

Impact of Preferential Flow on Radionuclide Distribution in Soil

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Migration of radionuclides in soils and their transfer to edible plants are usually estimated using volume-averaged bulk concentrations. However, radionuclides might not be homogeneously distributed in soils due to heterogeneous water flow and solute transport. One important cause of heterogeneous transport is preferential flow. The aim of this study was to investigate the spatial distribution of radionuclides in the soil in relation to preferential flow paths and to assess the possible consequences for their transfer from soil to plants. We identified the preferential flow paths in a forest soil by staining them with a blue dye, and we compared radionuclide activity in samples from the stained preferential flow paths with those from the unstained soil matrix. The activities of the atmospherically deposited radionuclides ¹³⁷Cs, ²¹⁰Pb, ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am were enriched in the preferential flow paths by a factor of up to 3.5. Despite their different depositional histories, the distribution of the radionuclides between preferential flow paths and matrix was similar. Our findings indicate increased transport of radionuclides through the preferential flow paths, representing a possible risk of groundwater contamination. Furthermore, enrichment of radionuclides in the preferential flow paths might influence the uptake by plants. The heterogeneous radionuclide distribution in the soil and the more intense rooting in the preferential flow paths can be incorporated into soil-to-plant transfer models. Taking the correlated radionuclide and root distribution between the two flow regions into account provides a more physical and biological basis for the calculation of plant activities with transfer models than using the homogeneously mixed bulk soil activities as input parameters.

Introduction

Migration of elements such as radionuclides and their transfer from soil to plants are estimated with bulk soil activities, although it is known that water flow and solute transport in soils is often heterogeneous (1-5). Such heterogeneities, one of them being preferential flow, are the rule rather than the

exception (6). Preferential flow is the rapid transport of water and solutes, bypassing a large part of the soil matrix and therefore affecting only a small portion of the whole soil volume. Short-term laboratory and field experiments showed that preferential flow plays an important role in the transport of highly sorbing solutes such as pesticides (7). However, to our knowledge, no long-term field experiments exist that assess the impact of preferential flow on the spatial distribution of sorbing substances. It affects the leaching process of such substances toward the groundwater as well as their accessibility by plant roots.

To analyze the long-term impact of preferential flow on the distribution of different tracers, we take advantage of the fallout of radionuclides with different depositional histories: the single-pulse deposition of ¹³⁷Cs in 1986, the continuous deposition of ²¹⁰Pb and the step-input deposition of ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am in the 1950s and 1960s. The aim of this study was to investigate the spatial distribution of these radionuclides in a soil profile in relation to preferential flow paths and to assess the consequences for the transfer from soil to plants.

Experimental Methods

The study presented here was conducted on four plots of 3 m by 7 m situated on the corners of a well-characterized site of approximately 2 ha in Central Northern Switzerland from April 1998 to May 1999. The site was planted in 1930 with Norway spruce (*Picea abies* (L.) Karst.) as the dominant tree species mixed with beech (*Fagus sylvatica* L.) and some other species. The soil is an acid brown forest soil (Dystric Cambisol (8)). Selected chemical and physical properties are given in Table 1.

To determine the total atmospheric deposition of radionuclides, we took samples (blocks with the following dimensions: 14 cm width, 25 cm length, and 10 or 20 cm depth, see Table 2) to a depth of 0.6 m at one of the four plots.

To stain the preferential flow paths in the soil we applied 45 mm of a dye solution (deionized water containing 3 g L⁻¹ Brilliant Blue FCF (CI 42090)) in 6 h with a portable sprinkling device (6, 9) at a constant rate that did not cause ponding. This corresponds approximately to a heavy thunderstorm shower. One day after dye application, we opened a trench to 1.2 m depth. A vertical soil profile of 1 m by 1 m was prepared within the irrigated plot, 30 cm away from the plots' border, and photographed. These photos (*n* = 100) were then used to quantify the proportion of the profile area stained with Brilliant Blue. This dye coverage is a measure for the volumetric proportion of preferential flow paths. In delineated depths of 0-9, 9-20, 20-50, and 50-100 cm, samples were taken with a small spatula from the stained regions, representing preferential flow paths, and from the unstained soil matrix. On each of the four plots we sampled five profiles with a separation distance of 10 cm. To determine the root biomass we used three small cores per depth and flow region (e.g., preferential flow path and matrix) with a volume of 9.07 cm³ each. The cores were pushed carefully by hand into the soil perpendicular to the exposed face. We repeated the same experimental protocol five times (04/98, 06/98, 10/98, 04/99, 05/99). After the soil had been oven dried and sieved (2 mm), ¹³⁷Cs and ²¹⁰Pb were measured using γ -spectroscopy, and ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am (the latter being a decay product of ²⁴¹Pu) were measured using α spectroscopy (10). Since preparation prior to analysis of ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am is excessively time-consuming, we only measured a limited set of samples, namely depth-wise mixed samples of the dates 10/98 and 04/99.

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TABLE 1 Soil Characteristics (Mean of the Four Plots) of the Experimental Site

depth [cm]	pH (CaCl ₂)	C _{org} [g kg ⁻¹]	bulk density [g cm ⁻³]	CEC _{org} ^a [mmol kg ⁻¹]	texture (s/u/c) ^b [g kg ⁻¹]
0-9	3.4	26.7	1.13	80.6	360/429/211
9-20	3.7	11.8	1.32	62.5	375/423/202
20-50	3.8	5.9	1.44	62.2	360/421/220
50-100	3.8	3.0	1.56	86.5	294/456/250

^a Effective cation exchange capacity expressed as the sum of cations extracted with 1 M NH₄NO₃ and a soil-solution ratio of 1.25 ^b s = sand u = silt c = clay

TABLE 2 Radionuclide Inventories (One Plot) of the Experimental Site

depth [cm]	¹³⁷ Cs (vd [%] ^a) [Bq m ⁻²]	²¹⁰ Pb _{unsupported} (vd [%]) [Bq m ⁻²]	^{239,240} Pu (vd [%]) [Bq m ⁻²]	²⁴¹ Am (vd [%]) [Bq m ⁻²]
0-10	6251 (69)	2144 (52)	43.3 (59)	15.9 (58)
10-20	1846 (20)	436 (10)	17.4 (24)	7.5 (27)
20-40	596 (7)	829 (20)	7.1 (10)	3.1 (11)
40-60	396 (4)	717 (17)	5.6 (8)	0.8 (3)

^a vd = vertical distribution as percent of the total inventory of the soil profile

Statistical analyses for the ¹³⁷Cs and ²¹⁰Pb and root biomass data were performed with the software S+ (MathSoft, Seattle, version 5). The differences of the log-transformed activities between the preferential flow path and soil matrix samples were tested using analysis of variance. The statistical model used was a split-plot-model with the plot as the block-factor, the depth and flow region as split-unit-factors, and the time of sampling as the main-unit-factor. The data were log-transformed to approximate a normal distribution of the residuals. The residuals were checked for normality and independence using the normal-probability-plot and the Tukey-Anscombe-plot. For ²¹⁰Pb we used a paired *t*-test to test the difference between preferential flow paths and matrix within individual depth zones, although we are aware that samples within one plot may be spatially dependent.

Results and Discussion

Radionuclide Inventory The total ¹³⁷Cs inventory (Table 2) compared well to the sum of the ¹³⁷Cs fallout following the atomic bomb testing in the 1960s (~2.7 kBq m⁻²) and the Chernobyl accident in 1986 (~6 kBq m⁻², both calculated for May 1, 1986) (11). Most of the ¹³⁷Cs (69%) can still be found in the uppermost 9 cm. The agreement between measured and published results is also satisfactory for ^{239,240}Pu with a measured inventory of 73.4 Bq m⁻² and published data of 81 ± 18.5 Bq m⁻² (12) and for ²⁴¹Am with a measured inventory of 27.3 Bq m⁻² compared to the published inventory of 31 ± 7 Bq m⁻² (13). ²¹⁰Pb is an intermediate member of the decay chain of ²³⁸U. Its presence in soil samples relates to the decay of U present in the soil (i.e., supported ²¹⁰Pb, analogous to soil background) and to atmospheric deposition of ²¹⁰Pb formed from gaseous ²²²Rn escaped to the atmosphere (unsupported ²¹⁰Pb). To estimate the portion of supported ²¹⁰Pb, we subtract the mean activity of matrix soil samples from a depth below 9 cm (Table 3), assuming virtually no atmospherically derived ²¹⁰Pb could have reached the sorbing matrix below the surface horizon. We chose this method rather than derivation on the basis of the precursor ²²⁶Ra and an assumed ²²²Rn loss history. The latter can hardly be quantified as it depends on the soil water content (14). To test the validity of this simplified procedure, we compared the measured unsupported ²¹⁰Pb inventory (Table 2) with a modeled inventory. Knowing the atmospheric deposition rate

of ²¹⁰Pb ($P = 140 \text{ Bq m}^{-2} \text{ y}^{-1}$ (15) for Zurich, located approximately 20 km SE of our site), we can model the ²¹⁰Pb inventory (I) with the following equation

$$I(t) = \frac{P}{\lambda}(1 - e^{-\lambda t}) \quad (1)$$

where λ is the decay constant of ²¹⁰Pb (0.03108 y⁻¹) and t the time period of deposition and decay. The saturation level of 4500 Bq m⁻², reached after 100 y, compares well with the measured inventory of 4100 ± 200 Bq m⁻² (Table 2).

Preferential Flow Paths An example of water flow heterogeneity is depicted by the dye distribution (Figure 1). The proportion of preferential flow paths, expressed as dye coverage, decreases with depth, being 0.69, 0.38, 0.13, and 0.02 in the four different depth zones from top to bottom.

There is a significantly higher root biomass in the preferential flow paths than in the matrix ($p < 0.001$, see also Table 1). Roots often show a preference for growing in macroporous interstices rather than in the denser soil matrix or aggregates (16-19).

Radionuclide Distribution in Relation to Preferential Flow Paths ¹³⁷Cs The ¹³⁷Cs activity is significantly higher in the preferential flow paths as compared to the matrix ($p < 0.001$). In 0-9, 9-20, and 20-50 cm depth the ¹³⁷Cs activity in the preferential flow paths exceeds that of the matrix by factors of 2.2, 2.4, and 3.5, respectively. The differences in the individual depth zones are statistically significant as well (Table 3). From 50-100 cm, the ¹³⁷Cs activity is close to the detection limit in both matrix and flow path soil with no statistically significant difference between the two flow regions.

The ¹³⁷Cs activities are tracing the recent flow paths and highlight the importance of preferential flow for the spatial distribution of radionuclides and the temporal stability of the observed flow paths over a period of at least 13 y. Most of the ¹³⁷Cs originates from the Chernobyl accident on April 28, 1986, and was deposited mainly during a single rainstorm on May 1, 1986 (20). High rainfall intensities are known to promote preferential flow and the transport of rather immobile compounds such as pesticides (7).

²¹⁰Pb The distribution of ²¹⁰Pb between preferential flow paths and matrix is almost the same as in case of ¹³⁷Cs. The activities of ²¹⁰Pb are significantly higher in the preferential flow paths than in the matrix ($p = 0.001$), by a factor of 1.9 in 0-9 cm, 1.3 in 9-20 cm, and 1.3 in 20-50 cm depth (Table 3).

Atmospheric ²¹⁰Pb is removed from the atmosphere by dry and wet deposition. This represents a steady input into the soil. Despite temporal variations of the ²¹⁰Pb deposition, on time scales of a year or more, the atmospheric flux remains fairly constant (21, 22). Therefore the ²¹⁰Pb input was continuous and more or less uniform whereas ¹³⁷Cs was introduced into the system approximately as a single pulse input. The similarity in their distributions in the soil in relation to the preferential flow paths suggests that the flow paths that were actively conducting water during the time period of the Chernobyl fallout are largely the same ones that were "active" over the course of years.

^{239,240}Pu and ²⁴¹Am In contrast to ¹³⁷Cs and ²¹⁰Pb, the activities of ^{239,240}Pu and ²⁴¹Am seem to be only slightly enriched in the preferential flow paths of the topsoil (0-9 cm). To compare the various radionuclides, we calculated the activity differences between preferential flow paths and matrix samples and normalized them with the inventory given in Table 2. Doing this, the differences between preferential flow paths and the matrix of ^{239,240}Pu and ²⁴¹Am are only slightly smaller than those of ¹³⁷Cs and ²¹⁰Pb. In 9-20 cm depth, ^{239,240}Pu and ²⁴¹Am activities are markedly larger in the preferential flow paths. At even greater depths, both

TABLE 3. Activities of Radionuclides and Root Biomass in Stained Flow Paths and Unstained Soil Matrix

depth [cm]	root biomass ^a [g m ⁻²]	¹³⁷ Cs ^b [Bq kg ⁻¹]	²¹⁰ Pb ^c [Bq kg ⁻¹]	²⁴¹ Am ^d [Bq kg ⁻¹]	m.e. ^e	^{239,240} Pu ^d [Bq kg ⁻¹]	m.e.	²³⁸ Pu ^d [Bq kg ⁻¹]	m.e.
Flow Path									
0-9	1654**	82.6**	60.09**	0.33	0.03	0.78	0.04	0.032	0.003
9-20	859*	12.7**	21.22*	0.08	0.01	0.23	0.01	0.008	0.002
20-50	645**	4.4**	18.49*	0.015	0.004	0.02	0.01	<LD	
50-100	nm ^f	0.9	20.09	0.006	0.002	0.01	0.01	<LD	
Matrix									
0-9	1022	37.9	32.13	0.27	0.03	0.66	0.03	0.021	0.003
9-20	509	5.3	15.94	0.045	0.01	0.11	0.01	0.003	0.001
20-50	226	1.26	14.64	<LD ^g		<LD		<LD	
50-100	nm ^f	0.85	22.32	<LD		<LD		<LD	

*** Difference between preferential flow paths and matrix is statistically significant ($p < 0.01$ and $p < 0.05$ respectively) * Fine roots (<2 mm)
^b ¹³⁷Cs activities are decay corrected to March 1 1998 ^c Measured ²¹⁰Pb (supported and unsupported) ^d α measurements are decay corrected to the date of measurement (February 1999) ^e Measurement error ^f Not measured ^g Limit of detection

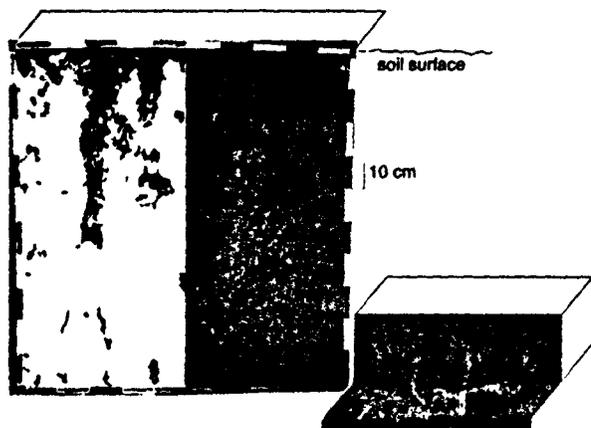


FIGURE 1 Soil profile with stained preferential flow paths. The image on the left shows the black and white image of stained flow paths and unstained soil matrix. The image was digitized and processed with the software Adobe Photoshop 5.0. The inset exhibits details from 90 to 100 cm depth and shows the three-dimensionality of flow paths.

^{239,240}Pu and ²⁴¹Am were only detected in flow-path material. The ratio ^{239,240}Pu/²⁴¹Am is remarkably stable in all samples (2.5 ± 0.2), excluding samples with activities close to the detection limit (Table 3). These ratios lie in the same range as those of other samples of Swiss soils obtained in 1999 (Froidevaux, unpublished data). This implies a similar migration rate of Pu and Am.

Plutonium was emitted into the atmosphere as a consequence of atmospheric nuclear bomb tests and the SNAP-9A (Systems for Nuclear Auxiliary Power generator) satellite re-entry in the 1950s and early 1960s (12). It reached the ground both as dry and wet deposition in a variety of rain events, attached to leaf litter, and as particles. The importance of the depositional history for radionuclide migration and distribution in the soil is documented by other studies (13, 23).

The elevated activities of ¹³⁷Cs, ²¹⁰Pb, ^{239,240}Pu, and ²⁴¹Am in preferential flow paths highlight the importance of preferential flow for the mobility of strongly sorbing substances and are an expression of the temporal stability of the preferential flow structures. Generally, all radionuclides investigated in this study are rapidly immobilized by soil particles and show a slow transport behavior in soils and sediments once they are adsorbed (13, 23, 24). Diffusion is probably negligible. For example, Pu occurs in soils and sediments mainly in the form of tightly bound plutonium dioxide (25) and has a high K_d (>500 mL g⁻¹) (26) and a low

diffusion coefficient (<10⁻¹⁰ cm² s⁻¹) (27). By comparing nuclear weapon-derived cesium in soils between 1977 and 1992, Smith et al. found no vertical mobility during these 15 years (28).

The occurrence of radionuclides at greater depths implies an initial mobility during the first hours after deposition, when water flow and thus physical rather than chemical processes control their behavior. After the initial displacement, redistribution between phases and surface reactions become more important. We cannot rule out remobilization and translocation of radionuclides by co-transport with dissolved or colloidal ligands (1, 26, 29-31), but even in case of facilitated transport the main movement of radionuclides would still occur in preferential flow paths. If we assume only an initial mobility of radionuclides immediately after deposition, we can estimate the temporal stability of flow paths to be at least 40 y.

Relevance. Radionuclides travel more rapidly through the supposedly strongly sorbing soil toward groundwater than expected based on the chromatographic transport concept, therefore providing a risk for the groundwater. Recently, activities of ¹³⁷Cs above the tolerance level were found in aquifers around Chernobyl (Ukraine) despite a thick soil layer that should have served as a natural filter (Shestopalov and Bohuslavsky, 2000 oral communication).

The second important pathway from the soil to the human food chain is the uptake by plants. The transfer of elements such as ¹³⁷Cs from soil to plants is in most transfer models described by a transfer factor relating plant to soil concentrations. The transfer factor is obtained from greenhouse experiments using well mixed, unstructured soil materials (32). Commonly used transfer models, e.g., the radionuclide dose estimation program ECOSYS, which is the basis of the emergency management systems in Germany, Austria, and Switzerland and an integral part of the European real time dose assessment system, assumes homogeneous uptake from a defined soil volume (2). In the original equation

$$C_{\text{plant}} = C_{\text{soil}} F \quad (2)$$

the plant concentration C_{plant} is expressed in terms of the bulk concentration in the soil and the transfer factor F . The bulk concentration C_{soil} is obtained by mixing the entire radionuclide inventory into the top 10 cm (grassland) or 25 cm (arable land) of the soil. Using the ¹³⁷Cs deposition found at our study site and a transfer factor of 0.14 (32) yields a ¹³⁷Cs activity in fresh grass of 3.5 Bq kg⁻¹ for 25 cm mixing depth or 8.7 Bq kg⁻¹ for 10 cm mixing depth. Equation 1 yields highly variable results depending on the mixing depth over which we assume homogeneous distribution of radionuclides (Figure 2, right-hand side).

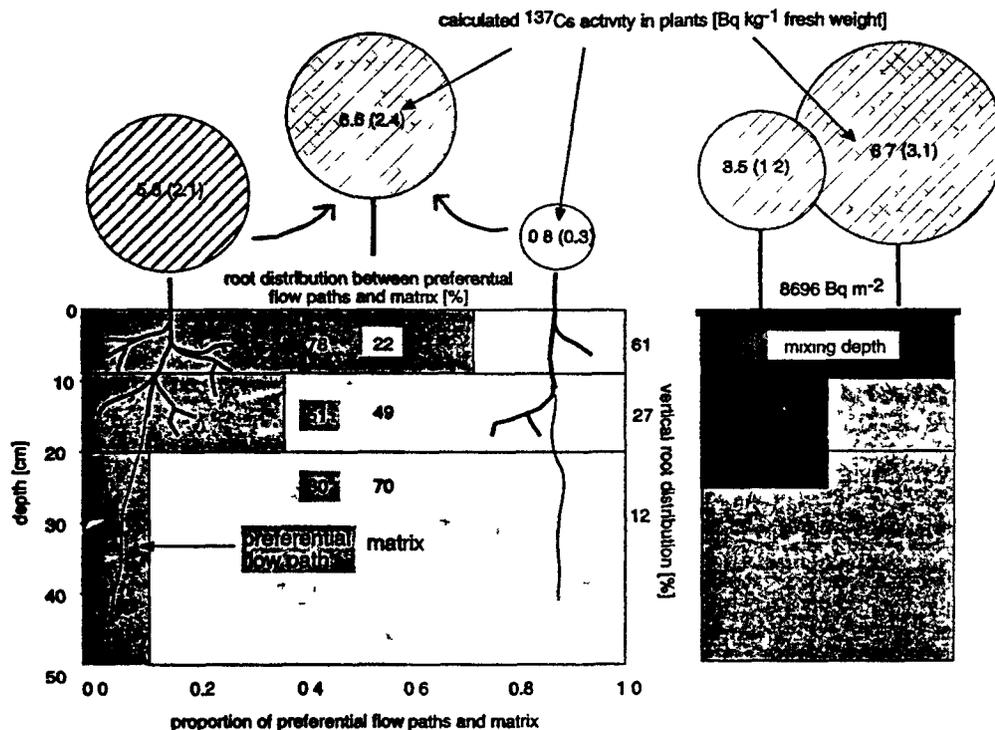


FIGURE 2. Grass ^{137}Cs activities calculated with the commonly used transfer model ECOSYS (eq 2, right-hand side) and with our transfer model including preferential flow and inhomogeneous root distribution (eq 3, left-hand side). ECOSYS calculates the soil ^{137}Cs activity by mixing the deposition of $8696 \text{ Bq } ^{137}\text{Cs m}^{-2}$ homogeneously to a soil depth of 25 and 10 cm, respectively, with a bulk density of 1.4 g cm^{-3} . Transfer factors for grass are taken from Ng, 0.14 (32), and Müller and Prühl, 0.05 (numbers in parentheses) (2).

Knowing the radionuclide and root distribution in the soil, we can estimate the plant concentrations on a more physical and biological basis. Since root biomass is enriched in the preferential flow paths as compared to the soil matrix (Table 3), the elevated radionuclide activities in the preferential flow paths represent higher concentrations in the rhizosphere. The following extended equation illustrates how preferential radionuclide distribution and inhomogeneous rooting can be incorporated into soil-to-plant transfer models

$$C_{\text{plant}} = \sum_{i=1}^{n_h} \sum_{j=1}^2 C_j(\Delta z_i) R_j(\Delta z_i) F_j \quad (3)$$

where $j = 1$ for preferential flow paths, $j = 2$ for soil matrix, n_h refers to the number of soil horizons, Δz to the depths of the individual horizons, C to the radionuclide concentration, F to the transfer factor, and R to the relative root density expressed as

$$R_j(\Delta z_i) = \frac{r_j(\Delta z_i) A_j(\Delta z_i)}{\sum_{i=1}^{n_h} \sum_{j=1}^2 r_j(\Delta z_i) A_j} \quad (4)$$

with r_j and A_j being the root density and the area of flow region j . Thus, the root density in the two flow regions (preferential flow paths and matrix) is weighed with the area of the respective flow region to account for the "actual" root distribution in the soil profile (for the distribution of the root densities between preferential flow paths and matrix within the individual soil horizons, see Figure 2). The radionuclide activity in plants can be calculated on the basis of eq 3 with measured root and radionuclide concentrations in the preferential flow path and matrix compartments, the areas of the two compartments (Table 3, Figure 2), and published transfer factors (in our example, they were assumed to be

equal for the two compartments). With measured ^{137}Cs activities of preferential flow paths and matrix and the respective root concentrations, we can estimate the contribution of the two flow regions to the calculated plant activities. By adding the contributions of both flow regions, we calculate the plant activity to be 6.6 Bq kg^{-1} (Figure 2, left-hand side). For the calculation, we included the three depth zones from 0 to 0.5 m. However, since the root biomass is relatively small in 20–50 cm depth and ^{137}Cs activities are low, the contribution of the root uptake from 20 to 50 cm depth to the total plant activity is smaller than 1%.

This example shows that the substantially different elemental concentrations of preferential flow paths and matrix may be a critical factor for calculating plant concentrations with a transfer model. The differentiation of the flow regions is important, because the root distribution, which is a key factor for plant uptake, is biased toward higher root densities in the preferential flow paths. Taking the correlated radionuclide and root distribution between the two flow regions into account provides a more physical and biological basis for the calculation of plant activities than mixing the entire deposition homogeneously over an arbitrarily chosen depth. This concept can also be applied to other elements such as heavy metals or nutrients.

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