Migration of radionuclides in undisturbed soil profiles

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Abstract
The vertical distribution of ^{137}Cs and natural radionuclides in undisturbed soil profiles in agricultural, bog and forest areas was studied. Samples were collected at four sites in central Sweden: Skogsvallen (permanent pasture land on a silty clay soil), peatland (raised bog, and low pine) and surrounding forest. The soils samples were collected in 1987, 2005 and 2007 and slides in 1 cm thickness. At Skogsvallen site the relative ^{137}Cs activity in the 0-5 cm layer decreased from 98 to 57 % between 1987 and 2007. The migration rate of ^{137}Cs in 1987 and 2007 was estimated as 0.7 and 0.4 cm y^{-1} respectively. The activity concentrations of ^{40}K and ^{239}Pu/^{240}Pu almost constant within the soil profile. The highest activity concentration was found for ^{40}K (982-1080 Bq kg^{-1}) and lowest for ^{239}Pu/^{240}Pu (0.02-0.48 Bq kg^{-1}). In the open bog peat profile peak of ^{137}Cs activity was found in the uppermost 1–4 cm of Sphagnum layers, whereas at the low pine site ^{137}Cs was mainly found in deeper (10–12 cm) layers. The migration rate was 0.6 cm y^{-1} at the open bog site and the migration centre of ^{137}Cs was at a depth of 10.7, while the rate at the low pine site was 0.8 cm y^{-1} and the migration centre was at 14.9 cm. In the forest peak of ^{137}Cs activity has moved from the surface to about 5 cm and about 50% of the ^{137}Cs found within the depth interval 0-18 cm was retained within the upper 6 cm

KEYWORDS: Sphagnum sp., migration, ^{137}Cs, ^{240}Pu/^{239}Pu, ^{40}K, ^{232}Th, ^{226}Ra

Introduction

After Chernobyl accident large areas of agricultural land, forest and peatland in central and northern Sweden were heavily contaminated with radiocaesium. Knowledge of the dynamics of radiocaesium deposition and downward migration through the soil is important for predicting the radionuclides transfer to man. It is important to know how long radiocaesium will remain within the upper soil horizons where it is readily available for root uptake and transfer to plants. The migration and distribution of radiocaesium in the soil profile varies depending on type of ecosystem, soil properties and land use. In agricultural soils with high amounts of clay, radiocaesium (^{137}Cs) is bound to clay minerals. In forest and peatland soils with a high organic matter content ^{137}Cs can be bound either to organic matter or to soil biomass components (mostly fungal hyphae).

Distribution of radiocaesium in undisturbed agricultural soils have been investigated earlier in several studies, in both semi-natural and natural grassland areas (e.g. Rosén et al., 1999) when less attention was given in forests and peatland (Rosén et al., 2009). In Sweden, the area covered by natural or near natural mires comprises about 4.9 million ha or 11.0% of total land area (Rydin et al. 1999). The surface of a raised bog is a mixture of water pools, raised hummocks dominated by mosses and flatter lawns, and is colonised by plants adapted to low pH and low levels of nutrients. Since rainfall is merely source of water, raised bog is an extremely nutrient-poor ecosystem, only a few vascular species such as bog rosemary (*Andromeda polifolia* L.), sundew (*Drosera* sp.) black crowberry (*Empetrum nigrum* L.), hare's-tail cotton grass (*Eriophorum vaginatum* L.), cranberry (*Vaccinium* sp.,) and heather (*Calluna vulgaris* L.) grow on the open bog.

Sphagnum mosses are common on raised bogs and together the Sphagnum species form one of the most important groups of nonvascular plants found on bogs and other types of wetland. The only trees that are usually found on an open bog site are birch (*Betula* sp.) and Scots pine (*Pinus sylvestris* L.), which height rarely exceed 1 m.

Raised bog ecosystems deserve more research in radioecology, since processes that determine caesium uptake and binding in a raised bog are not well understood. The transfer of ^{137}Cs within a raised bog ecosystem is different from that in forest or on agricultural land. In soils with high clay content, there is low bioavailability and low vertical migration rate of radiocaesium due to its binding to certain clay minerals (Wauters et al. 1996). In nutrient-poor but organic matter-rich forest soils, the vertical migration rate of ^{137}Cs is also low but the bioavailability is often high, particularly for mycorrhizal fungi (Vinichuk & Johanson, 2003; Vinichuk et al. 2004). In forests and pastures, extensive fungal mycelium counteracts the downward transport of ^{137}Cs by an upward translocation flux (Rafferty et al. 2000), which results in very slow net downward transport of ^{137}Cs in the soil profile.

Ombrotrophic bogs lack of fungal mycelium, and the downward migration of ^{137}Cs in the Sphagnum layers is faster than in forest soil, but still rather slow. Thus, as one of the most nutrient poor ecosystems, raised bogs are characterised by a relatively high ^{137}Cs bioavailability to bog vegetation and mosses in particular (Orlov et al. 1999). Recent studies (Schleich et al. 2000) show that in Sphagnum layers, Cs is translocated continuously towards to the growing apex of the Sphagnum shoots, where it is accumulated.

The aim of the present study was to estimate and compare ^{137}Cs and other natural radionuclides migration with time in undisturbed soil profiles of different soils and to study effect of soil type and mineralogy affecting the re-distribution of radionuclides in forest and agricultural land. The studies were carried out during the period 1987–2007.

Materials and methods

There were three study areas: Skogsvallen (permanent pasture land on a silty clay soil), peatland (raised bog, and low pine) and surrounding forest. The Skogsvallen site is located 50 km northwest of Uppsala in the Västmanland county (59°57′33″ N, 17°14′13″ E). The field is located in a narrow valley (about 50-100 m wide) situated between an esker and a moraine hill. The parent material consists of lacustrine silty clay sediments deposited in former stages of the Baltic Sea. The soil has not been ploughed or fertilised since 1960 and is used as permanent pasture for cattle grazing. The vegetation is dominated by grass species, and roots were frequently observed to about 75-cm depth.

Sampling in Skogsvallen was done on 3 circles (A, B and C) which centres are separated by 20 m. Soil sampling for ^{137}Cs was done according to Rosén et al., (1999). At each circle 5 soil samples were taken to depth of 25 cm. The top 10 cm were sampled with a cylinder corer with a diameter of 57 mm and for 10-25 cm samples were taken with a core of 23 mm in diameter. The soil from 1-10 cm depth was sliced into 1 cm layers and from 10 to 25 cm into 2.5 cm layers. Samples taken from the same circle were then put together to form a batch sample for each layer. An aliquot of soil was dried at ca. 40 °C before measurement by gamma spectrometry.

For natural radionuclides each of the circles was divided into 4 sectors. In each sector 3 soil samples (all together 12) were taken. Soil samples were sliced in the same way as for ^{137}Cs. Activity concentrations obtained for each of the studied radionuclides in the sampling site are the arithmetic mean of the measurements corresponding to the three circles.

The small raised bog (Palsjömossen) located within a coniferous forest in Heby commune, about 35 km NW of Uppsala (60°03′04″ N, 17°07′47″ E). The bog can be divided on the basis of the vegetation present into two different parts, open bog and low pine sites. The open bog site has a typical structure of hummocks and hollows and the vegetation consists mostly of Sphagnum species (*S. balticum, S. tenellum, S. fuscum* and *S. rubellum*) and a sparse cover of vascular plants such as rosemary, sundew and cranberry. In the dry part of the open bog site, a few Scots pine trees less than 1.5 m high can be found.

On the low pine site, slow growing Scots pine is rather frequent. The field layer is dominated by crowberry, cloudberry (*Rubus chamaemorus* L.), heather and Labrador tea (*Ledum palustre* L.).
On the forest site vegetation consisted mainly of the conifers Scots pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* L.), with some intermixture of deciduous trees such as birch (*Betula spp*.). In the forest understorey, bilberry (*Vaccinium myrtillus* L.) lingonberry (*Vaccinium vitis-idaea* L) and heather (*Calluna vulgaris* L.) were rather common. The forest floor consisted of a shallow surface layer (3-10 cm) which mainly originated from partially decomposed tree litter and other plant debris which had accumulated on top of the mineral substratum which to a great extent consisted of large stones and, in some locations, of solid rocks.
Four peat soil profiles were sampled at the open bog site, two at the low pine site and one on the forest site in 2004-2005 (Rosén et al., 2009). Samples were collected from the bog by digging with a spade and cutting into 10–10 cm^2 blocks with a saw to a total depth of between 30 and 40 cm. In June 1989, three peat profiles from the open bog site were sampled using a cylindrical steel bore (diameter 57 mm) to a depth of about 20 cm. Sampled material was kept in a refrigerator at a temperature of +4° C before the laboratory analyses. Wet profile samples were sectioned into 1 or 2 cm layers by means of a measuring scale and a sharp knife, placed in paper bags, weighed and dried at 105 °C to constant weight. Dried samples were milled to a grain size of about 2 mm to get a homogeneous and well-mixed material.
Activity concentration (Bq kg^{-1}) of ^{137}Cs and natural radionuclides in layers of soil profiles was determined using calibrated HPGe detectors. All estimates of ^{137}Cs activity concentrations were recalculated to the sampling date and expressed on a dry matter (DM) basis. $^{239}Pu/^{240}Pu$ was measured according to Matisoff et al., (2011). In order to obtain deposition-independent values for comparisons of ^{137}Cs depth distribution in profiles, a relative ^{137}Cs distribution was calculated as:

$$q_i = A_i \div A_{tot}$$

where q_i is the nuclide fraction in the layer of interest, A_i is the activity content (Bq m^{-2}) in the layer and A_{tot} is total activity content (Bq m^{-2}) in the peat profile. The radionuclide migration rate was calculated using the equation (Arapis et al. 1997):

$$\sum (X - X_i) q_i = 0$$

where X_i is the depth (cm) from the surface to the centre of the peat layer in question and q_i is the nuclide fraction in that peat layer. The migration rate is then obtained as X (cm) divided by the time (years) since the deposition took place.

Results

In Skogsvallen soil has not been cultivated or fertilized since the 1960s. The silty clay soil was classified as a Dystric Cambisol in the FAO-system and a Typic Dystrochrept in Soil Taxonomy. Results for ^{137}Cs were obtained in 1987 and 2007 and for natural radionuclides in 2009. Between 1987 and 2007 the relative ^{137}Cs activity in the 0-5 cm layer decreased from 98 to 57 % (Fig. 1). In 2007 peak of the ^{137}Cs activity moved downwards and was mainly located in the upper 0-10 cm layers. The migration rate of ^{137}Cs in 1987 and 2007 was estimated as 0.7 and 0.4 cm y^{-1} respectively. The mean estimate of ^{137}Cs deposition calculated back to 1987 at Skogsvallen was 97 kBq m^{-2}.
The natural radionuclides (^{226}Ra, ^{232}Th, ^{40}K and $^{239}Pu/^{240}Pu$) showed only slightly different distribution within the depth. The values for those radionuclides from the natural series (^{226}Ra and ^{232}Th) somewhat increased with depth. The activity concentrations for ^{40}K and $^{239}Pu/^{240}Pu$ almost constant within the soil profile. The highest activity concentration was found for ^{40}K (982-1080 Bq kg^{-1}) and lowest for $^{239}Pu/^{240}Pu$ (0.02-0.48 Bq kg^{-1}) (Fig. 1).

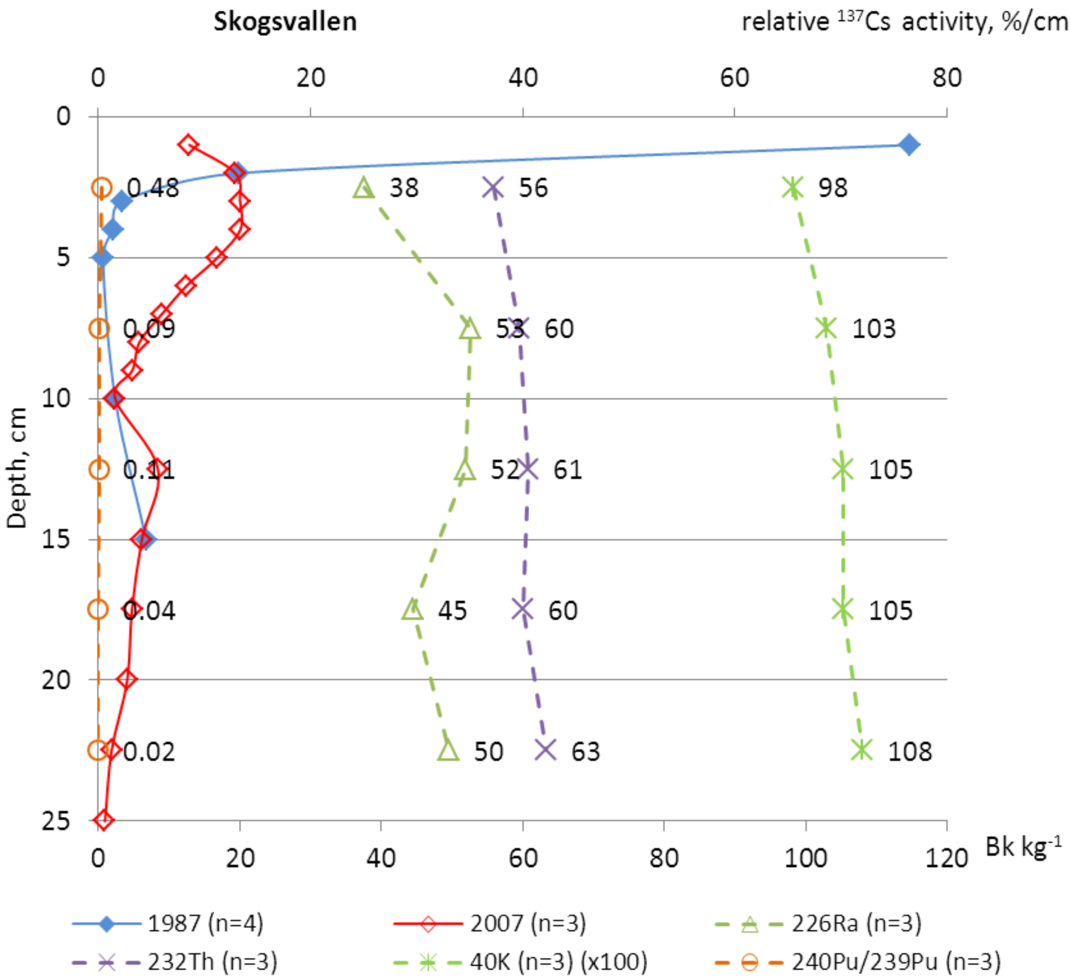

Fig. 1. Vertical distribution of ^{137}Cs, ^{226}Ra, ^{232}Th, ^{240}Pu/^{239}Pu and ^{40}K (x10) deposition on a grass land clay soil, 1987-2009 (solid lines, relative activity %, dotted lines with values, Bq kg^{-1}). ^{40}K values should be multiplied by 100. ^{137}Cs deposition 97 kBq m^{-2}.

In the peat profile on open bog a strong retention of ^{137}Cs in the upper 0–2 and 2–6 cm layers were observed where the respective values were 6000 and 3000 Bq m^{-2}. The ^{137}Cs deposition then dropped to about 1000 Bq m^{-2} and remained more or less constant down to the bottom of the profile (40 cm). The highest ^{137}Cs deposition on the open bog site was 5970 and the lowest 800 Bq m^{-2}.

A similar trend was observed at the low pine site but the distribution of ^{137}Cs within this profile was slightly different (Fig 2). The ^{137}Cs deposition in the upper 0–2 cm layer was 4000 Bq m^{-2} and decreased to about 2000 Bq m^{-2} at the depth between 2 and 10 cm. Below this layer down to a depth of 40 cm there was a slowly decreasing trend, with values less than 1000 Bq m^{-2}. The highest ^{137}Cs deposition at the low pine site was 3820 and the lowest 210 Bq m^{-2}.

About half of the ^{137}Cs activity was located within 0-5 cm depth on the open bog and 0-11 cm depth on the low pine sites (Fig 2).

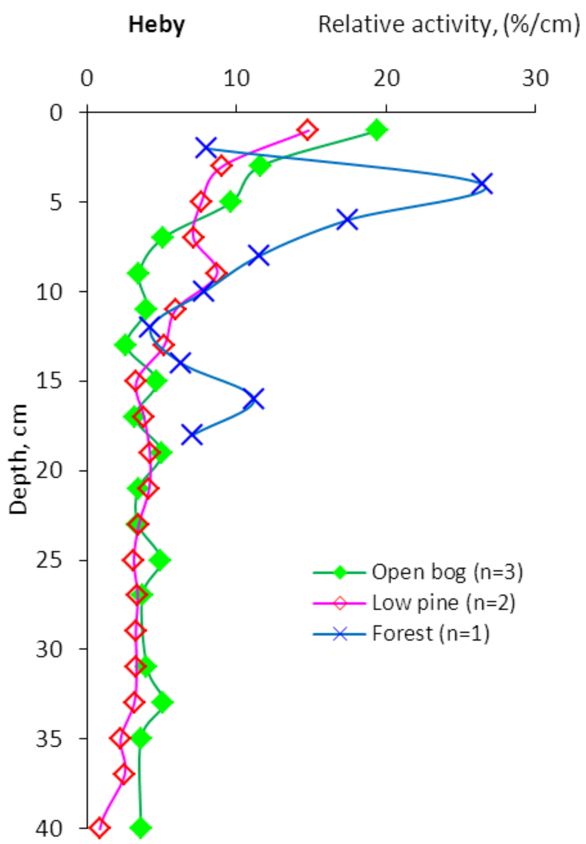

Fig. 2 Vertical distribution of ^{137}Cs deposition (relative activity, %/cm) on the open bog, low pine and forest sites (Heby, 2005). ^{137}Cs deposition 23 kBq m^{-2}.

In the forest peak of ^{137}Cs activity has moved from the surface to about 5 cm and about 50% of the ^{137}Cs found within the depth interval 0-18 cm was retained within the upper 6 cm (Fig 2).
The estimated mean ^{137}Cs deposition in 2005 was about 23 k Bq m^{-2} at both open bog and low pine sites and about 18 k Bq m^{-2} at forest site. The migration centre of ^{137}Cs in 2005 was 10.7, 14.9 and 7.5 cm on the open bog, low pine and forest sites respectively. The ^{137}Cs migration rate was 0.6, 0.8 and 0.4 cm y^{-1} at open bog, low pine and forest sites respectively.

Discussion

In mineral soils clay binds ^{137}Cs rather efficiently, resulting in a very slow vertical migration rate of radionuclides and consequently low uptake in most plant species (Rosén et al. 1999). Since the Skugsvallen site illitic clay minerals are dominant in, the low mean migration rates at this site (silty clay) was as could be expected because of the soil texture.
There are clear differences in distribution of ^{137}Cs between years within soli profile. This also has been shown by others studies (Forsberg et al., 2000). The differences in natural radionuclides distribution in the soil profile seems to be not significant as expected.
In forest soil the vertical migration of radiocaesium in 2005 is also rather slow and similar to that we have in Skugsvallen in 2007, indicating some binding of ^{137}Cs. Such slow migration can be explained by the uptake and accumulation of ^{137}Cs in fungal mycelium. Rather high ^{137}Cs activity seems to be accumulated in fungal mycelium (Vinichuk et al., 2004) and there seems also to be an upward migration of ^{137}Cs in the mycelium.

The open bog is very nutrient-poor compared with forest and agricultural systems. Thus, fungal mycelium as well as clay only marginally will affect the migration rate of ^{137}Cs in the open bog. Nevertheless, the vertical migration of ^{137}Cs in the bog is similar to low pine area. The radionuclide is bound to living material. Most of the ^{137}Cs activity in the peat profiles was found in living moss plants, mainly in the apical few centimetres of the Sphagnum mosses, which has formed since the Chernobyl fallout occurred (Rosén et al., 2009).
Reason for the peak of the ^{137}Cs activity at about 5 cm depth in the forest site is very likely due to the presence of a large amount of fungal mycelium in the organic rich upper soil layers. Fungi are shown to be rather efficient system for retention (Vinichuk & Johanson 2003; Vinichuk et al. 2004) and upward (Rafferty et al. 2000) migration of ^{137}Cs in the upper layer of forest soil profile.

Thus, the dynamics of ^{137}Cs in the soil depend on physico-chemical interactions with inorganic and organic soil constituents, but also on biological interactions in the soil-plant system, including microbial activity.

Conclusions

Migration rates of ^{137}Cs within soil profiles are slow. After 23 yrs about 50% remains in the upper 5 cm, except for an open bog where it was in the top 9 cm. Retention of radionuclides is high, probably due to biological activity (*Sphagnum,* fungal mycelium, earthworms) and soil mineralogy (clay minerals).

Acknowledgements

The studies were funded and supported by Swedish University of Agricultural Sciences (SLU), Swedish Radiation Safety Authority (SSM), Nordic Nuclear Safety Research (NKS) and Universidad de Cantabria and Gobierno de Cantabria.

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