Influence of the nature of clay minerals on the fixation of radiocaesium traces in an acid brown earth–podzol weathering sequence

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Summary

The magnitude of radiocaesium fixation by micaceous clay minerals is affected by their transformation, which depends on weathering in soil. The net retention of radiocaesium traces was quantified by sorption–desorption experiments in the various horizons of four sandy soils forming an acid brown earth–podzol weathering sequence derived from sandy sediments and characterized by marked changes in mineral composition. The features of the 2:1 minerals of the four soils, resulting from an aluminization process in depth and a desaluminization process towards the surface, had a strong influence on Cs⁺ fixation. Beneath the desaluminization front, which deepens from the acid brown earth to the podzol, hydroxy interlayered vermiculite was dominant and the ¹³⁷Cs⁺ fixation was the weakest. At the desaluminization front depth, vermiculite was responsible for the strongest ¹³⁷Cs⁺ fixation. In the upper layers, smectite appeared in the podzolized soils and the ¹³⁷Cs⁺ fixation decreased. The magnitude in Cs⁺ fixation therefore appeared as a tracer of the transformation process affecting the 2:1 clay minerals in the acid brown earth–podzol weathering sequence. This magnitude was positively correlated with the vermiculite content of the studied soil materials estimated by the rubidium saturation method.

Introduction

Soils retain radiocaesium because they contain a few highly selective sites associated with the presence of micaceous minerals (Sawhney, 1972; Eberl, 1980; Cremers et al., 1988). Micas are common in some soils derived from sediments and weathered rocks, particularly of the sialic type. In soils where the transformation of micas is active these minerals undergo changes in their structure and properties. The K⁺ depletion, gain in hydrated exchangeable cations and oxidation of structural Fe²⁺ cause the transformation of biotite into vermiculite and smectite (Fanning et al., 1989) in which Al interlayering may occur (Barnhisel & Bertsch, 1989). The retention of ¹³⁷Cs⁺ traces is strongly influenced by these changes, as reported by Maes et al. (1999) in a laboratory weathering sequence biotite → vermiculite → oxidized vermiculite → hydroxy interlayered vermiculite (HIV). These authors found that the interlayer occupancy by potassium in biotite and hydroxy-Al groups in HIV strongly limited Cs⁺ fixation. Vermiculite fixed large amounts of radiocaesium, but the oxidized vermiculite retained much more radiocaesium in acid conditions, because of its dioctahedral character or a greater resistance to acid weathering or both. It was concluded that Cs⁺ fixation in the studied weathering sequence occurred on vermiculitic sites associated with micaceous wedge zones.

Such observations suggest that weathering in soil strongly influences the fixation of Cs⁺. Large differences may be expected not only in the different profiles of a soil weathering sequence but also in the various horizons at each site. In acid forest soils strong mineralogical variations are characteristic. They are related both to horizon differentiation and to the distribution of plant roots, which are dynamic weathering agents in soils (Courcestes & Gobran, 1997).

This paper describes the ¹³⁷Cs⁺ retention properties of the A and B horizons in a soil weathering sequence acid brown earth–podzol developed in sandy sediments. The soil sequence is represented by four distinct profiles in which vermiculite, smectite and hydroxy interlayered 2:1 clay minerals are derived from the weathering of mica and chlorite. The ¹³⁷Cs⁺ retention properties are assessed in relation to the mineralogical variations and the quantification of vermiculitic minerals.
Materials and methods

The acid brown earth–podzol weathering sequence

Four soil profiles studied earlier by Herbauts (1982) were sampled on the Lower Lias outcrop in southeast Belgium. The altitude ranges between 320 and 335 m, the annual rainfall amounts to 1100 mm, and the mean annual temperature is 7.7°C. All the soils were sampled in deciduous forest with *Fagus sylvatica*, *Quercus petraea* and *Quercus robur* as dominant species.

The bedrock (calcareous sandstones of Lower Lias age) is covered by a two-layered sheet: an autochthonous sandy layer formed by the dissolution of the calcareous bedrock is overlain by a mixture of this sandy material with loessic silt-sized particles. The soil sequence developed in the upper material is as follows: acid brown earth → ochreous brown earth → brown podzolic soil → podzol, i.e. Dystric Cambisol → Haplic Podzol according to the World Reference Base for Soil Resources (WRB) classification (FAO, 1998). Following a gradient of increasing podzolization, this sequence of soil coincides with the evolution of the humus from mull to moder-mor. This evolution coincides with the morphological development of the profile from Ah-Bw to Ah-Bh and eventually Ah-E-Bh. The increasing podzolization is well illustrated by (i) selective chemical extraction of Fe and Al and (ii) the mineralogical composition of the clay fraction (<2 μm) (Herbauts, 1982).

The general evolution of the clay minerals in the sequence is illustrated in Figure 1 (Herbauts, 1982). In the moderately acid B horizons, Al interlayering affects the 2:1 clay minerals produced by the weathering of mica and chlorite: 2:1–2:2 hydroxy-Al intergrades are the dominant clay minerals. In the more acidic surface horizons, aluminium is complexed by organic compounds and removal of Al interlayers occurs. The latter process has been called ‘dechloritization’ by Frink (1969), ‘de-aluminization’ by Vicente et al. (1977) and ‘desaluminization’ by Herbauts (1982). In the surface horizons, desaluminization occurs with increasing intensity towards the more weathered soils, resulting in the dominance of smectite-like minerals. These smectite-like minerals present a 1.7–1.8-nm reflection in their X-ray diffraction after saturation with Mg²⁺ and ethylene-glycol solvation, but collapse to 1.0 nm when heated (200°C) after saturation with K⁺. These minerals are identified as degradation smectites derived from micaceous minerals. From the ochreous brown earth to the podzol, the deepening of the desaluminization front is therefore associated with the advent of smectitic clays. Whatever the soil weathering stage, the sand and silt fractions have homogeneous mineralogical compositions dominated by quartz (Herbauts, 1982).

Soil characterization

The following horizons were sampled in the four soil profiles of the sequence: the Ah and B horizons of the acid brown earth, the Ah, AB and Bw1 horizons of the ochreous brown earth, the Ah, Bh, Bw1 and Bw2 horizons of the brown podzolic soil, and the Ah, E, Bhs, Bs and C horizons of the podzol. The samples from mineral horizons were air-dried and sieved to <2 mm. The samples from organic-rich Ah horizons were sieved to <2 mm without drying and kept in closed bags at 4°C to avoid their becoming hydrophobic. The
particle size analysis was done after dispersion using an ultrasonic probe (50 W, 15 min) and Na⁺ resins as a dispersing agent (Rouiller et al., 1972). Total carbon (Walkley-Black), exchangeable bases and cation exchange capacity (CEC) (1 M NH₄OAc at pH7), and KCl-extractable Al and H were determined using the procedures outlined by Page et al. (1982, pp. 570, 160, 163, respectively); pH was measured in both H₂O and 1 M KCl (10 g:25 ml). Elemental analyses were made in HF–H₂SO₄ digests of calcined <2 mm soil samples (Page et al., 1982, p. 7) by atomic absorption spectrophotometry.

Radioceasium sorption–desorption experiments

Soil samples (air-dried in the case of mineral horizons, sieved to pass 2 mm) were equilibrated with a mixed KCl–CaCl₂ solution with a total chloride concentration of 10⁻³ M, a potassium adsorption ratio (PAR) value of 0.057 mmol kg⁻¹ (PAR is defined as (K)/Ö(Ca) where () refers to ion concentration in (mm) and a K:Ca molar ratio of 0.082. Dialysis bags containing 1 g of soil and 5 ml of the mixed KCl–CaCl₂ solution were placed in 95 ml of ¹³⁷CsCl-labelled KCl–CaCl₂ solution (carrier free ¹³⁷Cs, ¹³⁷Cs⁺ concentration 1 × 10⁻¹⁰ M representing a γ activity of 44 kBq dm⁻³). The extent of the ¹³⁷Cs⁺ sorption, which continued for 7 days, was measured by γ counting of the solution phase using a NaI scintillator γ counter (Auto-gamma 5000 series, Canberra) for 5 min. At the end of the sorption the relative quantity of sorbed radioceasium, the distribution coefficient Kₐs characterizes the sorption properties of the soil. The distribution coefficient is defined by the following equation: Kₐs = Xcₛ/[Cs], where Xcₛ is the ¹³⁷Cs γ activity on the adsorbed phase in Bq kg⁻¹ and [Cs] is the ¹³⁷Cs γ activity in the liquid phase in Bq dm⁻³.

Desorption was done using a method adapted from Wauters et al. (1994) in which 100 ml of the mixed KCl–CaCl₂ solution with 5.5 g of K⁺–Ca²⁺-saturated resin and 1 g of soil was shaken end-over-end. Both sorption and desorption were carried out in an identical K⁺–Ca²⁺ electrolyte background because both the soil materials and the desorbing resin had previously been equilibrated with the same mixed KCl–CaCl₂ solution. The desorption was monitored for 25 days: both the solution and the resin were replaced after 1, 2, 4, 7, 11 and 18 days, and counted for their γ activity. The ¹³⁷Cs⁺ net retention was evaluated by the relative quantity of radioceasium retained at the end of the desorption process. This relative quantity is expressed as a percentage of the initial ¹³⁷Cs⁺ input.

Quantification of the vermiculitic sites

A rubidium fixation method adapted from Ross et al. (1989) was used to quantify the vermiculitic sites of the soil materials. In a 15-ml centrifuge tube, 1 g of soil (<2 mm) was washed three times with 10 ml of 0.5 M RbCl and once with 80% methanol. The sample was heated at 110°C overnight, cooled in a desiccator and then washed four times with 10 ml of 0.5 M NH₄Cl. Supernatants were discarded. The sample was then digested in HF–HNO₃–HClO₄ (Lim & Jackson, 1982). The Rb content was determined by atomic absorption spectrophotometry. The vermiculite content in g kg⁻¹ was evaluated by Rb content (cmolkg⁻¹)/0.154; the evaluation is based on a CEC of the vermiculite of 154 cmolkg⁻¹ (Ross et al., 1989).

Results and discussion

Soil characteristics

Table 1 presents major characteristics of each horizon of the four soils. All the horizons are acidic and depleted of exchangeable bases, with base saturation less than 5% except in the case of the Ah horizon in the acid brown earth (16%). Aluminium extractable in KCl represents 30–60% of the effective cation exchange capacity (ECEC) in the organic-rich horizons and 65–94% in the mineral horizons.

The depth distribution of organic carbon reveals a regular decrease in the profile of the acid brown soil (59), the ochreous brown soil (57) and the brown podzolic soil (29). In the last profile (79), the organic carbon decreases sharply in the E horizon and increases in the Bh₅, which is typical of a well-developed podzol.

Sand is the dominant (73–96%) particle size fraction in all the horizons. This reflects the siliceous character of the soil parent material. The clay content is very small (0.7–9.2%) and decreases from the acid brown soil to the podzol.

The total reserve in bases (TRB) sums to the total content of basic cations and is an estimator of the content of weatherable minerals (Herbillon, 1988). The total reserves are invariably small in all the soil horizons (<23 cmolkg⁻¹). They markedly decrease, however, from the acid brown earth to the podzol. Potassium is the dominant cation in the TRB (37–53%) because of the presence of K-bearing primary minerals such feldspars and micas (Herbauts, 1982). The increase in the proportion of K in the TRB from the acid brown soil (37–40%) to the podzol (48–53%) illustrates the greater resistance of the K-bearing minerals to acid weathering. The distribution of both clay content and TRB suggests increased weathering of primary silicate minerals from the acid brown earth to the podzol.

Cs sorption–desorption data

The ¹³⁷Cs⁺ sorption–desorption data for each soil horizon are presented in Figure 2. Except for the Ah and the E horizon in the podzol, the sorption was fast and large in all cases, exceeding 95% of the initial ¹³⁷Cs⁺ input after 7 days. The expression of the sorption by the Kₐs gives more pronounced differences between the various horizons,
Table 1 Major properties of the four soils forming an acid brown earth–podzol weathering sequence developed in sandy material (59, acid brown earth; 57, ochreous brown earth; 29, brown podzolic soil; 79, podzol)

<table>
<thead>
<tr>
<th>Horizon</th>
<th>Depth /cm</th>
<th>pH</th>
<th>H₂O</th>
<th>KCl</th>
<th>Organic C /g kg⁻¹</th>
<th>Exchange complex</th>
<th>KCl-extractable Al /cmol kg⁻¹</th>
<th>H /cmol kg⁻¹</th>
<th>TRB b /cmol kg⁻¹</th>
<th>Total K /cmol kg⁻¹</th>
<th>Sand %</th>
<th>Silt %</th>
<th>Clay %</th>
</tr>
</thead>
<tbody>
<tr>
<td>59 Ah</td>
<td>0–10</td>
<td>4.2</td>
<td>3.7</td>
<td>25.7</td>
<td>2.35</td>
<td>14.87</td>
<td>1.33</td>
<td>0.64</td>
<td>19.25</td>
<td>7.22</td>
<td>82.0</td>
<td>9.5</td>
<td>8.5</td>
</tr>
<tr>
<td>59 B</td>
<td>10–20</td>
<td>5.0</td>
<td>4.3</td>
<td>4.9</td>
<td>0.24</td>
<td>4.78</td>
<td>1.29</td>
<td>0.16</td>
<td>20.14</td>
<td>8.07</td>
<td>80.7</td>
<td>10.1</td>
<td>9.2</td>
</tr>
<tr>
<td>57 Ah</td>
<td>0–5</td>
<td>4.0</td>
<td>3.3</td>
<td>33.7</td>
<td>0.29</td>
<td>11.10</td>
<td>1.48</td>
<td>0.63</td>
<td>22.25</td>
<td>11.04</td>
<td>76.9</td>
<td>17.2</td>
<td>5.9</td>
</tr>
<tr>
<td>57 AB</td>
<td>5–15</td>
<td>4.5</td>
<td>3.5</td>
<td>15.0</td>
<td>0.13</td>
<td>5.27</td>
<td>1.89</td>
<td>0.55</td>
<td>17.54</td>
<td>6.79</td>
<td>76.0</td>
<td>17.6</td>
<td>6.4</td>
</tr>
<tr>
<td>57 Bw1</td>
<td>15–30</td>
<td>4.9</td>
<td>4.6</td>
<td>7.6</td>
<td>0.02</td>
<td>3.54</td>
<td>0.87</td>
<td>0.03</td>
<td>22.33</td>
<td>8.28</td>
<td>73.1</td>
<td>20.9</td>
<td>6.0</td>
</tr>
<tr>
<td>29 Ah</td>
<td>0–4</td>
<td>4.8</td>
<td>3.4</td>
<td>61.4</td>
<td>0.95</td>
<td>23.70</td>
<td>1.43</td>
<td>0.55</td>
<td>12.19</td>
<td>4.88</td>
<td>88.1</td>
<td>7.9</td>
<td>4.0</td>
</tr>
<tr>
<td>29 Bh</td>
<td>4–12</td>
<td>4.3</td>
<td>3.3</td>
<td>21.3</td>
<td>0.10</td>
<td>8.39</td>
<td>1.99</td>
<td>0.98</td>
<td>11.73</td>
<td>4.46</td>
<td>84.9</td>
<td>10.4</td>
<td>4.7</td>
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<td>29 Bw1</td>
<td>12–24</td>
<td>4.9</td>
<td>4.3</td>
<td>11.9</td>
<td>0.08</td>
<td>5.65</td>
<td>1.35</td>
<td>0.59</td>
<td>12.97</td>
<td>4.88</td>
<td>83.7</td>
<td>11.2</td>
<td>5.1</td>
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<td>29 Bw2</td>
<td>24–40</td>
<td>4.9</td>
<td>4.6</td>
<td>4.7</td>
<td>0.02</td>
<td>3.20</td>
<td>0.73</td>
<td>0.03</td>
<td>11.36</td>
<td>4.88</td>
<td>83.5</td>
<td>11.4</td>
<td>5.1</td>
</tr>
<tr>
<td>79 Ah</td>
<td>0–10</td>
<td>4.9</td>
<td>3.3</td>
<td>8.3</td>
<td>0.29</td>
<td>7.84</td>
<td>0.59</td>
<td>0.35</td>
<td>6.50</td>
<td>3.18</td>
<td>95.7</td>
<td>3.1</td>
<td>1.2</td>
</tr>
<tr>
<td>79 E</td>
<td>10–30</td>
<td>5.1</td>
<td>3.9</td>
<td>1.4</td>
<td>0.04</td>
<td>1.18</td>
<td>0.24</td>
<td>0.09</td>
<td>4.86</td>
<td>2.55</td>
<td>93.4</td>
<td>5.9</td>
<td>0.7</td>
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<tr>
<td>79 Bhs</td>
<td>30–45</td>
<td>4.3</td>
<td>3.6</td>
<td>5.7</td>
<td>0.12</td>
<td>5.00</td>
<td>2.17</td>
<td>0.57</td>
<td>8.87</td>
<td>4.25</td>
<td>92.2</td>
<td>4.3</td>
<td>3.5</td>
</tr>
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<td>79 Bs</td>
<td>45–60</td>
<td>4.7</td>
<td>4.2</td>
<td>4.6</td>
<td>0.09</td>
<td>3.90</td>
<td>1.66</td>
<td>0.34</td>
<td>9.55</td>
<td>5.09</td>
<td>93.4</td>
<td>4.1</td>
<td>2.5</td>
</tr>
<tr>
<td>79 C</td>
<td>60–80</td>
<td>4.9</td>
<td>4.7</td>
<td>1.6</td>
<td>0.02</td>
<td>1.58</td>
<td>0.38</td>
<td>0.08</td>
<td>10.44</td>
<td>5.52</td>
<td>92.7</td>
<td>2.6</td>
<td>4.7</td>
</tr>
</tbody>
</table>

a Total exchangeable bases. b Total reserve in bases.
with $K_d$ ranging between 252 and 26216 dm$^3$ kg$^{-1}$. These values, when plotted on a log scale (Figure 3), are correlated with the $^{137}$Cs$^+$ net retention value, which is the relative quantity of radiocaesium retained at the end of the desorption process ($r = 0.70$).

The $^{137}$Cs$^+$ net retention values are plotted in relation to soil horizons in Figure 4. These values range from 14.3 to 96.9% of the initial $^{137}$Cs$^+$ input. The net $^{137}$Cs$^+$ retention value in the Ah horizons decreased gradually from the acid brown soil (80%) to the ochreous brown earth and the brown podzolic soil (73% and 60%, respectively) and then strongly to the podzol (14%). In each profile of the sequence, the maximum net $^{137}$Cs$^+$ retention value is observed at successively greater depth amounting to 80, 97, 91 and 93% in, respectively, the Ah horizon of the acid brown soil, the AB horizon of the ochreous brown soil, the Bh horizon of the brown podzolic soil and the Bhs horizon of the podzol.

Cs fixation and soil characteristics

The decrease of the Cs$^+$ net retention in the Ah horizons from the brown soil to the podzol could be associated with a decrease in clay content (Table 1). This relationship could not be confirmed, however, when all the horizons were taken into account ($r = 0.34$). Similarly, the net $^{137}$Cs$^+$ retention value seemed unrelated to the organic carbon content ($r = 0.25$). Soil organic matter plays no part in specific Cs$^+$ retention (Cremers et al., 1988). Even in highly organic soils, the mineral fraction, however small it may be, is responsible for the Cs$^+$ fixation in soils (Shand et al., 1994; Hird et al., 1995).
Comparison of the depth distribution of the Cs\(^+\) net retention value (Figure 4) with the particle size data (Table 1) also illustrates that clay content alone provides insufficient information for predicting the adsorption by the soil, as previously reported by Livens & Loveland (1988). On the other hand, a comparison of Figures 1 and 4 suggests that the variation of the Cs\(^+\) net retention might be associated with the nature of the clay minerals present in each soil horizon and therefore with the weathering stage characterizing each soil in the sequence.

Cs\(^+\) fixation and soil clay mineralogy

Fixation of Cs by clay minerals has been widely studied. This fixation occurs on a few highly selective sites present at the edge or in the interlayer of micaceous minerals of illitic type and accounting for about 1–2% of their cation exchange capacity (Sawhney, 1972; Francis & Brinkley, 1976; Cremers et al., 1988). In this study we found no significant correlation between total K content and Cs\(^+\) fixation (\(r = 0.28\)). This poor relation could be because (i) total K does not strictly reflect illite content (mica and K feldspar are present) and (ii) Cs\(^+\) fixation is not solely associated with illitic sites.

Various clay minerals were identified in the soil clay fraction of the sequence: kaolinite, illite, hydroxy interlayered vermiculite (HIV), vermiculite and degradation smectite. Their occurrence varies with soil type and depth (Figure 1). Beneath the desaluminization front, which deepens from the acid brown earth to the podzol, HIV minerals are present. Just above this line in each profile vermiculite is present. In the ochreous brown earth and the podzolic soils of the sequence vermiculite progressively gives way to smectite towards the soil surface.

When the depth distributions of the net \(^{137}\)Cs\(^+\) retention value (Figure 4) are compared with the mineralogical evolution in each profile (Figure 1) it is evident that maximum \(^{137}\)Cs\(^+\) net retention coincides with the depth of the desaluminization front and in the horizon in which the clay fraction contains vermiculite without Al interlayering and is devoid of smectitic clay minerals. Weaker Cs\(^+\) retention is associated with the presence of HIV minerals in deeper horizons and with degradation smectites in the surface horizons.

Maes et al. (1998) have already observed weak Cs\(^+\) retention by soil dominated by HIV in an acid brown forest soil derived from siliceous schist. They attributed this poor retention to the blocking effect of the hydroxy-Al interlayers which impede the collapse of vermiculitic layers and therefore Cs\(^+\) fixation in the interlayer position.

The degradation smectites identified in the Ah horizons of the podzolic soils likely derive from the desaluminization of HIV (Herbauts, 1982) as reported by others (Malcolm et al., 1969; Carnicelli et al., 1997). During formation from the parent mineral HIV, these degradation smectites have lost layer charge mainly by loss of Al from the tetrahedral sheet and oxidation of octahedral Fe\(^{3+}\).
Layer charge is a major determinant of Cs⁺ fixation by 2:1 minerals (Cornell, 1993), and so the weaker Cs⁺ fixation capacity in horizons dominated by degradation smectites could arise from the smaller layer charge. However, degradation smectites are reported to have substantial capacity to fix K (Robert, 1973; Badraoui et al., 1987; Borchardt, 1989), particularly after wetting and drying (Badraoui et al., 1987). This capacity has been related to collapse of layers on heating after saturation with K, which has been observed in the Ah horizons of podzolic soils (Herbauts, 1982). The weaker Cs⁺ fixation in the Ah horizons dominated by degradation smectites could therefore be related to the competition between K⁺ and Cs⁺ for specific sorbing sites or to turbostratic disorder of the 2:1 layers (Mamy & Gaultier, 1976; Maes et al., 1985).

**Quantification of vermiculitic layers**

Vermiculite content was quantified using the rubidium methodology adapted from Ross et al. (1989). Table 2 presents both fixed Rb⁴⁺ and computed vermiculite contents as well as the values of the ratio fixed Cs⁺/fixed Rb⁴⁺. The Rb and vermiculite contents range from 0.07 to 1.18 cmolₑ⁻¹ kg⁻¹ and 0.05 to 0.76%, respectively. For each of the four soils, the largest vermiculite content is invariably observed in the horizon below that in which degradation smectite was identified and above that in which HIV is dominant. This observation is supported by X-ray data which indicates that vermiculite is the dominant 2:1 clay mineral in this type of horizon (Figure 1). Comparing vermiculite and clay contents (Tables 1 and 2) reveals that the proportion of vermiculite in the clay fraction ranges between 3 and 19%, assuming that vermiculite content of coarser fractions is negligible. The smallest vermiculite/clay ratio occurs in the horizons where HIV or degradation smectite are dominant.

Figure 5 illustrates the relation between 137Cs⁺ net retention and vermiculite content. This strong linear (r = 0.95) relation suggests that vermiculitic sites dominate radioacesium retention. In acid soils vermiculites have a dioctahedral structure (Douglas, 1989), which probably enhances Cs⁺ fixation as reported by Maes et al. (1999). The relation between Cs⁺ fixation and vermiculite content also confirms that weaker Cs⁺ retention is associated with either degradation smectite or HIV minerals. Moreover, it highlights the fact that the fixation of Cs⁺ traces in soils strongly depends on the nature of the clay mineral rather than on clay content. These results suggest that the specific sites for Cs⁺ fixation in soils are not illitic *sensu stricto*: they are vermiculitic sites, most probably associated with micaceous interlayers.

Table 2 also indicates that the values of the fixed Cs⁺ × 10⁻⁷ /Rb⁴⁺ ratio range between 7.7 and 47.4. Most of the values for this ratio fall within a rather narrow range (7.7–13.8), which suggests that the number of highly specific Cs⁺ sorbing sites, though small, represents a fairly constant proportion of the vermiculitic sites.

**The weathering model**

The maximum value of the 137Cs⁺ net retention deepens with increasing weathering stage. For each profile, this value is associated with the greatest vermiculite content, located at the desaluminization front (Figure 1). In the soil sequence, the fixation of radioacesium traces therefore provides a valuable
the acid brown soil to the podzol. The magnitude of Cs+ fixation is therefore observed in different horizons from the sandy sediments is characterized by a strong mineralogical heterogeneity arising from the transformation of vermiculite. The weathering stage strongly affects the Cs+ fixation properties. The maximum net 137Cs+ retention value occurs at increasing depth with increasing weathering stage and is therefore observed in different horizons from the acid brown soil to the podzol. The magnitude of Cs+ fixation is positively and strongly related to the soil vermiculite content estimated by the rubidium method. Fixation of Cs+ decreases in soil horizons with clay fractions dominated by hydroxy interlayered vermiculite or smectite. In a constant electrolyte background, Cs+ fixation appears as a good tracer of the transformation processes affecting vermiculite in the soil sequence. These results suggest that Cs+ is fixed on vermiculitic sites most probably associated with micaceous interlayers.

Conclusions

The soil weathering sequence acid brown earth → ochreous brown earth → brown podzolic soil → podzol developed from sandy sediments is characterized by a strong mineralogical heterogeneity arising from the transformation of vermiculite. The weathering stage strongly affects the Cs+ fixation properties. The maximum net 137Cs+ retention value occurs at increasing depth with increasing weathering stage and is therefore observed in different horizons from the acid brown soil to the podzol. The magnitude of Cs+ fixation is positively and strongly related to the soil vermiculite content estimated by the rubidium method. Fixation of Cs+ decreases in soil horizons with clay fractions dominated by hydroxy interlayered vermiculite or smectite. In a constant electrolyte background, Cs+ fixation appears as a good tracer of the transformation processes affecting vermiculite in the soil sequence. These results suggest that Cs+ is fixed on vermiculitic sites most probably associated with micaceous interlayers.

References


