Migration of radiocaesium in six Swedish pasture soils after the Chernobyl accident

A comparison with earlier studies 1987-2005

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Abstract

This study comprises six pasture soils in central and northern Sweden which were strongly affected by the Chernobyl fallout in 1986. The aim of the study was to investigate the vertical migration and plant uptake of radiocaesium as related to soil texture and other soil properties. The results were compared with earlier studies made on the same sites in 1987 to 2005. The soils were sampled down to 60 centimetres depth and analysed in layers. 21 years after the accident the main part of the activity (61-97 %) was still present in the upper ten centimetres of the soil. The migration rate as a whole decreased during the sampling period but varied considerably between the sampling occasions at each location. The mean migration rates were in the range of 0.2-0.5 cm year^{-1}. There is a clear decrease in plant uptake with time, the decrease was however greatest in the first years after fallout. In 2007 the transfer of ^{137}Cs to the vegetation was highest in an organic soil and in a gravely sandy loam in the mountain region and lowest in a clay soil. In this study there was no clear connection between migration and soil properties 21 years after the accident. Instead the differences in migration seam to be a product of biological and hydrological factors at the individual locations.

Keywords: Radiocaesium; ^{137}Cs; Migration; Migration depth; Soil-plant transfer; Chernobyl fallout; Field study; Pasture soils

Sammanfattning

Migration av radiocesium i sex svenska betesmarker efter Tjernobylolyckan - En jämförelse med tidigare studier 1987-2005

Studien omfattar sex jordar som i varierande grad använts som betesmarker sedan Tjernobylolyckan 1986. Jordarna är alla belägna i de områden som drabbades värst av det radioaktiva nedfallet. Syftet med studien var att undersöka den vertikala migrationen och växtupptaget av radiocesium och att sätta dem i förhållande till jordart och andra markegenskaper. Resultatet jämfördes med tidigare studier gjorda på de utvalda platserna under perioden 1987 till 2005. Jordarna provtogs ned till 60 centimeter och analyserades i skikt. 21 år efter olyckan återfanns huvuddelen av aktiviteten (61-97 %) fortfarande i de översta tio centimeterna. Migrationshastigheten minskade generellt under provtagningsperioden. Det fanns dock en betydande variation mellan provtagningstillfällena på de olika platserna. Medelhastigheten i de olika jordarna varierade mellan 0,2 och 0,5 cm år $^{-1}$. Växtupptaget minskade också tydligt under provtagningsperioden, störst var minskningen mellan första och andra provtagningstillfället. År 2007 återfanns den högsta överföringen till vegetationen i en organogen jord och i en näringsfattig jord i ett fjällnära område. Den minsta överföringen återfanns på en lerjord. 21 år efter olyckan går det i den här studien inte att visa på något tydligt samband mellan jordart och migration. Migrationen tycks istället vara beroende av biologiska och hydrologiska faktorer på de enskilda platserna.

Contents

1. Introduction

The accident at the Chernobyl power plant in 26 April 1986 resulted in a vast release of radionuclides to the environment. The effect was not only restricted to the immediate surroundings, large areas of Europe and the former Soviet Union was affected. Parts of central and northern Sweden were heavily contaminated with windborne fission products, deposited as wet fallout.

Numerous radioactive nuclides were released during the course of the Chernobyl accident. The composition of the fallout varied greatly with distance from the reactor and climatic conditions at the time of deposition. Cesium (^{137}Cs, ^{134}Cs), iodide (^{131}I) and strontium (^{90}Sr) were of greatest interest from a Swedish perspective. Most research has been carried out on ^{137}Cs. Because of its relatively long half-life, ^{137}Cs will remain in the environment for decades. ^{137}Cs is also readily taken up by plants thereby entering the food chain.

It is important to understand the dynamics of deposited radiocaesium in order to minimise the degree of external radiation and food chain contamination. It is also important to have knowledge of the vertical distribution in soils to plan effective countermeasures. ^{137}Cs is known to be strongly fixed to specific sites on clay minerals, e.g. interlayer positions of micas. ^{137}Cs also form relatively weak and reversible bonds to organic material. An organic soil can therefore be expected to have a higher migration rate and a higher transfer of ^{137}Cs to plants than a clay rich soil. Information like this is essential in order to point out high risk areas and allocate resources in case of a nuclear accident.

2. Aim

The aim of this study was to:

- Investigate the vertical distribution of radiocaesium in six pasture soils, situated in parts of the country that were strongly affected by the Chernobyl fallout in 1986.
- Calculate the migration depth and migration rate at each site.
- Calculate the transfer factor from soil to plant at each site.
- Compare 2007 years result with earlier studies conducted at the different sites in 1987-2005 and relate them to soil texture and other soil properties.

3. Background

3.1. Basic principles of nuclear power and radioactivity

Nuclear power harvests the energy released from nuclear fission in order to produce electricity. A controlled chain reaction is started by bombarding uranium (^{235}U) with neutrons (Isaksson, 2002). ^{235}U is split in to daughter nuclides which in their turn decay in a chain of successive breakdown towards a stabile element. ^{137}Cs is an intermediate element in one chain of reactions and decays into stabile barium (^{137}Ba). The physical half-life time for this decay is 30 years. In the table below the physical half-life for all radionuclides mentioned in this study are summarised as well as the activity released from the Chernobyl accident given both as absolute activities and percent of the reactor content.

Table 3.1. Physical half-lives for all nuclides mention in this study as well as the activity released from the Chernobyl accident, given as PBq and percent of the total reactor content. Modified from IAEA (2006) and Devell et al. (1996)

	Half-life days (d) years (y)	Activity released (PBq)	Released material in percent of the total reactor content
^{137}Cs	30.0 y	85	33
^{134}Cs	2.06 y	47	33
^{131}I	8.04 d	~1760	50-60
^{239}Pu	24065 y	0.013	3.5
^{90}Sr	29.12 y	10	4-6
^{133}Xe	5.25 d	6500	100
^{235}Ur	$4 * 10^9$ y	No data	No data

Decaying nuclides emit energy either in form of particles or electromagnetic waves (Isaksson, 2002). α-particles are identical to helium-nucleuses, a heavy particle with low penetration easily stopped by a piece of paper. Because of its low ability to penetrate material the α-radiation is concentrated onto a smaller area making it more harmful but easier to protect against. β-particles, i.e. electrons or positrons, have a higher ability to penetrate material but are still stopped by a centimetre layer of wood. The emission of electromagnetic waves or γ-radiation can only be stopped by a thick layer of lead. Decay of ^{137}Cs into stable ^{137}Ba is a two step process resulting in both β- and γ-radiation (Fig 3.1). Becquerel (Bq) is the SI derived unit for measuring radioactivity. One Bq corresponds to one nucleus decay per second.

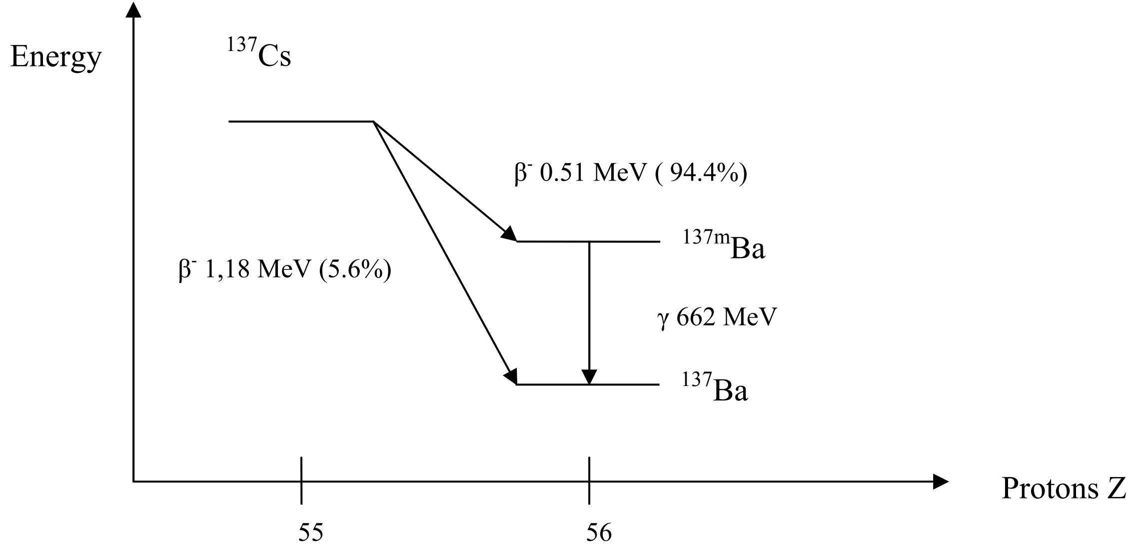

Figure 3.1. Decay scheme for ^{137}Cs. Modified from Isaksson (2002)

3.2. Radioactivity in our immediate environment and food

The mean radiation dose to the non-smoking Swedish population in general is 2.4 millisievert a year (mSv y^{-1}) (Andersson et al., 2007). The two major sources are medical examinations and natural background radiation from the ground and construction material. Naturally occurring nuclides in our food and water make for a mean radiation dose of 0.2 mSv y^{-1}. The dose from caesium is in comparison very low (0.01 mSv y^{-1}), almost exclusively from external radiation.

Relatively large subgroups of the population are however exposed to higher levels of radiation (Andersson et al., 2007). One such group is smokers and former smokers. Smokers are to a higher extent exposed to radon in indoor air. Radon in drinking water also makes an important contribution to the total dose in parts of the country. Frequent flyers are exposed to higher levels of cosmic radiation than the general population. Finally people involved in reindeer husbandry still have an elevated intake of radiocaesium. All these subgroups conduce to a higher mean dose.

Sievert is the SI-derived unit of dose equivalent. It reflects the biological effect of radiation by multiplying the adsorbed dose (J kg^{-1}) with a dimensionless unit that measures the relative biological effectiveness (RBE) of each radiation source.1 mSv corresponds roughly to 80 000 Bq from ^{137}Cs (Jansson & Rydén, 2000). 1 mSv will lead to 1 statistical case of radioactively induced cancer in 20 000 people with a normal age distribution. In case of a nuclear accident the International Commission on Radiological Protection (ICRP) has stated that no person should be exposed to more than a 5 mSv increase in radiation due to the accident, during the first year. During the following years an increase of 1 mSv can be accepted.

After the Chernobyl accident the Swedish authorities used these threshold values to instil threshold limit values in foodstuffs (Åhman, 2006; Johanson, 2006). The limit value for all foodstuffs was set at 300 Bq kg^{-1} in 1986. In 1987 a new limit value for game, reindeer meat, mushrooms, wild berries and fish from freshwater lakes was set at 1500 Bq kg^{-1}. The activity in these foodstuffs was generally much higher than 300 Bq kg^{-1}, making the limit practically useless. Another reason for changing the limit value was that none of these none agricultural food sources are consumed to any great extent, making them a lesser contribution to the total ingested activity. The activity in game, mushrooms and fish from nutrient poor lakes in the worst affected areas are still to this day a cause of concern.

In case of another nuclear accident, the European Union has decided on new threshold limit values which immediately will replace the old ones. The new threshold values, found in Table 3.2 are over all higher than the existing Swedish ones.

Table 3.2. Threshold limit values for foodstuffs set up by the European Union. Modified from Persson & Preuthun (2002)

Nuclides	Example	Baby food (Bq kg^{-1})	Dairy products and liquid foodstuffs (Bq kg^{-1})	Other foodstuffs (Bq kg^{-1})	Less important foodstuffs (Bq kg^{-1})
Alfa-emitting	^{239}Pu	1	20	80	800
Strontium isotopes	^{90}Sr	75	125	750	7500
Iodide isotopes	^{131}I	150	500	2000	20 000
All non natural nuclides with a half-life > 10 d	^{134}Cs ^{137}Cs	400	1000	1250	12 500

3.3. The Chernobyl accident

The Chernobyl accident is by far the largest nuclear accident of all times. Earlier incidents like the Windscale or Harrisburg accidents never resulted in any substantial emissions although they both generated a public opinion against nuclear power. The Chernobyl accident was the disaster everybody feared would happen.

On the night of 26 April 1986 the personnel at the power plant in Chernobyl, Ukraine was investigating how reactor four could be used to supply power to the power plant itself, in case of power failure (UNSCEAR, 2000). In order to do this, the normal security regulations were overruled. The reactor was overheated to a point were the cooling water finally vaporized and exploded. The loss of a cooling system quickly led to vaporization of parts of the fuel and a second explosion followed. This explosion blew the core apart and destroyed most of the building. Approximately 20 hour after the initial explosion the heat of the reactor ignited combustible gases released from the core. As a result a fire started in the graphite rods. The heat of the fire greatly contributed to the spread of fission products.

Over 300 000 people in Ukraine, Belarus and the Russian Federation were evacuated from their homes and some five million people still live in the contaminated areas (Moberg, 2001). A restricted zone with a radius of 30 km around the power plant was created. The restricted zone is completely closed for civilians and is not likely to be opened for a long time. During the first year part of the pine forest within the restricted zone died as a result of the extreme radiation. Decline in the populations of rodents and some species of insects in the restricted area, during the first years, have also been reported. 28 fire-fighters involved in the extinction of the fire in the reactor died within months after the accident due to a lacking immune system. White blood cells involved in the body's defense against alien organisms are being produced in the bone marrow. The rapid mitosis is sensitive to radiation and is the most critical part of the human body (Johansson, 1996). Cancer is the most serious none acute effect. An increase in thyroid cancer in children in the most affected areas has been detected. There is however no significant increase in any other form of cancer.

The radioactive cloud contained numerous different radioactive nuclides. The nuclides were released in different physio-chemical forms (IAEA, 2006). The heat of the explosion and the fire vaporized volatile elements (e.g. ^{137}Cs and ^{131}I). Nuclides with lower volatility (e.g. ^{90}Sr) were also to a great extent vaporized although they later condensed on to soot, dust or heavier fuel particles (e.g. Plutonium, ^{239}Pu). The nuclides with intermediate volatility were carried further of by the wind and the most volatile particles and inert gasses (e.g. Xenon, ^{133}Xe) were spread all over Europe. Heavy fuel particles were deposited in the vicinity of the nuclear plant.

The fire in the reactor continued for ten days. During this time the wind pattern changed a number of times causing distribution of fission products in all directions. The main fallout over Sweden occurred three days after the accident. The fallout was initially dominated by volatile nuclides with short half-lives (Devell, 1991). After just a couple of weeks when the activity of the short lived nuclides had declined, it became evident that ^{134}Cs (2.06 y) and especially ^{137}Cs (30.0 y) were the main nuclides of interest. IAEA estimated the total release of ^{137}Cs to 85 PBq of which about five percent was deposited over Sweden (IAEA, 2006). Caesium is mainly deposited as wet fallout and the deposition was therefore very dependent on the precipitation at the time. The deposition of ^{137}Cs over Sweden ranged from 3 to 200 kBq m^{-2} (Edvarson, 1991) and was concentrated to the central and northern parts of the country (Fig 3.2). A second minor peak in dry deposition occurred on the 8th of May due to core heat up a few days earlier. Only southern and western parts of Sweden were affected.

The nuclear weapon testing in the 1960s resulted in a release of 600 PBq ^{137}Cs to the atmosphere (UNSCEAR, 1982) i.e. a considerably larger activity than the fallout from the Chernobyl accident. The debris from nuclear weapon testing was however distributed over the entire globe. The mean global fallout for central Sweden has been estimated at 2.8 kBq m^{-2}.

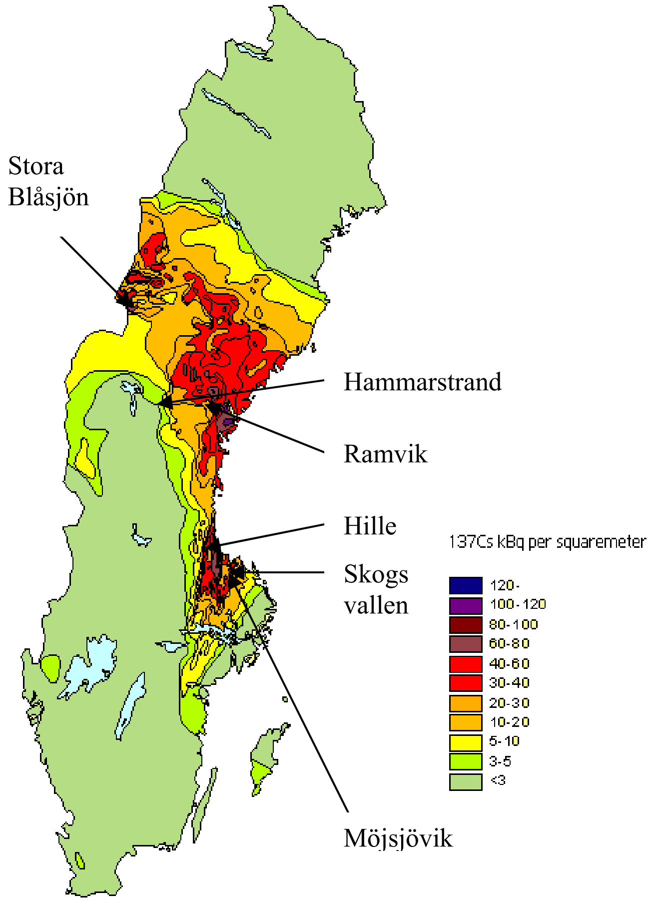

Figure 3.2. The deposition of ^{137}Cs over Sweden after the Chernobyl accident. The map is based on measurements preformed in May-October 1986. The locations for the six investigated sites have been added.
Modified (Sveriges Nationalatlas, 2008).

From a Swedish perspective the most important lesson from the accident was that there are more to nuclear preparedness than to avoid accidents in our national power plants. We also need to consider fallout from a source outside our own borders. There are over hundred reactors located closer to us than the reactor in Chernobyl (Persson & Preuthun, 2002). Many of these reactors are situated in the former Soviet Union were the current economical situation have made nuclear safety less of a priority.

Another important lesson was that authorities need reliable information at the earliest possible state. The Soviet Union did not recognize the accident until two days after, when big parts of Europe already had been subject to deposition (Bengtsson, 2006). If the accident had been recognized earlier on, farmers would have had time to stable their animals. Pregnant women all over Europe would also have benefited from better information. In other European countries there is a definite peek in number of abortions that correspond in time with the accident. Contradicting information and distrust in governmental authorities is a probable reason for this totally unfounded action. The Swedish people also suffered from contradicting information of which much could have been avoided if the authorities had been better prepared.

3.4. Radiocaesium in the environment

3.4.1. Radiocaesium adsorption in soils

Caesium is very strongly bound to clay minerals (Francis & Brinkley, 1976). This can be attributed to the fact that caesium has a high affinity for frayed edge sites (FES) in partly weathered 2:1 phyllosilicates (e.g. illite) (Cremers et al., 1988). Weathering of 2:1 phyllosilicates is a slow process were the layered mineral slowly expands and the potassium trapped in the interlattice is replaced with other positive ions. Cs^+ is a small monovalent ion with low hydration energy and is therefore strongly bound to these sites. The process is shown in Figure 3.2. The fixation increases with time as more caesium moves from the edge of the mineral and penetrates the interlattice; this process is often referred to as ageing (Krouglov et al., 1998). Also in soils where 2:1 phyllosilicates only constitute a small part of the clay fraction their high binding capacity for radiocaesium is of great significance (Cremers et al., 1988). Caesium is also along with other cations attracted to negatively charged sites on the sheet-like clay minerals and oxides in the soil. These attractions are however none specific and readily reversible.

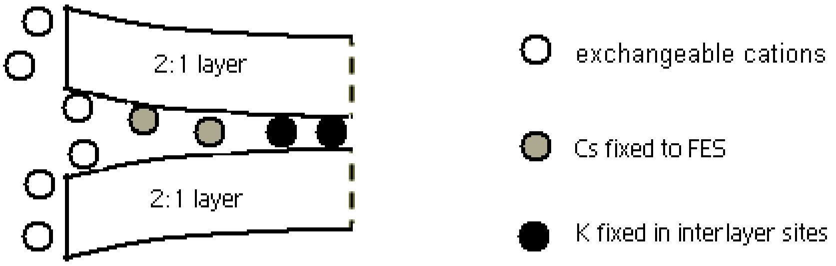

Figure 3.2. Weathering of 2:1 phyllosilicates releases potassium and enables caesium fixation to frayed edge sites (FES) and interlattice binding sites. Modified from Eriksson et al. (2005).

Repeated wetting and drying cycles cause swelling and shrinking behaviour in layered clay minerals. Studies conducted by Rosén et al. (2006) show that this process triggers the penetration of radiocaesium and increases fixation. Wauters et al. (1994) suggest that a high (Ca+ Mg / K) ratio also have a positive influence on fixation. Hydrated calcium and magnesium are too big to replace potassium in the interlattice. They might however contribute to the weathering by binding to negatively charged areas at the very edge of the mineral thereby expanding the gap between the two layers.

In a resent study Giannakopoulou et al. (2007) found that caesium sorption had a maximum at pH 8, in four different mineral soils. The high negative charge of the exchangeable binding sites at this pH enables more caesium ions to be sorbed. Exchangeable binding sites are however less important than the pH-independent FES for the overall retention (Cremers et al., 1988). Wauters et al. (1994) found no direct connection between sorption and pH-level. pH is however related to the cation exchange capacity (CEC) and might therefore indirectly affect the long term sorption behaviour in the soil.

Humic substances and to a lesser extent also negatively charged functional groups in organic material have the ability to form weak and reversible bonds to alkali metals, including caesium (Bonn & Fish, 1993). The CEC of an organic soil increases with increasing pH. This is because the negatively charged binding sites will be protonated, i.e. neutrally charged, and unable to bind cations at low pH-values. The sorption to none specific exchangeable binding sites in organic matter inhibits some of the fixation to mica clays by means of competition (Valcke & Cremers, 1994; Shand & Cheshire 1994). Organic matter also decreases the affinity of caesium for FES, possibly by stabilizing the layered structure counteracting the collapse of the frayed edge (Stauton & Dumat, 2002). The partitioning between stabile and exchangeable binding sites can be investigated by sequential extractions (Tessier et al., 1979). This method, with slight modifications, is widely acknowledged in adsorption studies.

3.4.2. Migration

It is generally agreed that caesium migrates very slowly in most soils. The soil itself act as a shield from radiation, a slow migration therefore results in a slow decrease in external radiation. Arapis et al. (1999) found that the reduction in dose rate due to vertical migration ten years after the accident ranged between 3.5 and 17.5 %, i.e. less than the reduction due to physical decay. A slow migration rate also keeps the activity within the root zone and can thereby contribute to a greater transfer of activity to the vegetation. A faster migration could instead have posed a threat to the groundwater supply. Long-term field studies on migration of radiocaesium originating from Chernobyl conducted by several authors (e.g. Arapis & Karandinos, 2004; Isaksson et al., 2001; Forsberg, 2000; Rosen et al., 1999; Arapis et al., 1997) shows that most of the contamination resides in the upper five to ten centimetres of the soil. The authors also report a higher migration rate in peat, gley and coarser sandy soils.

The strong fixation to FES and interlattice binding sites in mineral clay soils prevent caesium from taking mobile form (i.e. ionic, exchangeable or water-soluble) (Arapis & Karandinos 2004). Organic soils with a small mineral fraction have a higher proportion of mobile water-soluble caesium. The loose structure of many organic soils is another reason for their high migration rate (Kudelsky et al., 1996). In wet gley soils caesium associate to mobile ferrous oxides which have an increasing effect on the migration (Arapis et al., 1997).

The migration rate in soils is known to decrease with time as a result of increased overall sorption and fixation to clay particles. The decrease is greatest during the first years after deposition and will thereafter level out (Smith et al., 1999; Rosén et al., 1999).

Physico-chemical form and fallout conditions also affect the migration. Caesium associated to heavy fuel particles deposited in the vicinity of the nuclear plant are rather insoluble and less mobile than radiocaesium attached to natural aerosols, the main mode of deposition over Sweden (Bunzl & Schimmack, 1995; Krouglov et al., 1998).

There has been no proof of horizontal migration although it probably takes place to a lesser extent. The large variation in deposition ranging over just a few meters makes it hard to prove any horizontal movement in the field (Isaksson & Erlandsson, 1995). Higher activities in low parts of the landscape are likely to be a result of surface run-off at the time of deposition rather than horizontal migration (Arapis & Karandinos, 2004).

3.4.3. Radiocaesium uptake in plants

Plants extract nutrients from the soil, doing so they also extract small amounts of radionuclides. The uptake of caesium is, in the short term, related to the amount of plant available caesium in the soil solution. The plant available caesium is in turn governed by the partitioning between specific and exchangeable binding sites in the soil. Ageing, discussed above, has for this reason a profound impact on plant uptake. The uptake decreases as the fixation increases. This decrease in transfer factors (TFg) with time is demonstrated by Rosén et al. (1999). Parekh et al. (2007) found that microorganisms in the soil have a key role in enhancing the retention by incorporating and recycling the activity, thereby reducing the plant uptake. Fungal mycelium has an equally important role in retention in forest ecosystems (Vinichuk et al., 2004). Water content in the soil is another factor that might affect the plant uptake (Absalom et al., 2001).

Experiments conducted by van Bergeijk et al. (1992) show that pH, within natural limits, have no direct effect on plant uptake on mineral soils. This corresponds with the results of Wang et al. (2003). The indirect effect especially on CEC might however be significant (Absalom et al., 2001).

Caesium and potassium are both alkali metals and are in some aspects chemically similar. For this reason the two elements have analogous plant uptake behaviour (Roca & Vallejo, 1995). A similar relationship exists between strontium and calcium. Potassium is taken up at a higher rate than caesium. The uptake of caesium in potassium rich soil will therefore be limited. In soils permanently or temporarily low in plant available potassium on the other hand, caesium will readily be taken up. Ammonium (NH_4^+) has also been shown to compete with potassium and caesium fore plant uptake (Absalom et al., 2001).

Mineral soils with high clay content have a high proportion of stabile binding sites. Clay soils are also often rich in potassium. We can therefore expect a low plant uptake and a low transfer factor in these soils. Organic soils low in potassium and clay minerals can be expected to have a high uptake and consequently a high transfer factor. Soils in between these two extreme categories can be expected to behave in accordance with their clay content i.e. a gravelly or sandy soil is likely to have a higher uptake than a silty loam soil (Grytsyuk et al., 2006a).

3.5. Countermeasures

When the Swedish government learned about the accident they quickly recommended farmers in the whole country to keep their animals inside (Rosén, 2006). Deposition of especially ^{131}I onto grasslands used for pasture can quickly lead to elevated activity in grazing animals and thus contaminate the milk within days after deposition. In some parts of the country fodder became scares and grazing animals finally had to be allowed to eat the contaminated grass. Milk from such farms was discarded. The government also recommended an immediate harvest of growing green crop in order to remove the deposited activity from the site. The recommended action was however not carried out to any large extent. Instead the farmers harvested the green crop at the normal time of the year, cutting the grass a bit higher in order to avoid the lower part of the straw which was directly exposed to the fallout. This was a generally successful action.

To postpone harvest in order for natural processes like wash of, physical decay and dilution due to biomass production to decrease the activity might be a successful solution (Persson & Preuthun, 2002). It depends on the time of fallout. Another possibility is to harvest the crop and then store it while waiting fore the activity of short-lived nuclides (e.g.^{131}I) to decline.

Removal of the crop followed by immediate planting of a new crop is a countermeasure that focuses on removing the deposition from the site. Another way to remove the activity is to remove the top layer of the soil (Persson & Preuthun, 2002). If the fallout occurs outside the growing season this might be the only way to decontaminate the site. Both of these actions generate large quantities of waste material that needs to be taken care of. For this reason, especially removal of the top soil is not practical in any large scale. Another drawback with this method is that a lot of organic material is removed from the soil. This might have a negative effect on the productivity. A possible scope of use however can be small areas used for vegetable or fruit production. Small areas with high intensity production can also be covered so that the nuclides do not contaminate the soil, given that there is sufficient time fore preparation before the fallout. Run off might however lead to elevated activities at the edges of the cover.

Swedish countermeasures have generally focused on minimizing the root uptake of radionuclides to the crop. Ploughing and potassium fertilization have proven to be vey effective countermeasures. Ploughing homogenize the soil and distribute the nuclides over a much greater depth. This removes much of the nuclides from the top layer where the root activity usually is highest. It also speeds up the fixation to clay particles by increasing the contact area. Potassium fertilization together with ploughing has been shown to minimize the plant uptake of radiocaesium five to ten times (Rosen, 1991). Experiments with deep ploughing, a procedure were the top soil is skimmed and buried beneath the normal ploughing depth, have been conducted with various results (Bréchignac, 2000 mentioned in Forsberg, 2000). The technique is however not practical in other than lighter sandy soils.

Another approach to minimize root uptake is adding strong adsorbents to the soil. By adding five percent by weight of clay to a peat soil the uptake of radiocaesium has been shown to decrease more than ten times (Rosen et al. 2006). Experiments with zeolites have also been conducted, generally resulting in increased fixation (Valcke et al., 1997; Fawaris & Johanson, 1995). Adsorbents are only effective in soils that lack natural adsorbents, i.e. soils with low clay content.

Productive and well managed agricultural soils are easy targets for effective countermeasures. Less productive soils with low pH and high organic matter content are on the other hand not as easy to contend with. Because of their lower productivity these soils are often used for pasture or leygrass production. Pasture grass has generally higher caesium content than leygrass and both types of grass crops are generally higher in activity than cereal grain (Rosén, 1996). This combined with a lower management profile makes for a relatively high transfer of caesium to the vegetation.

4. Materials and Methods

Samples were taken at six locations in four different counties, Uppsala County, Gävleborg County, Västernorrland County and Jämtland County. All sampled fields have to different extent been used for pasture and have therefore not been ploughed since the Chernobyl accident. This makes it possible to study the undisturbed migration. Earlier studies have been conducted at the sites at several occasions 1987 to 2005. The sites were originally chosen to represent different soil types and are all situated in areas that received high deposition during the fallout in 1986. All sites were sampled in August and September 2007, except for Skogsvallen which was sampled in May earlier the same year.

4.1. Sampling techniques

4.1.1. Soil samples

The soil sampling was done in accordance with earlier studies (Rosén et al., 1999; Jansson, 2004), using the same sampling method in order to compare the results. At each site fifteen samples were collected in three circles, with a radius of five meters. The circles were placed in a linear manner with 20 meters between the centre points of the circles (Fig 4.1).

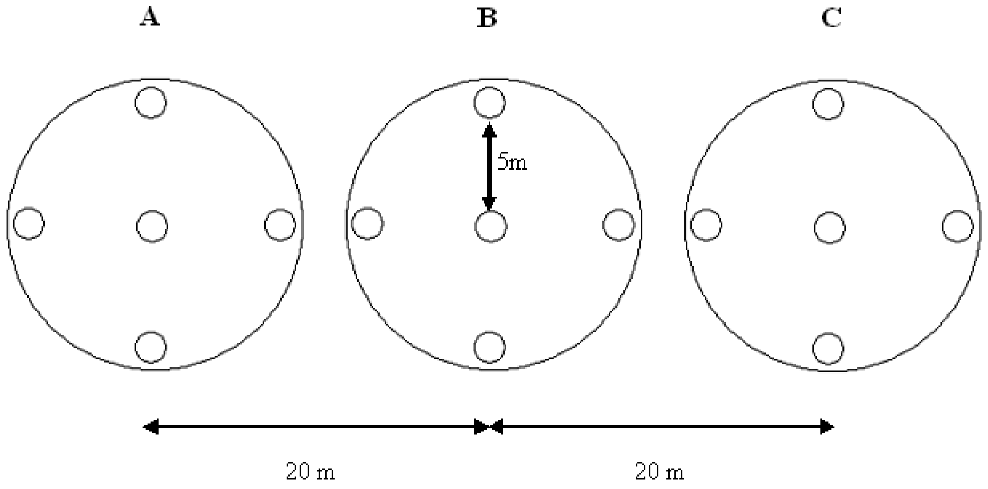

Figure 4.1. Five samples were taken in each circle. Samples taken from the same circle were put together to form a batch sample for each layer. The three circles were placed in a line with 20 meters between the centre points of the circles in accordance with earlier studies.

The soils were sampled down to 60 centimetres depth (Fig 4.2). The top ten centimetres were sampled with a cylinder corer with a diameter of 57 mm. The core were put into plastic bags and handled with care during transport to the laboratory where it was cut in one-centimetre slices. The 10-60 cm samples were sampled with an Ultuna core sampler, 23 mm in diameter. The core was sliced into 2.5 cm layers at the field site.

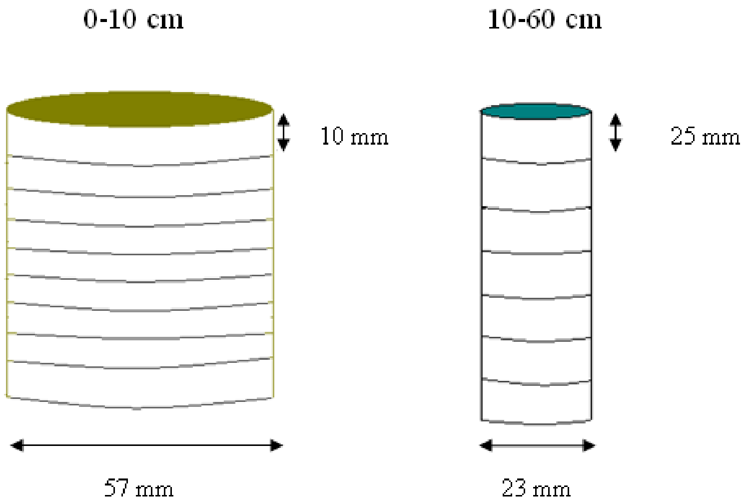

Figure 4.2. Sampling depth and layer thickness for the two sampling procedures.

Samples taken from the same circle were then put together to form a batch sample for each layer. All sites would then have three batch samples for all layers, one from each circle. The samples were dried at a temperature of 30° C for at least a week. Coarse material was taken away using a 2 mm sieve before a representative amount of soil was taken out for analysis.

4.1.2. Grass samples

In each circle five 0.25 m^2 micro plots were placed over the sampling positions for the soil samples. Grasses and herbs were cut five centimetres above the ground in order to avoid contamination by soil particles. Old grass from previous year was not collected. Plant material from the same circle was then put together into a batch sample covering a total area of 1.25 m^2. The samples were dried at 70-80° C for a minimum of 24 h then weighed, milled and homogenised before a representative amount was taken out for analysis.

4.2. Analyses and calculations

4.2.1. Analyses

All samples were put into plastic vials prior to analysis. The samples were then analysed with a High-purity geranium detector housed in a low background laboratory. A geranium detector measures gamma emission and is able to separate the gamma emission from different elements using the specific energy emitted by each element. Less than ten percent of error was considered to be acceptable. No samples were, due to lack of time, analysed for more than 12 hours regardless of percentage of error.

Activity data for all soil samples were corrected for decay back to the time of fallout (1/5-1986). This was done in order to compare results from earlier years regardless of physical breakdown. Since ^{137}Cs originating from global fallout is present only in background concentrations there have been no attempts to distinguish between the two sources. The detector is calibrated for full vials. For this reason two different vial sizes were used, 35 and 60 ml.

4.2.2. Migration depth and migration rate

The migration can be described by calculating the migration depth i.e. the weighted mean value for the activity in the soil X, as described in Arapis et al. (1997). The weighted mean value uses the relative activity in each layer q_i. The relative activity in each layer is described as a fraction between the absolute activity A_i (Bq m^{-2}) and the total activity at the site A_{tot} (Bq m^{-2}).

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

The weighted mean value is calculated using the formula below.

$$\sum_{i=1}^{n}(X - X_i)q_i = 0$$

X_i represents the centre of each layer. The centre X corresponds to the depth where the sum of gravity points of all radionuclide quotas is equal to zero. The migration rate is then calculated by dividing the change in migration depth (cm) with the time (y) since fallout or previous sampling occasion.

4.2.3. Transfer factors

When comparing the uptake of radionuclides in plant material at different sites the deposition level needs to be taken into consideration. The idea of using transfer factors (TFg) is to describe the uptake as related only to soil properties and environmental factors. For this reason the activity in the plant material is divided by the ground deposition instead of comparing the absolute values. Ground deposition is here defined as the total activity at the site. The small part of the activity that might been lost due to gazing or harvest is considered neglect able. The concept of transfer factors has been widely used after the Chernobyl accident. The formula is presented below (Rosén et al., 1999).

$$TF_g = \frac{Activity\ in\ plant\ dry\ matter}{Ground\ deposition}\left[\frac{Bq_{veg}\,kg^{-1}}{Bq_{soil}\,m^{-2}} = \frac{m^2}{kg_{d.w.}}\right]$$

4.2.4. Statistics

The change in migration depth with time at each site as well as the difference in migration depth between the sites in 2007 were analysed with ANOVA (General linear model). Pair-wise comparisons were made with the Tukey method. The change over time in Stora Blåsjön was however analysed with a non parametrical test, Moods median test. This was because the data could not be assumed to be normally distributed. The transfer factors in year 2007 were analysed with an unpaired 2-t test. The migration rates as well as the connection between the transfer factor and the plant available and non-exchangeable potassium concentrations were analysed with regression analysis. The significance level for all test was set at 5 % ($\alpha = 0.05$).

4.3. Site description

The sites are all located in the central and northern part of the country. Climatic conditions vary slightly between the sites. There is a general decrease in annual mean temperature to the north, due to an increase in both latitude and altitude. Precipitation is fairly evenly distributed over the year with a maximum in the end of the summer. Mean temperature, precipitation and altitude are presented in Appendix 1, along with GPS-positions and brief facts on the sampled sites.

The selected soils include two peat soils, two silty loam soils, one silty clay soil and one gravelly sandy loam soil. Complete chemical and physical data for all soils modified from Rosén et al., (1999) are found in Appendix 2. Brief descriptions for all soils are presented below.

Möjsjövik, pasture land on a fen-peat soil: Möjsjövik is located 25 km west of Uppsala in Uppsala County. The top organic horizon is 0.85 meter deep and overlays a sulphide rich gyttja (Rosén et al., 1999). The soil was formed in a lacustrine environment when the area lay below the coast line of that time. The sampled field has been used for pasture, but there had clearly not been any grazing animals at the site this year. The vegetation consists of herbs and broad leaf grasses. The field has been ploughed but only prior to the Chernobyl fallout. The soil was fertilized with 50 kg ha^{-1} potassium in 1987 and 1988, respectively. The soil is classified as a Thionic Histosol in the FAO-system and a Typic Sulfihemist in Soil Taxonomy.

Skogsvallen, permanent pasture land on a silty clay soil: Skogsvallen is located 50 km north west of Uppsala close to the village Östervåla in Uppsala County, although it used to belong to the County of Västmanland. The clay was deposited when the soil was a part of the Baltic basin (Rosén et al., 1999). The sampled field lies in a narrow valley between an esker and a moraine hill. Roots, mainly from various grass species, are present through out the sampled depth. The soil has not been cultivated or fertilized since the 1960s. The soil is classified as a Dystric Cambisol in the FAO-system and a Typic Dystrochrept in Soil Taxonomy.

Hille, temporary grassland on a fen-peat soil: Hille is located 10 km north of Gävle in the County of Gävleborg. The soil consists of a 35 cm thick organic horizon, formed from fen-peat (Rosén et al., 1999). Below the peat lies a layer of sulphide-rich gyttja. Traces of iron sulphide can be found in the gyttja layer. The soil was formed in a lacustrine environment in the former Baltic basin. The field is drained and was before the Chernobyl accident used for crop production, ploughed and fertilized with regularity. The current vegetation consists of various species of grass and small shrubs. The soil is classified as a Thionic (Terric) Histosol in the FAO-system and a Terric Sulfihemist in Soil Taxonomy.

Ramvik, permanent pasture land on a silty loam soil: Ramvik is located 23 km north north-west of Härnösand in the County of Västernorrland. The parent material is alluvial deposits of silt and clay layered in 1-3 cm thick layers (Rosén et al., 1999). The sampled site is situated in a steep (30%) slope. The vegetation consists of various grass species and herbs and was heavily grazed by horses at the time of sampling. The soil is well drained with roots down to one meters depth and has not been ploughed or fertilized since the Chernobyl accident. The soil is classified as Eutric Regosol in the FAO-system and Typic Udorthent in Soil Taxonomy.

Hammarstrand, permanent pasture land on a silty loam soil: Hammarstrand is situated 80 km east of Östersund in the County of Jämtland. The parent material is alluvial deposits originating from when the site was under water and a part of Lake Ragunda (Rosén et al., 1999). Due to a failed attempt to construct a timber flume in 1796, the lake was drained altogether in just four hours. The sampled field is situated on a gentle slope just by the Indals River. The soil has not been cultivated or fertilized during the past thirty years and are currently used for pasture. The vegetation consists of various grass species and herbs. The soil is classified as a Eutric Regosol in the FAO-system and a Typic Udorthent in Soil taxonomy.

Stora Blåsjön, semi natural grassland on a gravelly sandy loam soil: Stora Blåsjön is located 200 km north east of Östersund in the County of Jämtland close to the Norwegian border. The area is quite mountainous and the sampled field is situated in a steep slope at the base of Mesklumpen (924 m) (Rosén et al., 1999). The parent material is colluvial deposits and consists mainly of mica schist. The soil has been ploughed at least once, more than fifty ears ago and has been used for pasture since the 1950s. Various spices of grass and herbs grow on the site. The soil is classified as a Dystric Regosol in the FAO-system and a Typic Cryorthent in Soil Taxonomy.

5. Results

5.1. Deposition and distribution of ^{137}Cs in the soil profiles

5.1.1. Deposition

The deposition was defined as the total activity in the soil corrected for decay back to the time of fallout (1/5-1986). This definition considers the soil, the activity in the grass and the possible loss of activity due to grazing were considered negligible. The highest total activity in year 2007 was found in Hille in Gävleborg County and the lowest in Hammarstrand situated in the County of Jämtland. The total activity for all sites and years of sampling are given in Table 5.1.

Table 5.1. Distribution of ^{137}Cs (kBq m^{-2}) in three layers for all locations and years of sampling. All values were corrected for decay back to the time of fallout (1/5 1986). Values from 1987-1995 taken from Rosén et al. (1999). Values from 2000 taken from Hermansson (2001) except for Hille and values from 2003 taken from Jansson (2004) except for Hille and Möjsjövik

	Möjsjövik					**Skogsvallen**						
Year	**1987**	**1994**	**2000**	**2004**	**2007**	**1987**	**1992**	**1994**	**2000**	**2003**	**2004**	**2007**
0-5 cm	70.9	52.3	48.9[b]	47.2	48.8	89.2	91.4[b]	85.2[b]	75.8[b]	80.5	53.2	64.7
5-25 cm	6.4	11.9	16.1	37.7	25.9	2.1	3.4	7.4	13.9	35.1	25.4	44.8
25-60 cm	—	—	4.9	2.7	3.9	—	—	—	1.1	8.1	2.4	3.4
Total	77.2	64.2	70.0	81.6	78.6	91.3	94.8	92.57	90.8	116.4	90.0	113.0
Mean value[a]					**74.3**							**97.1**

	Hille							**Ramvik**				
Year	**1987**	**1990**	**1994**	**2000**	**2002**	**2005**	**2007**	**1987**	**1994**	**2000**	**2003**	**2007**
0-5cm	153.8	124.9	108.9[b]	101.4	84.8	86.4	80.8	45.3	40.3	35.3[b]	8.0	33.7
5-25 cm	18.1	17.5	99.3	131.5	122.5	160.8	142.2	2.7	16.5	45.9	21.2	55.9
25-60 cm	—	—	—	8.5	3.2	32.0	22.7	—	—	1.0	1.0	1.0
Total	171.8	142.4	208.2	241.4	210.5	279.3	245.7	48.0	56.8	82.1	30.2	90.6
Mean value[a]							**214.2**					**61.5**

	Hammarstrand					**Stora Blåsjön**				
Year	**1989**	**1994**	**2000**	**2003**	**2007**	**1989**	**1995**	**2000**	**2003**	**2007**
0-5 cm	40.1[b]	20.8[b]	44.4[b]	25.0	31.0	39.9	17.9	39.6[b]	28.4	39.1
5-25 cm	5.4	6.6	15.9	12.8	9.7	5.1	3.5	5.4	4.8	6.0
25-60 cm	—	—	2.61	0.3	0.04	—	—	1.9	—	0.7
Total	45.6	27.4	62.8	38.0	40.8	45.1	21.5	46.8	33.2	45.9
Mean value[a]					**42.9**					**38.5**

[a] Mean value for the sampling occasions.

[b] The 0-5 cm layer include the activity in residual organic material that did not pass the 2 mm sieve.

The total activity at each site varied between the different sampling occasions. For this reason a mean value for the whole sampling period was calculated to make a better estimate of the deposition at each location. Just as in year 2007, Hille had the highest mean for the total activity. The lowest estimate of deposition was calculated for Stora Blåsjön, the second location in Jämtland. In Skogsvallen, Hille and Ramvik there seamed to be an increase in the total activity over the sampling period, although this was not statistically verified. Complete activity data for all sites in year 2007 are found in Appendix 3.

5.1.2. Distribution of ^{137}Cs within the soil profiles.

There was a general translocation of radiocaesium from the 0-5 cm layer to the 5-25 cm layer at all locations, with exception for Stora Blåsjön. This trend was however much more pronounced in Hille and Ramvik than at the other sites. All sites had activity below 25 centimetres but there was no clear pattern of translocation between the 5-25 cm layer and the 25-60 cm layer at any site, with a possible exception for Hille. This depth has however only been sampled since 2000. Table 5.2 shows the perctage distribution of the ^{137}Cs-activity in the six different soils.

Table 5.2. Depth distribution of ^{137}Cs, percentage per soil layer, in three layers for all locations and years of sampling. Values from 1987-1995 taken from Rosén et al. (1999). Values from 2000 taken from Hermansson (2001) except for Hille and values from 2003 taken from Jansson (2004) except for Hille and Möjsjövik

	Möjsjövik					**Skogsvallen**						
Year	**1987**	**1994**	**2000**	**2004**	**2007**	**1987**	**1992**	**1994**	**2000**	**2003**	**2004**	**2007**
0-5 cm	91.8	81.3	69.9	57.8	62.0	97.7	96.4	92.0	83.5	69.1	65.7	57.3
5-25 cm	8.2	18.7	23.1	38.9	33.0	2.3	3.6	8.0	15.3	30.2	31.4	39.7
25-60 cm	—	—	7.0	3.3	5.0	—	—	—	1.2	0.7	2.9	3.0

	Hille							**Ramvik**				
Year	**1987**	**1990**	**1994**	**2000**	**2002**	**2005**	**2007**	**1987**	**1994**	**2000**	**2003**	**2007**
0-5cm	89.9	87.6	52.3	42.0	40.3	30.9	32.9	95.7	70.6	43.0	26.6	37.2
5-25 cm	10.1	12.4	47.7	54.5	58.2	57.6	57.9	4.3	29.4	55.8	70.2	61.7
25-60 cm	—	—	—	3.5	1.5	11.5	9.2	—	—	1.2	3.2	1.1

	Hammarstrand					**Stora Blåsjön**				
Year	**1989**	**1994**	**2000**	**2003**	**2007**	**1989**	**1995**	**2000**	**2003**	**2007**
0-5 cm	88.1	75.8	70.6	65.7	76.1	90.5	82.6	84.5	85.4	85.2
5-25 cm	11.9	24.2	25.3	33.6	23.8	9.5	17.4	11.5	14.6	13.2
25-60 cm	—	—	4.1	0.7	0.1	—	—	4.0	—	1.6

Figure 5.1 a-e shows the relative distribution of radiocaesium at the sampled locations for all years of sampling. Figure 5.1 is together with Table 5.1 and Table 5.2 the basis for the following review of the deposition and vertical distribution at the individual sites.

Möjsjövik, pasture land on a fen-peat soil: Between 1987 and 2007 the relative activity in the top five centimetres decreased from 92 to 62 %. The relative activity in the 5-25 cm layer increased during the same period from 8 to 33 %. In 1987 there was a clear activity front within the first centimetre. The activity front has since then moved downward and levelled out slightly. In 2007 the activity decreased sharply around ten centimetres. The activity front in 2004 was actually somewhat deeper and more levelled than the activity front in 2007. This corresponds with an increase in activity in the 0-5 cm layer (58-62 %) and a decrease in the 5-25 cm layer (39-33 %) between the same years. This goes against the general downward movement of the previous years. The mean estimate of deposition at the site was 79 kBq m^{-2}.

Skogsvallen, permanent pasture land on a silty clay soil: Between 1987 and 2007 the relative activity in the 0-5 cm layer decreased from 98 to 57 %. During the same period the activity in the 5-25 cm layer increased from 2 to 40 %. The sharp shallow activity front in the first two sampling occasions has successively moved downward and levelled out. This trend was visible also between 2004 and 2007. Just as in Möjsjövik the activity decreased sharply around ten centimetres. The mean estimate of deposition at the site was 97 kBq m^{-2}.

Hille, temporary grassland on a fen-peat soil: In 1987 the relative activity in the top five centimetres was 90 %, by 2007 the activity in the layer had decreased to 33 %. During the same period the activity in the 5-25 cm layer increased from 10 to 58 %. There were only small differences between the sampling occasions 2005 and 2007. The relative activity in the

0-5 cm layer increased slightly from 31 to 33%. The activity in the 5-25 cm layer was however quite constant. Instead the relative activity in the 25-60 cm layer decreased from 11 to 9 %. In 2007 there were only weak signs of a front structure left in the soil; instead the activity decreased gradually with dept down to about 25 centimetres. The mean estimate of deposition at the site was 214 kBq m^{-2}.

Ramvik, permanent pasture land on a silty loam soil: Between 1987 and 2007 the relative activity in the top five centimetres decreased from 96 to 37 %. The relative activity in the 5-25 cm layer increased during the same period from 4 to 62 %. In accordance, the front structure becomes less and less pronounced with time. Just as in Möjsjövik the general trend of translocation between the top five centimetres and the layer below was interrupted in 2007. The activity in the 0-5 cm layer increased from 27 to 37 % and the activity decreased from 70 to 62 % in the 5-25 cm layer. This can be seen in Figure 5.1 as an increase in relative activity in 2007 above 9 centimetres and a decrease below the same depth, matched against 2003. The mean estimate of deposition at the site was 62 kBq m^{-2}.

Hammarstrand, permanent pasture land on a silty loam soil: Between 1989 and 2007 the relative activity in the 0-5 cm layer decreased from 88 to 76 %. During the same period the activity in the 5-25 cm layer increased from 12 to 24 %. Just as in Möjsjövik and Ramvik the general trend with translocation between the top five centimetres and the layer below was interrupted in 2007. The activity in the 0-5 cm layer changed from 66 to 76 %. In accordance, the activity in the 5-25 cm layer changed from 34 to 24 %. In 2007 Hammarstrand had a clear activity front at three centimetres depth and a relatively steep decrease in activity down to about ten centimetres. The mean estimate of deposition at the site was 43 kBq m^{-2}.

Stora Blåsjön, semi natural grassland on a gravelly sandy loam soil: In 1989 the relative activity in the top five centimetres was 90 %, by 2007 the activity in the layer had decreased to 85 %. During the same period the activity in the 5-25 cm layer increased from 10 to 13 %. Stora Blåsjön was the only site where there was no clear pattern of translocation between the top two layers. The front structure, with a well defined activity front in the 1-2 cm layer was relatively intact over the whole sampling period. Activity values in 2003 were only measured down to 25 centimetres depth. This should be kept in mind when comparing the change between the layers. The mean estimate of deposition at the site was 38 kBq m^{-2}.

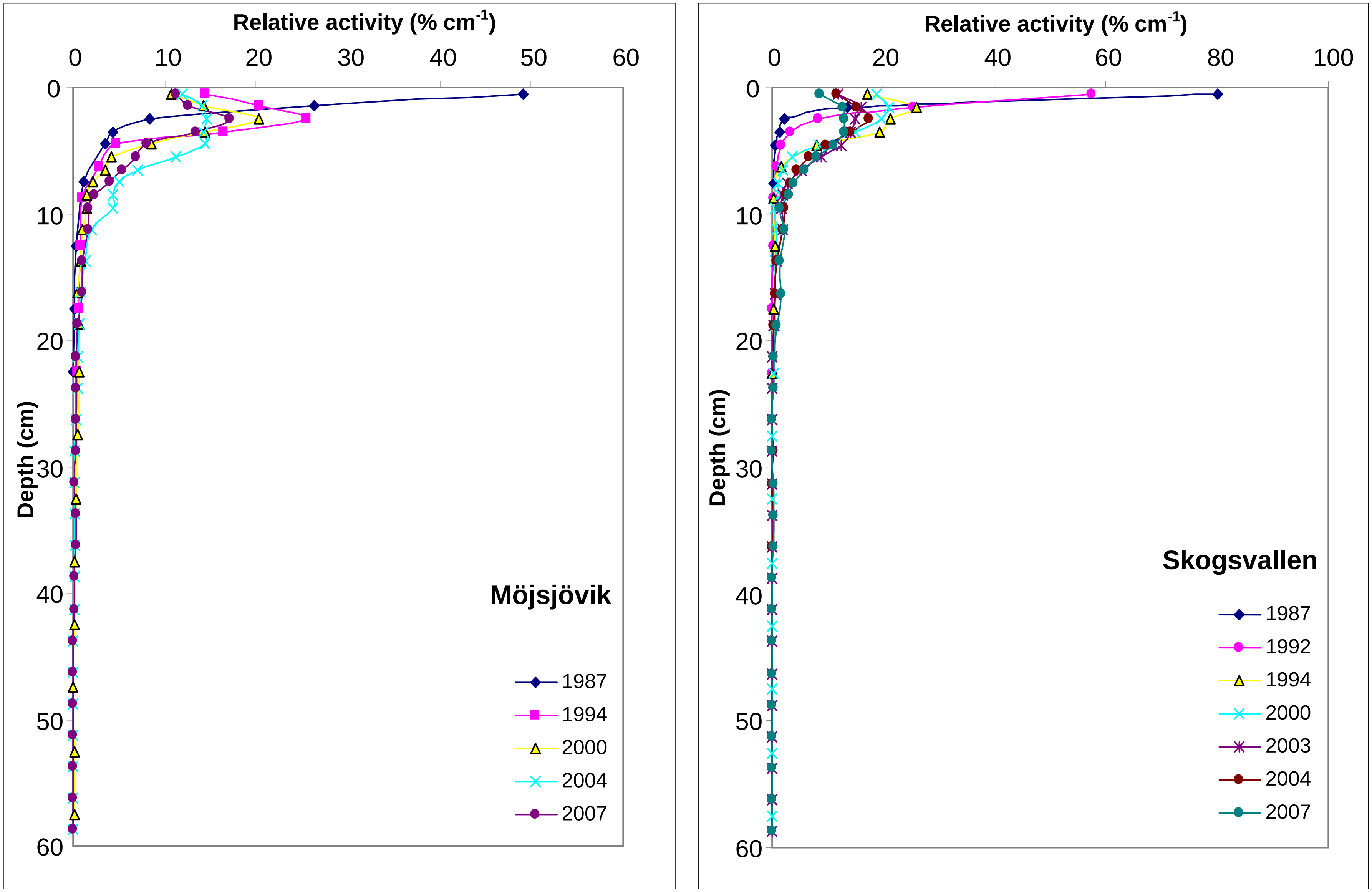

Figure 5.1. a, b The relative activity of ^{137}Cs per square metre (%cm^{-1}) in Möjsjövik and Skogsvallen for all years of sampling.

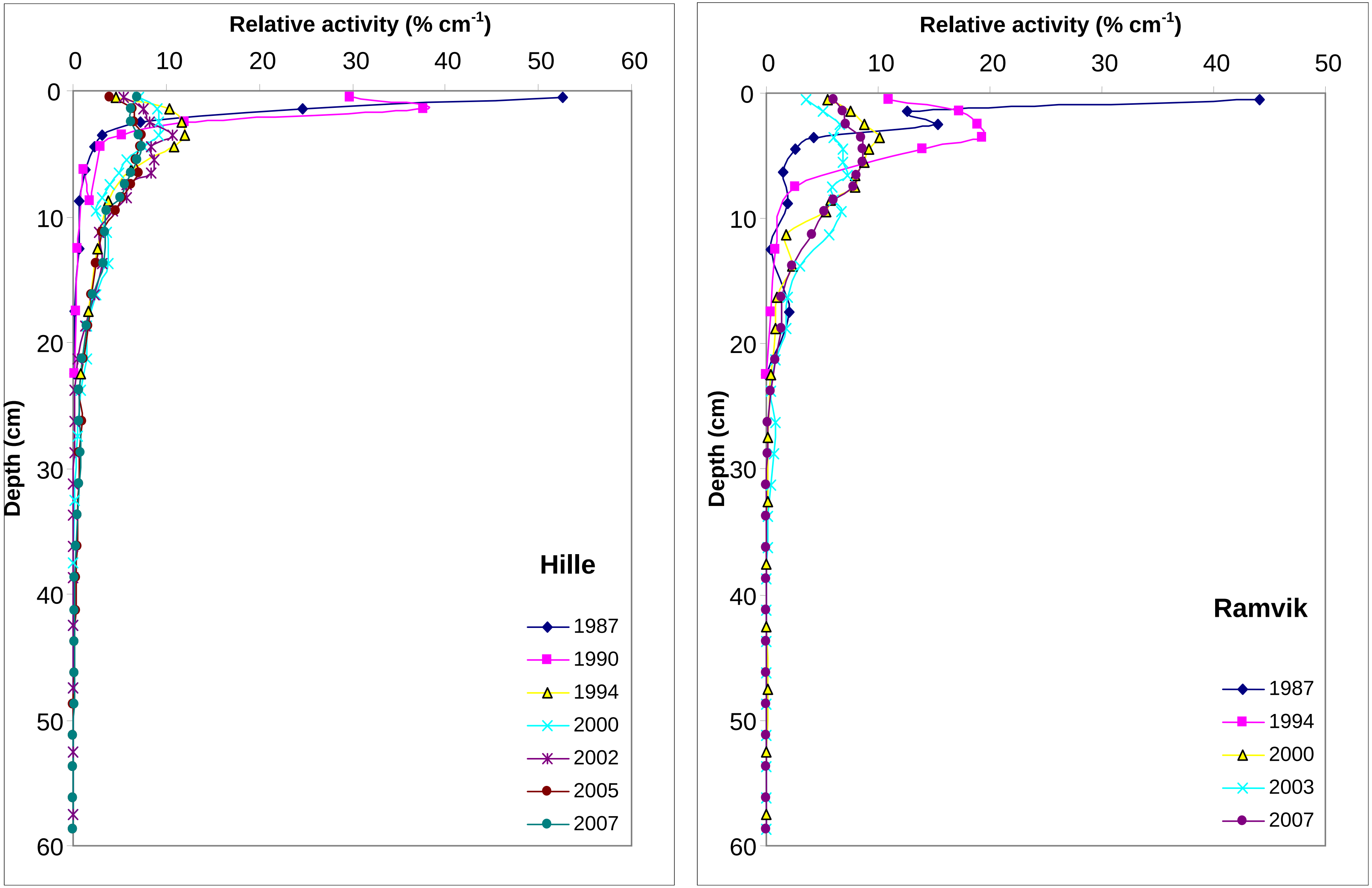

Figure 5.1. c, d The relative activity of ^{137}Cs per square metre (% cm^{-1}) in Hille and Ramvik for all years of sampling.

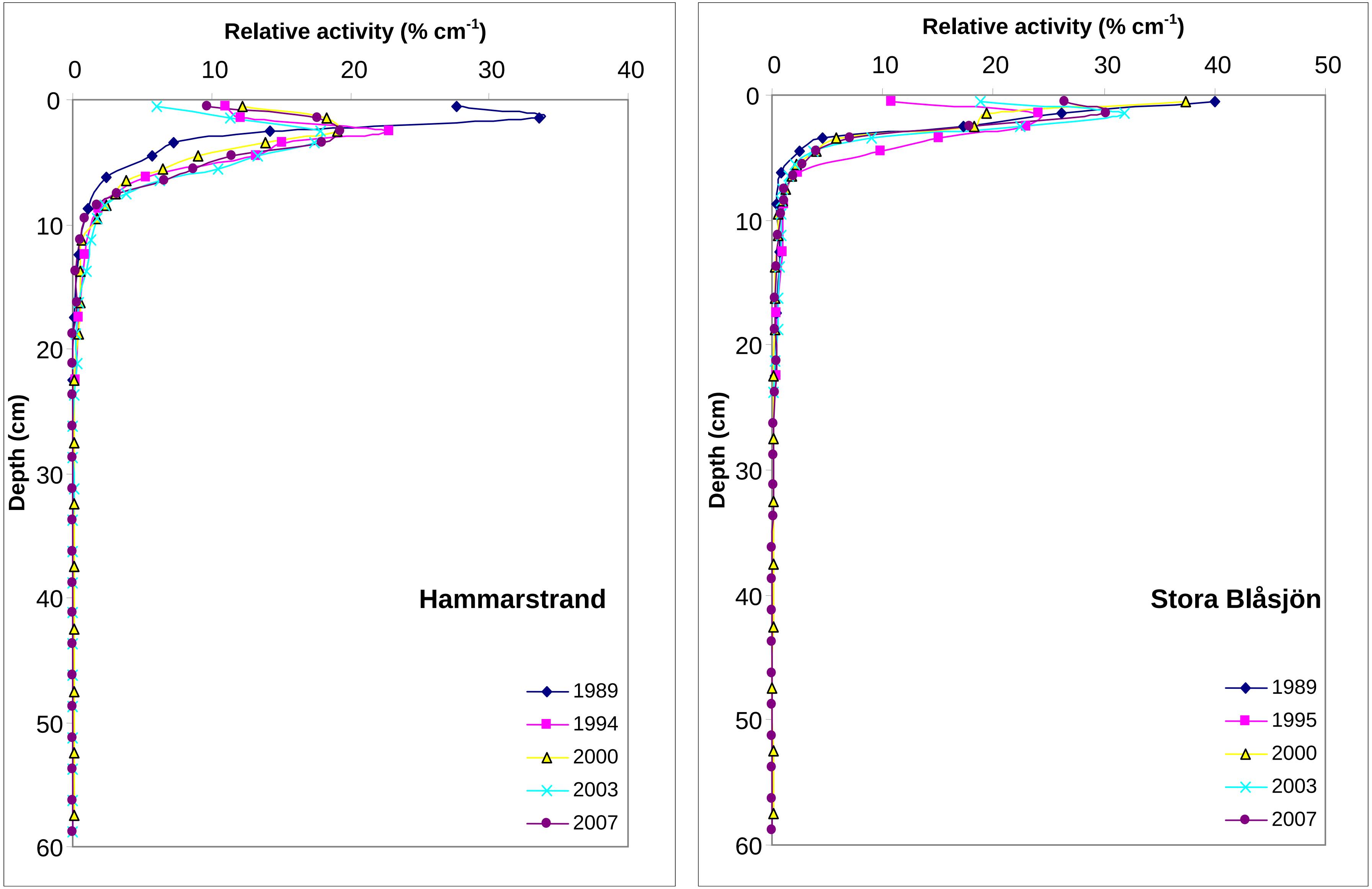

Figure 5.1. e, f The relative activity of ^{137}Cs per square metre (% cm^{-1}) in Hammarstrand and Stora Blåsjön for all years of sampling.

5.2. Migration

5.2.1. Migration depth

The migration depth in the soil tended to increase with time over the sampling period as a whole. This trend was however only visible in Skogsvallen in year 2007. Skogsvallen was the only location with an increase between every sampling occasion. Skogsvallen was also the only location with a significant change in migration depth between 2000 and 2007. In Möjsjövik, Hille and Ramvik the migration depth in 2007 was slightly shallower than at the previous sampling occasion. In Hammarstrand the migration depth was actually shallower than in both 2003 and 2000, the difference was however not statistically significant. Stora Blåsjön was the only site where there was no significant difference between any sampling occasion. Migration depth given as weighted mean values for the activity and migration rates for all locations and years of sampling are summarized in Table 5.3.

Table 5.3. Migration depth and migration rate for all locations and years of sampling. The migration depth was calculated as a weighted mean value for the ^{137}Cs activity in each profile according to Arapis et al. (1997). Migration depths with the same letter (a-e) were not significantly different (α=0.05), within each site. Values from 1987-1995 taken from Rosén et al. (1999). Values from 2000 taken from Hermansson (2001) except for Hille and values from 2003 taken from Jansson (2004) except for Hille and Möjsjövik

	Möjsjövik					**Skogsvallen**						
Year	**1987**	**1994**	**2000**	**2004**	**2007**	**1987**	**1992**	**1994**	**2000**	**2003**	**2004**	**2007**
Migration depth (cm)	2.0^{a}	4.0^{ab}	7.2^{b}	6.5^{b}	6.5^{b}	0.9^{a}	1.4^{ab}	2.6^{bc}	4.0^{cd}	4.6^{d}	5.4^{de}	6.6^{e}
Migration rate (cm y^{-1})	1.47	0.28	0.53	-0.18	-0.02	0.72	0.10	0.53	0.24	0.24	0.67	0.38

	Hille							**Ramvik**				
Year	**1987**	**1990**	**1994**	**2000**	**2002**	**2005**	**2007**	**1987**	**1994**	**2000**	**2003**	**2007**
Migration depth (cm)	2.1^{a}	2.7^{a}	7.3^{b}	8.9^{b}	7.6^{b}	10.9^{b}	10.5^{b}	1.5^{a}	4.0^{b}	7.1^{c}	8.5^{c}	7.7^{c}
Migration rate (cm y^{-1})	1.41	0.19	1.13	0.26	-0.82	1.09	-0.17	1.09	0.34	0.60	0.50	-0.19

	Hammarstrand					**Stora Blåsjön**				
Year	**1989**	**1994**	**2000**	**2003**	**2007**	**1989**	**1995**	**2000**	**2003**	**2007**
Migration depth (cm)	2.5^{a}	3.7^{ab}	5.5^{b}	4.9^{ab}	3.7^{ab}	2.5^{a}	4.0^{a}	3.9^{a}	3.3^{a}	3.3^{a}
Migration rate (cm y^{-1})	0.82	0.22	0.30	-0.19	-0.28	0.81	0.23	-0.01	-0.20	0.01

The greatest migration depth in 2007 was calculated for Hille, 10.5 cm and the lowest for Stora Blåsjön, 3.3 cm. Hille was significantly different from all the other sites except Ramvik. Stora Blåsjön was significantly different from all other sites except Hammarstrand. Hammarstrand was significantly different from Skogsvallen, Ramvik and Hille. Möjsjövik was significantly different from Hille and Stora Blåsjön. Migration depths given as weighted mean values for the ^{137}Cs activity, with plotted standard deviations for all locations 2007 are given in Figure 5.2.

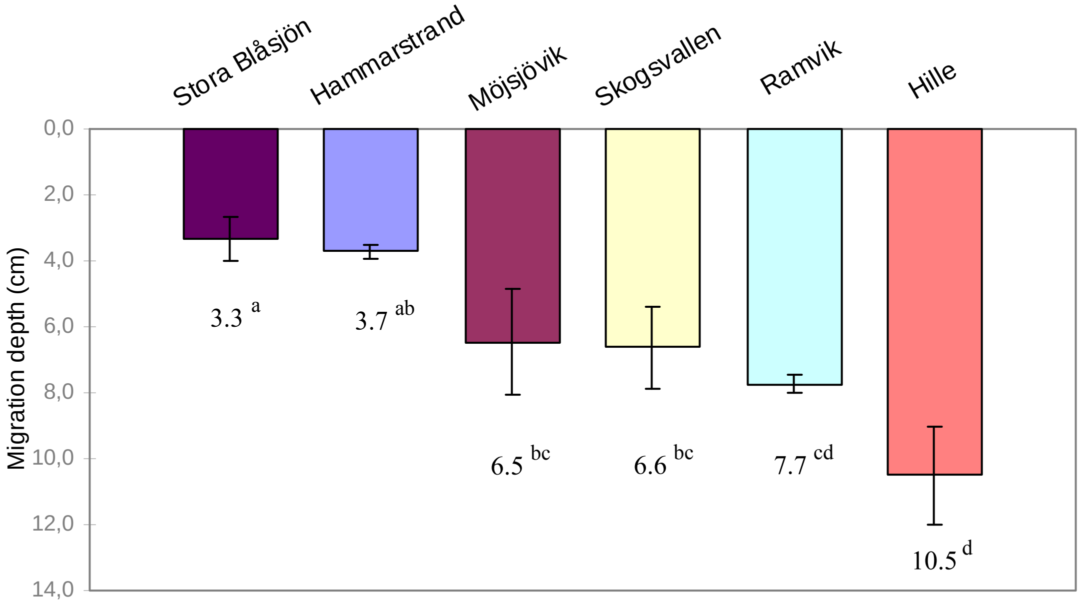

Figure 5.2. Migration depths (cm), calculated as weighted mean values for the ^{137}Cs activity 2007 according to Arapis et al. (1997), with standard deviation for all locations. Migration depths with the same letter (a-d) were not significantly different (α=0.05).

5.2.2. Migration rate

The initial migration rate was the highest at all sites and ranged between 0.72 and 1.47 cm y^{-1}. The migration rate as a whole decreased significantly with time although it varied considerably within each individual location. The migration rates in Möjsjövik, Hille, Ramvik and Hammarstrand in 2007 were actually negative. Changes in the migration rates for all locations 1987-2007 are shown in Figure 5.3.

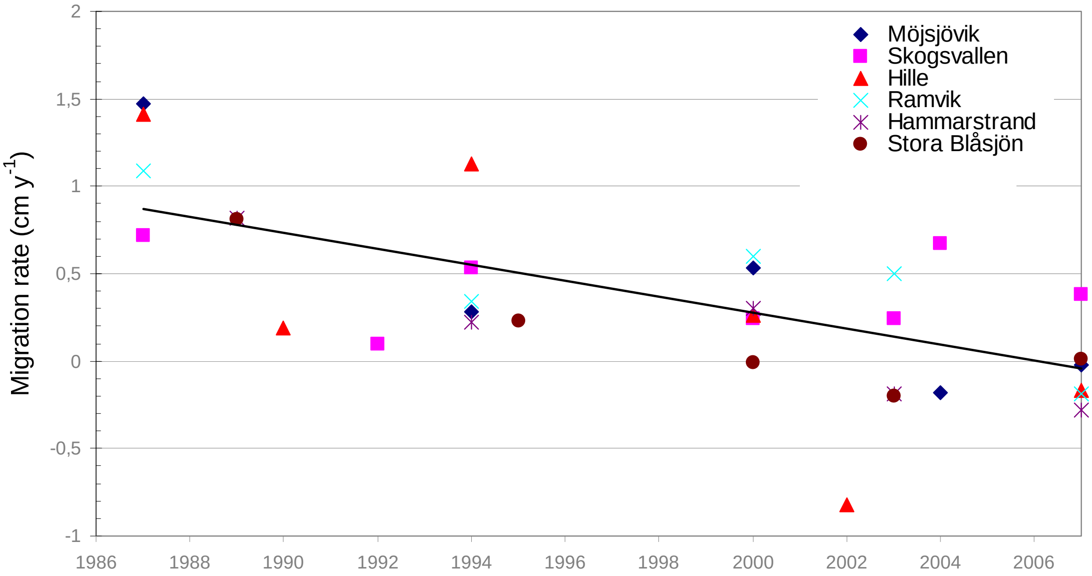

Figure 5.3. Change in migration rates (cm y^{-1}) for all locations over the sampling period 1987-2007. The trend line (y=91.7-0.0457x, R^2 = 0.38) were produced with values from all locations and years of sampling. Values from 1987-1995 taken from Rosén et al. (1999). Values from 2000 taken from Hermansson (2001) except for Hille and values from 2003 taken from Jansson (2004) except for Hille and Möjsjövik.

In order to overcome the considerable variation in migration rates the mean migration rate for each sampling location was calculated. Mean migration rates and initial migration rates are summarized in Table 5.4.

Table 5.4. Mean and initial migration rates (cm y^{-1}) for all locations and years of sampling

	Möjsjövik	**Skogsvallen**	**Hille**
Mean migration rate (cm y^{-1})	0.30	0.32	0.52
Initial migration rate (cm y^{-1})	1.47	0.72	1.41

	Ramvik	**Hammarstrand**	**Stora Blåsjön**
Mean migration rate (cm y^{-1})	0.38	0.18	0.16
Initial migration rate (cm y^{-1})	1.09	0.82*	0.81*

*These sites were not sampled until 1989.

5.3. Transfer factors

There was a clear decrease in the uptake of radiocaesium during the sampling period 1987-2007. There was a decrease in both radiocaesium content in the grass and transfer factors for all investigated locations. The largest decrease was found in Skogsvallen where the transfer factor had decreased by 99 % by 2007 as compared with 1987. The smallest decrease was found in Möjsjövik where the transfer factor had decreased by 75% over the same period.

There were relatively small changes between the previous sampling occasion 2003/2004/2005 and 2007. The largest relative change occurred in Ramvik where the transfer factor decreased from 0.5 to 0.2 m^2 kg d.w.$^{-1}$ The transfer factor also decreased slightly in Möjsjövik, Skogsvallen and Stora Blåsjön. In Hille and Hammarstrand there was actually a small increase. Figure 5.4 illustrates the change in transfer factors over the sampling period. The change between the first and the second sampling occasion accounted for the main decrease at all locations.

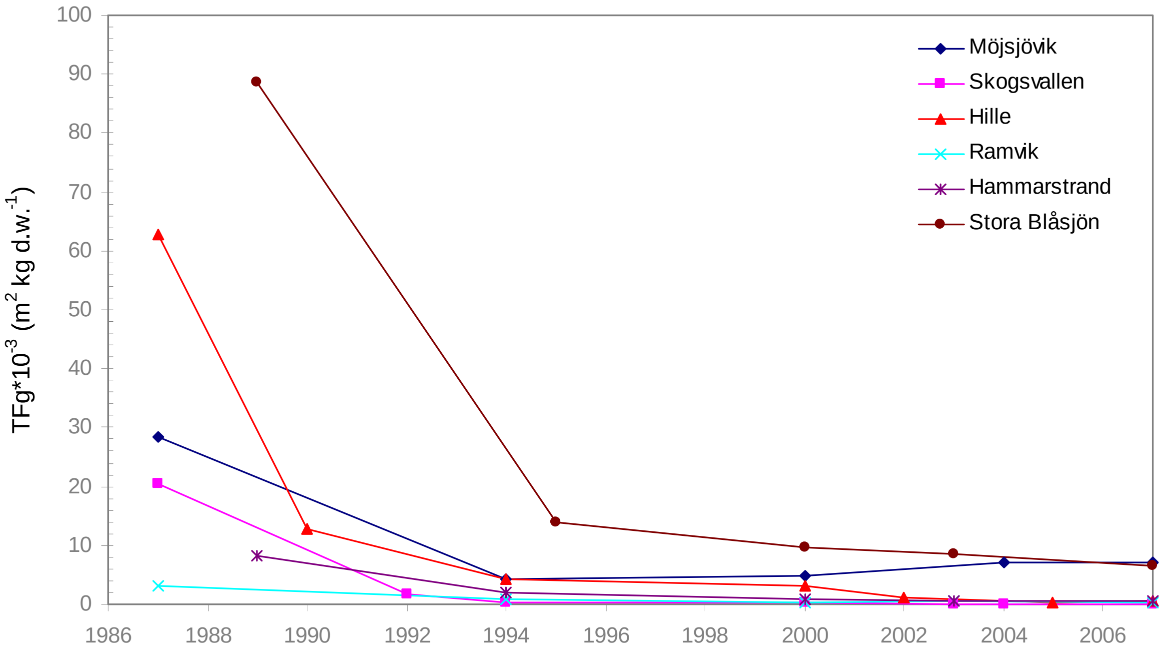

Figure 5.4. Change in transfer factors (m^2 kg d.w.$^{-1}$) for all locations over the sampling period 1987-2007. Values from previous years taken from Jansson (2004) except for Hille and Möjsjövik.

The highest transfer factor in 2007 was found in Möjsjövik (7.0 m^2 kg $d.w.^{-1}$) along with the highest radiocaesium content in the grass (333 Bq kg $d.w.^{-1}$). The lowest transfer factor was found in Skogsvallen (0.04 m^2 kg $d.w.^{-1}$) that also had the lowest activity in the grass (3 Bq kg $d.w.^{-1}$). The transfer factor in Möjsjövik was significantly different from that of Skogsvallen, Hille, Ramvik and Hammarstrand. Skogsvallen was significantly different from Ramvik and Hammarstrand. Stora Blåsjön was despite its high mean transfer factor not significantly different from any other site due to very high variability. Transfer factors for all locations 2007 are shown in Figure 5.5. Transfer factors and activity data for all years are found in Appendix 3.

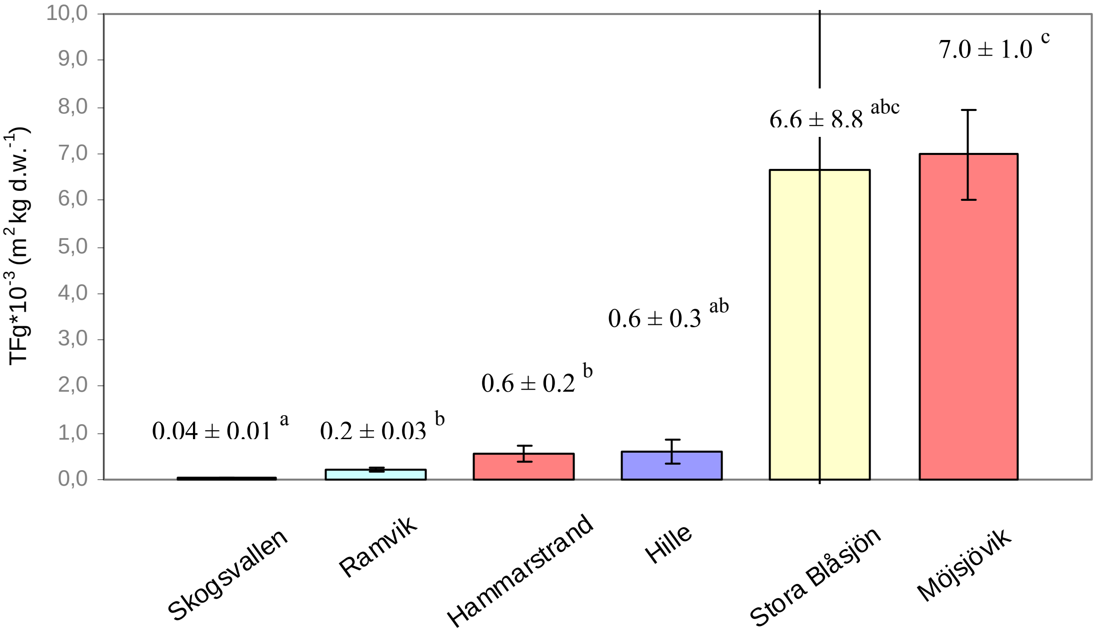

Figure 5.5. Transfer factors (TFg $*10^{-3}$ m^2 kg $d.w.^{-1}$) with standard deviations for all sampled sites 2007. Transfer factors with the same letter (a-c) were not significantly different (α=0.05).

Potassium and caesium are known to have analogous plant uptake behaviour. There should therefore be a negative correlation between plant available potassium and the uptake of radiocaesium. No such correlation could however be found in this study (Fig. 5.6). Neither was there a significant correlation between the non-exchangeable potassium content and the transfer factor (Fig. 5.7). The variables were plotted as logarithms in order to achieve a normal distribution within each data set.

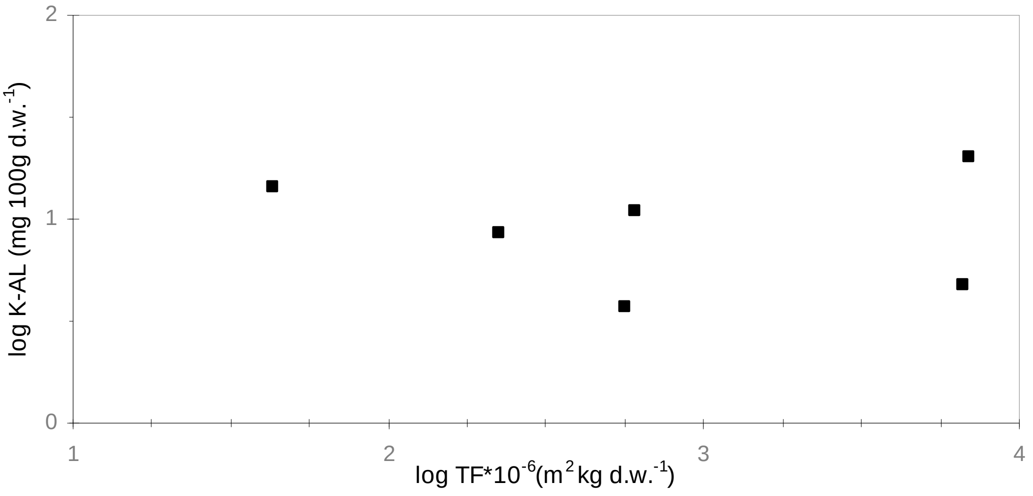

Figure 5.6. The non-significant relation between the logarithm of the plant available potassium content (log mg 100 g d.w.$^{-1}$) in the top twenty centimetres of the soil and the logarithm of the transfer factor (log TF*10^{-6} m^{2} kg d.w.$^{-1}$). Values on K-AL taken from Larsson (2008).

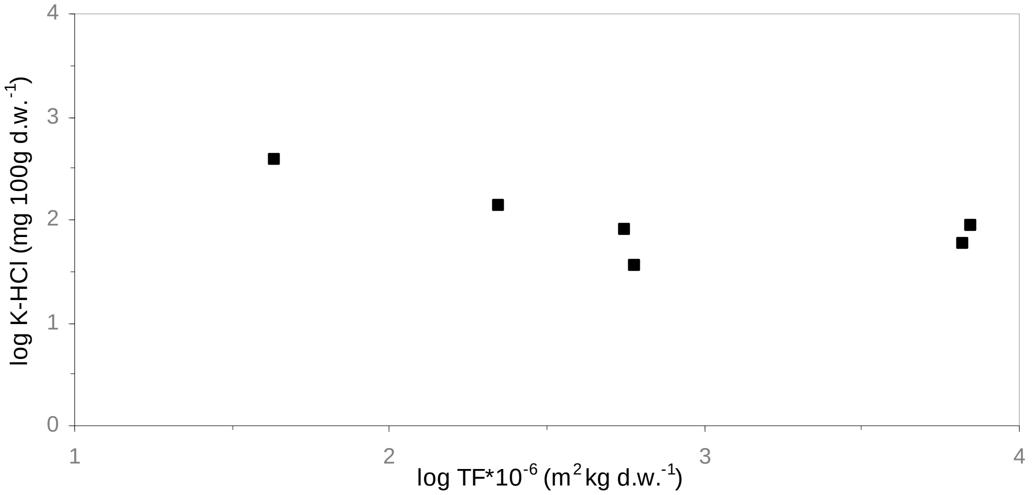

Figure 5.7. The non-significant relation between the logarithm of the non-exchangeable potassium content (log mg 100 g d.w.$^{-1}$) in the top twenty centimetres of the soil and the logarithm of the transfer factor (log TF*10^{-6} m^{2} kg d.w.$^{-1}$). Values on K-HCl modified from Rosén et al. (1999).

6. Discussion

6.1. Deposition

The differences in total activity between the sampling occasions should not been seen as an actual change in deposition. There is still to this day small fallout of activity originating from nuclear weapon testing. These activities are however hardly detectable and have no real effect on the total activity. There are great uncertainties involved in scaling up the activity in the soil cores to square meters. The differences in total activity are rather a product of our sampling procedure and natural variability.

The sites were only sampled down to 25 centimetres prior to year 2000. One may think that the increased sampling depth might have had a positive effect on the total activity. This is however only part of the explanation. The activity below 25 centimetres is very small, especially in the first years and is therefore unlikely to make such a difference to the total ground deposition. A more probable explanation is that the position of the sampling point has changed. The local conditions at the time of fallout such as level of precipitation, wind direction and pooling of water may considerably affect the ground deposition and cause it to vary within the same field. For this reason a shift in sampling position, even just a few metres, may result in an evident change in the total activity. This is known to have happened in Hille and Ramvik and it is probable that such a shift also has taken place in Skogsvallen. The sampled location in Ramvik is located in a steep slope. The site is also heavily grazed by horses. Both these factors make the site extra sensitive to erosion and horizontal redistribution of activity and offer a second possible explanation to the change in total activity.

6.2. Vertical distribution and migration

6.2.1. Vertical distribution and migration depth

There was a slow vertical migration of radiocaesium between 1986 and 2007 at all locations, except at Stora Blåsjön. The translocation of activity from the top 0-5 cm layer to the 5-25 cm layer below corresponds with the increase in migration depth over the sampling period as a whole. The translocation of activity was generally more pronounced during the first half of the sampling period. With exception for Skogsvallen, there were no significant changes in migration depth after year 2000.

The weighted mean value was estimated as the product between the relative activity of each layer and the depth of that layer. This way of defining migration depth emphasizes the change in the deeper layers. Relatively small changes in the deeper layers have a major impact on the weighted mean value. This explains why the migration depth in Möjsjövik and Hammarstrand in year 2000 was deeper than in 2003 even though the vertical translocation between the top layer and the 5-25 cm layer was less pronounced.

In a study of a large range of Chernobyl contaminated soils in Belarus seven years after the accident, Arapis et al. (1997) found that at least 80 % of the ^{137}Cs activity still resided in the top ten centimetres of the soil. In a Swedish long term study of two sandy soils and one peat soil, the activity in the 0-10 cm layer ranged between 85 and 93 %, eleven years after the fallout (Forsberg, 2000). In this study the relative activity in the top ten centimetres ranged between 61 and 97 %.

Möjsjövik and Hille are both organic peat soils with similar soil properties. They had never the less quite different migration patterns. Möjsjövik had a less pronounced translocation between the 0-5 cm and the 5-25 cm layer and a significantly shallower migration depth. The still relatively clear front structure in Möjsjövik was almost completely levelled out in Hille (Fig 5.3). The vertical distribution in Möjsjövik actually had more in common with the mineral soils, Stora Blåsjön being the exception. The most probable reason fore this is the fact that the soil in Hille has been drained. Another possible explanation is the notably deeper and denser root zone in Möjsjövik. The microorganisms in the root zone are known to enhance retention of radiocaesium (Parekh et al., 2007).

Skogsvallen was the only clay soil in the study. Clay soils are because of fixation to FES in clay minerals, expected to have a shallow migration depth and a limited translocation of activity. This was true for Skogsvallen during the first sampling occasions (1987-1994). Unlike the other sites however, the migration showed no tendency to level off towards the end of the sampling period. The vertical distribution was more pronounced with every sampling occasion and so was the increase in migration depth. Skogsvallen was the only site with a significantly deeper migration depth in 2007 than in 2003. The limited migration during the first sampling occasions and the low transfer factor at the site still gives us reason to believe that fixation to clay minerals takes place in the soil. The most probable reason for the ongoing migration would then be bio diffusion due to earthworms (personal communication, Nicholas Jarvis). This possibility is currently subject for further research undertaken at the Department of Soil and Environment.

Ramvik and Hammarstrand have quite similar soil properties. Both soils have clay contents just above ten percent. The migration pattern in the two soils was quite similar in the first and second sampling occasion. In the third sampling occasion the main part of the activity in Ramvik was found in the 5-25 cm layer while the vertical distribution in Hammarstrand was relatively unchanged. In 2007 the migration depth in Ramvik was significantly deeper than that of Hammarstrand. Ramvik has more sand in the top soil than Hammarstrand. The coarser soil particles make the soil more porous, this might explain the different migration patterns.

Stora Blåsjön was the only site where there was no significant increase in migration depth over the sampling period. There was, in agreement with the unchanging migration depth, no real trend of translocation between the top five centimetres and the deeper horizons. The fact that the soil was not sampled until 1989 is of course important to take into consideration. There was considerable variance in migration depth on several sampling occasions, indicating a large heterogeneity of the migration at the site. The clay content in Stora Blåsjön is comparable with that of Ramvik and Hammarstrand, about ten percent in the top soil. The clay content can therefore not alone be responsible for the fixation. Recycling between living and dead parts of the vegetation also keeps the activity from migrating further down the profile. A third factor that might add to the unchanging vertical distribution is the topography. The sampled site is located in a mountainous area in the lower part of a steep slop. It is possible that the site has been subject to upwelling of water from higher up in the slope during spring flood or heavy rainfall. Rocky subsoil and the fact that the soil is frozen a greater part of the year also limit the migration.

There was, in this study, no clear connection between migration and soil properties 21 years after the accident. The clay content in the mineral soils was expected to hinder vertical migration. Skogsvallen, the only clay soil in the study, did not have a significantly shallower migration depth than the other mineral soils. On the contrary the vertical translocation in

Skogsvallen even preceded that of Möjsjövik, one of the organic soils. Organic soils were expected to have a greater migration depth than mineral soils. Hille the second organic soils in the study, did have a significantly greater migration depth than the mineral soils. The migration depth in Hille was however also significantly deeper than that of Möjsjövik. The difference in migration seams instead to be a product of biological and hydrological factors at the individual locations.

6.2.2. Migration rate

The migration rate as a whole decreased with time at all locations, although it varied considerably and even increased between sampling occasions at all locations. The R^2-value for the trend line in Figure 5.3 indicate that only 38 % of the variation in migration rate between the different sites and sampling occasions arose due to change in time. The rest of the variance was a result of other factors related to the individual locations. There are great uncertainties involved in calculating the migration rates for every sampling occasion. Each migration depth is afflicted with uncertainties and the difference between two migration depths divided by a relatively short period of time is therefore even more uncertain.

There seams to be a correlation between soil properties and initial migration rates. The highest initial migration rates were found in the two organic soils Möjsjövik and Hille (1.5 and 1.4 cm y^{-1}). Ramvik stood out among the mineral soils with a comparably high initial migration rate of 1.1 cm y^{-1} while the initial migration rate in the other minerals soils ranged between 0.7 and 0.8 cm y^{-1}. The lower clay content and higher proportion of sand in the top soil is probably the reason for the higher value in Ramvik as compared with Skogsvallen. Hammarstrand and Stora Blåsjön had despite their similar soil properties with Ramvik an initial migration rate more similar to that of Skogsvallen. These two soils were however not sampled until 1989. The difference in time between fallout and the first sampling occasion is likely to be the reason for the difference in initial migration rates.

Ivanov et al. (1997) measured initial migration rates between 0.6 and 1.1 cm y^{-1} in a large study of eastern European soils, conducted in 1987. The migration depth in that study was however defined as the median depth. The analogous initial migration rates in this study ranged between 0.5 and 1.0 cm y^{-1} (Rosén et al., 1999).

Arapis & Karandinos (2004) measured mean migration rates between 0.1 and 0.3 cm y^{-1} in a study of sloping semi-natural soils in northern Greece 10 years after the accident. These rates are within the lower range of the mean migration rates in this study (0.2-0.5 cm y^{-1}). The mean migration rates in this study were however lower than those reported from a study of a large range of soil types in Belarus (0.4-1.2 cm y^{-1}) conducted seven years after the accident (Arapis et al. 1997). Much of the difference between this study and that of Arapis et al. (1997) can however be explained by the different sampling periods. The mean migration rate in this study after just eight years ranged between 0.3 and 0.9 cm y^{-1}.

The relatively constant migration depths in the last sampling occasions lead to slow migrations rates. Some migration rates especially in 2007 were actually negative. There is a vast uncertainty imbedded in these values and they are therefore hard to interpret. Errors and uncertainties in the sampling technique, like a small shift in sampling location or contamination of the deeper layers with activity from higher up in the profile, have a greater impact on small changes. There was no significant decrease in migration depth at any site. Negative migration rates are therefore not to be seen as an actual upward transport but rather as an indication on that the downward migration has levelled out. That is not to say that there is no upward transport of activity. The activity in the soil solution moves with the pore water in both directions. Bio diffusion due to earth worms also works in two ways. There is however no support for a net upward transport of activity in this study.

6.3. Transfer factors

There was a clear decrease in uptake with time. This is in agreement with what could be expected as the over all fixation in the soil increases. The decrease was greatest during the first years directly after fallout. The sharp decrease in transfer factors between the first sampling occasions had by 2007 levelled of at all sites. In 2007 there was actually a higher transfer at two of the sampled locations, as compared with the previous sampling occasion. This should not be seen as an actual increase in uptake but rather as a product of natural variability.

There was no correlation between uptake of radiocaesium and plant available or non-exchangeable potassium concentrations in the soil, in this limited study. Plant available potassium varies considerably and is often almost depleted at the end of the growing season (Eriksson et al., 2005). The values for 2007 were sampled in the end of August which is probably why the values were generally low. The pool of non-exchangeable potassium is slowly weathered and more stable over time (Eriksson et al., 2005).

The two organic soils in the study Möjsjövik and Hille had significantly different transfer factors. Organic soils have low radiocaesium fixating capabilities and are therefore expected to have a high plant uptake. There is no obvious explanation for why Hille have such a low transfer factor. Hille received a greater deposition than Möjsjövik. The transfer factor is calculated by dividing the absolute activity in the grass with the ground deposition. There is however a clear difference also in absolute activities. The activity front has migrated further down in Hille than in Möjsjövik but this is probably not enough to explain the dissimilarities. Unfortunately there is no data on the clay content for these soils. Difference in clay content or clay mineralogy affects the uptake even in organic soils. Different peat composition is another possible explanation, suggested by Rosén et al. (1999).

The transfer factor in Skogsvallen was a factor ten lower than the transfer factors at the other sites. There was however no significant difference in transfer factors between Skogsvallen and Hille, probably because of the high standard deviation at the latter. Skogsvallen is a clay soil with high concentration non-exchangeable potassium in the top soil. There is a strong connection between clay content and non-exchangeable potassium concentration in soils (Eriksson et al., 2005). It is therefore often difficult, especially in such a limited study as this, to separate and grade the effects of fixation to FES and competition between caesium and potassium for uptake by plants.

Ramvik and Hammarstrand are quite similar both in respect to clay content and potassium concentrations. Although there were large differences in the migration pattern between the sites, there was no significant difference in transfer factors. Ramvik had an exceptionally low transfer factor in 1987 compared to the other sites. One possible explanation for this is that the site was sampled at the very end of the growing season.

The transfer factor in Stora Blåsjön was not significantly different from any other site. This was probably because of the large variation at the site. The high mean value was however in agreement with the previous sampling occasions. The top horizon, where much of the activity still resides is rich in organic matter which reduces fixation and increase the plant available caesium fraction. Nutrient poor soils in upland pastures are known to have high transfer factors. Recycling between living and dead plant material helps to explain the high plant availability combined with the limited migration (Gastberger et al., 2000). Rocky sub soil often results in a shallow root system and since the main part of the activity is concentrated in the top soil this may in turn result in a higher plant uptake.

The transfer factors in this study were, with exception for Hille, in agreement with what could be expected regarding the different soil properties and bio/hydrological factors at the locations.

There are relatively few long-term plant uptake studies conducted on pasture soils regarding radiocaesium originating from the Chernobyl accident. Grytsyuk et al. (2006b) studied the uptake in radio polluted lands in northern Ukraine. The highest transfer factors in this study were within the lower range of the transfer factors at the comparable Ukrainian sites. The level of deposition in the investigated part of Ukraine was however greater than at the Swedish sites. Gastberger et al. (2000) studied the soil-to-plant uptake of fallout caesium in Austrian lowland and Alpine pastures. The lowest transfer factor in this study was within the range of the highest Austrian values. The deposition levels in the Austrian study were generally lower than in this study.

6.4. Conclusion

There was a slow vertical migration of radiocaesium between 1986 and 2007 at all sites, except Stora Blåsjön where no migration could be confirmed. The translocation of activity from the top centimetres of the soil to the deeper layers was generally more pronounced during the first half of the sampling period. With exception for Skogsvallen, there was no significant change in migration depths after year 2007. The main part of the activity (61-97 %) was after 21 years still present in the upper ten centimetres of the soil. The migration rate as a whole decreased with time although it varied considerably within each individual location. The initial migration rate was the highest at all locations. In this study there was no clear connection between migration and soil properties 21 years after the accident. Instead the differences in migration seam to be a product of biological and hydrological factors at the individual locations. The influence of soil properties on the migration seams to be greatest in the first years after fallout. This is in direct disagreement with Forsberg, (2000) who found that the soil properties are of lesser importance during the first 5-10 years where after their effect on the migration increase.

There was a clear decrease in plant uptake with time. The change between the first and the second sampling occasion accounted for the main decrease at all locations. There were relatively small changes between the previous sampling occasion and 2007. The highest transfer factor in 2007 was found in Möjsjövik an organic soil and Stora Blåsjön a nutrient poor upland soil with high organic matter content in the top horizon. The lowest transfer factor was found in Skogsvallen, a silty clay soil. There was no clear correlation between potassium content and plant uptake of radiocaesium in this study.

References

Absalom, J.P., Young, S.D., Crout, N.M.J., Sanchez, A., Wright, S.M., Smolders, E., Nisbet, A.F. & Gillett A.G. 2001. Predicting the transfer of radiocaesium from organic soils to plants using soil characteristics. *J. Environ. Radioactivity* Vol. 52 p. 31-43.

Andersson, P., Carlsson, M., Falk, R., Hubbard, L., Leitz, W., Mjönes, L., Möre, H., Nyblom, L., Söderman, A-L., Larsson, K.Y., Åkerblom, G. & Öhrlén, E. 2007. Strålmiljön i Sverige. SSI rapport 2007:02 ISSN 0282-4434.

Arapis, G., Petrayev, E., Shagalova, E., Zhukova, O., Sokolik, G. & Ivanova, T. 1997. Effective Migration of ^{137}Cs and ^{90}Sr as a function of the Type of Soils in Belarus. *J. Environ. Radioactivity* Vol. 34 p. 171-185.

Arapis, G., Chesnokov, A., Ivanova, T., Potapov, V. & Sokolik, G. 1999. Evaluation of dose equivalent rate reduction as a function of vertical migration of ^{137}Cs in contaminated soils. *J. Environ. Radioactivity* Vol. 46 p. 251-263.

Arapis, G.D. & Karandinos, M.G. 2004. Migration of ^{137}Cs in the soil of sloping semi-natural ecosystems in Northern Greece. *J. Environ. Radioactivity* Vol. 77 p. 133-142.

Bengtsson, G. 2006. Strålskyddsnytt nr 1 2006 p. 7-8. ISSN 0280-0357.

van Bergeijk, K.E., Noordijk, H., Lembrechts, J. & Frissel, M. J. 1992. Influence of pH, Soil Type and Soil organic Matter Content On Soil-to-Plant Transfer of Radiocesium and – Strontium as Analyzed by a Nonparametric Method. *J. Environ. Radioactivity* Vol. 15 p. 265-276.

Bonn, B.A. & Fish, W. 1993. Measurement of electrostatic and site specific associations of alkali metal cations with humic acid. *J. Soil Sci.* Vol. 44 p. 335-45.

Bréchignac, F., Vallejo, R., Vandecasteele, C., Shaw, G., Forsberg, S. & Madoz-Escande, C. 2000. Improving the short and long term prediction ability of radiocontaminant behavior in agro-ecosystems. Rapport Scientifique et Technique. IPSN – RST 2000.

Bunzl, K., Schimmack, W., Krouglov, S.V. & Alexakhin, R.M. 1995. Changes with time in the migration of radiocaesium in the soil, as observed near Chernobyl and in Germany 1986-1994. *Sci. Total Environ.* Vol. 175 p. 49-56.

Cremers, A., Elsen, A., De Preter, P. & Maes, A. 1998. Quantitative analysis of radiocaesium retention in soils. *Nature* Vol. 335 p. 287-96.

Devell, L. 1991. Composition and properties of plume and fallout materials from the Chernobyl accident. In: Moberg, L. (ed). The Chernobyl fallout in Sweden. p. 29-46. Stockholm. ISBN 91-630-0721-5.

Devell, L. & Powers, D.A. 1996. The Chernobyl reactor accident source term. Development of a consensus view. At the international conference; One decade after Chernobyl: Summing up the consequences of the accident. Vienna 8-12 April paper number: IAEA-Cn-63/155. Vienna.

Edvarson, K. 1991. Fallout over Sweden from the Chernobyl accident. In: Moberg, L. (ed). The Chernobyl fallout in Sweden. p. 47-65. Stockholm. ISBN 91-630-0721-5.

Eriksson, J., Nilsson, I. & Simonsson, M. 2005. Wiklanders Marklära. Studentlitteratur. Lund. p. 226. ISBN 91 44-02482-7.

Forsberg, S. 2000. Behavior of 137Cs and 90Sr in agricultural soils: influence of ageing and soil type on availability, migration and plant uptake. Sveriges Lantbruksuniversitet. Uppsala. *Acta Universitatis Agriculturare Sueciae*. Agraria 212.

Fawaris, B.H. & Johanson, K.J. 1995. Sorption of 137Cs from undisturbed forest soil in a zeolite trap. *Sci Total Environ.* Vol. 72 p. 251-256.

Francis, C.W. & Brinkley, F.S. 1976. Preferential adsorption of 137Cs to micaceous minerals in contaminated freshwater sediment. *Nature* Vol. 260 p. 511-513.

Gastberger, M., Steinhäusler, F., Gerzabek, M.H., Lettner, H. & Hubmer, A. 2000. Soil-to-plant transfer of fallout caesium and strontium in Austrian lowland and Alpine pastures. *J. Environ. Radioactivity* Vol. 49 p. 217-233.

Giannakopoulou, F., Haidouti, A., Chronopoulou, A. & Gasparatos, D. 2007. Sorption behavior of caesium on various soils under different pH levels. *J. Hazard. Mater.* (2007), doi:10.1016/j.jhazmat. 2007.06.109.

Grytsyuk, N., Arapis, G. & Davydchuk, V. 2006a. Heavy metals effects on forage crops yields and estimation of elements accumulation in plants as affected by soil. *Sci. Total Environ.* Vol. 354 p. 224-231.

Grytsyuk, N., Arapis, G. & Davydchuk, V. 2006b. Root uptake of ^{137}Cs by natural and semi-natural grasses as a function of texture and moisture of soils. *J. Environ. Radioactivity* Vol. 85 p. 48-58.

Hermansson. I. 2001. Migration av radiocesium i sju olika markprofiler i Tjernobyldrabbade områden i Sverige år 2000-En jämförelse med tidigare studier 1987-1995. Examens och seminariearbeten nr 44. Institutionen för markvetenskap. SLU. Uppsala.

IAEA. 2006. Environmental consequences of the Chernobyl accident and their remediation: twenty years of experience / report of the Chernobyl Forum Expert Group 'Environment'. — Vienna: International Atomic Energy Agency. 2006. p.; 29 cm. — (Radiological assessment reports series, ISSN 1020-6566), STI/PUB/1239, ISBN 92–0–114705–8.

Isaksson, M. 2002. Grundläggande strålningsfysik. Studentlitteratur. Lund. p. 60. ISBN 91-44-01528-3.

Isaksson, M. & Erlandsson, B. 1995. Experimental determination of the vertical and horizontal distribution of ^{137}Cs in the ground. *J. Environ. Radioactivity* Vol. 27 p. 141-160.

Isaksson, M., Erlandsson, B. & Mattsson, S. 2001. A 10-year study of the ^{137}Cs distribution in soil and a comparison of Cs soil inventory with precipitation-determined deposition. *J. Environ. Radioactivity* Vol. 55 p. 47-59.

Ivanov, Y.A., Lewyckyj, N., Levchuck, S.P., Prister, B.S., Firsakova, S.K., Arkhipov, N.P., Arkipov A.N., Kruglov, S.V., Alexakhin, R.M., Sandalls, J. & Askbrant, S. 1997. Migration of ^{137}Cs and ^{90}Sr from Chernobyl Fallout in Ukrainian, Belarussian, and Russian Soils. *J. Environ. Radioactivity* Vol. 35 p.1-21.

Jansson, L.T. & Rydén, B.E. (ed). 2000. Strålskydd. Natur och kultur i samarbete med Uppsala universitet, tredje upplagan. Borås. ISBN 91-27-08232-6.

Jansson, M. 2004. Migration av radiocesium i olika markprofiler samt förekomst av radiocaesium i grundvattnet i Sverige år 2003 efter Tjernobyl olyckan - En jämförelse med tidigare studier 1987-2000. Examens och seminariearbeten nr 59. Institutionen för markvetenskap. SLU. Uppsala.

Johanson, K.J. 1996. Strålning människan och miljön. Uppsala. ISBN 91-576-5096-9.

Johanson, K.J. 2006. Cesium 137 i svamp, bär och vilt. Strålskyddsnytt nr 1 2006 p. 25 & 31. ISSN 0280-0357.

Krouglov, S.V., Kurinov, A.D. & Alexakhin, R.M. 1998. Chemical fractionation of ^{90}Sr, ^{106}Ru, ^{137}Cs and ^{144}Ce in Chernobyl-contaminated soils: an evolution in the course of time. *J. Environ. Radioactivity* Vol. 38 p. 59-76.

Kudelsky, A.V., Smith, J.T., Ovsiannikova, S.V. & Hilton, J. 1996. Mobility of Chernobyl derived 137 Cs in a peat bog system within the catchment of the Pripyat River, Belarus. *Sci. TotalEnviron.* Vol. 118 p. 101-113.

Larsson, M. 2008. Soil factors influencing transfer of ^{137}Cs from soil to crop –results from a field study made 21 years after the Chernobyl accident. Institutionen för markvetenskap. SLU. Uppsala.

Moberg, L. 2001 Kärnkraftsolyckan i Tjernobyl en sammanfattning femton år efter olyckan. SSI rapport 2001:7

Moberg, L. 2006. Strålskyddsnytt nr 1 2006 p. 10-11. ISSN 0280-0357.

Parekh, N.H., Poskitt, J.M., Dodd, B.A., Potter, E.D. & Sanchez, A. 2007. Soil microorganisms determine the sorption of radio nuclides within organic soil systems. *J. Environ. Radioactivity* (2007) dio:10.1016/j.jenvrad.2007.10.017.

Persson, K., & Preuthun, J. (ed). 2002. Livsmedelsproduktion vid nedfall av radioaktiva ämnen. Västerås. ISBN 91-7056-113-3.

Roca, M.C.& Vallejo, V.R. 1995. Effect of soil potassium and calcium on caesium and strontium uptake by plant roots. *J. Environ. Radioactivity* Vol. 28 p. 141-159.

Rosen, K. 1991. Effects of potassium fertilization on caesium transfer to grass, barley, and vegetables. In: Moberg, L. (ed). The Chernobyl fallout in Sweden. p. 305-22. ISBN 91-630-0721-5.

Rosén, K. 2006. Tjernobyl olyckan och jordbruket. Strålskyddsnytt nr 1 2006 p. 20-21. ISSN 0280-0357.

Rosén, K., Eriksson, Å. & Haak., E. 1996. Transfer of radiocaesium in sensitive agricultural environments after the Chernobyl fallout in Sweden. I. County of Gävleborg. *Sci Total Environ.* Vol. 368 p. 795 -803.

Rosén. K., Öborn, I. & Lönsjö, H. 1999. Migration of radiocaesium in Swedish soil profiles after the Chernobyl accident. 1987-1995. *J. Environ. Radioactivity* Vol. 46 p. 45-66.

Rosén, K., Shand, C.A., Haak, E. & Cheshire, M.V. 2006. Effect of clay content and wetting- and-drying on radiocaesium behavior in a peat and a peaty podzol, *Sci Total Environ.* Vol. 182 p. 117-133.

Shand, C.A., Cheshire, M.V., Smith, S., Vidal, M. & Rauret, G. 1994. Distribution of radiocaesium in organic soils. *J. Environ. Radioactivity* Vol. 23 p. 285-302.

Smith, J.T., Fesenko, S.V., Howard, B.J., Horrill, A.D., Sanzharova, N.I. & Alexahin, R.M. 1999. Temporal Change in Fallout ^{137}Cs in Terrestrial and Aquatic Systems: A Whole Ecosystem Approach. *Environ. Sci.Technol.*, Vol. 33 p. 49-54.

Stauton, S., Dumat, C., 2002, Possible role of organic matter in radiocaesium adsorption in soils, *J. Environ. Radioactivity* Vol. 58 p. 163-173.

Sveriges Nationalatlas webpage. http://www.sna.se/webbatlas/kartor/kopia/cesium_137_efter_tjernobyl_86.html. 2008-05-12.

Tessier, A. Campbell, P.G.C. & Bisson, B. 1979. Sequential extraction procedure for the speciation of particulate trace metals. *Anal. Chem.* Vol. 51 p 844-850.

UNSCEAR. 1982. Ionizing radiation: Sources and biological effects. United Nations Scientific Committee on the effect of Atomic Radiation 1982. Report to the General Assembly with annexes. United Nations New York.

UNSCEAR. 2000. Report Vol. II. Sources and effects of ionizing radiation Annex J Exposures and effects of the Chernobyl accident. United Nations New York.

Valcke, E. & Cremers, A. 1994. Sorption-desorption dynamics of radiocaesium in organic matter soils. *Sci. Total Environ.* Vol. 157 p. 275-283.

Valcke, E., Elsen, A. & Cremers, A. 1997. The use of zeolites as amendments in radiocaesium- and radiostrontium- contaminated soils part I-IV: A soil-chemical approach. *Zeolites* Vol. 18 Issues 2-3. February-March 1997. p. 205-231.

Wauters, J., Sweeck, L., Valcke, E., Elsen, A. & Cremers, A. 1994. Availability of radiocaesium in soils: a new methodology. *Sci. Total Environ.* Vol. 157 p. 239-248.

Vinichuk, M.M., Johanson, K.J., Rosén, K. & Nilsson, I. 2004. Role of the fungal mycelium in the retention of radiocaesium in forest soils. *J. Environ. Radioactivity* Vol. 78 p. 77-92.

Wang, X., Rambung, Th. & Geckeis, H. 2003. Effect of pH and humic acid on the adsorption of caesium onto γ-Al_2O_3. *J. Radioanalytical Nuclear Chem.* Vol. 258. No 1 83-87.

Åhman, B. 2006. Renskötsel 20 år efter Tjernobyl. Strålskyddsnytt nr 1 2006 p. 20-21 ISSN 0280-0357.

Other sources

Personal communication: Prof Nicholas Jarvis. Dept. Soil and Environment. SLU. Uppsala. 2008-04-08.

Appendix 1

Brief facts about the sampled sites. Modified from Rosén et al. (1999)

Site	GPS Coordinates	Altitude (m)	Precipitation[a] (mm)	Mean Temperature[a] (◦C)	Parent material	Soil Taxonomy (1992)	FAO legend (1998)	Mean Dep.^{137}Cs kBq m^{-2}	Sampling date
Möjsjövik	171413E 595733N	20	563	5.3	Fen-peat	Typic Sulfihemist	Thionic Histosol	74	
A									070818
B									070818
C									070906
Skogsvallen	171057E 601005N	40	566	5.3	Lacustrine silty clay sediments	Typic Dysrochrept	Dystric Cambisol	97	
A									070511
B									070514
C									070521
Hille	171181E 604453N	20	617	5.2	Fen-peat overlaying limnic material	Terric Sulfihemist	Thionic Histosol	214	
A									070914
B									070914
C									070918
Ramvik	175020E 624946N	25	703	3.8	Alluvial silty and sandy loam	Typic Udorthent	Eutric Regosol	62	
A									070828
B									070828
C									070828
Hammarstrand	162158E 630555N	125	539	2.2	Alluvial silty loam	Typic Udorthent	Eutric Regosol	43	
A									070819
B									070830
C									070830
Stora Blåsjön	140856E 645039N	480	699	1.4	Colluvial deposits, gravely sandy loam	Typic Regosol	Dystric Cryorthent	38	
A									070820
B									070820
C									070821

[a] The mean annual precipitation and air temperature for the period 1961-1990. The climatic stations used were: Tärnsjö, Uppsala air field, Gävle, Härnösand, Krångede and Stora Blåsjön.

Appendix 2

Selected physical and chemical properties of the studied soil profiles: Soil texture, bulk density, total C, N and S (% dry weight) pH (H_2O, 1:2.5) and potassium extracted by ammonium lactate (pH 3.75) and HCl (2 M boiling). Modified from Rosén et al. (1999). Values on K-AL taken from Larsson (2008)

Site/ horizon	Depth cm	Bulk density g cm^{-3}	Particle size distribution (mm) %			C%	N%	S%	pH (H_2O)	K-AL mg 100 g d.w.$^{-1}$	K- HCl mg 100 g d.w.$^{-1}$	CEC$_{eff}$	CEC$_{pH7}$
			sand 2-0.05	silt 0.05-0.002	clay < 0.002								
Möjsjövik, Typic Sulfihemist (Soil Survey Staff, 1992); Thionic Histosol (FAO,1998)													
Hap	0-20	0.33	—	—	—	37.30	2.29	0.22	5.5	20.3[a]	87	—	—
He1	20-50	0.15	—	—	—	37.40	1.90	0.49	5.4		34	—	—
He2	50-75	0.12	—	—	—	40.90	1.93	0.68	5.3		52	—	—
Hi	75-85	0.10	—	—	—	42.50	1.79	0.66	5.3		64	—	—
Har	85-100	0.14	—	—	—	28.60	2.08	1.4	5.0		149	—	—
2Cr	100-110	0.39	—	—	—	—	—	—	5.4		402	—	—
Skogsvallen, Typic Dystrochrept (Soil Survey Staff, 1992); Dystric Cambisol (FAO,1998)													
Ah1	0-3	0.72	15[b,c]	43[b,c]	42[b]	12.70	1.04	0.015	5.2	14.4[a]	317	—	—
Ah2	3-5	1.40	—	—	—	3.40	0.33	0.002	5.3		363	—	—
Bw1	5-20	1.32	15	42	42	1.90	0.18	<0.001	4.7		398	—	—
Bw2	20-35	1.37	13	40	46	1.30	0.13	<0.001	6.2		467	—	—
BC1	35-50	1.39	12	40	48	1.30	0.13	<0.001	6.5		460	—	—
BC2	50-75	1.35	6	42	52	0.70	0.09	<0.001	7.4		606	—	—
C	75-95	—	1	45	54	0.20	0.04	<0.001	7.7		637	—	—
Hille, Terric Sulfihemist (Soil Survey Staff, 1992); Thionic (Terric) Histosol (FAO,1998)													
Hap	0-25	0.34	—	—	—	26.70	1.74	0.14	5.9	11.0[a]	36	—	—
He1	25-33	0.16	—	—	—	36.90	1.88	0.29	5.5		28	—	—
Hi	33-47	0.19	—	—	—	44.70	2.07	0.39	4.3		15	—	—
2Cg	47-55	0.29	—	—	—	11.50	1.20	0.66	3.1		190	—	—
2Cr(g)	55-105	0.44	—	—	—	5.20	0.65	1.94	2.9		355	—	—

Appendix 2 (continued)

Site/ horizon	Depth cm	Bulk density g cm^{-3}	Particle size distribution (mm) %			C%	N%	S%	pH (H_2O)	K-AL mg 100 g d.w.$^{-1}$	K- HCl mg 100 g d.w.$^{-1}$	CEC$_{eff}$	CEC$_{pH7}$
			sand 2-0.05	silt 0.05-0.002	clay < 0.002								
Ramvik, Typic Udorthent (Soil Survey Staff, 1992); Eutric Regosol (FAO,1998)													
Ah1	0-7	—	47[b]	42[b]	12[b]	4.20	0.30	<0.001	5.4	8.6[a]	145	—	—
Ah2(Ap)	7-23	1.27	44	48	8	1.80	0.15	<0.001	5.6		130	—	—
Bw	23-24	1.41	32	57	11	0.80	0.07	<0.001	5.6		165	—	—
CBg	34-35	1.38	52	40	8	0.30	0.03	<0.001	5.8		164	—	—
Cg1	55-57	1.55	21	66	13	0.20	0.02	<0.001	5.8		231	—	—
Cg2	75-100	1.56	16	71	13	0.20	0.02	<0.001	5.6		228	—	—
Hammarstrand, Typic Udorthent (Soil Survey Staff, 1992); Eutric Regosol (FAO,1998)													
Ah1	0-6	0.68	34	56	10	4.13	0.29	0.037	6.3	3.7[a]	102	10.9	21.0[d]
Ah2(Ap)	6-20	1.12	38	57	5	1.33	0.11	0.02	6.1		70	5.8	10.3[d]
Ah3	20-30	1.24	47	51	2	0.53	0.03	0.002	6.2		60	4.0	6.1[d]
C1	30-50	1.34	47	51	2	0.25	0.01	0.01	6.4		46	3.1	4.4[d]
C2	50-100	1.27	49	49	2	0.21	0.01	0.003	6.6		44	3.2	4.4[d]
Stora Blåsjön, Typic Cryorthent (Soil Survey Staff, 1992); Dystric Regosol (FAO,1998)													
Oi	0-2	0.91	—	—	—	16.60	0.97	0.104	5.9	4.7[a]	103	—	—
Ah	2-4	1.29	54	36	10	4.33	0.32	0.041	6.2		52	9.2	—
Ah/E/B(Ap)	4-15	1.12	56	39	5	1.62	0.12	0.017	6.5		51	3.1	9.6[e]
Bs(Bw)	15-35	1.32	58	39	3	1.05	0.08	0.011	6.7		66	1.9	7.4[e]
BC	35-60	1.55	58	39	3	0.56	0.00	0.006	6.0		69	1.0	6.2[e]
CG	60-100	—	63	36	1	0.13	0.00	0.002	6.1		96	0.4	1.6[e]

[a] The analysis was carried out on the horizon 0-20 cm

[b] The Skogsvallen and Ramvik soils were analysed by the pipette method; sand 2-0.06 mm; silt 0.06-0.002 mm; clay < 0.002 mm.

[c] The analysis was carried out on the horizon 0-5 cm.

[d] CEC$_{pH7}$ = Ca(OAc)2 (pH 7).

[e] CEC$_{pH7}$= Ca + Mg + K + Na+ titratable acidity.

Appendix 3

Transfer factors TFg *10^{-3} (m^2 kg^{-1} d.w.) and absolute activity in grass (Bq kg^{-1} d.w.) for all locations and years of sampling. Values from previous years taken from Jansson (2004) except for Hille and Möjsjövik

	Möjsjövik		**Skogsvallen**		**Hille**		**Ramvik**		**Hammarstrand**		**Stora Blåsjön**	
Year	TFg* 10^{-3}	Bq/kg d.w.	TFg* 10^{-3}	Bq/kg d.w.	TFg* 10^{-3}	Bq/kg d.w.	TFg* 10^{-3}	Bq/kg d.w.	TFg* 10^{-3}	Bq/kg d.w.	TFg* 10^{-3}	Bq/kg d.w.
1987	28.3	2120	20.5	1862	62.7	12094	3.0	155	—	—	—	—
1989	—	—	—	—	—	—	—	—	8.2	298	88.5	2946
1990	—	—	—	—	12.7	1865	—	—	—	—	—	—
1992	—	—	1.6	152	—	—	—	—	—	—	—	—
1994	4.4	232	0.4	40	4.2	746	0.9	50	2.1	77	—	—
1995	—	—	—	—	—	—	—	—	—	—	13.8	435
2000	4.7	312	0.2	16	3.1	461	0.3	19	0.8	49	9.8	405
2002	—	—	—	—	1.1	165	—	—	—	—	—	—
2003	—	—	0.04	5	—	—	0.5	14	0.5	19	8.6	285
2004	7.1	381	0.05	5	—	—	—	—	—	—	—	—
2005	—	—	—	—	0.4	69	—	—	—	—	—	—
2007	7.0	333	0.04	3	0.6	92	0.2	12	0.6	14	6.6	170

The activity of ^{137}Cs (kBq m^{-2}) in each sampled layer for all sites in 2007, with standard deviations. Data corrected for decay back to the time of fallout (1/5 1986)

Depth (cm)	**Möjsjövik**	**Skogsvallen**	**Hille**	**Ramvik**	**Hammarstrand**	**Stora Blåsjön**
0-1	8862 ± 956	9346 ± 654	16152 ± 2052	5 481 ± 790	3 961 ± 1013	11 947 ± 1341
1-2	9523 ± 5278	14054 ± 1232	14842 ± 381	6 279 ± 765	7 238 ± 1616	13 664 ± 1328
2-3	13363 ± 646	14538 ± 2819	15009 ± 531	6 506 ± 1154	7 905 ± 1999	8 302 ± 2629
3-4	10627± 2456	14571 ± 3288	16849 ± 2935	7 636 ± 829	7 269 ± 782	3 309 ± 1406
4-5	6388 ± 860	12215 ± 1865	17942 ± 2264	7 762 ± 95	4 653 ± 786	1 894 ± 1007
5-6	5320 ± 537	9060 ± 1536	17236 ± 5500	7 807 ± 746	3 483 ± 178	1 307 ± 721
6-7	4293 ± 1646	6527 ± 986	15549 ± 5485	7 359 ± 87	2 692 ± 601	894 ± 496
7-8	3172 ± 1402	4227 ± 527	14302 ± 6624	7 040 ± 186	1 338 ± 556	502 ± 255
8-9	1828 ± 507	3492 ± 405	12413 ± 2924	5 431 ± 894	721 ± 272	502 ± 340
9-10	1270 ± 405	1725 ± 1160	9004 ± 2143	4 660 ± 417	343 ± 133	381 ± 172
10-12.5	3355 ± 1487	6207 ± 1710	20796 ± 3983	9 193 ± 376	539 ± 323	684 ± 130
12.5-15	1956 ± 543	4092 ± 2062	21055 ± 10141	5 460 ± 1746	281 ± 288	481 ± 144
15-17.5	2172 ± 1266	4925 ± 5551	12924 ± 4337	3 115 ± 1002	305 ± 278	257 ± 51
17.5-20	1151 ± 600	2717 ± 3239	9117 ± 2839	3 034 ± 508	17 ± 15	339 ± 181
20-22.5	816 ± 278	1244 ± 1054	5963 ± 1778	1 948 ± 617	0 ± 0	437 ± 376
22.5-25	564 ± 272	626 ± 516	3803 ± 2124	854 ± 642	0 ± 0	252 ± 48
25-27.5	549 ± 288	233 ± 122	3747 ± 670	462 ± 74	0 ± 0	197 ± 43
27.5-30	571 ± 236	247 ± 198	4451 ± 1920	206 ± 41	27 ± 46	168 ± 119
30-32.5	490 ± 385	501 ± 463	4048 ± 3759	96 ± 166	1 ± 1	120 ± 138
32.5-35	623 ± 622	560 ± 724	3060 ± 3026	108 ± 41	0 ± 0	100 ± 48
35-37.5	814 ± 958	497 ± 784	2156 ± 1988	90 ± 155	8 ± 14	53 ± 93
37.5-40	231 ± 145	238 ± 310	1202 ± 569	52 ± 91	2 ± 3	24 ± 20
40-42.5	188 ± 141	321 ± 321	812 ± 291	0 ± 0	0 ± 0	45 ± 70
42.5-45	132 ± 60	64 ± 110	662 ± 473	0 ± 0	0 ± 0	27 ± 46
45-47.5	93 ± 84	54 ± 65	791 ± 625	28 ± 48	0 ± 0	0 ± 0
47.5-50	74 ± 20	105 ± 93	907 ± 861	0 ± 0	0 ± 0	0 ± 0
50-52.5	50 ± 74	217 ± 95	372 ± 238	0 ± 0	2 ± 4	0 ± 0
52.5-55	30 ± 34	66 ± 16	229 ± 224	0 ± 0	0 ± 0	0 ± 0
55-57.5	41 ± 71	190 ± 91	108 ± 106	0 ± 0	0 ± 0	0 ± 0
57.5-60	42 ± 14	131 ± 114	163 ± 154	0 ± 0	0 ± 0	0 ± 0
Total	**78586 ± 22272**	**122991 ± 32100**	**245665 ± 70944**	**90606 ± 11471**	**40787 ± 8910**	**45886 ± 11203**