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Spatial variability of the vertical migration of fallout ^{137}Cs in the soil of a pasture, and consequences for long-term predictions

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Abstract Various transport models are presently used to predict the long-term migration behaviour of fallout radiocesium on the soil. To examine to what extent the uncertainty of these predictions is influenced by the spatial variability of the migration rates, we determined the depth profiles of Chernobyl-derived ^{137}Cs at 100 plots in a 100 m×100 m pasture. These data were used to obtain the frequency distributions of the characteristic transport parameters of three widely used transport models (e.g. dispersion-convection model, residence time model, and back-flow model). The results show that these transport parameters are generally log-normally distributed with a coefficient of variation of about 80%. Finally, each transport model was employed to predict the resulting frequency distribution of the ^{137}Cs inventory in the main root layer (0–7 cm) of the pasture, 20, 50, and 100 years after the deposition. If only the spatial variability of the transport parameters is taken into account, this analysis revealed that the dispersion-convection model and the back-flow model always predicted rather similar, but significantly higher median inventories than those obtained with the residence time model. If, in addition, the spatial variability of the amount of ^{137}Cs deposited is also taken into account, the frequency distributions of the ^{137}Cs inventories in the root layer become so wide that differences in the median inventories predicted by the three models become statistically significant only after 100 years. Several statistically significant correlations between the transport parameters of the three models were also detected.

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Introduction

The long-term transfer of fallout ^{137}Cs to man via terrestrial pathways (e.g. plant uptake, resuspension) depends considerably on the residence time of this radionuclide in the root zone of agricultural and grassland sites. For this reason, numerous studies were carried out to quantify the vertical migration rate of radiocesium in the upper layers of different soil types (for a recent review see, for example, [1]). Usually, migration rates (or residence half-times) of artificial radionuclides are obtained by evaluating the depth profiles observed in field, lysimeter, or column studies after given time periods following the deposition of the radionuclide with a suitable transport model. However, due to the spatial heterogeneity of the soil properties, one has to realize that the migration rates of fallout radionuclides may vary considerably in horizontal directions, even on a small scale. To predict, therefore, realistically, for example, the ^{137}Cs inventory in the root zone of a given pasture several decades after deposition, quantitative information on the spatial variability of the migration rates at this site is required.

Migration rates (or mean residence times) of a radionuclide in a given soil layer can usually not be measured directly in the field, but rather are evaluated from observed depth profiles after preselected time periods by using a transport model. However, because the underlying assumptions and the parameters of each transport model differ, the value of the migration rate thus obtained depends to some extent on the transport model used. Consequently, the resulting shape of the depth profile of the radionuclide in the soil predicted for a period of 50 years, for example, after deposition will also depend on the model selected. At present, however, a study on this subject, which also involves the effect of spatial variability of the model parameters, does not seem to be available. This is not too surprising, because for each site it involves the assessment of the vertical distribution of fallout ^{137}Cs in the soil at a large number of sampling plots, and its subsequent evaluation with various transport models.

The purpose of the present investigation thus was:

- to assess the vertical distribution of Chernobyl-derived ^{137}Cs in the root zone of the soil at 100 plots of a pasture (100×100 m)
- to evaluate these depth profiles employing three transport models to obtain the frequency distribution of the corresponding model parameters
- to use the results obtained for each transport model to predict the frequency distribution of the ^{137}Cs inventory in the root zone of this pasture 20, 50, and 100 years after deposition

For this purpose, the effect of the spatial variability of the transport parameters and that of the amount of ^{137}Cs deposited at each sampling plot on the uncertainty of the inventory prediction was evaluated separately.

The transport models considered here were:

1. the well-known convection-dispersion model, where the transport of the radionuclide is characterized by the two parameters D_s (dispersion coefficient) and v_s (solute velocity)
2. the widely used simple compartment model, where the soil layers are considered as a series of compartments, and the transport of the radionuclide from one compartment to the next one is characterized by the corresponding first order rate constant (or residence half-time)
3. a back-flow model, where the transport of the radionuclide in the root zone is described by one downward flow rate and one upward flow rate

Presently, it is not possible to decide which of the above transport models is superior with respect to long-term predictions. A comparison of the root zone ^{137}Cs inventory predictions, as presented here for one type of soil will, however, at least reveal whether the differences in the long-term predictions are statistically significant or whether they are identical within the uncertainties already resulting from the spatial variability of the transport parameters. In addition, we also evaluated the combined effect of the spatial variability of the transport parameters and of the observed ^{137}Cs deposition on the uncertainty of the inventory prediction.

Finally, to test the presence of a spatial dependence (or autocorrelation) between the transport parameters from adjacent sampling plots, the corresponding semi-variograms were calculated.

Materials and methods

Site

The upland pasture site investigated was located about 180 km southeast of Munich in the Berchtesgaden National Park between the mountains Jenner (1800 m), Watzmann (2700 m) and Hagengebirge (2200 m). The area belongs to the Kalkalpen (Jurassic limestone), and the slopes are built up by micaceous and marl limestones. The mean precipitation rate is about 1600 mm per year and approximately 50% of the annual precipitation is snow. The exact location of the pasture is 13° 00’ 47’’ E; 47° 33’ 14’’ N, the altitude about 1500 m. The orientation of the slope is westward, with an inclination of about 15°. The site has been utilized for many years as a pasture for grazing cows from the first weekend in July to the last weekend in August.

Soil types

The soil can be classified as a distric cambisol with an organic matter content of 19%, clay content of 33%, sand 19%; silt 48%, cation exchange capacity 116 $mmol_c\ kg^{-1}$, carbon to nitrogen ratio 9.4, and a pH of 4.5.

From a recent determination of the vertical distribution of ^{137}Cs and ^{134}Cs in the soils at various plots along a transect of this pasture, it was found that Chernobyl-derived radiocesium is still very close to the soil surface and is practically not found below a depth of 10 cm (<2%) [2]. In the present study, where we were interested only in the migration rates of Chernobyl-derived radiocesium, we had, however, to distinguish between Chernobyl-derived ^{137}Cs and ^{137}Cs from the fallout of global weapon testing, because radiocesium from this source is also still present mainly in this layer. For this reason, ^{134}Cs was also determined in all soil samples. The fraction of Chernobyl-derived ^{137}Cs from the ^{137}Cs and ^{134}Cs measured in the samples can then be obtained for each soil sample by using the ratio of $^{137}Cs/^{134}Cs=1.75$, as observed on 30 April 1986 in the deposition in Bavaria/Germany [3].

Vegetation

As is typical for pasture sites, a large variety of different herbage plants are growing. The plant association was classified as *Alchemillo-Cynosuretum* [4], the main species being *Alchemilla vulgaris, Cynosurus cristatus, Trifolium repens, Phleum alpinum, Poa alpina*, and *Crepis aurea*.

Even though the main part of the plant roots (>70%) was found in the upper 5 cm of the soil, the thickness of this root zone was found to vary somewhat from plot to plot due to the presence of different plant communities. For the same reason, the differentiation between the root zone and the deeper soil layers depends to some extent on the plant community present at a given plot.

Sampling and sample preparation

For soil sampling, a 100×100 m square with grid lines at 10-m-intervals was laid out. Within each of the resulting 10×10 m squares a 1×1 m plot was selected at random (using a random number generator) and marked. In this way a total of 100 plots were selected within the 100×100 m area in 1997. This type of sampling (random sampling within blocks) has the advantage that each point in the area has a chance to be selected, but that, nevertheless, an uneven coverage of the area is avoided [5, 6]. Soil samples were collected with a corer (7.5 cm in diameter and 10 cm long) from the upper 10 cm of the A_h horizon at each plot where the main part (>80%) of the root mass was present. The cores were subsequently divided in layers of 0–3.5 cm, 3.5–7 cm, and 7–10 cm. All soil samples were air dried, ground to pass through a 2 -mm-sieve, and homogenized.

Measurements

The elements ^{137}Cs and ^{134}Cs were determined by direct gamma-spectrometry using high purity Ge-detectors and a multichannel analyzer. Detectors were calibrated with standards made up to the same geometry as the samples (100-ml-cylinder), in the case of ^{134}Cs, losses by sum coincidences during counting were taken into account. Counting times were adjusted to provide 1-σ errors of less than about 3% for ^{137}Cs and 10% for ^{134}Cs. All measured activities were corrected for radioactive decay to the reference date 1 June 1996.

Transport models

The following transport models were used to obtain values for the model parameters by fitting the observed depth profiles of ^{137}Cs in the soil and to predict the inventory of ^{137}Cs in the main root layer 0–7 cm of the pasture 20, 50, and 100 years after the deposition.

Convection-dispersion model

In this one-dimensional convective-dispersive, local equilibrium, mass transport model the total concentration $C^t(x, t)$ of the radionuclide in the soil (mobile and sorbed) at time t and distance x with respect to the concentration C_o^t of the radionuclide deposited as a single pulse at time t=0 to the soil surface (x=0) is given [7, 8, 9] as:

$$C^t(x,t)/C_0^t \exp[-\lambda t]$$
$$= \frac{1}{\sqrt{\pi D_s t}} \exp\left[-\frac{(x-v_s t)^2}{4D_s t}\right] - \frac{v_s}{2D_s} \exp\left[\frac{v_s}{D_s} x\right] \operatorname{erfc}\left[\frac{x+v_s t}{2\sqrt{D_s t}}\right] \quad (1)$$

Here λ is the decay constant of ^{137}Cs, erfc(ξ)=1–*erf*(ξ), where erf(ξ) is the error function,

$$D_s = \frac{D}{1+\frac{\rho}{\varepsilon}K_d} \quad \text{and} \quad v_s = \frac{v_w}{1+\frac{\rho}{\varepsilon}K_d} \quad (2)$$

where K_d is the distribution coefficient of ^{137}Cs in the soil, ρ the bulk density and ε the porosity of the soil. D and v_w are the dispersion coefficient and the mean pore water velocity, respectively.

Equation (1) will be used in the following to obtain for each observed depth profile of ^{137}Cs in the soil the values of D_s and v_s, using a non-linear least-squares fitting algorithm (Levenburg-Marquardt; Table Curve 2D). Even though the values for *Kd* cannot be obtained with this procedure, the knowledge of D_s and v_s is sufficient to predict with Eq. (1) the depth profile of total ^{137}Cs in the soil at any time t.

Compartment model

In a compartment model the soil is divided in N layers of arbitrary thickness, usually 1–2 cm (see, for example, [9, 10, 11, 12, 13, 14, 15]). It is assumed that the transport of the radionuclide from layer (i) into the next layer (i+1) is proportional to the amount of activity in the layer i, but independent of the activity in the layer i+1. Within each layer no concentration gradient of the radionuclide shall exist. The transfer of activity A_i (Bq m^{-2}) of a radionuclide in the compartment i in a small time interval Δt (day) is thus given for the first layer as

$$dA_1/dt = RACD - [\lambda + k_{1,2}]\, A_1(t) \quad (3)$$

and for the layers i=2,....., N as

$$dA_i/dt = -[\lambda + k_{i,i+1}]\, A_i(t) + k_{i-1,i}\, A_{i-1}(t) \quad (4)$$

where $k_{i,j}$ (day^{-1}) is the fractional rate of transfer from compartment i to compartment j and λ is the decay constant of the radionuclide. *RACD* is the rate of activity deposited per unit area (in Bq cm^{-2} day^{-1}), which must be known as a function of time. In the present case, a single input event at 30 April 1986 (Chernobyl fallout) was used, corresponding to the total Chernobyl-derived ^{137}Cs inventory at each plot of the pasture. To obtain the values of k_i for a given radionuclide in a given soil layer, the differential equation system of Eqs. (3–4) is integrated numerically. The values for k_i are varied until the compartment activity matches the observed activity in this compartment. The residence half-time of a radionuclide in layer i is finally obtained as τ_i=0.693/k_i. (For details, see [16]). All residence half-times evaluated here do not include the radioactive decay of the radionuclide (i.e., they are ecological residence half-times). Equations (3–4) will be used in the following to obtain for each observed depth profile of ^{137}Cs in the soil the values of τ_i .The regression method used for fitting an observed depth profile to Eqs. (3–4) is the Marquardt method, as provided by the software package ModelMaker. Once the residence half-times are evaluated for a given sampling plot, the compartment model can be used to predict there with Eqs. (3–4) the depth profile of total ^{137}Cs in the soil at any time t.

Back-flow model

The simple compartment model described above by Eqs. (3) and (4) is applicable only when the water and radionuclide transport are dominated by convective processes [17]. In the present study, however, we observed that at many plots the radiocesium concentration in the soil declined even 10 years after deposition monotonically, indicating that convective transport is small as compared to diffusion. In this case it is more appropriate to use a linear compartment model, where the soil is divided into N layers and both downward and upward transfer between adjacent soil layers are included. The system of differential equations is then given for the first layer as (see [17, 18]:

$$dA_1/dt = RACD - [\lambda + k_{1,2}]\, A_1(t) + k_{2,1}\, A_2(t) \quad (5)$$

for the next layers:

$$dA_i/dt = -[\lambda + k_{i,i+1} + k_{i,i-1}]\, A_i(t) + k_{i-1,i}\, A_{i-1}(t) + k_{i+1,i}\, A_{i+1}(t) \quad (6)$$

where i=2,......, N–1 and for the last layer:

$$dA_N/dt = -[\lambda + k_{N,N+1} + k_{N,N-1}]\, A_N(t) + k_{N-1,N}\, A_{N-1}(t) \quad (7)$$

Thus, downward transport can be due to diffusive and convective processes, whereas the upward transport is due to diffusion only. In the present case, where only three layers were available for analysis, all $k_{i,i+1}$ were assumed to be equal and will be denoted in the following as the *downward transfer rate*. Analogously all $k_{i+1,i}$ were also assumed to be equal and are denoted as the *upward transfer rate*.

Application of Eqs. (5–7) does not imply that the transfer by diffusion has to be physical diffusion only. It is well known that diffusional transport of radionuclides in the soil, which is due to biological processes (e.g., mixing by soil organisms, transport by plant roots) can be also treated as a diffusive transport [19].

Equations (5–7) will be used in the following to obtain for each observed depth profile of ^{137}Cs in the soil the values of the downward and upward transfer rates between the compartments. The regression method used for fitting an observed depth profile to Eqs. (5–7) is the Marquardt method, as provided by the software package ModelMaker. Once the downward and upward transfer rates between the compartments are evaluated for a given sampling plot, the compartment model can be used to predict with Eqs. (5–7) the depth profile of ^{137}Cs in the soil at any time t.

Results and discussion

Evaluation of the model parameters and their variability

The profiles of Chernobyl-derived ^{137}Cs at the 100 plots (three soil layers each, not shown separately here) exhibited in general a continually decreasing trend with increasing depth. Only in some cases was the ^{137}Cs activity in the second layer greater than in the first layer. At two plots the ^{137}Cs activity in the deepest (third) layer was, however, so small that it was below our detection limit

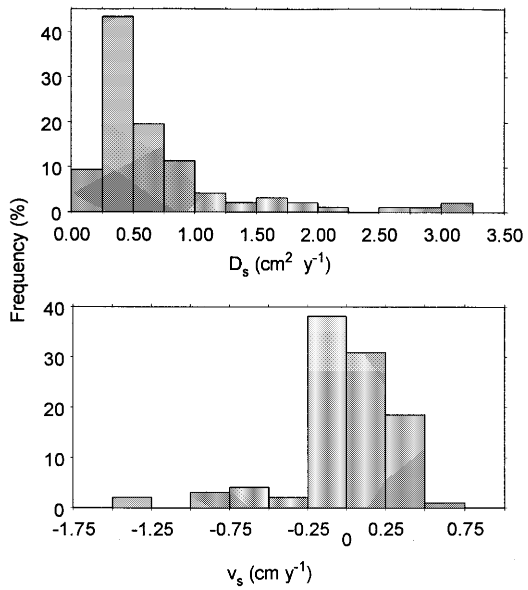

Fig. 1 Frequency distribution of the two parameters D_s and v_s of the dispersion-convection model, as resulting from evaluating 97 depth profiles of Chernobyl-derived ^{137}Cs in the soil of a 100×100 m field

(1 Bq/kg). Thus, a total of 98 ^{137}Cs depth profiles was available for evaluation with the above three models.

Convection-dispersion model

If the convection-dispersion model is used to describe the depth profiles of Chernobyl-derived ^{137}Cs as observed 11 years after deposition at the various plots, a good fit is generally obtained. The coefficient of determination r^2 was usually >0.99, and a satisfactory fit of the experimental data was not possible at only one plot. The resulting values for D_s and v_s are shown as histograms in Fig. 1. As evident from this graph, both of these model parameters varied within a wide range: for D_s a minimum value of 0.17 cm^2 year^{-1} and a maximum of 3.1 cm^2 year^{-1} (median 0.48 cm^2 year^{-1}) is obtained. Application of the Kolmogorov-Smirnov goodness-of-fit test shows that a log-normal frequency distribution of D_s cannot be rejected at the 0.05 level. The corresponding coefficient of variation, calculated according to Gilbert [5] for a log-normal frequency distribution is 0.77.

About 50% of the ^{137}Cs depth profiles could be fitted only if negative values for the parameter v_s were admitted (see Fig. 1, bottom). These depth profiles were characterized by a steep drop of the ^{137}Cs concentration from the first to the second layer and rather similar concentrations in the second and third layer. It is interesting to note that for these depth profiles the back-flow model also suggests that the upward transfer rate of radiocesium exceeds the downward transfer rate (see below). The frequency distribution of all data follows neither a normal nor a log-normal distribution. The minimum value is –1.42 cm year^{-1}, the maximum 0.52 cm year^{-1}. The median is 0.001 cm year^{-1}. Because neither a normal nor a log-normal is adequate to characterize the data, the width of the distribution is not quantified by a coefficient of variation, but rather by the lower and upper quartiles: the corresponding values were found to be –0.083 and 0.21, respectively. Negative values of v_s at a given plot do, however, not necessarily imply that also an upward water flux v_w is occurring there on average. It only indicates that an upward convectional flux v_s of ^{137}Cs is occurring. The underlying process of this upward component (perhaps bioturbation) can, however, not be evaluated here.

In order to examine whether the values obtained for v_s and D_s are independent of each other or to some extent associated, the Spearman correlation coefficient for the 49 data pairs where v_s is positive (downward convectional transport) was calculated. A significant *negative* correlation (P=0.0015) between these two parameters is apparent. In contrast, if we calculate this correlation coefficient for the 48 data pairs where v_s is negative (upward convectional transport), a significant *positive* correlation (P<0.0001) between v_s and D_s is evident. Thus, because at the study site all ^{137}Cs was after 10 years still present essentially only in the 0–1-m layer, increasing downward ^{137}Cs fluxes have to be counterbalanced by smaller diffusional components. If an upward flux is needed for the best fit of the depth profile, increasing absolute values of v_s will require larger values of the D_s.

Finally, it is interesting to test whether values of v_s (or D_s) from positions near one another in the field tend to be more similar than others. For this purpose the corresponding semi-variograms were calculated. Because D_s was log-normally distributed (see above), these values were transformed correspondingly before calculating the semi-variogram. These variograms (not shown separately) do, however, not suggest the presence of a smaller variance of v_s (or (D_s) at smaller separation distances (lags) as compared to the overall sample variance (i.e., flat shape of the variogram; all the variation appears to be nugget). Anisotropic effects of the variograms were not observable either. Obviously, any spatial dependence of the transport parameters occurs on this pasture only over distances smaller than the shortest sampling interval (here about 10 m).

Compartment model

To fit the depth profiles of Chernobyl-derived ^{137}Cs as observed 11 years after deposition at the various plots, the corresponding mean residence half-times in the first (0–3.5 cm) and second soil layer (3.5–7 cm) were adjusted until the calculated depth profile agreed with the observed one. The frequency distributions of the resulting residence half-times are shown in Fig. 2. As evident from this graph, both of these model parameters vary within a wide range: For the layer 0–3.5 cm a minimum value 4.5 years and a maximum of 154 years (median 17 years) is obtained for the mean residence half-time. Application of the Kolmogorov-Smirnov goodness-of-fit test showed

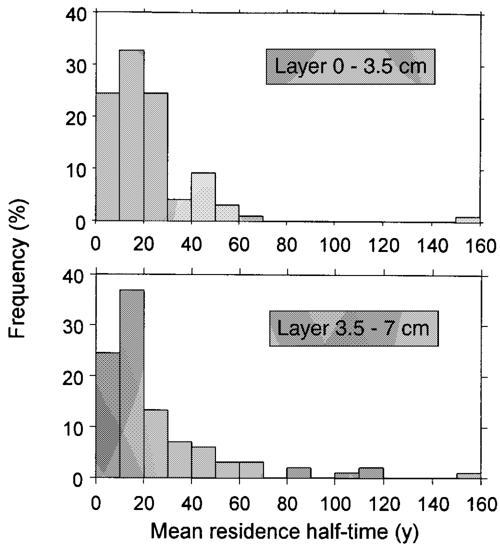

Fig. 2 Frequency distribution of the two parameters mean residence times of radiocesium in the layer 0–3.5 cm and 3.5–7 cm of the compartment model, as resulting from evaluating 98 depth profiles of Chernobyl-derived ^{137}Cs in the soil of 100×100 m field. Residence times do not include radioactive decay (ecological residence half-times)

that these data follow a log-normal distribution at the 0.05 level. The corresponding coefficient of variation is 0.78. Very similar values were found for the residence half-time of ^{137}Cs in the layer 3.5–7 cm (Fig. 2, bottom): the minimum value is 4.8 years, the maximum value 157 years, and the median 17 years. Again a log-normal distribution is adequate to characterize the distribution. The corresponding coefficient of variation is 0.92 and thus somewhat higher than for the first layer.

To test whether any association between the residence half-times in these two layers at the various plots is detectable, we calculated first the Spearman correlation coefficient ρ_{Sp} for all 98 data pairs. In this case ρ_{Sp}=–0.1095 and is thus not significant (*P*-level: 0.2829). However, if for this calculation we use only the data pairs from those plots where positive values for v_s (downward convectional flux of ^{137}Cs) were obtained (see above), a highly significant correlation is apparent (ρ_{Sp}=0.468, n=49, P<0.001). This signifies that at these plots elevated values of the residence half-times of ^{137}Cs in the first layer are generally also associated with elevated residence half-times of ^{137}Cs in the second layer. From this we may conclude that if the transport of ^{137}Cs is strongly influenced by a downward convectional component, the corresponding residence half-times in the first two layers will be correlated to some extent. On the other hand, if we calculate the Spearman correlation coefficient for the residence half-times of ^{137}Cs in the upper two layers at those plots, where v_s is negative (upward convectional flow of ^{137}Cs), we did not observe any correlation (ρ_{Sp}=0.177, n=45, P=0.222).

Another interesting relationship becomes evident when we examine for each plot the sign of the difference between the ^{137}Cs-residence half-time of the first and second layer. If we apply for this purpose the sign-test (or the Wilcoxon matched pair test), we find for those plots where v_s is positive (downward convectional flux of ^{137}Cs) that in 94% of all cases the ^{137}Cs-residence half-time in the second layer is larger than in the first layer (*P*-level of the sign-test and of the Wilcoxon matched pair-test <0.0001). In contrast to that, we observe for the plots where v_s is negative (upward convectional flux of ^{137}Cs) that in the vast majority (>90%) of the cases the ^{137}Cs-residence half-time in the second layer is smaller than in the first layer (*P*-level of the sign-test and of the Wilcoxon matched pair-test <0.0001). This may be explained by considering that an upward convectional flux retards the diffusional ^{137}Cs transport from the first to the second layer. As a result, ^{137}Cs will remain for a comparatively longer time period in the first layer than in the second layer, because a loss of activity from the first layer to a further upper layer is not possible.

Similarly, the semi-variograms of the ^{137}Cs residence half-times revealed that they were spatially independent.

Back-flow model

To fit the depth profiles of Chernobyl-derived ^{137}Cs as observed 11 years after deposition at the various plots, the corresponding downward and upward transfer rates in the soil layer 0–7 cm were adjusted until the calculated depth profile agreed within experimental error with the observed one. However, only 79 depth profiles could be fitted adequately, i.e., within the experimental error of the data. The values for χ^2, calculated to characterize the goodness of the fit, were in these cases between 0.002 and 173 (mean 31). The frequency distributions of the resulting downward and upward transfer rates are shown in Fig. 3. As evident from this graph, both of these model parameters vary within a wide range: for the downward transfer rate a minimum value of 0.017 year^{-1} and a maximum value of 0.17 year^{-1} (median 0.059 year^{-1}) were obtained. Application of the Kolmogorov-Smirnov goodness-of-fit test showed that these data follow a log-normal distribution at the 0.05 level. The corresponding coefficient of variation is 0.46. For the upwards transfer rate (Fig. 2, bottom) the minimum value is 0.00 year^{-1}, the maximum value 3.6 year^{-1}, and the median 0.063 year^{-1}. Again, the distribution of the data is log-normal. The corresponding coefficient of variation is 2.25 and thus considerably higher than for the downward transfer rate.

In order to examine for a possible relationship between the back-flow model and the convection-dispersion model, we plotted the downward transfer rate versus the convectional flow v_s obtained from the convection-dispersion model. A very strong positive correlation was found if we selected only those plots where v_s is positive, i.e., downward convectional component prevailing (Spearman correlation coefficient=0.7449, n=31, P<0.0001). In addition,

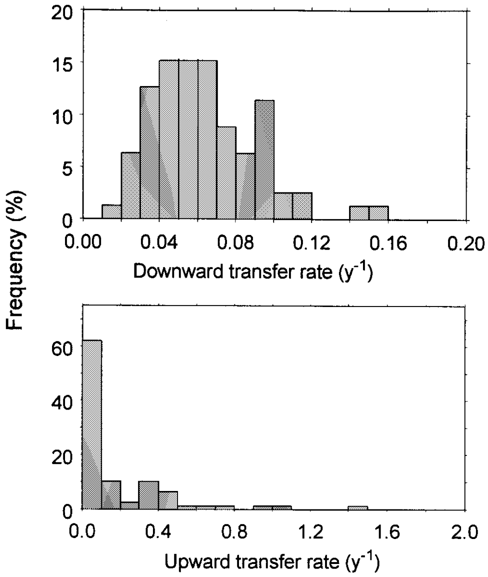

Fig. 3 Frequency distribution of the two parameters downward transfer rate and upward transfer rate of the back flow model, as resulting from evaluating 79 depth profiles of Chernobyl-derived ^{137}Cs in the soil of 100×100 m field

one observes that at these plots the downward transfer rate is in 97% of the cases larger than the upward transfer rate (*P*-level of sign-test <0.0001). Conversely, at those plots where v_s obtained form the convection-dispersion model is negative (upward convectional component), one finds that in 84% of the cases the upward transfer rate is larger than the downward transfer rate (sign-test, n=49, P<0.0001). From this we may conclude that if application of the convection-dispersion model suggests that the transport of ^{137}Cs is strongly influenced by a downward convectional component, the back-flow model will indicate this as well.

After logarithmic transformation, the semi-variograms were calculated also for the downward and upward transfer rates. A spatial dependence of the above transport parameters was, however, not detectable.

Long-term predictions by the three models

Transport models are frequently developed to also estimate the long-term migration behavior of radionuclides in the soil. In this connection, information on, for example, the radiocesium inventory in the root layer of the soil after a given time period is of interest. In principle, all three models considered here permit such a long-term prediction, provided that the corresponding transport parameters are independent of time. Within the first few years after a deposition event, this assumption is hardly valid for fallout ^{137}Cs (see e.g. [8, 20]). In the present study, however, where we are interested mainly in the effect of the spatial variability of the transport parameters on the uncertainty of a prediction, we will disregard this source of uncertainty. The uncertainties considered here for each model will thus be due to (1) the spatial variability of the model parameters and (2) the spatial variability of the amount of radiocesium deposited at the study site. In the present case, information on these two sources is available separately. For this reason we will estimate with each of the above transport models:

a) For each of the 100 plots the percentage ^{137}Cs activity in the 0–7 cm soil layer, at t=20, 50, and 100 years after deposition. This quantity is defined here as the ^{137}Cs activity observed after a time t in this layer at a given plot with respect to the total deposited ^{137}Cs activity present at the same time t at the same plot (therefore radioactive decay of ^{137}Cs does not need to be considered). The percentage activity defined in this way thus accounts only for the spatial variability of the transport parameters. It can be calculated for each plot with the values of the two transport parameters of each model.
b) The normalized inventory of ^{137}Cs in the layer 0–7 cm after a given time. This quantity we define for each plot as the ^{137}Cs activity at time t in this layer at a given plot with respect to the total median ^{137}Cs activity deposited at all plots at the same time t (this quantity is also independent of the radioactive decay of ^{137}Cs). The normalized inventory at a plot thus accounts for the spatial variability of the transport parameters and of the amount of ^{137}Cs deposited.

The frequency distributions of the percentage ^{137}Cs activity in the 0–7 cm layer predicted by the three models for the time periods 20, 50, and 100 years after deposition are shown in Figs. 4, 5 and 6. Figure 4 shows, for example, that after 20 years the dispersion-convection model and the back-flow model predict that at about 40% of the plots between 90% and 100% of the activity deposited there will be still present in the 0–7-cm-layer. According to the residence time model, this will be the case only at 21% of the plots. All models, however, predict that at many plots much smaller fractions of the deposited ^{137}Cs (down to about 30%) will also be observable for this layer. Differences between the models become more evident if one looks at the predictions for 50 and 100 years. Figure 5 shows that after 50 years all models predict that the percentage activity can vary between 0% and 100% rather evenly, but while for the dispersion-convection model and the back-flow model the corresponding medians are both about 64%, the median predicted by the residence time model is significantly smaller (47%, difference significant at the 0.05% level, Wilcoxon rank sum test). Similarly, after 100 years (Fig. 6) the dispersion-convection and the back-flow model predict that the median percentage activities in the 0–7-cm-layer are 48% and 39%, respectively, while the residence time model predicts a median of only 15%. Again, however, Fig. 6 demonstrates that, as a result of the spatial variability of the transport parameters, the frequency distributions of the percentage ^{137}Cs activity in this layer are, even after 100 years, still rather wide.

Finally, the corresponding frequency distribution of the normalized inventory in the 0–7-cm layer are shown in Figs. 7, 8 and 9. As mentioned, this quantity takes also

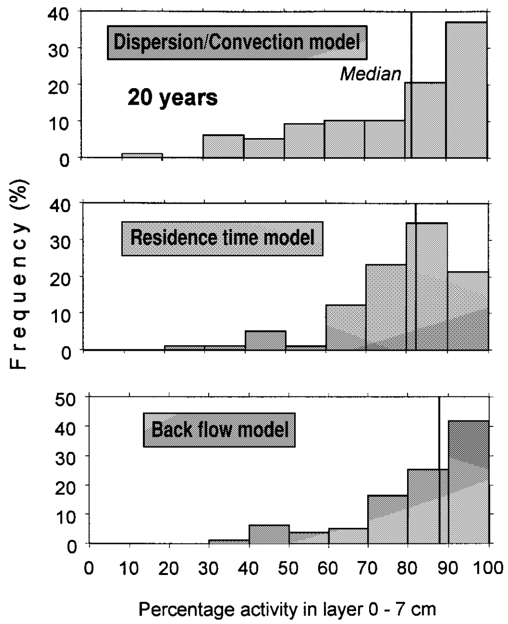

Fig. 4 Frequency distribution of the percentage activity of ^{137}Cs in the layer 0–7 cm, as predicted by the three transport models for the time period 20 years after deposition. In this case, only the spatial variability of the corresponding transport parameters is taken into account

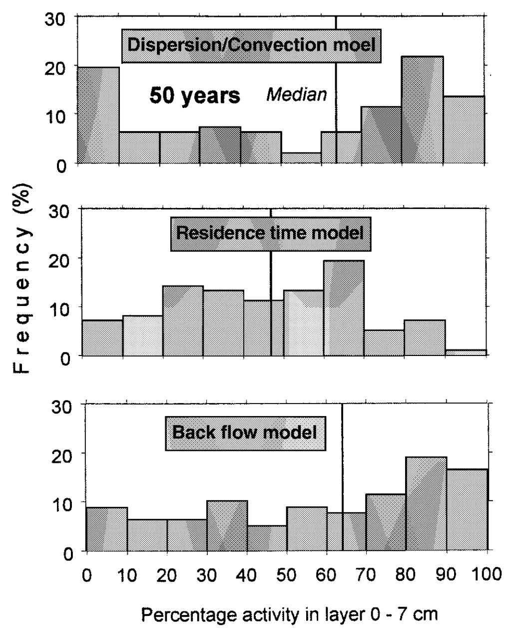

Fig. 5 As Fig. 4, but for the time period 50 years after deposition

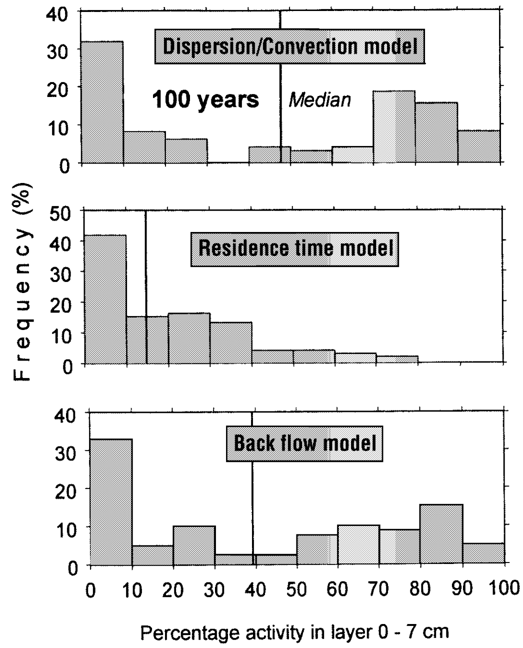

Fig. 6 As Fig. 4, but for the time period 100 years after deposition

the spatial variability of the ^{137}Cs deposition into account. The total coefficient of variation (CV) of this deposition at the sampling site was found as 45%. The normalized ^{137}Cs inventory, as defined above, can exhibit values above 100%, because on a given plot the total ^{137}Cs inventory can be larger than the mean or median ^{137}Cs inventory of the whole pasture. Because the normalized inventory includes the spatial variability of the transport parameters and that of the total inventory, the frequency distributions obtained from all models are quite wide, especially after 20 years, when very little ^{137}Cs has yet been leached from this layer. After longer times, when a substantial fraction of ^{137}Cs has been eluted from the 0–7-cm layer, the frequency distributions become smaller. Nevertheless, even after 50 years, according to the dispersion-convection model, only 0–20% of the normalized ^{137}Cs activity deposited will still be present in the 0–7-cm layer at 24% of the plots, while at 11% of the plots 80–100% of the normalised deposited activity will still be found (Fig. 8, top). Again, one can examine to what extent the predictions by the various models disagree. While after 20 years (Fig. 7) the medians and the shape of the distributions are very similar, Fig. 8 indicates that after 50 years the median normalized inventory predicted by the residence time model (41%) is somewhat smaller than that from the dispersion-convection model (49%) and from the back flow model (53%). Application of the Wilcoxon rank sum test shows, however, that these differences are not statistically significant (P>0.05). Only after 100 years (see Fig. 9) is the median normalized activity in the 0–7-cm layer as predicted from the residence time model (15%) significantly smaller (P<0.01) than that obtained by the dispersion-convection model (37%) and by the back-flow model (34%). Thus, because the normalized inventory includes the spatial variability of two sources, the corresponding frequency distributions of this quantity become so wide that significant differences in the prediction obtained by the various models will show up only after very long time periods.

In conclusion, there can be no doubt that the quality of a long-term prediction of the vertical migration of fallout

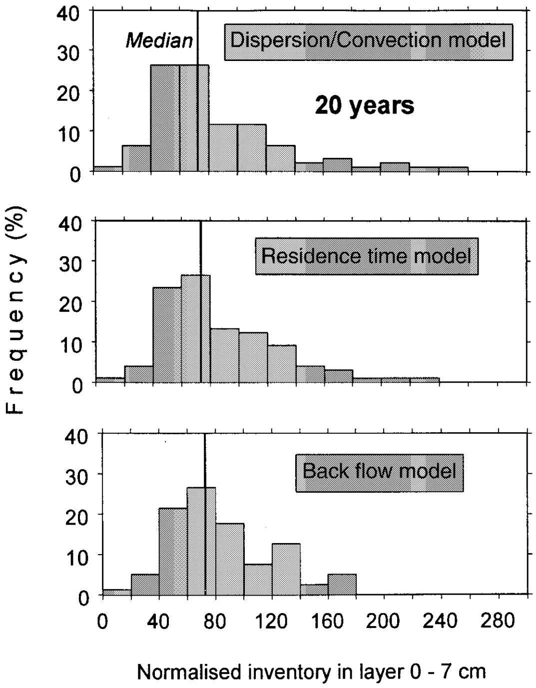

Fig. 7 Frequency distribution of the normalized inventory of ^{137}Cs in the layer 0–7 cm, as predicted by the three transport models for the time period 20 years after deposition. In this case, the spatial variability of the corresponding transport parameters and that of the amount of ^{137}Cs deposited is taken into account

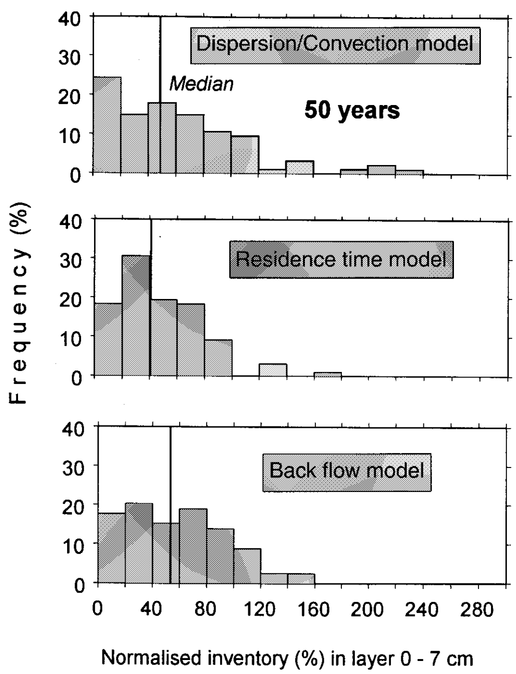

Fig. 8 As Fig. 7, but for the time period 50 years after deposition

^{137}Cs in the soil depends in the first place on the choice of the appropriate transport model. It was, however, not the purpose of this investigation to examine which of the three transport models considered here is superior in this

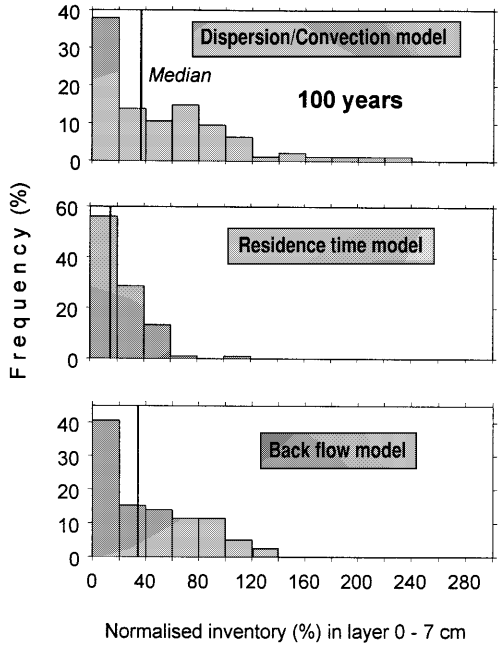

Fig. 9 As Fig. 7, but for the time period 100 years after deposition

respect. Nevertheless, the present analysis revealed that if only the spatial variability of the transport parameters is taken into account, the resulting uncertainty is not large enough to disguise significant difference in the predictions of the three models. In particular, the dispersion-convection model and the back-flow model always predicted rather similar but significantly higher long-term median ^{137}Cs inventories in the 0–7-cm soil layer than those obtained with the residence time model. If, however, in addition, the spatial variability of the amount ^{137}Cs deposited is also taken into account, the frequency distributions of the ^{137}Cs inventories in the 0–7-cm layer become so wide that differences in the median inventories predicted by the three models only become statistically significant after 100 years.

It should be noted, however, that the spatial variability of the ^{137}Cs-deposition as observed here (CV=0.45) might well be considerably smaller at other sites. Even though Ulsh et al. [21] observed similarly high values (CV=0.40 and 0.41, respectively) for alpine and montane sites in Colorado (USA) as compared to our site, on a meadow in the City of Salzburg (Austria), Lettner et al. [22] observed for the spatial variability of Chernobyl-derived ^{137}Cs fallout on a 1-ha area a value of only CV=0.21. Similarly, Sutherland and de Jong [23] found for a native grassland a CV=0.18, and McGee et al. [24] for the integrated ^{137}Cs deposition at an upland peat site a value of 0.24. For a recent review, see Sutherland [25]. In such cases, it might be expected that the differences predicted in the ^{137}Cs inventories in the 0–7-cm layer by the three models may become significant even after rather short time periods.

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