Migration of Fallout $^{239+240}$Pu, $^{241}$Am and $^{137}$Cs in the Various Horizons of a Forest Soil Under Pine

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ABSTRACT

At five plots in a pine stand the vertical distributions of $^{239+240}$Pu, $^{241}$Am and $^{137}$Cs from the global fallout of weapons testing in the sixties, and of $^{137}$Cs from the Chernobyl fallout, in the soil (podzol) were determined. To obtain the migration rates of the radionuclides in each soil horizon from these depth profiles, a compartment model was used. On average, the vertical migration rates of Pu and Am from the global fallout were similar and low (in general $<1$ cm year$^{-1}$) in all soil horizons. The smallest mobility ($0.1$ cm year$^{-1}$) was observed in the O$fh$ horizon, i.e. the horizon where the pine needles are already decomposed and humic substances have been formed. In the underlying mineral horizon the migration rates increase in general with increasing depth. Radiocaesium from the global fallout is also enriched in the O$fh$ horizon and exhibits rather similar migration rates to the two actinides, except in the O$fh$ horizon, where it is more mobile by a factor of about five. While the enrichment of the actinides in the organic soil layer seems to be the result of complex formation with humic substances, the corresponding enrichment of radiocaesium in this layer is probably due to temporary immobilization and recirculation processes induced by the soil microflora. Chernobyl-derived $^{137}$Cs is presently significantly more mobile in the mineral soil than radiocaesium from the global fallout by about a factor of two.
INTRODUCTION

To estimate the long-term radiation dose to man due to artificial radionuclides in the environment, a detailed knowledge of their behaviour in the biosphere is indispensable. Because most fallout-radionuclides are deposited eventually on the soil surface, an information on the migration behavior, especially of the long-lived radionuclides, in the various layers of a given soil type is necessary. Otherwise, it will not be possible to estimate the time during which the deposited radionuclides can be resuspended into the air from the top soil layer, or to assess the residence-time of a radionuclide in the root zone. In addition, the external $\gamma$-radiation dose at the soil surface will depend strongly on the depth distribution of the corresponding radionuclide in the soil. The present information available on this subject is, however, mainly restricted to the migration behavior of radiocaesium in agricultural soils (see, e.g. Coughtrey & Thorne, 1983). Much less is known about the highly radiotoxic $\alpha$-emitters plutonium and americium, which can be determined in soil samples only after elaborate radiochemical separation procedures (Muller & Sprugel, 1977; Rai & Serne, 1977; Jakubik, 1979; Wildung & Garland, 1980; Frissel et al., 1981; Bondietti, 1982; Yamamoto et al., 1982; Cawse, 1983; Gee et al., 1983; Coughtrey et al., 1984a, b; Peirson, 1988; Bunzl et al., 1992). This is especially true for forest soils, where very few data are available, even though forest products, such as mushrooms, berries and game, can contribute significantly to the radiation exposure of man.

The purpose of the present investigation was, therefore, to obtain quantitative information on the migration rates of fallout $^{239+240}\text{Pu}$, $^{241}\text{Am}$ and $^{137}\text{Cs}$ in the various horizons of a typical sandy podzol soil in a pine stand.

Plutonium-239+240, $^{241}\text{Am}$ and $^{137}\text{Cs}$ were deposited on the soil surface in Germany in the sixties as a result of the global fallout from weapons testing, and again in 1986 after the reactor accident at Chernobyl. By evaluating the presently observed depth profiles of these radionuclides in a soil it is possible to obtain, for each soil horizon, information on the corresponding mean migration rate since the time of their deposition. For this purpose, the data at which a given fallout-radionuclide was deposited on the soil surface has to be known. Because $^{239+240}\text{Pu}$ and $^{137}\text{Cs}$ were deposited during two time periods, it is necessary to determine separately, in each soil layer, the corresponding fractions originating from the global fallout in the sixties, and from the Chernobyl fallout in 1986. This can be achieved by measuring in each soil layer the isotopic ratios $^{238}\text{Pu}/^{239+240}\text{Pu}$ and $^{134}\text{Cs}/^{137}\text{Cs}$, because these ratios were significantly
different in the fallout from these two sources. In this way the migration rates of Pu and radiocaesium from both sources in the various soil layers can be compared.

Americium-241 is present in German soils mainly due to ingrowth from global fallout-241Pu during its passage through the soil rather than by direct deposition on the soil surface. The migration rate of Am in each soil layer can, therefore, only be determined after evaluation of the migration rate of Pu in each layer and subsequent consideration of the Am-ingrowth.

In a recent study on the migration of these radionuclides in a podzolic parabrown earth under spruce (Bunzl et al., 1992) it was observed that the migration rates of Pu, Am and radiocaesium from global fallout were rather similar in most soil horizons and increased with depth in the mineral horizons. A considerably higher migration rate was, however, observed for radiocaesium from the Chernobyl fallout. It was, therefore, of particular interest to see whether such a behaviour was also observable in other forest soils.

MATERIAL AND METHODS

Site and soil

The pine (Pinus sylvestris) plantation 'Taxöldener Forst' is located 200 km northeast of Munich, Germany. The height above sea level is 400 m, the annual precipitation 710 mm, and the average annual temperature 7.9 °C. The soil is classified as a podzol (FAO: Podzol; US system: Spodosol) and exhibited the following horizons: Olf (5.5–1 cm); Ofh (1.5–0 cm); Aeh (0–2 cm); Ahe (2–9 cm); Bhs (9–20 cm); Bsv (>20 cm). Some characteristics of this soil are given in Table 1.

Sampling

At five places in the forest, always located approximately in the middle between the trees (i.e. about 3 m from the trunks), the horizons Olf, Ofh, Aeh were sampled separately with a frame in 1991. The size of the frame was 40 × 60 cm in the top layer. To minimize contamination of the deeper soil layer by dropping particles from the upper layers, successively smaller frame sizes were used for sampling the deeper soil layers. Samples from the Ahe horizon were taken in layers of 2–5 and 5–9 cm, from the Bhs horizon in layers 9–15 and 15–20 cm. From the Bsv horizon the 20–30 cm layer was sampled.
<table>
<thead>
<tr>
<th>Soil horizon (depth (cm))</th>
<th>Olf (4–1.5)</th>
<th>O fh (1.5–0)</th>
<th>Aeh (0–2)</th>
<th>Ahe (2–9)</th>
<th>Bhs (9–20)</th>
<th>Bsv (&gt;20)</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH (CaCl₂)</td>
<td>3.1</td>
<td>2.7</td>
<td>2.3</td>
<td>3.3</td>
<td>4.0</td>
<td>4.3</td>
</tr>
<tr>
<td>Clay, &lt;2 µm (%)</td>
<td>—</td>
<td>—</td>
<td>3</td>
<td>3</td>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td>Silt, 2–63 µm (%)</td>
<td>—</td>
<td>—</td>
<td>17</td>
<td>17</td>
<td>11</td>
<td>7</td>
</tr>
<tr>
<td>Sand, &gt;63 µm (%)</td>
<td>—</td>
<td>—</td>
<td>80</td>
<td>80</td>
<td>84</td>
<td>90</td>
</tr>
<tr>
<td>Organic matter (%)</td>
<td>84</td>
<td>51</td>
<td>8.9</td>
<td>2.6</td>
<td>5.8</td>
<td>1.4</td>
</tr>
<tr>
<td>N total (%)</td>
<td>1.5</td>
<td>0.85</td>
<td>0.13</td>
<td>0.05</td>
<td>0.10</td>
<td>0.03</td>
</tr>
<tr>
<td>CEC (Mehlich) (meq/kg)</td>
<td>861</td>
<td>811</td>
<td>108</td>
<td>53</td>
<td>155</td>
<td>38</td>
</tr>
<tr>
<td>Exchangeable ions (meq/kg)</td>
<td>Ca</td>
<td>83</td>
<td>19</td>
<td>1.4</td>
<td>1.3</td>
<td>2.9</td>
</tr>
<tr>
<td></td>
<td>Mg</td>
<td>9</td>
<td>4</td>
<td>0.5</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>K</td>
<td>10</td>
<td>6</td>
<td>0.6</td>
<td>0.3</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>Na</td>
<td>2</td>
<td>2</td>
<td>0.1</td>
<td>0.3</td>
<td>&lt;0.1</td>
</tr>
<tr>
<td></td>
<td>Fe (mg/100g)</td>
<td>2.4</td>
<td>21</td>
<td>6.6</td>
<td>3.1</td>
<td>7.0</td>
</tr>
<tr>
<td></td>
<td>Mn (mg/100g)</td>
<td>2.8</td>
<td>0.4</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
</tr>
</tbody>
</table>
Procedures

Before radiochemical analysis, the air dried soil from each layer of the mineral soil was sieved to 2 mm for the removal of stones and roots (c. 10% in the 0–2 cm layer; < 3% in the deeper layers), and carefully mixed. The dry weight was determined at 105°C. To obtain some information on the spatial variability of the migration rates of the radionuclides in the soil, each layer from each of the five plots was analysed separately.

Determination of radiocaesium

Caesium-137 and 134Cs were determined by direct γ-spectrometry, using a high-purity germanium detector and a multichannel analyser. With 134Cs, losses by sum-coincidences during counting were taken into account.

Determination of Pu and Am

Plutonium and Am were isolated from the soil matrix and purified by radiochemical separation procedures. Special care was taken in removing 228Th, because it interferes in the α-spectrum with 238Pu, and is present in the mineral soil layers in a very large excess with respect to 238Pu. For details see Bunzl et al. (1992, 1994). From the purified Pu fraction the plutonium isotopes were electrodeposited on stainless steel discs. Subsequently, 239+240Pu and 238Pu were determined by α-spectrometry, using a 300 mm² silicon surface barrier detector (alpha efficiency 10–20%). The resolution was 40–50 keV FWHM at 4–6 MeV. Similarly, from the purified Am-fraction, Am was electrodeposited and 241Am again determined by α-spectrometry. The detection limit for a counting time of 10,000 min was about 0.1 mBq of 238Pu, 239+240Pu and 241Am. Quality control was assured by determining Pu- and Am-isotopes in certified reference samples.

Evaluation of the origin of 137Cs and 239+240Pu

Caesium-137 was deposited in the sixties by global fallout and in 1986 by Chernobyl fallout. Because 134Cs is only Chernobyl-derived and its ratio 134Cs/137Cs is well known for the sampling area (1.75 ± 0.07; reference date 27 April 1986; Hötzl et al., 1987), it is possible to calculate the 137Cs fraction from each source separately for each soil layer from the measured 137Cs and 134Cs activities. This fractionation is also possible for 239+240Pu, because the ratio 238Pu/239+240Pu was c. 0.03 in south Germany for the
global fallout (Bunzl & Kracke, 1988) and for the Chernobyl fallout 0.42 (Hötzl et al., 1987). Occasionally, a greater mobility of $^{238}\text{Pu}$ from the global fallout as compared to $^{239+240}\text{Pu}$ is reported (Hanson, 1975; Cigna et al., 1988). In the present case, however, such a behaviour is not indicated. Except in the top horizon (Olf), where Chernobyl-derived Pu can be expected, the ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ was within the range reported for global fallout (3–5%) in each soil layer. Because, there was only one value slightly above 6% observed for this ratio in the top Olf horizon, in the following any contribution of Chernobyl-derived Pu to the observed Pu activity in the soil will be neglected.

**Evaluation of the rates of migration**

The residence halftimes $\tau$ of Pu, Am and Cs in the various soil layers were evaluated with a compartment model (Frissel et al., 1981; Boone et al., 1985, Velasco et al., 1993). The advantage of this compartment model is that no information on the actual transport processes of the radionuclide in the soil (e.g. sorption properties, speciation, water infiltration, etc.) is needed. The disadvantage of this model is, of course, that one can only evaluate the residence times of a radionuclide in the various soil layers, but can not obtain any information about the physico-chemical processes controlling the migration process. Nevertheless, the residence times obtained are useful in comparing quantitatively the mobility of different radionuclides in different soils. In the present case, for the transfer of activity $A_i$ (Bq m$^{-2}$) of a radionuclide in the compartment ‘i’ in a small time interval $\Delta t$ (day)

$$\frac{\Delta A_i}{\Delta t} = K_{i-1} A_{i-1} - K_i A_i - \lambda A_i$$

where $K_i$ (day$^{-1}$) is the fractional rate of transfer from compartment ‘$i - 1$’ to compartment ‘$i$’, and the $\lambda$ is the disintegration constant of the radionuclide. In the first compartment eqn (1) has to be written as

$$\frac{\Delta A_1}{\Delta t} = D(t) - K_1 A_1 - \lambda A_1$$

where $D(t)$ is the rate of deposition (in Bq cm$^{-2}$ day$^{-1}$), which must be known as a function of time. To obtain, for each radionuclide, in each soil layer the corresponding value of $K_i$, the system of eqns (1) and (2) is integrated numerically. The initial condition for the integration is the depth profile of the activity $A_{i0}$, measured at $t = 0$ for each radionuclide. In the case of the global fallout, $D(t)$ is known (see below); and for the initial
depth profile $A_{i0}$ we assume (in accordance with Frissel et al. (1981) and Jakubik (1979)) that the activity of the radionuclide remains, initially, completely in the first layer. In the case of the Chernobyl-fallout this assumption is not justified for south Germany (see below). For this situation we measured the initial depth profile $A_{i0}$ after the deposition period and put $D = 0$ thereafter.

Because the $A_i$ in eqn (1) were determined experimentally, $K_i$ is the only unknown parameter for each layer $i$ and can be evaluated by an iterative procedure to any accuracy desired. In practice, however, it is sufficient to evaluate the $K_i$ only with an accuracy corresponding to the analytical error of the depth profile. The residence half-time of the radionuclide in layer $i$ is given then by $\tau_i = 0.693/K_i$ (Boone et al., 1985). To compare the values of $\tau$ observed in soil horizons of different thicknesses, one can, for example, report for each horizon the value of $\tau/L$ (Frissel et al., 1981). Because, however, the reciprocal of this ratio can be interpreted as a velocity, in the following, the rate of migration of a radionuclide in a given horizon $i$ will be characterized by $v_i = L_i/\tau_i$.

Initial conditions

Radioceasium

For $D(t)$ of the $^{137}\text{Cs}$ deposition from the global fallout to the soil surface in the sixties the values given by Bachhuber et al. (1982), and Frissel et al. (1981) will be used. Most of the deposition from Chernobyl-derived radioceasium occurred at the sampling location a short time after a heavy thunder shower (this in contrast to the radioceasium from the global fallout, where only a very small fraction of the total activity was deposited during thunder showers). As a result, a significant fraction of the Chernobyl-radioceasium deposited was immediately infiltrated into the soil via channels and cracks at a rate that exceeded the long-term Cs-migration by a few orders of magnitude (Schimmack et al., 1989). To eliminate this anomalous initial infiltration effect for the evaluation of the long-term migration rate, the Chernobyl-derived $^{137}\text{Cs}$-depth profile in the soil, determined from one soil core at the sampling site shortly after this event (July 1986) was used as the initial depth profile at $t = 0$ in the compartment model.

To check, whether at this time most of the radioceasium activity had actually arrived at the soil surface, we determined in 1991 again, for comparison, the total activity of Chernobyl-derived radioceasium in the soil of the pine stand. Within experimental error, it was identical to that observed in July 1986. This shows that obviously all of the Chernobyl-
derived radiocaesium in the pine canopy was deposited on the forest floor within about two months. It should be noted, however, that for other tree species a quite different deposition record of Chernobyl-derived radiocaesium on the soil surface might occur. Thus, in an old stand of Norway spruces it was observed, for example, that it took about two years before all the Chernobyl-radioactivity was transferred from the canopy to the forest floor (Bunzl et al., 1989a).

**Plutonium**

The deposition history of $^{239+240}$Pu from the global fallout was obtained from that of $^{137}$Cs (see above) and also the observed ratio of $^{239+240}$Pu/$^{137}$Cs in the soil was obtained. Because the migration rate of these two radionuclides in the soil may be different, this ratio has to be obtained from their total activity per unit area of soil (0-40 cm). In the present case, on average, (five profiles) a value of $0.04 \pm 0.02$ was found. Within experimental error, this is similar to measurements shortly before the Chernobyl accident, where values around $0.018$ were found (Cawse & Baker, 1985; Bunzl & Kracke, 1988). As a result of $^{137}$Cs-decay, this ratio was of course correspondingly lower during the main fallout period in the sixties.

**Americium**

As mentioned previously, $^{241}$Am is in the soil as a result of the ingrowth from $^{241}$Pu and to a much smaller extent from direct deposition. Equation (1) has to be extended for this radionuclide to

$$\frac{\Delta A_{i,Am}}{\Delta t} = K_{(i-1),Am} A_{(i-1),Am} - K_{i,Am} A_{i,Am} - \lambda_{Am} A_{i,Am}$$

Evaluation of eqn (3) requires the depth profile of $^{241}$Pu in the soil as a function of time. This quantity was not measured. However, because it can be assumed that the $K_{i}$ of $^{239+240}$Pu and $^{241}$Pu are identical (no isotope effect for the migration rate), $A_{i,241Pu}$ can be calculated as a function of time from the $A_{i,239+240Pu}$ obtained first and the known deposition history of $^{241}$Pu in the first compartment. This latter quantity is obtained from the deposition history of $^{239+240}$Pu (see above) and the ratio $^{241}$Pu/$^{239+240}$Pu $= 16$ in fresh weapons testing fallout (Holm, 1988). The very small fraction of directly deposited $^{241}$Am as a function of time was obtained from the amount of $^{241}$Pu present in the fallout.
RESULTS AND DISCUSSION

Accumulated deposition of $^{239+240}$Pu, $^{241}$Am and $^{137}$Cs in the soil

The total activities observed presently in the forest soil per square metre and down to a depth of $30\,\text{cm}$ are given in Table 2. Because these radionuclides were not observed below that depth, the values given represent the accumulated activity of each radionuclide deposited at that site by the weapons-testing and by the Chernobyl fallout. While the contribution of Chernobyl-derived $^{239+240}$Pu and $^{241}$Am to the previous inventory of these radionuclides in the soil from the global fallout is negligible, this is not the case for $^{137}$Cs (see Table 2). For this radionuclide the Chernobyl-derived deposition was higher by a factor of about five as compared to global fallout. The mean ratio $^{241}\text{Am}/^{239+240}\text{Pu}$ in the soil from global fallout is about 0.3 and thus agrees well with other observations on the total deposition by the global fallout.

Depth profiles

As an example of the five depth profiles of $^{137}$Cs, $^{239+240}$Pu and $^{241}$Am determined in the pine stand, the values observed at Site A are shown in Fig. 1. Qualitatively similar profiles were found at the other sites. For $^{239+240}$Pu, $^{241}$Am and $^{137}$Cs from the global fallout the highest activity concentrations (in Bq per square metre and per cm thickness of the soil layer) are found about 30 years after the deposition in the Ofs horizon, i.e. in the organic layer where the pine needles are largely humified. Below that layer the activity concentrations decreased rapidly and were negligible for depths $>30\,\text{cm}$.

Migration rates

To obtain more quantitative information on the vertical transport of the radionuclides, the depth profiles were evaluated with the compartment

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Global fallout</th>
<th>Chernobyl-derived</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>$1830 \pm 700$</td>
<td>$6449 \pm 1879$</td>
</tr>
<tr>
<td>$^{239+240}$Pu</td>
<td>$55 \pm 9$</td>
<td>$\leq 0.4$</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>$21 \pm 4$</td>
<td>—</td>
</tr>
</tbody>
</table>
model described above to obtain the migration rates for each radionuclide and for each soil layer from the five depth profiles. Even though Pu, Am and Cs from the global fallout were observable down to a depth of 30 cm in the soil, it is not possible to obtain with the compartment model the migration rates for the deepest compartment. Therefore, the rates are given for these radionuclides only to a depth of 20 cm, and for Chernobyl-derived radiocaesium to a depth of 9 cm. To illustrate the spatial variability of these data, they are plotted separately in Fig. 2. The considerable variability of the migration rates at the various plots is clearly visible, especially in the deeper soil layers and shows that at least four to five depth profiles should be evaluated to obtain a representative average rate for each soil layer. These average rates, calculated for each soil layer as the median values from the five individual values, are given in Fig. 3.

**Plutonium and americium**

Figure 3 shows that the migration rates of $^{239+240}\text{Pu}$ and $^{241}\text{Am}$ from the global fallout are small in all soil horizons (in general $< 1 \text{ cm year}^{-1}$) and that they are rather similar. To test statistically, whether a significant difference between the migration rates of Pu and Am exists in any soil layer, the ratio $r = (\text{migration rate of Pu})/(\text{migration rate of Am})$ was
Fig. 2. Individual vertical rates of migration of fallout $^{239+240}\text{Pu}$, $^{241}\text{Am}$, and $^{137}\text{Cs}$ from the global fallout in the sixties and of $^{137}\text{Cs}$ from the Chernobyl fallout in the soil as observed in each soil layer at the five sites in the pine forest.

calculated for each layer of the five plots. The $t$-test was then applied to detect whether, for a given soil layer, this ratio was significantly different from unity. A value for $r$, which is significantly different from unity ($P < 0.05$, two sides) was, however, not found for any soil horizon. This does not imply that differences in the mobilities of Pu and Am do not exist. They might be present, but they are obviously too small to be detected from the limited data available (five depth profiles in the present case).

Figure 3 illustrates that for Am and Pu the migration rates decreased from about 0.5 cm year$^{-1}$ in the Olf horizon to 0.1 cm year$^{-1}$ in the Ofh horizon. The strong retention of these actinides in the Ofh horizon is obviously the result of complex formation between these elements and humic substances (Coughtrey, 1984a; Kim et al., 1989; Pavlotskaya et al., 1991a,b). In the underlying mineral horizon the migration rates of the actinides increases with increasing depth from about 0.3 cm year$^{-1}$ in the 0–2 cm layer to about 1.7 cm year$^{-1}$ in the 15–20 cm layer, with exception of the 9–15 cm layer, where these rates are only around 0.4 cm year$^{-1}$. Again, humic substances with the ability to form complexes dominate in the organic fractions of this horizon.
Fig. 3. Average vertical rates of migration of fallout $^{239+240}$Pu, $^{241}$Am, and $^{137}$Cs from the global fallout in the sixties and of $^{137}$Cs from the Chernobyl fallout in the soil as observed in each soil layer in the pine forest.

It is interesting to compare these results with a recent investigation in the parabrown earth soil of a spruce stand ($Picea abies$), where it was observed that fallout Pu and Am were enriched strongly in the top layer of the mineral layer, rather than in the organic horizons as observed here (Bunzl et al., 1992). The migration rates of $^{239+240}$Pu under spruce were $0.2 \text{ cm year}^{-1}$ in the Oh horizon and $0.08 \text{ cm year}^{-1}$ in the top mineral layer. The migration rates of $^{241}$Am were similar to those of $^{239+240}$Pu.

A detailed explanation for this different behaviour cannot be given presently. Because actinides are hardly taken up even by any plants (Lux et al., 1993), it is rather unlikely that recirculation of actinides in the organic horizons, as observed for nutrient ions and radiocaesium (see below) will be responsible for this behaviour. In earlier investigations on the soil-to-plant transfer in cereals (Bunzl & Kracke, 1987), however, it is observed that Fe seems to behave in a way that is similar in some respects to Pu and Am. Probably, because of the similar charge/radius ratio of Fe(III) and the tetravalent actinides, the formation of organic complexes is similar for these elements. The enrichment of iron in the Ofh-horizon of the pine stand ($51 \text{ mg m}^{-2}$) as compared to the much lower corresponding
value in the spruce stand (11 mg m\(^{-2}\)) demonstrates the comparatively high concentration of organic ligands for iron in the Oh-layer of the pine stand. The analogous enrichment of Pu and Am in this layer thus indicates strongly that Fe, Pu and Am exhibit a similar tendency for complex formation with soil organic matter.

Migration rates of actinides in forest soils are hardly available for comparison. Bertha and Choppin (1984), concluded that Am should be more mobile than Pu as hydrolysis of Am(III) occurs more readily than hydrolysis of Pu(IV). Similarly, Routson et al. (1977) observed in acidic soils that Pu showed one order of magnitude greater sorption coefficients than Am. Coughtrey \textit{et al.} (1984b), suggested also that the use of Pu as a homologue for Am might underestimate the mobility of Am in the soil. These present investigations on fallout actinides in a forest soil cannot support those predictions in any soil horizon. If differences are present between the rates of Pu and Am, they are obviously rather small. This is also in agreement with the investigation on the migration rates of fallout actinides in soil under spruce, where a slightly enhanced mobility of Am with respect to Pu was observed only in the Oh horizon.

**Radiocaesium**

While the migration rates of radiocaesium from the global fallout and from the Chernobyl fallout in the organic soil horizons are similar and rather low, they differ significantly in the mineral horizons (see Fig. 3). In each of the mineral layers Chernobyl-derived radiocaesium is more mobile by a factor of about two. This behavior has been observed before for other soils (Bunzl \textit{et al.}, 1989b, 1992). It indicates that the fixation of this radionuclide by the clay minerals in the soil is a rather slow process which obviously extends over many years. Recent speciation studies also revealed that Chernobyl-derived Cs is comparatively labile-bound to the soil (Livens & Baxter, 1988). In the layers below 9 cm the activity concentration of Chernobyl derived \textsuperscript{137}Cs is presently too low to yield useful migration rates.

Compared to the pine stand we found in the organic horizons of a podzolic parabrown earth soil of a spruce stand (Bunzl \textit{et al.}, 1992), similar migration rates for \textsuperscript{137}Cs from global fallout, whereas Chernobyl-derived radiocaesium was faster by a factor of 3–6. As a consequence, 41% of the total Chernobyl-derived \textsuperscript{137}Cs activity was found in 1990 in the mineral soil of the spruce stand, compared to only 25% in the pine stand in 1991.

To check whether different sorption properties of the organic matter for \textsuperscript{137}Cs are the reason for this behavior, for the distribution coefficients \(K_d\)
for this radionuclide were determined in the organic layers of both soils (Bunzl & Schimmack, 1989). The resulting values were, however, rather similar, about 30 cm$^3$ g$^{-1}$ in the Of-horizons and about 100 cm$^3$ g$^{-1}$ in the Ofh-(resp. Oh) horizon. The comparatively long residence time of Chernobyl-derived $^{137}$Cs in the Ofh-horizon of the soil from the pine stand must, therefore, be the result of other mechanisms.

Recently, it has been shown (Olsen et al., 1990; Clint et al., 1991; Bürgmann et al., 1994) that the microflora, especially the fungal biomass, strongly contributes to the temporary immobilization of $^{137}$Cs in forest soils. In addition, it has been demonstrated by Brückmann and Wolters (1994) that radiocaesium can also be actively transported upwards into the litter layer above the roots of the trees by the microflora (recycling). Both immobilization and recycling are determined by three major factors: $^{137}$Cs availability, growth conditions of the microflora and biotic interactions. In addition, a short-cut recirculation of the nutrients, especially of K and Ca, via roots—needles—litter—roots will also retain the nutrients and radiocaesium in the organic soil layers (Ronneau et al., 1991). The uptake of the nutrient ions and radiocaesium by the roots is, of course, tightly connected with the presence of the mycorrhizal fungi, which are symbionts with plant roots and which supply them with nutrients but also with radiocaesium.

The longer retardation of Chernobyl-derived $^{137}$Cs in the organic layers of the pine stand as compared to the spruce stand thus indicates that the above processes are more effective in the pine stand. A possible reason might be the fact that in the soil of the pine stand less nutrients are available for plant uptake. This is evident, e.g. from the different amounts of exchangeable K$^+$ in these soils in the upper 30 cm layer, where for the spruce stand twice the amount present in the pine stand was observed. Of that, only 12% was in the organic layers under the pine as compared to 35% under the spruce. Obviously, in order not to loose any nutrients by leaching, the nutrient cycling processes in the surface soil of the pine stand, where also a large amount of roots is present, are very efficient.

**CONCLUSIONS**

The vertical migration rates of Pu and Am from the global fallout in the podzol soil of a pine stand are in all horizons similar and low (in general < 1 cm year$^{-1}$). The smallest mobility (0.1 cm year$^{-1}$) was observed in the Ofh horizon, i.e. the horizon where the pine needles are mostly decomposed and humic substances have been formed. In the underlying mineral horizon the migration rates increase in general with increasing depth.
Radiocaesium from the global fallout exhibits rather similar migration rates as the two actinides, except in the Ofo horizon, where it is more mobile by a factor of about five. Chernobyl derived $^{137}$Cs, however, is presently significantly more mobile in the mineral soil than radiocaesium from the global fallout by about a factor of two.

The migration rates as evaluated above yield no information about the mechanism of the transport process of a radionuclide in the soil. Even though the migration of reactive solutes in the soil is frequently due to chromatographic leaching processes of the sorbed species, this may not be the predominant transport mechanism in the present case. The very long residence time of radiocaesium in the organic soil layers is probably due to temporary immobilization and recycling processes originating from the presence of the soil microflora, especially mycorrhizal fungi. Bioturbation by edaphic invertebrates will also promote recycling of radiocaesium. Even though Pu and Am exhibit similar long residence times in the organic layers as radiocaesium, their very slow migration rates are probably the result of complexation reactions with soil substances. The similar vertical distribution of iron and the actinides in the organic horizons suggests these elements exhibit an analogous tendency for complex formation with soil organic matter.

DEDICATION

This work is dedicated to Professor J. Klein on his 60th birthday.

REFERENCES


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