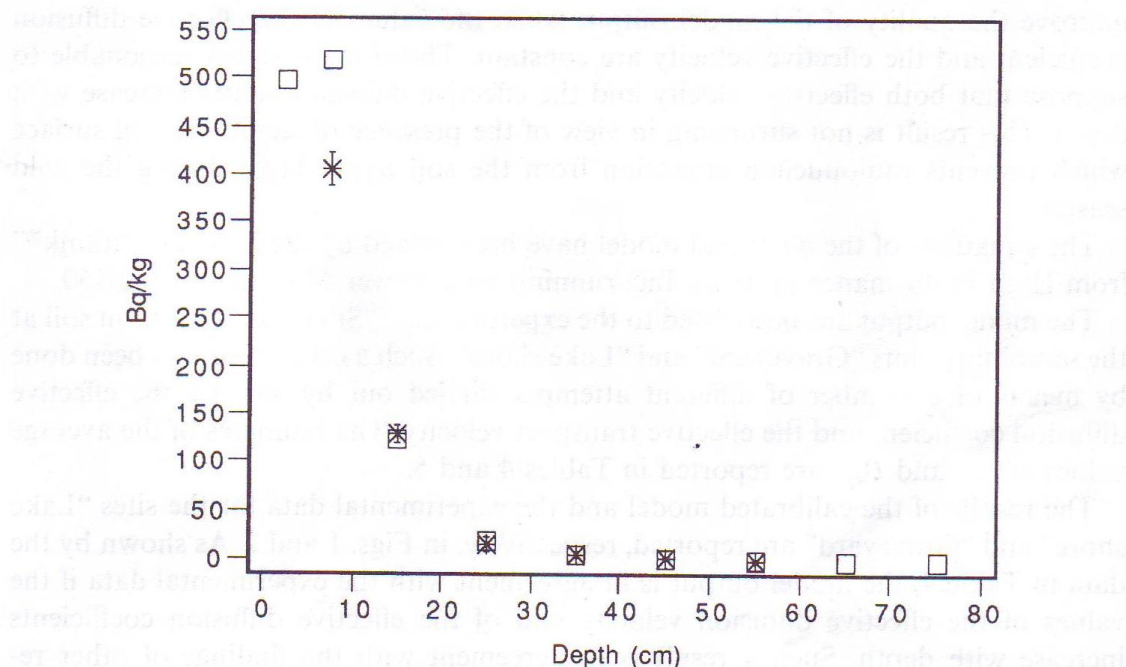

Fig. 1. Comparison of model output (squares) and experimental ^{90}Sr concentrations in soil (stars) at the "Graveyard" sampling point. The model data have been obtained by using the values of v_{eff} and D_{eff} reported in Table 4.
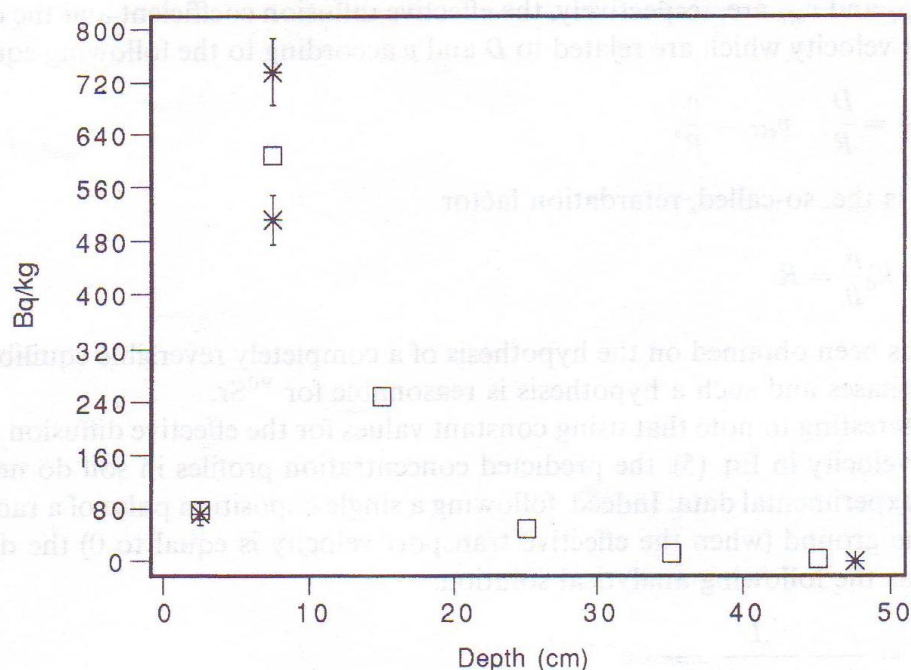
Fig. 2. Comparison of model output (squares) and experimental ^{90}Sr concentrations in soil (stars) at the "Lake shore" sampling point. The model data have been obtained by using the values of v_{eff} and D_{eff} reported in Table 4.

As the transport term in Eq. (5) corresponds to a spatial translation, it cannot improve the quality of the model output when the values of the effective diffusion coefficient and the effective velocity are constant. Therefore, it seems reasonable to suppose that both effective velocity and the effective diffusion values increase with depth. This result is not surprising in view of the presence of ice on the soil surface which prevents radionuclide migration from the soil upper layer during the cold season.

The equations of the described model have been solved by the Software "ithink®" from High Performance Systems, Inc. running on a Power Macintosh 8500/150.

The model output has been fitted to the experimental ^{90}Sr concentrations in soil at the sampling points "Graveyard" and "Lake shore". Such a calibration has been done by means of a number of different attempts carried out by varying the effective diffusion coefficient and the effective transport velocity. The estimates of the average values of v_{eff} and D_{eff} are reported in Tables 4 and 5.

The results of the calibrated model and the experimental data for the sites "Lake shore" and "Graveyard" are reported, respectively, in Figs. 1 and 2. As shown by the data in Table 4, the model output is in agreement with the experimental data if the values of the effective diffusion velocity and of the effective diffusion coefficients increase with depth. Such a result is in agreement with the findings of other researchers (Bachhuber et al., 1982).

The higher values of v_{eff} and D_{eff} in the upper layer of the site "Lake shore" compared with the values for "Graveyard" are due to the different physical and

Table 4
Effective transport velocity and effective diffusion coefficient evaluated by calibrating the model for the site "Graveyard"

Layer (cm)	v_{eff} (cm yr^{-1})	D_{eff} (cm^2 yr^{-1})
0–12.5	0.1	0.4
12.5–25	0.6	2.2
25–40	1.0	4.0

Table 5
Effective transport velocity and effective diffusion coefficient evaluated by calibrating the model for the site "Lake shore"

Layer (cm)	v_{eff} (cm yr^{-1})	D_{eff} (cm^2 yr^{-1})
0–5	0.4	1.8
5–12.5	0.1	0.4
12.5–25	0.4	1.8
25–40	1.0	3.4

chemical characteristics of the soils (Table 1). Indeed, a higher water content reduces the value of the retardation factor (see Eq. (6) and (7)) and consequently enhances the radionuclide migration.

On the other hand, a higher concentration of Ca (Table 2) may compete with the adsorption of Sr on soil particles (Sheppart & Thibault, 1990). The consequent reductions of the k_d and R values imply the increase of D_{eff} and v_{eff}.

11. Conclusions

Obviously, the estimated values of D_{eff} and V_{eff} are relevant to the specific environmental conditions of the site investigated and to a migration period of some decades. On the other hand, it is of importance to carry out studies for evaluating the effective radionuclide velocities and diffusions in field conditions. The information obtained is, indeed, useful for improving the reliability of simple generic models for predicting radionuclide transport in soil.

In the case considered in this paper it was possible to trace the ^{90}Sr propagation during nearly 40 years, into an undisturbed soil to á depth of 80 cm.

The model which has been developed to describe the phenomenon may be useful to forecast the evolution of a similar contamination in a soddy meadow soil over a rather long time interval.

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