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Migration of fallout radiocaesium in a grassland soil from 1986 to 2001
Part II: Evaluation of the activity–depth profiles by transport models

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Abstract

The vertical migration of ^{134}Cs, deposited by the Chernobyl fallout (1986), and ^{137}Cs, deposited by the Chernobyl and the global fallout, in the soil of an undisturbed Bavarian grassland in Germany was investigated from 1986 to 2001. The activity–depth profiles of both isotopes at ten sampling dates were evaluated by the classical convection–dispersion equation and a random walk particle model. In both models, the apparent migration velocity v and the apparent dispersion coefficient D were assumed to be independent of time. However, optimized values of v and D were significantly different for the different locations sampled at different times. If nevertheless constant values of v and D were used, the simulated activity densities per soil layer were out of the range of the spatial variability of the observed activity densities determined in 2001. It is concluded that without further simultaneous investigations e.g. on bioturbation at the study site, migration parameters of radiocaesium determined by classical transport models based on convection and dispersion during the first years after the deposition of the activity cannot be used for predictive purposes.

Keywords: ^{134}Cs; ^{137}Cs; Soil; Convection; Dispersion; Bioturbation; Spatial variability

1. Introduction

Among the anthropogenic radionuclides in the soil, radiocaesium plays a major role because it contributes significantly to the long-term radiation dose to man. To estimate this dose realistically, the migration of radiocaesium in the soil has to be quantified by transport models. In radioecology, the classical convection–dispersion equation (CDE) is mostly used for this purpose. In the CDE, the transport of the solute is described by two mechanisms: convection with the percolating water as characterized by the mean pore water velocity v_w, and hydrodynamic dispersion, i.e. molecular diffusion and mechanical dispersion caused by the spatial variation of v_w. The dispersion coefficient D_w is related to v_w by the longitudinal dispersivity $\alpha = D_w / v_w$. Since radiocaesium is strongly sorbed to soil particles, its migration is retarded against the percolating

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water: $v = v_w / R$, $D = D_w / R$, v being the 'apparent' or 'effective' migration velocity (cm a^{-1}), D the 'apparent' or 'effective' dispersion coefficient (cm^2 a^{-1}) of radiocaesium, and R the retardation factor. Assuming a linear adsorption isotherm for the very small mass concentrations of fallout radiocaesium in the soil, R depends on its distribution coefficient K_d: $R = 1 + (\rho / \Theta) \cdot K_d$, where ρ is the dry bulk density and Θ the water content of the soil. Then, the CDE of the one-dimensional convective–dispersive transport of radiocaesium is written as

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - v \frac{\partial C}{\partial x} \tag{1}$$

where $C(x,t)$ is the concentration of radiocaesium in the soil solution (Bq cm^{-3}), t the time (a) and x the soil depth (cm). All concentrations are resident concentrations (Jury and Roth, 1990). The activity density[1] A (=activity per unit area) of radiocaesium determined in a soil layer of thickness dx is related to C by $A = R \cdot \Theta \cdot C \cdot \mathrm{d}x$.

In general, the parameters v and D are assumed to be constant over x and t when integrating Eq. (1) (Smith and Elder, 1999; Bossew and Kirchner, 2004). However, even in homogeneous soils both parameters may apparently or actually depend on t, for instance due to a time dependent water flow (v_w, D_w), a time dependent sorption of radiocaesium (K_d), or the presence of mechanical mixing, e.g. by bioturbation. Many field and laboratory studies with undisturbed soils have revealed that D_w and α, respectively, are scaling values, i.e. they increase with increasing scale or, equivalently, with increasing time period of observation (e.g. Silliman and Simpson, 1987; Khan and Jury, 1990; Starret et al., 1996). This 'dispersion scale effect' is a consequence of the assumption that the dispersive flow can be quantified analogously to molecular diffusion by Fick's law (Jury and Roth, 1990; van Dam et al., 2004). The effect depends, however, on the soil structure and the water flow rate and can disappear at low flow rates (Khan and Jury, 1990; Vanderborght et al., 2001).

A time dependence of the sorption of radiocaesium to the soil is possible due to 'ageing' or 'fixation' processes during the first years after the deposition (Smith et al., 1999, 2000). According to laboratory studies of Comans and Hockley (1992) and Konoplev et al. (1996), an initial rapid adsorption of radiocaesium on illite and soils, respectively, is followed by slower uptake processes with time scales of weeks and months. Results of field studies on temporal trends in the radiocaesium migration (e.g. Bunzl et al., 1995; Rosén et al., 1999; Isaksson et al., 2001) also indicate a retardation during the first decade after 1986 by a factor of 2–3. However, due to the empirical approaches of these authors using simpler evaluation methods than the CDE with only one migration parameter not related to transport mechanisms, the relevance of the results for predictive purposes is not clear.

Physical mixing by bioturbation, i.e. mixing of the soil by earthworms and other burrowing animals, is one of the most important transport processes not related to the water flow (Müller-Lemans and Van Dorp, 1996). As pointed out by Anspaugh et al. (2002), physical, rather than chemical, processes in the bulk soil (and not only in the soil solution) can dominate the transport of radionuclides, at least in some types of soil. A consequence of physical mixing is that the activity–depth profiles of radionuclides with different chemical and sorption properties are similar. Actually Bunzl et al. (1994) observed at the grassland site of the present study that the depth distributions and the residence halt-times of ^{137}Cs from global fallout and $^{239+240}$Pu, both deposited at the same time but chemically completely different, were rather similar. As outlined by Boudreau (1986), bioturbation can be treated as a diffusive process covered in the CDE by the dispersion term. In this case, D is composed of three parts, molecular diffusion, hydrodynamic dispersion and physical mixing. The overall dispersive flux then depends on x and t because bioturbation decreases with depth (Müller-Lemans and Van Dorp, 1996) leading to a decreasing D with time due to the downward migration of radiocaesium and, thus, to an apparent time dependence of the total dispersion. In addition, the hydrodynamic part of the dispersion will also be affected due to the possibility of preferential water flows through earthworm burrows (Edwards et al., 1993).

Considering these potential complications of the radiocaesium migration it is not clear whether migration parameters obtained by applying the classical CDE can be used for calculating activity–depth profiles of radiocaesium in the future. Such profiles are needed for the long-term assessment of the radiation dose to man, particularly for the uppermost soil layers between 0 and about 15 cm depth, because the above-ground radiation of radiocaesium is attenuated by orders of magnitude by the accumulated soil mass above the source in deeper soil layers (Saito and Jacob, 1995). Therefore, the activity–depth profiles of ^{134}Cs and ^{137}Cs presented in the first part of this study (Schimmack and Schultz, 2006-this issue) were evaluated in the second part by the CDE. The profiles were determined at ten sampling dates between 1986 and 2001 in the soil of an undisturbed grassland in Bavaria. In order to detect

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[1] Quantities, units, and terms are used according to ICRU 65 (2001).

whether uncertainties due to numerical dispersion and discretisation seriously affect the results of the CDE the migration parameters were also determined by a random walk particle model (RWPM) developed by Bunzl (2001, 2002a,b) on the basis of the same transport processes as in the CDE, convection and dispersion.

2. Materials and methods

The study site and the determination of the caesium isotopes ^{134}Cs and ^{137}Cs are described in detail in the first part of this study (Schimmack and Schultz, 2006-this issue).

2.1. The convection–dispersion equation (CDE)

Assumptions of Eq. (1) not already mentioned in the Introduction are that the soil parameters ρ and Θ have also to be independent of x and t. Since this condition is not fulfilled, $\rho(x,t)$ and $\Theta\ (x,t)$ have to be substituted by their mean values averaged over the soil depth as well as over the observation period (for a deeper discussion see Shinonaga et al., 2005). The boundary condition of the integration Eq. (1) was that no activity input occurred at the soil surface during the observation period up to 2001. The initial conditions of the integration are explained in Section 3.2. Eq. (1) was solved numerically using a finite difference method by dividing the soil into simulation layers of uniform thickness of 1 cm. The calculations were performed with the Advanced Continuous Simulation Language (ACSL, version 11.7, AEgis Technologies Group, Austin, USA), a tool for modelling and simulating dynamic systems described by linear or nonlinear differential equations. The numerical dispersion was considered according to Duynisveld (1983).

2.2. The random walk particle model (RWPM)

The RWPM used in this study was developed by Bunzl (2001, 2002a,b). In this model, the transport of radiocaesium in the soil is represented by a set of independent particles moving one-dimensional paths through the soil. The paths are decomposed into small, stochastically independent displacements downwards and upwards. If the transport is only by dispersion (and diffusion), at each step the probability for the particle to move in any of these two directions is equal. The elementary displacement δ_D due to dispersion modified by sorption processes during a time step Δt is $\delta_D = Z\sqrt{6D\Delta t}$, where Z is a random

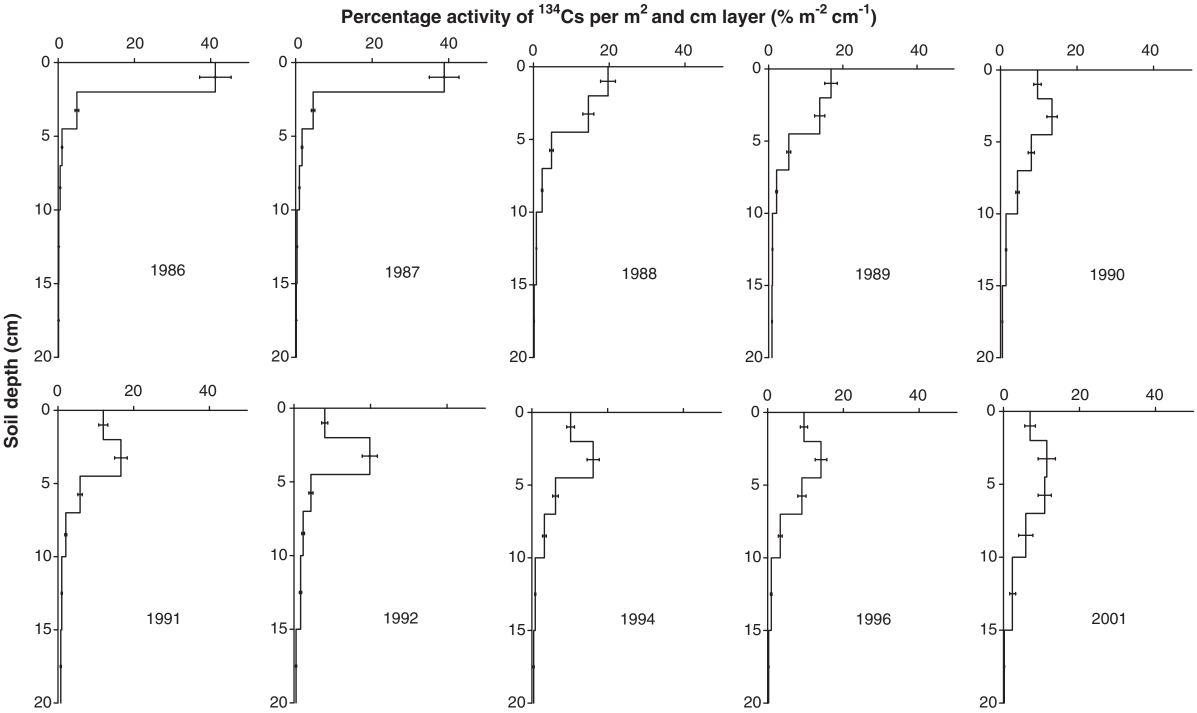

Fig. 1. Depth distribution of the volumetric activity concentration (=activity per squaremeter and per cm soil depth) of ^{134}Cs for 10 sampling dates between 1986 and 2001 at an undisturbed Bavarian grassland site, given as percentage (±experimental uncertainty) of the inventory of ^{134}Cs (% m^{-2} cm^{-1}).

Table 1
'Individual' evaluation of the ^{134}Cs migration in seven soil layers between 0 and 30 cm depth at a grassland site in Bavaria using the convection–dispersion model (CDE) and a random walk particle model (RWPM) with v = apparent vertical migration velocity of ^{134}Cs, D = apparent dispersion coefficient of ^{134}Cs, and RSS = residual sum of squares

Sampling year	CDE			RWPM		
	v	D	RSS	v	D	RSS
	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2
1987	0.11	0.00	19	0.06	0.00	22
1988	0.03	2.7	13	0.00	2.5	25
1989	0.00	2.6	44	0.00	2.2	57
1990	0.47	1.8	25	0.40	2.0	48
1991	0.34	0.71	67	0.30	0.40	89
1992	0.36	0.22	135	0.30	0.15	181
1994	0.25	0.44	64	0.20	0.40	114
1996	0.21	0.43	15	0.20	0.35	24
2001[a]	0.21	0.48	12	0.18	0.65	38
Range	0.00–0.47	0.00–2.7	12–135	0.00–0.40	0.00–2.5	24–181
Mean	0.22	1.04	44	0.21	0.93	67
SD[b]	0.16	1.04	40	0.11	0.93	53
CV[c] (%)	71	100	92	51	100	79
Median	0.21	0.48	25	0.20	0.40	48

The ^{134}Cs migration started at 13.6.1986 with the depth distribution of ^{134}Cs at this date as initial condition.

[a] The mean depth distribution of five plots was evaluated.

[b] Standard deviation.

[c] Coefficient of variation.

number uniformly distributed in [−1, +1] interval. The elementary step length δ_C for the vertical transport by convection modified by sorption processes during each time interval is given by $\delta_C = v\ \Delta t$. In contrast to δ_D, δ_C is always in the downward direction. The particle jumps randomly on its path along the x-axis (soil depth) beginning from $x=0$ at $t=0$. The upper (aerial) boundary ($x=0$) is fully impermeable (reflecting). After $n = t/\Delta t$ jumps, the final position x_{end} attained by the particle is recorded. Due to the random character of the process, different values for x_{end} will be obtained in general for different runs. If, however, this procedure is repeated k times (where k is about 10,000), the percentage fraction of the particles found in each soil layer of a given thickness, and thus the concentration depth profile of the particle in the soil after the time t, is obtained.

2.3. Parameter estimation

In both models, the best approximation of the experimental data was found by a least square fit minimizing the residual sum of squares RSS

$$\mathrm{RSS} = \sum_{l}^{1,n} \left[O_i - \sum_{J}^{k(i),k(i+1)-1} A_{ij} \right]$$

where O_i is the observed activity in the sampling layer i (total number of sampling layers: n), A_{ij} is the simulated activity in the simulation layer j, and $k(i+1)-1-k(i)$ is the number of simulation layers corresponding to the ith sampling layer ($k(1)=1$). Because the focus of this study was the simulation of the data for the soil layers between 0 and 15 cm depth (see 'Introduction'), the goodness of fit was tested for these layers by the Chi-square test, and

Table 2
'Individual' evaluation of the ^{134}Cs migration in seven soil layers between 0 and 30 cm depth at five plots of a grassland site in Bavaria, sampled in 2001, using the convection–dispersion model with v = apparent vertical migration velocity of ^{134}Cs, D = apparent dispersion coefficient of ^{134}Cs, and RSS = residual sum of squares

Sampling plot	v	D	RSS
	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2
A	0.23	0.58	24
B	0.28	0.32	17
C	0.17	0.30	57
D	0.15	0.57	28
E	0.21	0.52	33
Range	0.15–0.28	0.30–0.58	17–57
Mean	0.21	0.46	32
SD[a]	0.05	0.14	15
CV[b] (%)	23	30	47
Median	0.21	0.52	28

The ^{134}Cs migration started at 13.6.1986 with the depth distribution of ^{134}Cs at this date as initial condition.

[a] Standard deviation.

[b] Coefficient of variation.

Table 3
'Individual' evaluation of the ^{137}Cs migration in seven soil layers between 0 and 30 cm depth at a grassland site in Bavaria using the convection–dispersion model with v = apparent vertical migration velocity of ^{137}Cs, D = apparent dispersion coefficient of ^{137}Cs, and RSS = residual sum of squares

Sampling year	v	D	RSS
	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2
1987	0.06	0.00	6
1988	0.00	2.7	8
1989	0.00	2.4	22
1990	0.52	1.3	12
1991	0.34	0.41	20
1992	0.36	0.19	62
1994	0.24	0.37	34
1996	0.20	0.41	24
2001[a]	0.20	0.47	23
Range	0.00–0.52	0.00–2.7	6–62
Mean	0.26	0.62	32
SD[b]	0.12	0.58	22
CV[c] (%)	47	94	69
Median	0.23	0.37	26

The ^{137}Cs migration started at 13.6.1986 with the depth distribution of ^{137}Cs at this date as initial condition.

[a] The mean depth distribution of five plots was evaluated.

[b] Standard deviation.

[c] Coefficient of variation.

potential systematic deviations between observations and model by the Wald–Wolfowitz test. These tests and the correlation analysis were performed by the software Statistica, Version 7.1 (Statsoft, Tulsa, USA, 2005).

Table 4
'Individual' evaluation of the ^{137}Cs migration in seven soil layers between 0 and 30 cm depth at five plots of a grassland site in Bavaria, sampled in 2001, using the convection–dispersion model with v = apparent vertical migration velocity of ^{137}Cs, D = apparent dispersion coefficient of ^{137}Cs, and RSS = residual sum of squares

Sampling plot	v	D	RSS
	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2
A	0.22	0.57	31
B	0.26	0.27	21
C	0.17	0.28	35
D	0.16	0.69	24
E	0.20	0.45	60
Range	0.16–0.26	0.27–0.69	21–60
Mean	0.20	0.45	34
SD[a]	0.04	0.18	16
CV[b] (%)	20	40	46
Median	0.20	0.45	31

The ^{137}Cs migration started at 13.6.1986 with the depth distribution of ^{137}Cs at this date as initial condition.

[a] Standard deviation.

[b] Coefficient of variation.

3. Results

3.1. Activity–depth profiles of ^{134}Cs and ^{137}Cs

During the observation period 1986–2001, two radiocaesium isotopes were present in the soil: ^{137}Cs (half-life: 30.17 a) deposited by the global as well as by the Chernobyl fallout, and ^{134}Cs (2.06 a) deposited by the Chernobyl fallout only. Since Chernobyl-derived ^{137}Cs (CH-Cs) and ^{134}Cs exhibited a known ratio in the Chernobyl fallout (Hötzl et al., 1987), the migration behaviour of CH-Cs can be studied via ^{134}Cs. This is important because CH-Cs is more relevant for the long-term radiation dose than ^{134}Cs due to its longer half-life. At the study site, the inventory of total ^{137}Cs is a mixture of about 92% CH-Cs and 8% ^{137}Cs from global fallout (GF-Cs). The values for the activity density of ^{134}Cs and ^{137}Cs per soil layer can be found in Schimmack and Schultz (2006-this issue). In Fig. 1, the volumetric activity concentration of ^{134}Cs, i.e. the activity density per cm layer related to the inventory (% m^{-2} cm^{-1}), is

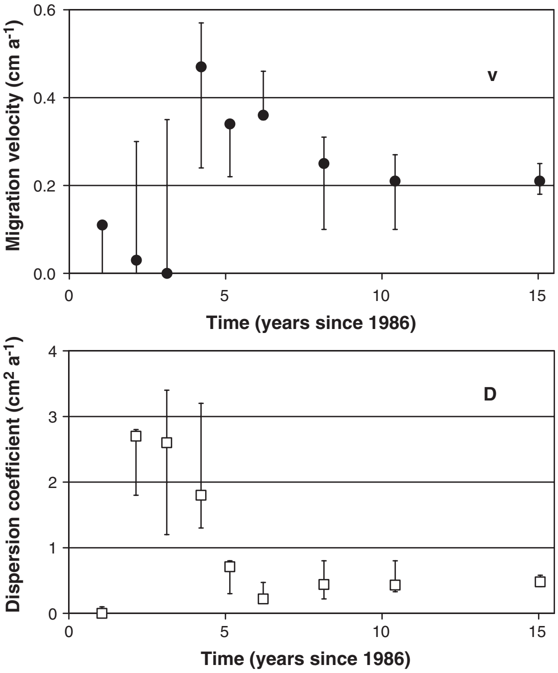

Fig. 2. Apparent vertical migration velocity v (above) and apparent dispersion coefficient D (below) of ^{134}Cs determined by the convection–dispersion model with respect to the first sampling date (13.6.1986) at the grassland site in Bavaria ('individual' evaluation). The uncertainty limits (bars) were obtained by allowing the simulated activity densities (=activities per unit area) to vary roughly within the range of the spatial variability of the observed activity densities. In 2001, the values refer to the evaluation of the mean activity–depth profile of five plots.

shown as a function of the soil depth. In this way, the integrated area between the curve and the axis of the soil depth represents 100% of the ^{134}Cs inventory and has to be the same for all graphs. The corresponding curves of ^{137}Cs (not shown) are rather similar to those of ^{134}Cs as outlined in Schimmack and Schultz (2006-this issue).

The activity–depth profiles of ^{134}Cs and ^{137}Cs were evaluated by the CDE and the RWPM. For a better comparison of the results at the various sampling dates, the evaluation was restricted to the soil layers between 0 and 30 cm. Due to the aim of this study (see 'Introduction'), the fits were optimized mainly with respect to the data of the soil layers down to 15 cm only. The profiles were evaluated by two approaches, (A) simulating each activity profile determined at a given sampling date at a given location of the grassland individually with respect to the start of the migration which was the same for all sampling dates ('individual' evaluation); and (B) using all measurements as one single data set ('single set' evaluation). For both approaches, the simulated profiles at the end of the integration period were obtained by using constant migration parameters for all sampling dates or different parameters optimized for each sampling date.

3.2. 'Individual' evaluation of the activity–depth profiles of ^{134}Cs and ^{137}Cs

At the study site, Chernobyl-derived radiocaesium was not sorbed completely within the first soil layer 0–2 cm at 30.4.1986, the main deposition date of the Chernobyl fallout in Bavaria. About 17% of the activity was transported into deeper soil layers (Schimmack et al., 1989). Therefore, the migration parameters of both caesium isotopes were determined in general with respect to the first sampling date (13.6.1986) because then the migration started with a known activity–depth profile as initial condition. In Table 1, the results for ^{134}Cs obtained by the CDE are shown in the left column, those obtained by the RWPM in the right column. Obviously, the parameter values agree fairly well. The value "zero" for D in Table 1 and the following tables does not mean that there is no diffusive transport at all because molecular diffusion is always present in soil. This diffusion,

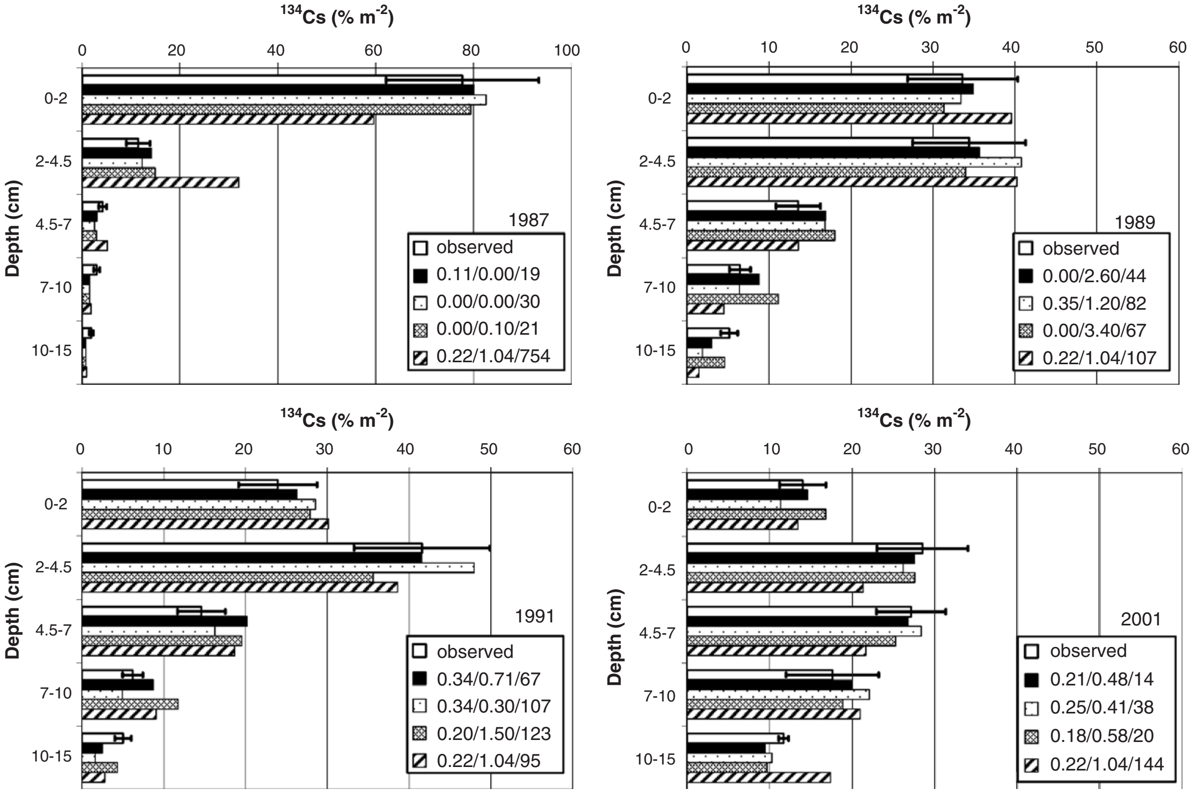

Fig. 3. Observed and simulated activity density (=activity per squaremeter) per soil layer of ^{134}Cs using the 'individual' evaluation for the first five soil layers in the sampling years 1987, 1989, 1991 and 2001 at the grassland site in Bavaria. The parameters for the simulated activity densities given in the legend are: v (cm a^{-1}) / D (cm^2 a^{-1}) / RSS ((% m^{-2})^2).

however, seems to be so small that it could not be detected by fits of the experimental values with minimal RSS values. Using the RWPM, it is necessary to repeat the calculations for a given set of parameters several times to get a reliable mean RSS. Thus, evaluations with this method were more time consuming than those with the CDE. Since the minimum RSS values were always greater than those of the CDE, the RWPM was not used furthermore. For the last sampling date (2001), the migration parameters in Table 1 refer to the mean activity distribution of the five plots. The values of the individual plots (Table 2) indicate a significant spatial variability (small scale) of the ^{134}Cs migration: the coefficient of variation (CV) was 23% for v and 30% for D. The value of D is clearly larger than the CV for the activity densities in the upper soil layers, which was about 20% (Schimmack and Schultz, 2006-this issue). The migration parameters of total ^{137}Cs obtained by the CDE are summarized in Tables 3 and 4. They were rather similar to those of ^{134}Cs, the spatial variability being also about the same.

In many migration studies published the migration parameters were not determined with respect to a reference date with a known activity–depth profile (e.g. first sampling date) but to the main or first deposition date assuming that the deposited activity was completely adsorbed to the surface soil during deposition. Therefore, the ^{134}Cs profiles were evaluated in addition with respect to the 30.4.1986 assuming that one day later the first soil layer 0–2 cm contained the totally deposited activity of ^{134}Cs. In this case, the values of D were considerably greater than those of Table 1, while v was rather similar with exception of 1987. In 1987, v of ^{134}Cs was 1.1 cm a^{-1} (D=0 cm^2 a^{-1}) instead of 0.11 cm a^{-1} (Table 1). For 2001, it should be noted that the parameter values in Tables 1 and 2 refer to ‘incomplete’ activity–depth profiles. Due to the short half-life of ^{134}Cs (2.06 years) its activity had disintegrated below the detection limit of the gamma detectors at 4 of 5 plots in the layer 15–20 cm and at all plots in the layers below 20 cm (Schimmack and Schultz, 2006-this issue). Rather likely the ^{134}Cs activity in these layers was not zero but only smaller than the detection limit. The missing activity can be estimated at each plot as described in Schimmack and Schultz (2006-this issue). Evaluating these ‘adjusted’ activity–depth profiles in 2001, only minor changes were found for the parameters. For the mean ‘adjusted’ profile, e.g., v was slightly reduced from 0.21 to 0.20 cm a^{-1} and D enhanced from 0.48 to 0.59 cm^2 a^{-1}. However, since these ‘adjusted’ profiles are speculative, they are not considered here furthermore.

For most years, the values in Table 1 were obtained by optimizing the fit of the experimental data. In 1988, 1991 and 1996, the optimized parameters were slightly changed in order to turn the Wald–Wolfowitz test to insignificance ($p>0.05$). In 1988, e.g., v=0.03 cm a^{-1} was used for this purpose instead of the optimized value v=0.00 cm a^{-1}. The Chi-square test was not significant for all years but 1992 due to the fifth layer 10–15 cm.

Table 5
‘Single set’ evaluation of the ^{134}Cs migration in seven soil layers between 0 and 30 cm depth at a grassland site in Bavaria using the CDE with v = apparent vertical migration velocity of ^{134}Cs, D = apparent dispersion coefficient of ^{134}Cs, and RSS = residual sum of squares

Sampling year	Calculated			Observed		
	v	D	RSS	v	D	RSS
	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2	cm a^{-1}	cm^2 a^{-1}	(% m^{-2})^2
1987	0.11	0.00	20	0.11	0.00	19
1988	0.00	4.9	12	0.00	4.4	4
1989	0.00	2.0	40	0.08	1.3	20
1991	0.25	0.00	62	0.31	0.00	20
1992	0.49	0.00	176	0.41	0.00	78
1994	0.00	0.09	30	0.00	0.35	23
1996	0.08	0.19	19	0.15	0.23	22
2001[a]	0.21	0.31	6	0.22	0.40	23
Range	0.00–0.49	0.00–4.9	6–176	0.00–0.41	0.00–4.4	4–78
Mean	0.14	0.94	46	0.16	0.84	26
SD[b]	0.17	1.74	55	0.15	1.50	22
CV[c] (%)	119	186	121	91	180	85
Median	0.10	0.14	25	0.13	0.29	21

The integration of the CDE was performed between successive sampling years starting with the optimized calculated (calculated) or the observed (observed) activity–depth profile of ^{134}Cs.

[a] The mean depth distribution of five plots was evaluated.

[b] Standard deviation.

[c] Coefficient of variation.

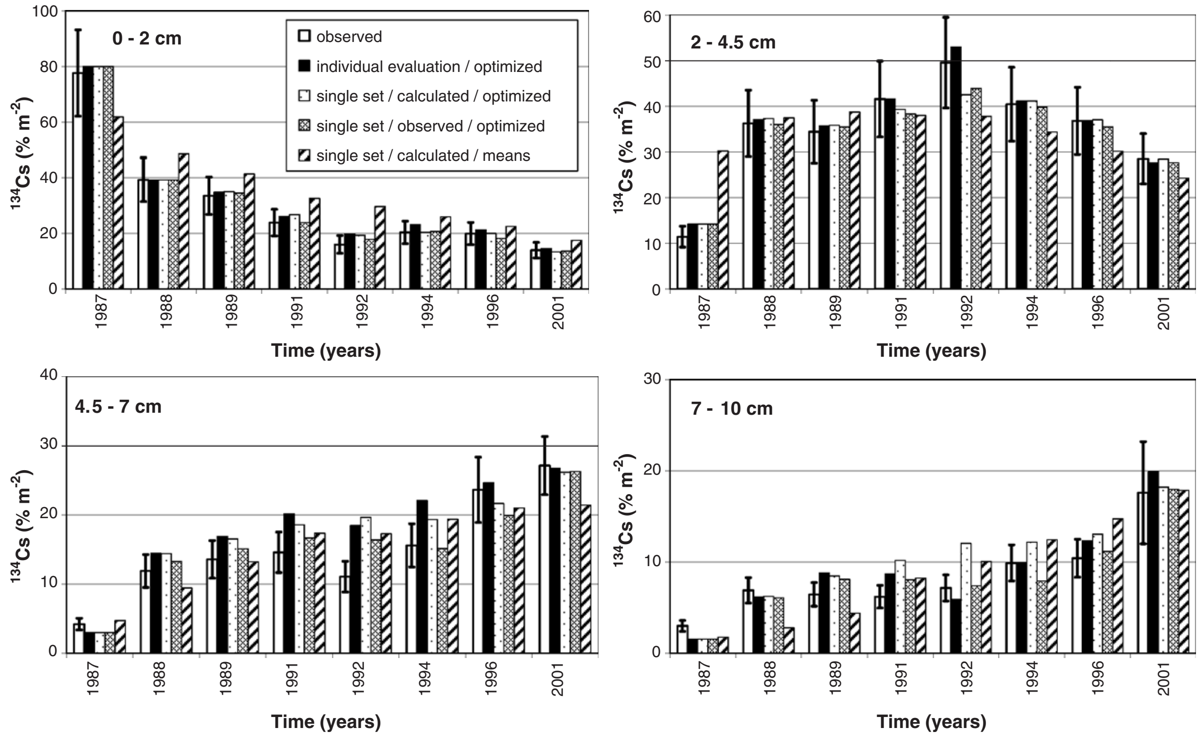

Fig. 4. Time course of the observed and simulated activity density (=activity per squaremeter) of ^{134}Cs between 1987 and 2001 for the soil layers 0–2, 2–4.5, 4.5–7 and 7–10 cm at the grassland site in Bavaria. The simulated activity densities were calculated using the 'individual' as well as the 'single set' evaluation. 'Optimized' = fitted by minimizing the residual sum of squares, 'calculated' = taking calculated initial depth profiles for the integration, 'observed' = taking observed initial depth profiles for the integration, 'means' = taking the mean migration parameters of Table 5, left column, for the simulation.

Because the unusually high activity in this layer could not be simulated by any combination of v and D, this was ignored for 1992. The values of v and D shown in Table 1 are not correlated (Pearson correlation: $r=-0.43$, Spearman rank correlation $R=-0.26$, both $p>0.05$, two sided). The uncertainties of the migration parameters of ^{134}Cs shown in Fig. 2 were obtained by changing the parameters as far as the simulated activity densities in all soil layers down to 15 cm were roughly within the limits set by the spatial variability (±20%, see above) not regarding the RSS. With a few exceptions, the Chi-square test and the Wald–Wolfowitz test were not significant for the "extreme" activity–depth profiles. However, using the mean migration parameters — $v=0.22$ cm a^{-1} and $D=1.04$ cm^2 a^{-1} (Table 1) — for the simulations (Fig. 3), the Chi-square test was significant ($p<0.05$) for most of the simulated profiles. Because in addition the simulated activity densities per soil layer in many cases exceeded the variability range of the measured values, the means of the migration parameters are not suitable for simulating all experimental profiles by constant values of the parameters. Several other combinations of constant values for v and D were also used, e.g. the medians, but the disagreement of the simulated and observed activity densities was even worse.

3.3. 'Single set' evaluation of the activity–depth profiles of ^{134}Cs

Evaluating all data as a 'single set', the CDE was integrated from the first sampling date, 13.6.1986, up to the last one in May 2001 using constant migration parameters for the total observation period or only for the integration periods between consecutive sampling dates. In the latter case, the observed activity–depth profiles at the end of the integration period were used for the optimization of the migration parameters as described above. As initial condition, the finally calculated activity–depth profile of the preceding integration (Table 5, left column 'calculated'), or the activity–depth profile observed at the start of the integration (Table 5, right column 'observed') were used. The Wald–Wolfowitz test was not significant for all years except 1991 for the evaluation using observed 'initial' profiles ($p<0.05$), and the Chi-square test was also not significant except 1992 for the evaluation using the calculated 'initial' profiles ($p<0.05$). As found for the 'individual' evaluation, the values of v and D (Table 5, both 'calculated' and 'observed') are not correlated ($r=-0.48$ and $r=-0.55$, respectively, $R=-0.70$ and $R=-0.67$, respectively, $p>0.05$, two sided).

In Table 5, the year 1990 was omitted. Considering this year, the migration parameters for 1990 (versus 1989) were rather high (v about 0.9 cm a^{-1}, D about 1.5 cm^2 a^{-1}) with low RSS values, those for 1991 (versus 1990) were zero with high RSS values for both parameters, using calculated as well as observed initial profiles. As a consequence, however, all "calculated" profiles from 1992 to 2001 exhibited much higher RSS values than given in Table 5, particularly in 1992 with RSS=344 (% $m^{-2})^2$ and $p<0.001$ for the Chi-square test, i.e. although optimized an extremely bad fit. The reason for these difficulties is that the sum of the activity densities of ^{134}Cs in the first two soil layers (0–4.5 cm) was 68.0% m^{-2} in 1989, 53.1% m^{-2} in 1990, 65.5% m^{-2} in 1991 and 65.6% m^{-2} in 1992. Therefore, simulating continuously the ^{134}Cs movement through the soil requires an upward flow of 12.4% activity per m^2 during one year from soil layers below 4.5 cm to the top soil. Upward flows of ^{134}Cs are possible in principle, e.g. by earthworms (Müller-Lemans and Van Dorp, 1996). For the migration period from 1990 to 1991, however, this would mean a net upward transport by earthworms of more than 25% of the activity below 4.5 cm. This is rather unlikely. It seems much more plausible that the difference between 1990 and 1991 was caused by the spatial variability of the ^{134}Cs activity. Therefore, the start of the integration period for the year 1991 was set to 1989 (Table 5).

In Fig. 4, the time course of the observed activity densities of ^{134}Cs in the soil layers between 0 and 10 cm depth is compared to that of the activities simulated by various methods. In the soil layers 0–2 and 2–4.5 cm, the optimized activities match the spatial variability of the observed activities throughout the observation period. In the layers 4.5–7 and 7–10 cm, larger deviations are present in the middle years 1989–1994. However, activities calculated by the mean migration parameters (Table 5) exceed the spatial variability in all soil layers for most of the years. The same holds for the medians and other combinations of constant values of v and D for the total observation period.

4. Discussion

The background of this study was the prediction of the long-term radiation dose to man caused by the presence of radiocaesium in the soil of grasslands. In most countries of Western Europe, this dose is dominated by the external radiation dose of radiocaesium depending essentially on its activity–depth distribution in the uppermost soil layers. For this reason, the migration parameters were determined by optimizing the fit of the data in the soil layers between 0 and 15 cm. In the soil layers below 15 cm depth the observed activity densities were only about 1–3% m^{-2}, but the simulated values were even much smaller and always <1% m^{-2}. This well known 'tailing' effect (see e.g. Kirchner, 1998; Bunzl, 2001) was not subject of the present investigation. The results presented reveal that the migration of total ^{137}Cs is dominated by the migration of CH-Cs as expected from the activity–depth profiles in the first part of this study (Schimmack and Schultz, 2006-this issue). The migration parameters of ^{137}Cs can be used as fair estimates for the migration of ^{134}Cs and CH-Cs, respectively, if total ^{137}Cs is dominated by CH-Cs as found at the study site. Moreover, the infiltration of activity into deeper soil layers during or immediately after the deposition event as observed at the study site (Schimmack et al., 1989) has to be considered for the determination of the migration parameters particularly during the first years after the deposition.

The main result of this study is, however, that the activity–depth profiles of radiocaesium determined for various locations at different times within 30 m^2 of a grassland site cannot be fitted by constant migration parameters. This result is independent of the evaluation method as well as of the transport model used, CDE or RWPM. Both models are based on the same physical processes, i.e. diffusion/dispersion and convection of the solute in the percolating soil water, but the determination of the migration parameters in the RWPM is completely different to the integration of the CDE. Because the results for v and D are rather similar in both methods uncertainties of the CDE integration due to the numerical dispersion and the discretisation of the soil depth into simulation layers of 1 cm thickness were not relevant.

The ten activity–depth profiles of radiocaesium were evaluated by two approaches: (A) determining individual migration parameters for each sampling location, and (B) evaluating the profiles as a single data set, i.e. considering each profile between 1986 and 2001 as a 'snap-shot' of the development of the radiocaesium activity as a function of soil depth and time. The results of both approaches clearly indicate that parameters determined by the classical CDE or an equivalent random walk model during the first years after the radiocaesium deposition are not useful for predictive purposes because they depend strongly on the location and the date of sampling, respectively. To obtain these results, the simulation uncertainties were quantified by means of the spatial variability of the activity densities determined in 2001. It should be noted that the experimental uncertainties of the activity densities were clearly smaller than the spatial variability. Considering only these experimental uncertainties at the location of sampling would result in 'experimental' uncertainties of the migration parameters smaller than shown in Fig. 2. Using the spatial variability is more adequate for estimating the 'variability' uncertainty of the radiocaesium migration within the sampling area of the grassland. However, in spite of the large range of the 'variability' uncertainties, the migration parameters were still significantly different. The dependence on the sampling location and the sampling year, respectively, seems to disappear about 6 years after the deposition of radiocaesium.

The situation is still more complicated for the 'single set' approach due to the interdependence of the results. Using the 'individual' approach the results for the migration parameters at the various sampling dates are independent of each other. Using the 'single set' approach with calculated initial depth profiles the parameters depend on each other if not set constant. The 'simulation pathway' through the time–depth space is increasingly different even for small differences during the first sampling years after 1986. This can easily be seen by the results mentioned above when the activity–depth profile of 1990 was included into the data set. The values of v and D shown in Table 5 represent only one particular 'simulation pathway', i.e. the optimized one. There are many other pathways, but changing any value in the left column of Table 5 has consequences for the other values. However, for all tested pathways the variation between the results for the various sampling dates was more pronounced than for the optimized one. If in addition the spatial variability is considered, and in this case also when using observed initial depth profiles, not only the finally simulated activity–depth profile is affected but also the profile at the beginning of the integration period. This situation is rather complex. Therefore, no attempt was made to calculate simulation uncertainties for the 'single set' migration parameters.

As yet, the different values of v and D cannot be explained unambiguously. Neither v nor D can be approximated by monotonous functions of time. For D, particularly the year 1987 drops out. This is surprising for two reasons. Firstly, in 1986/87 about 80% of the total activity of ^{134}Cs was present in the first soil layer 0–2 cm. This layer is characterized by a higher organic matter content (12%) and a smaller dry bulk density (0.7 g cm^{-3}) than in the deeper soil layer (see Table 1 in part I), thus favouring a higher dispersive and convective flux. Nevertheless, this soil layer showed a negligibly small D value even when considering the spatial variability. Secondly, the influence of the above mentioned 'ageing' or 'fixation' processes of freshly deposited radiocaesium in the soil on its migration behaviour should be most pronounced during the first year after 1986. Just during this year the activity–depth profile changed much less than later on. The variation of the hydrological conditions in the grassland soil is not likely to play a major role because the annual precipitation at the study site varied only moderately between 676 mm (1989) and 1029 (1993) up to 1995 (Schimmack and Schultz, 2006-this issue). The 'dispersion scale effect' mentioned in the Introduction would result in increasing values of D with increasing soil depth and migration time, respectively. This was not observed at the study site. Using both evaluation approaches (Tables 1 and 5), the maximum of D was in 1988.

There is, however, another result that has to be considered. In both evaluation approaches, the parameters v and D were not correlated. Such a correlation would have been expected if the dispersive flow was only caused by hydrodynamic dispersion. Obviously, other processes contributed to the total dispersion in addition. As outlined in the Introduction, physical mixing of the soil, e.g. by earthworms, can be treated mathematically like a dispersive flow. At the study site, earthworms were present in most soil layers throughout the observation period, particularly in the layers down to 20 cm. Unfortunately, the dependence of the earthworms' abundance and biomass on time as well as on the sampling locations within the sampling area were not determined. The values of D from 1986 to 2001 would be plausible if the abundance of the earthworms (i) was almost zero at the sampling location in 1987 but high at the other locations after 1987 and (ii) decreased with soil depth within the layers down to 10 cm. This would lead to a decreasing contribution of bioturbation to the total dispersion as radiocaesium moved down the profile and, thus, to an apparent time dependence of D. Moreover, this decrease would explain the decrease of the 'velocity' of the 50%-activity depth by a factor of about 3 from 1988 to 2001 (Schimmack and Schultz, 2006-this issue). Results of other authors using evaluation methods with only one migration parameter not related to transport mechanisms (e.g. Bunzl et al., 1995; Rosén et al., 1999; Isaksson et al., 2001) also indicate an 'overall' retardation of the radiocaesium migration during the first decade after 1986. It is not clear, however, whether the above assumptions on the earthworm abundance have been valid at the study site during this time period. Apart from that, the migration of radiocaesium was clearly dominated by dispersion only in 1988 and 1989 but not later on.

5. Conclusions

The radiocaesium migration in the soil of the undisturbed grassland investigated was characterized by significantly different migration parameters v and D for six locations sampled during the time period 1986–1992. Later on, the values were rather similar when using the 'individual' evaluation approach. A time dependence of the radiocaesium migration at the study site may be possible but cannot be proven. The 'dispersion scale effect' as well as 'ageing effects' are not likely to be responsible for the observed differences of v and D. Physical mixing by earthworms was observed at the study site and may have caused an apparent time dependence at least of D, but was not investigated in detail. However, the aim of this study was not to elucidate the causes for the diversity of the results but to examine the applicability of the classical CDE for obtaining migration parameters for predictive purposes. The main conclusion to be drawn from the results is that migration parameters determined by classical transport models based on convection and dispersion during the first years after a nuclear accident are only useful for calculating activity–depth profiles of radiocaesium in the future if at the same time further investigations as e.g. on the presence and depth dependence of bioturbation at the study site are performed. Otherwise, the dependence of the migration parameters on the sampling date makes it difficult, to give any recommendation for predicting *realistically* the long-term radiation exposure to man due to the presence of radiocaesium in the soil, particularly for the first years after an accident although this is the time predictions are needed urgently by regulating authorities. Whether other transport models as discussed e.g. by Kirchner (1998), Zhou (2002) and van Dam et al. (2004) are more appropriate for this purpose has to be investigated.

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