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Mechanisms Controlling Radionuclide Mobility in Forest soils

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1. Introduction

After a radioactive pollution of forest ecosystems, hot particles and aerosols are first intercepted by the tree canopy. Rainfall and throughfall transport the contaminants onto the soil surface. Runoff being limited, forest soils act as sinks for the radiopollutants. The fate of radionuclides in soils therefore influences the radioecological hazards for the entire food web through biological uptake. It depends upon the dissolution of the particles and the mobility of the radioactive soluble forms.

Soil processes strongly influence the radionuclide mobility in soils. The underlying mechanisms involve both abiotic and biotic factors. Three major processes are illustrated herebelow: sorption by soils colloids, complexation and/or association with organic matter, biological uptake and transport. A large attention is paid to radiocaesium.

2. Materials

The above-mentioned processes are characterized in some soils belonging to the soil collection illustrated hereafter.

Soil type/horizon	Soil net retention (%) ¹	% organic matter	% clay	% K ² (total)	pH (water)	CEC ³
I <i>Eutric cambisol</i> Ah	82	5.5	46	2.05	6.8	20
Bw	96	1.6	55	2.31	8.1	7
IIa <i>Dystric cambisol</i> OAh	62	30.2	—	1.45	3.7	38
Ah	81	24.3	26	1.68	3.7	32
AB	64	10.7	22	1.99	3.9	19
IIb <i>Podzoluvisol</i> Ah	57	23.5	8.5	0.96	3.9	32
E	86	6.3	13	1.00	4.0	14
III <i>Podzol</i> Ah	44	29.9	8	0.75	4.1	55
E	50	1.6	3	—	4.9	3
IV <i>Histosol</i> H	3	98	0	0.09	3.0	126

¹Percentage of the initial loading. ²Total K content is directly related to the content in micaceous clay minerals.

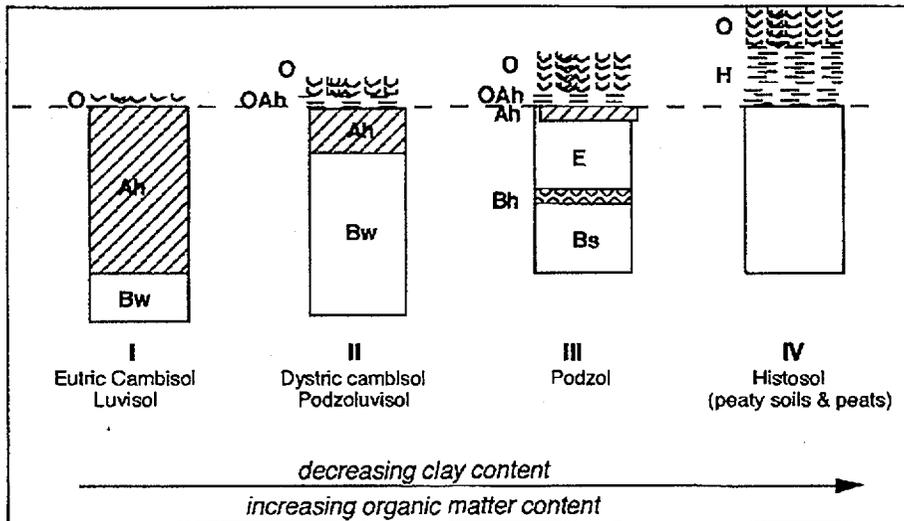
³Cation Exchange Capacity pH7 (cmol(+) / kg).

The figure illustrates a broad morphological schematic view of some major forest soil types observed under northern latitudes 50-60° N. the most distinctive property is the thickness of both the organic (O and H) and hemiorganic (Ah) horizons. Histosols (IV) are characterized by a large accumulation of organic matter (peats). An intense biological activity leads to the rapid decomposition of litter in Eutric Cambisols, Chernozems and Luvisols (I). Thicker O horizons are related to a poorer biological activity in acid soils such as Dystric cambisols, Podzoluvisols (II) and Podzols (III).

3. Results and discussions

Radiocaesium net retention in soils

Soil components adsorb the soluble forms of radionuclides. Under reversible conditions, desorption occurs: radionuclides can be leached or uptaken by biota. If adsorption is irreversible, radionuclides are fixed and become hardly available. Adsorption/desorption phenomena thus strongly influence the fate of radionuclides in soil environments. Sorption-desorption data for radiocaesium were obtained for five soil materials representative of the collection illustrated above (I→IV), using a recent methodology [1].



The selected soils differ in their content in clay, organic matter and micaceous clay minerals. The net retention of radiocaesium strongly discriminates the tested materials: in the Ah and H horizons, it largely decreases from 82 to 3 % (I>IIa>IIb>III>IV), with decreasing clay and total K content and increasing organic matter content.

Radiocaesium net retention is, from a *physico-chemical point of view*, under the full dependance of micaceous clay minerals and ionic conditions prevailing at the soil-solution interface [2,3].

In acid forest soils, major cations such as NH_4^+ and H^+ strongly influence radiocaesium desorption through respectively the trapping effect (interlayer collapse) and K depletion. Despite of its high CEC, organic matter does not directly contribute to irreversibly sorb the radiopollutant, but it may influence the reactivity of mineral surfaces for radiocaesium fixation through Al complexation. In the soil IIa, the net retention is higher in Ah than that in AB: aluminic cations block interlayer specific sites for Cs fixation in the AB horizon, while they are complexed by organic acids in the Ah. Organic acids may also contribute to K depletion, hence thereby Cs desorption.

Contamination hazards to above standing vegetation and other living organisms are therefore expected to be high on peaty soils and forest acid soils with thick organic layers. Greenhouse experiments using *Lolium multiflorum* L. indeed show that radiocaesium bioavailability largely increases with increasing organic matter content and decreasing content in clay and in micaceous clay minerals (IV>>III>IIbIIa>>I).

Association of radionuclides with mobile organic compounds

In some acid forest soils (II and III), the slow decomposition of organic materials accumulating in the upper layers produces soluble organic acids. These acids bear deprotonated groups and thus behave as anions in the soil solution. These anions can be associated with cationic radionuclides through weak electrostatic attraction (Cs), ionic bonding (Sr), or complexation (Ru, Ce, Am, Pu). Such associations have been characterized in the soil solution of the Of, Oh and (Oh + Ah/E) horizons of a heavily contaminated soddy podzolic soil (podzoluvisol: II), situated 6 km west of the nuclear reactor near Chernobyl (Novoshepelitchi, Ukraine).

Gel filtration isolates five "organic" fractions with decreasing molecular weight. $^{239+240}\text{Pu}$, ^{238}Pu and ^{241}Am are mostly associated with the highest molecular weight fractions. ^{90}Sr is concentrated in the lowest one (inorganic). ^{137}Cs is mostly associated with the intermediate fractions in the Of horizon and is more uniformly distributed with all the fractions in the deeper layers [4].

The different distributions of plutonium and americium, but particularly of strontium and caesium between the five fractions obtained by gel filtration of the soil solution from the three horizons clearly show that:

- the nature of soil organic matter present in the three horizons is different;
- these differences largely affect the association between organics and radionuclides; radiocaesium is most sensitive while plutonium is complexed very strongly.

In the soil solution of acid forest podzolic soils, organic compounds of various molecular weights are thus able to associate with cationic radionuclides. Such associations should undoubtedly have effects on their mobility and biological availability. In addition, the extent of this association is different for soil solutions sampled from distinct horizons: similar Kd values observed for a given radionuclide in various horizons do not necessarily imply a similar radioecological behaviour of this radiopollutant.

Radiocaesium biological uptake and upwards transport

Nutrient cycling is essential in forests ecosystems. An important process is the decomposition of contaminated litter material: radionuclides incorporated in dead plant remains are mobilized by other organism living in the surface organic horizons.

The decomposition of contaminated litter material in sandy podzolic soils (type II in the figure) had been studied *in situ* in two sites: in the Ukraine at Kopachi, a 60 years old stand of *Pinus sylvestris* situated 6 km south east of the Chernobyl NPP, in Waterford, Ireland, a 35 year old stand of *Pinus contorta*.

Litter material from O1 and Of organic horizons was used to fill nylon mesh litter bags that were further installed in their respective soil layers. Changes in weigh and radiocaesium content of the bags were recorded during one year.

The soil profile analysis reveals that the radiocaesium pool of the Of horizon is 35 (Ireland) and 70 (Ukraine) times greater than that of the O1. However, large increases in radiocaesium content are observed in the litter bags introduced in the O1.

The net importation of the radiopollutant in the litter O1 is not due to infiltration by canopy washings and litter leachate. A proposed explanation is that the decomposer fungi, which invade the O1 litter from the more contaminated Of, introduce radiocaesium to the litter: some fungi are indeed know as radiocaesium accumulators. Similar observations have been carried out under laboratory conditions [5].

By this mechanism radiocaesium is constantly transported upwards to the fresh litter layers: transport may partially explain the low downward migration of radiocaesium in the organic horizons of the forest soils.

Conclusions

The mobility of radionuclides in forest soils is governed by several processes involving both abiotic and biotic factors.

The sorption-desorption process chiefly governs the activity of radionuclides in the soil solution, hence thereby their mobility and biological availability. Radiocaesium exhibits a very low mobility in mineral soils. Both mobility and bioavailability however increase as the thickness of organic layers and their content in organic matter increases. Clay minerals of micaceous origin strongly act as sinks for radiocaesium in forest soils. The magnitude of Cs mineral fixation in topsoils is expected to be the highest in mineral soils of type I, and, to a lesser extent, of type II.

A low mobility of radiocaesium in the surface horizons of forest soils may also be partially explained by a biological mobilization: fungi absorb radiocaesium and transport it to upper layers, thereby contributing to constantly recycle the radioelement in the organic horizons. This mechanism is probably important in soils with thick organic layers (III, IV, and, to a lesser extent, II).

Radionuclides can be associated with soluble organic anions in the soil solution of forest acid soils. Such associations are highly mobile: they are stable in conditions of poor biological activity (low temperatures, acid soil infertility, water excess, etc.). Their magnitude is expected to be the highest in thick acid organic layers (soils of type III and IV).

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