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Downward migration and transfer to plants of radiocaesium in Scottish soil profiles
- A comparison with earlier studies

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Keywords: Radiocaesium, migration rate, migration depth, transfer factors, deposition, Scotland

Cover: My supervisors Klas and Charlie performing field work at Rora Moss, photo by author

Abstract

Following the accident at the nuclear power plant in Chernobyl in 1986, great volumes of ^{137}Cs were distributed over large parts of the former Soviet Union and Europe. Over Scotland, radiocaesium was deposited as wet deposition. Due to the relatively long physical half-life of 30 years, there are still measurable activity concentrations in the natural environment. This study was made on two organic soils, Knockandy Hill and Rora Moss, situated in the eastern part of the central highlands of Scotland. At these two sites, studies on migration and transfer between soil and plants were conducted. The aim of the thesis was to present the current state of radiocaesium in the Scottish soils and to compare these data with data from earlier studies, however, it was difficult to make any implications from this study due to variations in sampling techniques. Another aim was to study the transfer of ^{137}Cs from soil to plant. Caesium resembles potassium and is readily fixed to clay minerals in the soil and weakly bound to organic matter. Plants have a difficulty in discriminating between the two elements, hence the uptake of caesium can be considerable. This study has, in accordance with other similar studies, shown that the transfer of radiocaesium to heather (*Calluna vulgaris)* is significantly higher than the transfer to different grass species. Concerning the movement of caesium in the soils no significant difference could be found between the two organic soils. Their migration depths were 8.2 and 8.8 cm and their migration rates were 0.36 and 0.39 cm y^{-1} for Knockandy Hill and Rora Moss, respectively.

Keywords: Radiocaesium, migration rate, migration depth, transfer factors, deposition, Scotland

Sammanfattning

Kärnkraftsolyckan i Tjernobyl 1986 resulterade i att betydande mängder ^{137}Cs spreds över stora delar av det forna Sovjetunionen samt Europa. Över Skottland deponerades ^{137}Cs framförallt genom nederbörd. På grund av radiocesiums långa fysikaliska halveringstid på 30 år finns det fortfarande mätbara aktivitetsnivåer kvar i naturen. Den här studien innefattar migration i markprofilen och överföring av radiocesium mellan mark och växt på två organiska jordar, Knockandy Hill och Rora Moss. Dessa två jordar är belägna i den östra delen av det centrala skotska höglandet. Syftet med studien var att visa på det nuvarande cesiumtillståndet i de skotska jordarna och även att jämföra dessa data med data från tidigare studier, vilket visade sig vara svårt på grund av variationer i provtagningsteknik. Cesium liknar kalium och fixeras därmed med lätthet till lermineral och binds endast utbytbart till organiskt material. Växter har svårt att skilja mellan dessa ämnen och överföringen via rötterna kan bli betydande. Studien visade, i likhet med andra liknande studier, att överföringen var signifikant större till ljung (*Calluna vulgaris)* än till olika gräsarter. Angående hur cesium rör sig i marken kunde ingen signifikant skillnad konstateras mellan de båda organiska jordarna. Migrationsdjupen var 8,2 och 8,8 cm och migrationshastigheten var 0,36 och 0,39 cm $år^{-1}$ vid Knockandy Hill respektive Rora Moss.

Contents

1. Introduction

The Chernobyl accident which occurred on 26th of April 1986 did not only affect the immediate surroundings of the power plant, but large areas in Europe were contaminated by windborne fission products.

For the general public, ^{131}I and ^{137}Cs are the elements of most concern when it comes to radiation exposure in event of an accident. Generally, the ^{137}Cs contamination was the greatest problem after this accident. ^{137}Cs is of particular scientific interest, since it has a physical half-life of 30 years. Thus it will remain in the environment for several decades, while ^{131}I has a half-life of just 8 days. Once in soil, caesium is readily fixed to clay minerals and forms reversible bonds with organic matter. Hence, the clay and organic matter content of the particular soil affects the availability of caesium for plants. Because of its chemical similarity to potassium, caesium is readily taken up by plants. Thus, it can easily contaminate the food chain. Long-term exposure to high doses of radioactivity increases the risk of developing cancer.

Partly to improve the current countermeasures and also to get better knowledge of what to do and what interventions to apply in case of another accident, many studies have been conducted to enhance the knowledge of how caesium behaves in soil and how it is distributed to plants. However, there is a need for further investigations to improve the effectiveness of the countermeasures even more.

1.1 Aim

The aim of the project was to present the current status of Scottish soils and plants, regarding the radiocaesium originating from the accident in the nuclear power plant in Chernobyl, Ukraine in 1986 and to compare the results with earlier obtained data from Scottish sites sampled in 1990 and 1999. Another aim of the project was to study the migration of radiocaesium in the soil and the transfer from soil to plant.

2. Background

2.1 Radiation

Ionizing radiation has existed on earth ever since the Big Bang. The ionizing radiation that surrounds us today originates partly from cosmic radiation from outer space but also from radioactive isotopes in the ground, in the water and in both living and dead material. The most common sources of radiation come from the unstable isotopes of potassium, carbon, uranium and thorium and their disintegration products. ^{40}K and ^{14}C are special in the way that they commonly occur in the human body, just like their stable analogues. ^{40}K accounts for 0.01% of all naturally existing potassium (Jansson & Rydén 1994). Thorium and uranium occur in the ground both bound to minerals and as dissolved ions, ready to either be absorbed to plants or to be transported with the soil water flow. The levels of thorium and uranium in bedrock differ between places, hence the indoor activity concentration at a particular site depends on the underlying geology, building material, type of house and also the ventilation system. (Andersson *et al*. 2007)

Exposure to cosmic radiation increases with altitude, which means that individuals are subjected to enhanced levels of radiation during a flight, compared to travelling on the ground. Therefore, cabin crew constitutes a risk category. (Andersson *et al.* 2002)

There is also man-made ionizing radiation. The largest contribution to the world's population is the radiation remaining from the testings of nuclear weapons in the atmosphere from 1945 to the 1980´s. The testings resulted in vast release of radioactive materials that were widely dispersed in the atmosphere and deposited all over the surface of the earth. Worldwide monitoring networks have estimated that ^{90}Sr and ^{137}Cs were the main fission products released from the testings. The average annual doses received by the human population are about 10 % higher in the northern hemisphere than in the southern hemisphere because most of the testing took place there. (Andersson *et al*. 2002)

Another big contributor to the exposure of radiation is the use of x-rays in hospitals and dental care. The metal most commonly used for the production of x-rays is a tungsten isotope (Jansson & Rydén 1994). Other radio nuclides utilized in medicine are ^{99m}Tc (Sundell & Sundgren 2008), ^{11}C, ^{18}F and ^{75}Se; these nuclides have very short half-lives and are used as radioactive markers that can be attached to substances such as amino acids and traced inside the body (Leide-Svegborn *et al*. 2007). The amount of exposure per person is individual depending on health and teeth status. Apart from man-made radiation sources like hospitals, nuclear testings and accidents at nuclear power plants, some radiation come from research and the industry (Andersson *et al*. 2007).

2.2 Ionizing radiation

The radioactive, unstable, nuclides disintegrate in a process where one element is transformed into another, the excess energy of the nuclide being dissipated as electromagnetic radiation or as particles. Consequently, radiation is a kind of transport of energy. (Isaksson 2002)

Ionizing radiation is radiation that has enough energy to be able to eject electrons in the exposed material from their normal positions, thus, the atomic nucleus will no longer be uncharged. The phenomenon is naturally occurring in the environment. There are three different kinds of ionizing radiation; alpha, beta and gamma radiation. (Andersson *et al*. 2002)

2.2.1 Alpha radiation

Alpha radiation consists of a helium nucleus that is two neutrons and two protons. The spreading range of an alpha particle is a couple of centimeters and it can easily be blocked by *e.g*. a piece of paper. An example of an alpha radiating element is plutonium (Andersson *et al*. 2002). The alpha particle is a stable unit and when it hits the surrounding media it will pick up two electrons and become an uncharged helium atom. Because of the big difference in masses between the alpha particle and the original nucleus, the alpha particle will receive the highest velocity and the largest part of the kinetic energy. (Isaksson 2002)

2.2.2 Beta radiation

Iodine, caesium and strontium emit beta radiation, as well as gamma radiation. Beta radiation is constituted by an electron (β-) or a positron (β+). It has, in common with gamma radiation that the mass number of the nucleus does not change. However, emittance of *e*.g. β- radiation, will increase the atomic number by one unit, *e.g*; ^{137}Cs will decay into ^{137}Ba (Isaksson 2002). Beta radiation has a longer range than alpha radiation and travels about ten meters in air. The radiation can be blocked by *e.g*. a window pane. (Andersson *et al.* 2002)

2.2.3 Gamma radiation

Gamma radiation differs from alpha and beta radiation, since no particles are emitted. Instead, electromagnetic radiation, photons are emitted and they have the longest range. Gamma rays can travel hundreds of meters in air. Examples of gamma emitting elements are iodine and caesium. To reduce the radiation 0.1 m of lead, or 0.50 m of concrete is needed. (Andersson *et al*. 2002)

2.2.4 Becquerel, sievert and gray

Radioactivity concentration can be measured using different units. The most commonly used nowadays are the becquerel (Bq), sievert (Sv) and gray (Gy) units. Becquerel quantifies the number of decays per second, whereas gray is a unit for the amount of energy that has been absorbed in the body per unit of weight. Gray is also called adsorbed dose. One way of explaining gray is to say, if 100 people are exposed to 1 Gy, 5 % are likely to develop cancer. (Johansson 1996)

Since the three different kinds of radiation have varying biological effects, quality factors for alpha, beta and gamma radiation have been introduced. Thereby, you can get a measure of the biological effect of the dose. This is usually measured in mSv; 1 mSv equals 75 000 Bq taken orally. The effect of the radiation is also depending on what tissue that has been hit, different tissues are differently susceptible to radiation. (Johansson 1996)

2.2.5 Half-lives

Half-life is measured in time units. It is the period of time which it takes for 50 % of the nuclei in a sample to decay. The specific physiological half-life depends on the nature of the radionuclide and it can not be altered through any kind of management. However, the decay behavior of a single nucleus can not be predicted (Isaksson 2002). There are also biological and ecological half-lives. Biological half-life of an element refers to the time it takes for 50 % of the element to be excreted from a body. This reduction is influenced by the physical half-life, but also to biological processes in the body. These processes are dependent on age, sex and the physical condition of the exposed and can not be specified. Ecological half-life reflects the half-life of a radioactive substance in plants, animals and fungi living in a particular area. The ecological half-life depends on how the substance behaves in the soil, how easily it is entered in to the food-chain and also how fast it is excreted from that particular area. (Andersson *et al*. 2002)

2.3 Dispersion of anthropogenic radioactivity concentration in the environment

In case of an accident where radioactive particles are released to the atmosphere, the radioactive content of the reactor can be spread long distances, depending on the development of the temperature of the reactor core, the course of events and the weather conditions. Wet deposition is the most effective way of deposition of airborne contamination. When it rains or snows, the nuclides are washed out of the air and deposited onto the ground. Dry deposition occurs when the weather is dry and either the heavy particles falls to the ground due to the laws of gravity or when the cloud containing radioactivity passes the ground at low altitudes. When the cloud passes forests and fields, some of the nuclides are trapped in the vegetation and deposited there. Though, in a heavy rainfall, up to 90 % of the nuclides might be reallocated to the ground layer. (Johansson 1996)

After a fallout event, the growth stages and morphology of the plants determine the soil coverage and hence the proportion of the radionuclides that gets caught on the plants. Lichen, mosses and plants with spacious leaf areas and a significant coverage of the ground are examples of plants that will catch a large part of the fallout. The mode of deposition is of importance here. Dry deposition will leave a larger contribution of the fallout on the leaves than will wet deposition, since the latter will wash off the particles and hence transfer the fallout to the ground. This phenomenon is of big importance when it comes to the introduction of countermeasures to influence fallout on agricultural crops. (Andersson *et al*. 2002)

2.4 The Chernobyl accident

2.4.1 The turn of events

At the time of the accident, there were four nuclear energy producing reactors in Chernobyl. In one of them, experiments were taking place immediately before the accident. The reactor number four was run outside its authorized management and the safety systems had been switched off. The accident happened at 01:23, local time, on the 26th of March, 1986. The aim of the experiment was to study if and how the turbine could

deliver power in case of loss of the ordinary supply. The experiment went wrong and the effect in the reactor suddenly increased, leading to vaporization of the cooling-water. This resulted in an even higher temperature in the reactor and finally an explosion, tearing the reactor apart. The explosion caused fire in the graphite moderators and the building material of the reactor and it kept on burning for 10 days. (Blixt & Moberg 2005)

2.4.2 Dispersion of the content

Releases of radionuclides continued over a 10-day period, though with varying rates. On the first day the release was at its maximum, due to the explosion. The following days the radionuclides dispersed following the fumes and hot air coming from the burning graphite. (UNSCEAR 2002)

The total core load of the reactor at the time of the accident was 190.3 tons. An inventory of radionuclides left in the reactor core and analysis of air and fallout samples have led to estimations of the amounts of radionuclides released in the accident. Approximately 29 % of the core load was released to the surroundings, for estimated release of radionuclides see table 2.1. (UNSCEAR 2002)

Table 2.1. Half-life and estimated total release (PBq) of some of the radionuclides during the course of the Chernobyl accident (Modified from UNSCEAR, 2000)

Nuclide	Half-life	Activity concentration released
^{134}Cs	2.06 y	~54
^{137}Cs	30.0 y	~85
^{131}I	8.04 d	~1 760
^{133}Xe	5.25 d	6 500
^{90}Sr	29.12 y	~10
^{235}U	$4 \cdot 10^9$ y	no data
^{239}Pu	24 065 y	0.03

The contamination of radionuclides in the surroundings of the reactor differs from that far away. Volatile and light elements like caesium, iodine and tellurium were dispersed long distances, while the deposition in the vicinity of the reactor resembles the composition of the fuel and large particles and fuel elements e.g. uranium and plutonium were deposited there. (UNSCEAR 2002)

Caesium and the other radionuclides were spread in the environment after the Chernobyl accident, either because they were thrown thousands of meters up into the air due to the powerful generation of heat that was produced in the core or in any of the explosions that followed. These nuclides followed the wind and were swept away across Europe and the former Soviet Union (Johansson 1996). Apart from the neighboring countries, Ukraine, Russia and Belarus, mostly the Nordic countries were affected, but also Austria and Bulgaria. (Blixt & Moberg 2005)

There are sometimes interhemispheric air movements taking place between the southern and the northern hemisphere, but this seems not to be the case this time, since no

contamination from the Chernobyl accident have been detected in the southern hemisphere. (UNSCEAR 2002)

2.5 Nuclear power plants

Today, there are about 500 nuclear reactors spread all over the world, of which 39 are situated in United Kingdom (EA *et al*. 2008). They include a number of different thermal reactor types such as the pressurized water reactors, the boiling water reactors and the special reactor systems (Choppin *et al. 2002*).

In case of an accident in a nuclear reactor, caesium and iodine constitute the most dangerous fallout that reaches the ground. The radioactive noble gases that are produced in the reactor core are diluted into harmlessness in the atmosphere, when released into the air. (Andersson *et al.* 2002)

2.6 ^{137}Cs

Caesium is formed as an intermediate product in one of the chain reactions following the splitting of ^{235}U in a nuclear power plant, see reaction process below. ^{137}Cs has a half-life of approximately 30 years and decays in a two step process into stable barium. The decaying process of caesium results in emission of both β- and γ- radiation. (Isaksson 2002)

$$^{235}_{92}U \rightarrow {}^{1}_{0}n + {}^{236}_{92}U \rightarrow {}^{137}_{53}I + {}^{95}_{39}Y + 4\,{}^{1}_{0}n$$

$$^{137}_{53}I \xrightarrow{\beta-} {}^{137}_{54}Xe \xrightarrow{\beta-} {}^{137}_{55}Cs \xrightarrow{\beta-} {}^{137m}_{56}Ba \xrightarrow{\gamma} {}^{137}_{56}Ba\,(stable)$$

Caesium can easily be transferred via soil to plants to animals or humans. Caesium has similar chemical properties to potassium and follows its lookalike both in the soil and in the food web. Humans are very good at absorbing caesium, almost 100 % of what reaches the gastrointestinal tract is absorbed. (Andersson *et al*. 2002)

There is no known role for caesium in plants, but at soil solution levels exceeding 200μM it becomes slightly toxic, depending on the level of other ions in the solution. At extremely high levels it will produce necrosis in shoots and roots. (White & Broadley 2000)

2.6.1 Binding in soil

Caesium is a univalent cation and because of this, it is easily adsorbed to negatively charged constituents in the soil. Just like similar cations, especially potassium, but also ammonium, it has a high potential for adsorption to clay minerals, illites in particular. The absorption is related to the small hydration energy of the caesium ion. In event of adsorption, caesium ions lose their hydration water and form stable inner-sphere complexes. The strong affinity for illites comes from the presence of frayed edge sites (FES), which account for a minor part of the cation exchange capacity of the illites (Dumat *et al*. 1997). This has also been concluded by Cremers *et al*. (1988). Caesium has a high affinity for FES and above all in 2:1 phyllosilicate, like partly weathered illites.

The weathering of the layered clay mineral illite is a slow process where the layers expand and the potassium, which is usually trapped between the layers, in the interlattice, is exchanged for other positive ions, see figure 2.2. The fixation of cations increase with time as they penetrate the interlattice (Krouglov *et al*. 1998). It has become evident that even if the illites only make up for a small part of the clay fraction, its adsorption capacity of caesium is still of great importance (Cremers *et al*. 1988).

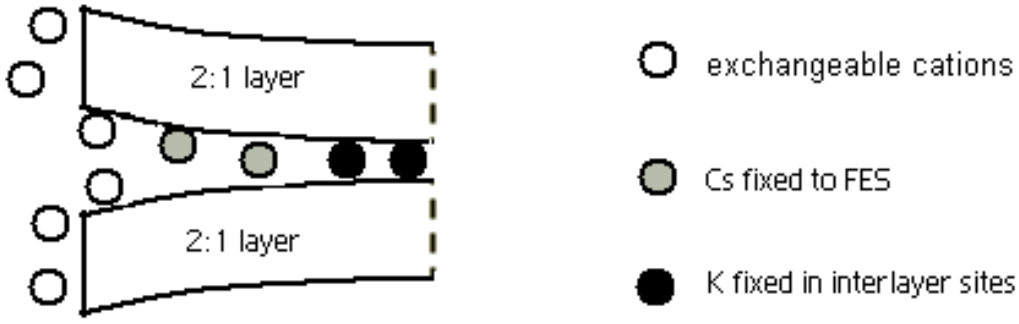

Figure 2.2. Schematic picture of weathering of illite. Potassium is released through weathering and caesium can be fixed to the original potassium binding positions at the frayed edge sites and the interlattice binding sites (Persson 2008).

Rosén *et al.* (2006) conducted a study concerning the fixation of caesium in clay added to a peat and a peaty podzol during repeated wetting and drying conditions. Repeated wetting and drying of incubated soils in a laboratory is a way of simulating natural conditions. The process triggers the penetration of radiocaesium into the interlattices of the clay minerals. The caesium fixation can be determinedby extraction with ammonium acetate (CH_3COONH_4). The study showed that a smaller fraction of caesium was available for exchange with ammonium acetate in treatments where clay was added.

In a study of sorption behaviour in different soils conducted by Shenber & Eriksson (1992), it was concluded that the transfer factor of caesium was strongest correlated to the clay content. But also the cation exchange capacity, pH-value and organic matter content influenced the distribution coefficient, though in a smaller extent. Studies have also shown that the proportion of sorbed caesium increases with the clay content. Though, in organic soils, there might not be sufficient illitic clay minerals to fix all the caesium and those not fixed will be competing with the other cations over ion exchange sites, hence the caesium concentration in the soil solution will depend on other ionic components being present (Kudelsky *et al*. 1996).

2.6.2 Binding to organic matter

Organic matter has only little selectivity for caesium since it is not likely to form complexes with organic molecules. The adsorption of caesium to organic matter is determined by the cation exchange capacity of the specific organic substance and thus the concentration of caesium in relation to other competing cations. Studies conducted by Dumat *et al*. (1997) suggest that clay minerals prevent caesium from binding to organic matter or that the binding to organic matter is so weak that it is easily reversible. This was also the opinion of Maguire *et al*. (1992) who studied three different clays; illite, kaolinite

and bentonite, and their role in the sorption of caesium to organic matter at different pH-values. They found that the humic acids or their functional groups bonded to the clays, preventing the sorption of caesium to the exchange sites of the humus. According to Shand *et al*. (1994) this is just an intermediate state in the journey from being exchangeable and available to the biota, to being fixed to minerals. Maguire *et al.* also stated that it is difficult to obtain organic matter that is free from mineral particles, hence they find it irrelevant to talk about caesium being bound to humic substances via complexation.

2.6.3 The influence of the pH-value

Wauters *et al*. (1994) found that there is no direct relation between sorption of caesium and the pH-value. On the contrary, Giannakopoulou *et al*. (2007) found that the caesium sorption had a maximum at a pH-value of 8 when studying mineral soils. Their explanation was that at this pH-value there is high negative charge at the exchangeable binding sites, making it possible for a larger number of ions to be sorbed. According to Cremer *et al*. (1998), the affinity of caesium to FES is of far greater importance than the fixation to exchangeable binding sites. Though, the pH-value is related to the capacity for ion exchange in the soil, the CEC of an *e.g*. organic soil increases with increasing pH-value, hence will the sorption behavior be influenced in the long run (Wauters *et al*. 1994).

In an experimental screening study on a sandy soil, a loamy sand, a loam and a clay soil, Wauters *et al.* (1994) concluded that in mineral soils only a fraction of caesium will be present at readily reversible ion exchange sites, while most of the caesium ions will be bound to specific sites. The authors also point out the importance of potassium and the positive relationship between high calcium and magnesium content versus the content of potassium resulting in an accelerating fixation of caesium.

2.6.4 Migration in the soil profile

Migration is the transport of a substance within the soil profile, either through ion transport in the percolating soil solution or as colloids or particulate matter (Forsberg 2000). The net flow downward with percolating water is counteracted by other processes *i.e*. mechanical mixing or bioturbation caused by earth living organisms and also uptake by roots or fungi in the soil (Bergman 1994). Adsorption is one factor deciding the speed of the downward migration of a substance (Gustafsson *et al*. 2007).

Long-term studies conducted by Rosén *et al.* (1999), Arapis *et al*. (1997) and Isaksson *et al*. (2001) show that most of the radiocaesium from the Chernobyl accident still lingers in the uppermost ten cm of the profile. These studies were conducted on a large range of soil types.

2.6.5 Uptake by roots

A consequence of the slow migration is that a large part of the caesium in a soil profile after fallout will be lingering in the root zone for many years to come and is thereby easily accessible for plant uptake. Because of the chemical resemblance to potassium, the roots of plants are unable to distinguish thoroughly between caesium and potassium.

However, plants do show a slight preference to potassium. The longer the duration in the soil, the more caesium becomes fixated to clay minerals, hence less will be available to plant uptake (Rosén *et al*. 1999). The uptake of caesium is related to the available amount in the soil, *i.e*. the amount of caesium being solved in the soil solution and the amount being exchangeable (Eriksson *et al.* 2005). Anyhow, there are similar mechanisms in the uptake process which makes potassium a competitive blocker of caesium uptake. (Zhu 2001)

When ^{137}Cs contaminates the soil, it can enter the food-chain through plant uptake and become a long lasting burden. However, the plant availability of ^{137}Cs in soils varies greatly between soil types. In trials conducted by Smolders *et al.* (1997), they found that the plant-soil concentration ratio for ^{137}Cs varied from 0.002 to 2.6 g g^{-1} between different soils types, and that the potassium concentration in the soil significantly affected the ^{137}Cs concentration ratio. The soil with the highest concentration ratio was a histosol, while the one with the lowest was a fluvisol. (Smolders *et al*. 1997)

The water content of the soil influences the plant uptake of nutrients and also the uptake of caesium. Bogs, for example, are rich in organic matter and are saturated with water, here caesium is mobile and available for plant uptake (Kudelsky *et al*. 1996). Hence a greater fraction of radiocaesium will be available for the plant. The study by Kudelsky *et al*. also showed that the runoff of radiocaesium from peat bogs, eight years after the accident in Chernobyl, were much greater than the runoff from unsaturated soils containing more minerals, hence a greater fraction of radiocaesium will be available for the plants.

Another factor affecting the root uptake is the weather conditions. The roots are always looking for moisture. During warm and dry years they will have to penetrate greater depths than usual to reach it. While during more humid conditions, the root system will be much shallower, leading to a greater uptake of radiocaesium. (Mascanzoni 1988)

2.7 Transfer to plants and animals

To be able to compare results from different sites, transfer factors are used. A transfer factor is simply the ratio between concentrations of a radionuclide in two media, *e.g.* plants and soil. The varying deposition levels between different sampling sites can be neglected. The transfer factor is calculated by using formula 3 shown in paragraph 3.4.3, where the ground deposition is presented as the total deposition at the site. By using transfer factors the transfer from *e.g*. soil to grass to cow to milk and finally to humans can be calculated and thereby estimations can be made to see if concentrations of radioactive elements in the different foodstuff are below the authority set limits. For the acceptable limits set by the European Union for different kinds of foodstuff, see table 2.3. (Rosén *et al.* 1999)

Table 2.3. Acceptable limits set by the European Union for foodstuff, in Bq kg^{-1} (Modified from Persson 2008)

Nuclides	Example	Baby food	Dairy products and liquid foodstuff	Other foodstuff	Less common foodstuff
Alfa-emitting	^{239}Pu	1	20	80	800
All non natural nuclides, half-life > 10 d	^{134}Cs ^{137}Cs	400	1 000	1 250	12 500
Strontium isotopes	^{90}Sr	75	125	750	7 500
Iodine isotopes	^{131}I	150	500	2 000	20 000

2.8 Biological influence of ionizing radiation

The influence of ionizing radiation is usually divided into stochastical and deterministical effects. The stochastic effects are also called late effects since they appear a relatively long while after the irradiation. It is the likelihood of the effect to appear that is dealt with, not the magnitude of it. Stochastic effects comprise cancer and genetic disorders. Deterministic effects on the other hand, deal with the magnitude of the effect and the aggravation of the effects along with higher radiation doses. It covers nausea, hair loss and damaged blood-producing tissues and is the consequence of powerful ionizing radiation of the whole body. A specific dose produces a specific deterministic effect of a tissue. (Isaksson 2002)

Because of the mending ability of the cells, short-term irradiation is more deleterious than long-term irradiation, when exposed to the same dosage. The dose of short-term irradiation where 50 % of the exposed population dies (LD_{50}) is about 4 Gy (Isaksson 2002). Though, occasions when these high dosages are reached are extremely rare. The average radiation dose for a typical person in the United Kingdom is 2.4 mSv per year. (EA 2008)

2.8.1 The reactive radicals of water

When ionizing radiation hits living cells, it usually interacts with a water molecule, since water constitutes 60 - 70 % of the cells. Usually, the water in the cell is split into an ionized water molecule and a free electron. The ionized water molecule is rapidly converted into the radicals of water; hydroxyl radical, hydrogen radical and electron. These radicals either form water, hydrogen peroxide, hydrogen gas or react with the biomolecules that are present in the cell. Even if the probability is low, the radicals might react with the DNA, which comprises 1 % of the cell content and this might result in mutations, hence being the beginning of cancer. (Johansson 1996)

2.8.2 The sensitive proliferate cells

Cells that often go through cell differentiation are more sensitive to radiation than other cells. An example of such tissues is the red bone marrow, spleen and gastro-intestinal tract. The red bone marrow produces granulocytes, which makes up the immune system in the body. If a person gets exposed to a dosage of 3 - 4 Gy, the immune system will be almost totally eliminated for about a week and the individual will be very susceptible to infections. The trombocytes, also called the red blood platelets, are also created in the red

bone marrow. Trombocytes make the blood coagulate in case of a wound. (Johansson 1996)

2.9 Caesium levels in Scotland

On the 3rd of May, 1986, a cloud containing radioactive materials drew past Scotland and deposited some of its content when it passed the country, see figure 2.4 (EA 2008). The measured levels of fallout subsequent to the accident varied from 0 Bq m^{-2} up to 3 000 Bq m^{-2}. The highest values were found in Wales, the Lake District and in the south west part of Scotland. The area including the sampling sites in this thesis had depositions from approximately 600 Bq m^{-2} up to 3 000 Bq m^{-2}. (Hart & Sly 1992; Bar'ykhtar *et al*. 1998)

2.9.1 The monitoring network, RIMNET

In United Kingdom, a monitoring network was installed and commissioned in 1988, following the accident. The purpose was to monitor the levels of radiation in the environment and to independently detect radioactivity concentrations coming from overseas accidents. The network is called RIMNET and it still has 27 monitoring sites spread all over Scotland that give hourly updates. These stations monitor the total radiation from both man-made and natural sources to the population and assess the dose rates across the country. The annual doses have fluctuated only slightly since the beginning of the monitoring in 1997 and still remains on 0.64 mSv. One fifth of this dose is generally agreed to come from man-made sources. (SEPA 2006)

Nowadays, the influence of the Chernobyl accident on the environment is of less concern. According to the Scottish Environment Protection Agency about 0.2 % of the average annual exposure to the population of Scotland in 2003 originated from the Chernobyl fallout. Most of the exposure, 43 %, is due to radon coming from the ground and the main artificial source is medical x-rays. (Landrock *et al.* 2008)

2.9.2 The effects of ^{137}Cs in Scotland

After the accident the action level was set to 1 000 Bq kg^{-1} in sheep. This was monitored in a programme called "the Mark and Release Scheme". The sheep were tested using an external monitor. Sheep with levels exceeding the limit were consequently marked with coloured paint and were not permitted to be sold to slaughter and the farmers' possibilities to move them around were restricted. (EA 2008)

On behalf of the British Government, general diet, milk, crops, airborne particulate, rain and fresh water and seawater surveys of radioactivity concentrations have been conducted. In none of them, except sheep, the limit was exceeded. For example the highest level found in fish 140 Bq kg^{-1} was a perch from Cumbria. (EA 2008)

Due to the high organic matter content in the Scottish soils, the radionuclide fallout has become a long term concern, *e.g*. increased body burden of radiocaesium in sheep grazing in the countryside. The years after the Chernobyl accident, 9 700 farms in the United Kingdom had restrictions concerning moving, selling and slaughtering their sheep. Most of the restrictions have now been lifted, but still in 2008, 369 farms in the

United Kingdom, five of which are situated in Scotland, have some restrictions left upon them. (EA 2008)

Other radio nuclides originating from the Chernobyl accident are no longer detectable in Scotland (EA 2008).

Figure 2.4. Schematic picture of the radio-active clouds passing the British Isles. One passed over Scotland on the 3rd of May 1986 (modified from MLURI 2009).

2.9.3 The Macaulay transects

In 1990, Macaulay Land Use Research Institute conducted a survey of the status of Scottish surface soils (Trends in Pollution of Scottish Soils, TIPSS). This was made in order to trace and assess the pollution of the soils due to atmospheric deposition. Four transects across Scotland, with a total of 30 sampling sites were established. The pollutants, apart from caesium, that were investigated were polychlorinated biphenyls, (PCB), polycyclic aromatic hydrocarbons (PAH), cadmium, copper, lead, nickel and zinc (MLURI, 2009). In 1999, the sampling was repeated but additional samples (termed north, south, east and west) were taken to assess the local variability. These samples were taken 5 - 300 metres distant in the direction from the recorded GPS position of the main sampling pit.

Macaulay Land Use Research Institute in Aberdeen, Scotland, divided Scotland up in four transects, see figure 2.5, of which transect number two is studied in this thesis. Transect two stretches through the central highlands of Scotland. The land use at all sites is extensive grazing, except for Rora Moss which is used as a peat land.

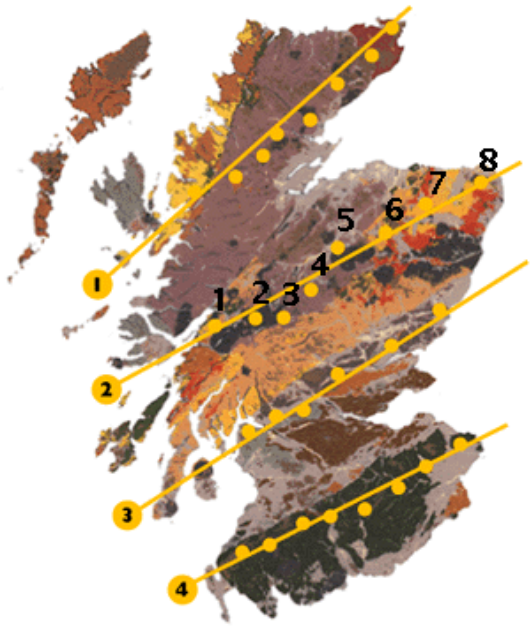

Figure 2.5. Map of the sampling sites on transect 2; 2:1 Loch Creran, 2:2 Glencoe, 2:3 Loch Rannoch, 2:4 Drumochter Pass, 2:5 Glen Feshie, 2:6 Lecht, 2:7 Knockandy Hill and 2:8 Rora Moss. (Modified from MLURI 2009)

Loch Creran is situated farthest to the west of the sampling sites. The soil is a Foudland association composed by blanket peat and the vegetation is moist Atlantic heather moor, typical of the west coast with a lot of suppressed heather.

Glencoe is the next site to the east after Loch Creran. It is a Darleith/Kirktonmoor association and the soil constitute of peaty podzols and peaty gleys. On top of this grows northern Atlantic heather moor.

Loch Rannoch is the third sampling site. Its soil is Countesswells/Dalbeattie/ Priestlaw association and it is composed of peaty gleys, some peaty podzols and peaty rankers. The vegetation is moist Atlantic heather moor.

Drumochter Pass is one of the middle sampling sites, situated almost in the middle of Scotland. It is an organic soil, composed of basin and valley peats on top of which northern Atlantic heather moor is found.

Glen Feshie is the other centrally situated site. It is Arkaig association composed of peaty podzols, some peaty gleys and humus-iron podzols. The vegetation is boreal heather moor.

Lecht is the third sampling site to the east. It is a Foudland composed of peaty podzols, peat, some peaty gleys and humus-iron podzols on top of which lichen rich boreal heather moor is to be found. (MLURI 2009)

Knockandy Hill is the second last site to the east. It is also a Foudland association, but this one is composed of peaty podzols; humus-iron podzols, peat and gleys. The vegetation is moist Atlantic heather moor.

Rora Moss is the sampling site farthest to the east. It is an organic soil composed of basin and valley peats. The vegetation is common lowland blanket bog. Information about carbon content, nitrogen content, bulk density, moisture, altitude and average annual rainfall is found in Appendix 1.

3. Materials and methods

In this thesis, a soil and vegetation sampling was conducted in 2008, for comparison with two studies, conducted by the Macaulay Land Use Research Institute in Aberdeen, Scotland, in 1990 and 1999. In the 2008 study, samples of soil and vegetation were collected at two different sites; Knockandy Hill and Rora Moss. GPS records were used to find the locations according to the National Grid References. These sites are the two eastern most sites along transect two, which stretches through the central highlands, from the middle west coast of Scotland to the middle east coast, see figure 2.5. In the 1990 and the 1999 soil was sampled at all sites along transect two.

3.1 Soil sampling in years 1990 and 1999

In the 1990 and the 1999 soil sampling a cylinder with a diameter of 25 mm and a height of 50 mm was used. In 1990 only one sample was taken at each site. In 1999 four samples were taken at each site, in the north, east, west and south direction, with a distance of 5 – 300 m between them. In both the years the samplings took place in September.

3.2 Soil sampling in year 2008

Due to the imprecise method used in 1990 and 1999, the soil sampling in 2008 was performed in accordance with methodology in other studies conducted by Rosén *et al.* (1999) on Swedish soils. This is a method that is easy to reproduce. Knockandy Hill was sampled on the 12th of December and Rora Moss on the 8th of December. At each location, three circles with a distance of 20 m and a radius of five m were made in the south-north direction; A, B and C, see figure 3.1. In each circle five core soil samples were collected.

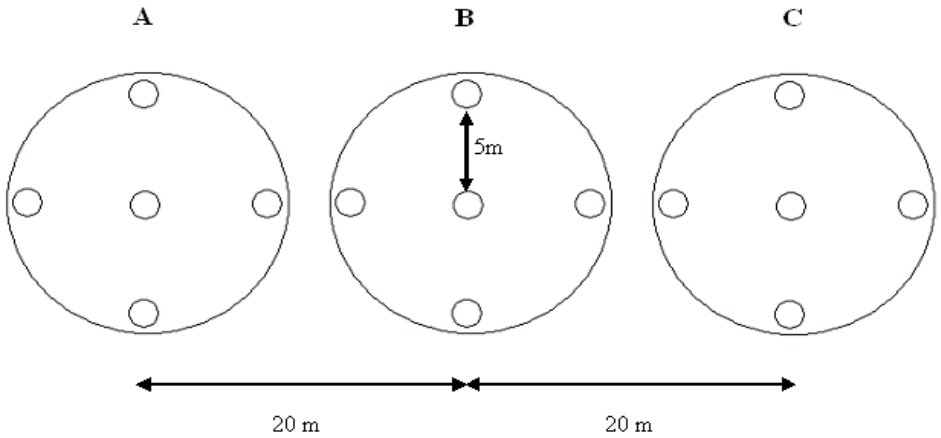

Figure 3.1. In each circle, five core samples were taken in the north, east, west, south direction and also one in the centre of the circle. The radius of the circle was five m and there was a distance of 20 m from centre point to centre point.

For the uppermost 10 cm of the soil, a cylinder corer with a diameter of 57 mm was used and the core was cut into 10 mm slices, see figure 3.2. In the middle of the first core pit,

an Ultuna core sampler with a diameter of 22 mm was put, and a core down to 50 cm depth was taken. This core was cut into 25 mm slices, starting the slicing at the bottom not to contaminate the samples above. All subsamples from the same depth in each circle were put together to form a composite sample.

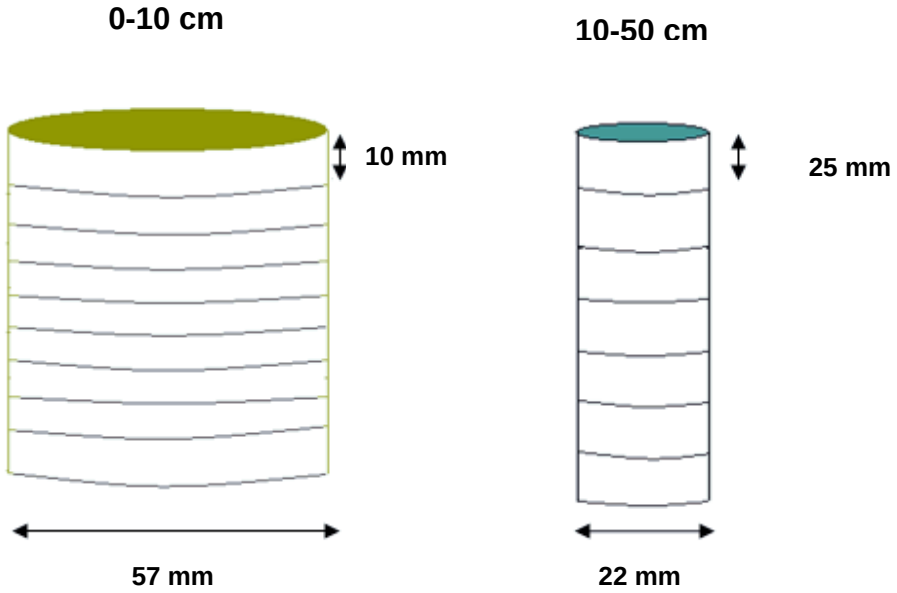

Figure 3.2. Schematic picture of the two cores sampled, showing sampling depth and the thickness of the layers for each sampler. The cylinder sampler to the left and the Ultuna core sampler to the right.

The soil samples were air-dried at 20 - 30 ºC for at least seven days. Then, the samples were weighed, if there were aggregates; they were crushed and subsequently sieved through a two mm sieve, all larger particles were discarded. The samples were homogenized and put into 35 mL or 60 mL vials, depending on the amount available, before analysis.

3.3 Vegetation sampling

Heather *(Calluna vulgaris)* and different grass species (*Deschampsia flexuosa, Deschampsia cespitosa, Agrostis canina* and *Phalaris arundinacea*) as well as some sedges (*Juncus acutiflorus* and *Juncus effusus*) were collected at Rora Moss and Knockandy Hill in 2008. They were gathered in the same circle as the soil samples. Five squares (0.25 × 0.25 m) were made in the north, east, west and south direction and also one in the centre of the circle, see figure 3.1. The vegetation was cut about five cm above the ground to avoid contamination by soil particles. Plant remains from previous growing seasons were also avoided. All vegetation samples from the same circle and category was put into the same bag.

The samples were dried at 60 ºC for 24 - 36 h. Thereafter they were weighted, milled and homogenized and representative amounts of the plants were taken out for analysis. The

samples were put into 35 mL or 60 mL vials, depending on the amount available, before analysis.

3.4 Analysis and calculations

3.4.1 Analysis

Samples from 1990 and 1999 were analyzed together with the new samples from 2008. The gamma emissions, representing the ^{137}Cs activity concentration in the samples, were measured using a computer aided germanium detector system, which was placed in a low background laboratory. The samples were measured for periods up to 24 hours. To minimize the uncertainty of the measurement, the samples that were expected to have lower activity concentrations were measured for 24 hours each. For the ones expected to have higher activity concentrations it was enough with 3 - 12 hours. Lower activity concentrations were expected in the vegetation samples and in the soil layers further down in the profile. All calculations of ^{137}Cs activity concentrations were made on dry weight (d.w.) basis.

3.4.2 Migration depth and migration rates

Migration depth can be described in different ways; one of them is as follows: the mean depth at which all caesium atoms are situated at a specific time. This is called the weighted mean value and is calculated using the formula 1 (Arapis *et al*. 1997).

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

X is the migration depth and is the depth where the sum of gravity points of all nuclides quotas equals zero. X_i is the centre of each layer in the soil profile. q_i represents the relative activity concentration in each layer and is calculated using formula 2. It is the activity concentration in each layer, A_i divided by the total activity concentration at the site, A_{tot}.

$$q_i = \frac{A_i}{A_{tot}} \qquad (2)$$

When having the migration depth, the migration rate can be calculated by dividing the depth with the time elapsed since the time of the fallout.

3.4.3 Transfer factors

Transfer factors make it possible to compare differences in uptake in plants between different sites that have received different fallout levels. While using transfer factors the uptake in the plants is only related to soil properties and environmental factors. A transfer factor is a quota between the levels in two different media *e.g.* soil and plant, plant and meat. A soil and plant transfer factor is calculated using formula 3 (Rosén *et al*. 1999). Here, ground deposition is the total activity concentration in the soil profile at the site.

$$TF_g = \frac{\text{Activity in plant}_{\text{d.w.}}}{\text{Ground deposition}} \left[\frac{Bq_{veg} kg^{-1}}{Bq_{soil} m^{-2}} = \frac{m^2}{kg_{d.w}} \right] \quad (3)$$

3.4.4 Statistics

The data were tested for normal distribution by a Kolmogorov-Smirnov test and thereafter tested for statistical significance in a paired-t test. The significance level was set to 5 %. Those parameters that had an uneven number of values were tested for statistical significance using ANOVA (General Linear Model).

4. Results

4.1 The activity concentration of ^{137}Cs in transect two

To present the activity concentration of ^{137}Cs in a meaningful way, the measured radioactivity concentration per kg was recalculated back to decay at the day of sampling.

Table 4.1. The mean ^{137}Cs activity concentration (Bq kg^{-1}) of the upper five cm of the soil profiles on the sampling dates

	1990 (n=1)	1999 (n=4)	2008 (n=3)
Loch Creran	120	109	-
Glencoe	108	295	-
Loch Rannoch	469	484	-
Drumochter Pass	94	147	-
Glen Feshie	159	108	-
Lecht	145	224	-
Knockandy Hill	163	94	194
Rora Moss	152	72	376

As can be seen in table 4.1, the detected activity concentration levels decreased at half of the sampling sites from the sampling in 1990 until the sampling in 1999. The activity concentration decreased in Loch Creran, Glen Feshie, Knockandy Hill and Rora Moss, while it increased in Glencoe, Loch Rannoch, Drumochter Pass and Lecht. The greatest decrease was found in Rora Moss, where the decrease was about 50 %. The greatest increase was found in Glencoe, where the activity concentration detected in 1999 was almost three times higher than that detected in 1990.

The highest activity concentration level of 1990 was found in Loch Rannoch, 469 Bq kg^{-1} and the lowest was 94 Bq kg^{-1} found in Drumochter Pass. The highest level in 1999 was also the detected in Loch Rannoch, 484 Bq kg^{-1}, but the lowest was from Rora Moss, 72 Bq kg^{-1}. For all measured values from the samplings in 1990 and 1999, see Appendix 2.

The present activity concentration levels measured in the upper five cm in Knockandy Hill and Rora Moss profiles differ significantly from one and other. Both also differ significantly from the levels detected previous years respectively, the levels have increased from 1999 until 2008. For all the measured values of Knockandy Hill and Rora Moss, see Appendix 3. If converting the detected values from 2008 from Bq kg^{-1} to Bq m^{-2} Knockandy Hill would have had 448 and Rora Moss 499 Bq m^{-2}.

4.2 The distribution of ^{137}Cs in the soil profiles sampled in 2008

The depth of the activity concentration of ^{137}Cs in the different circles varied within the sampled sites, the variation was at its biggest at a depth of 4 - 8 cm, and therefore a mean value for each site was calculated to make an easily comprehensible estimate of the activity concentration. As can be seen in figure 4.2, there was an increase of the relative ^{137}Cs activity concentration in the Knockandy Hill profile, from the soil surface and down to a depth of 7 - 8 cm, whereupon the relative activity conccentration decreased and

ceased at an approximate depth of 25 cm. This can also be seen in table 4.4, which shows the distribution of ^{137}Cs activity concentration in different layers.

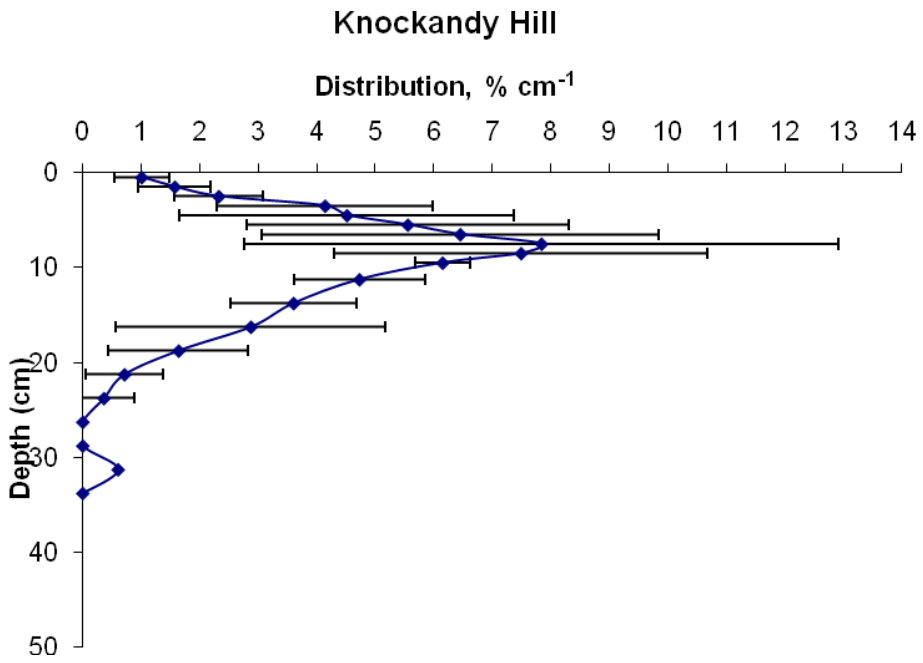

Figure 4.2. Distribution of ^{137}Cs in Knockandy Hill soil profile (% cm^{-1}) at the sampling day, means with standard deviation.

The depth of the distribution in the Rora Moss profile was similar to that in Knockandy Hill profile, see figure 4.3. The relative ^{137}Cs activity concentration increased down to a depth of about 6 cm and shows a wide peak in the figure 4.3 between 3 - 8 cm depth, from which it decreased and finally faded to almost nothing at a depth of 40 - 45 cm. All the estimated mean activity concentration values of all soil samples can be found in Appendix 4, as well as the total weight, number of replicates and Bq kg^{-1} at sampling date.

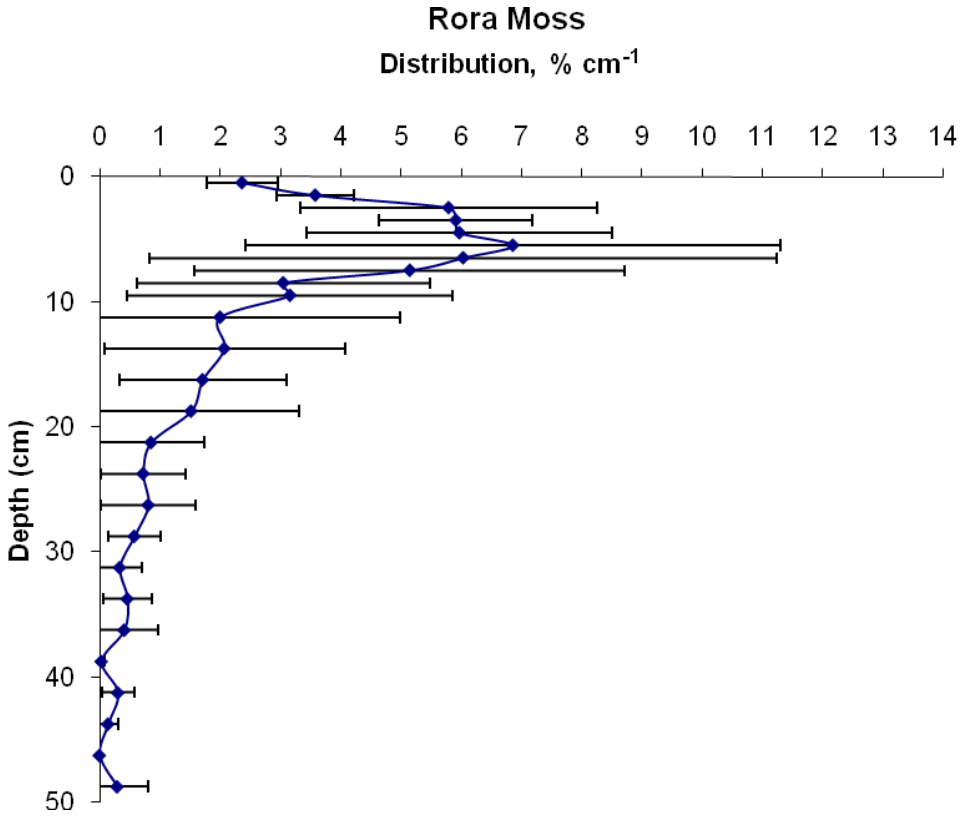

Figure 4.3. Distribution of ^{137}Cs in Rora Moss soil profile (% cm^{-1}) at the sampling day, means with standard deviation.

In table 4.4, the migration of ^{137}Cs is presented in terms of different layers of both the profiles. It was evident that in both, most of the caesium was left in the upper 25 cm, but there were still some activity concentration in the bottommost layers. The proportion of ^{137}Cs in the 0 - 5 cm layer was practically twice as high in Rora Moss as the same layer in Knockandy Hill. In the second layer, the percentage of ^{137}Cs activity concentration was higher in Knockandy Hill compared to in Rora Moss. Though, in the deepest layer, the percentage of the ^{137}Cs activity concentration was almost six times higher in Rora Moss than in Knockandy Hill and it was only this, bottommost, layer that showed a significant difference between the two sites.

Table 4.4. The distribution of ^{137}Cs (%) deposition in Knockandy Hill and Rora Moss per soil layer at the sampling day in 2008

Knockandy Hill	**%**	**Rora Moss**	**%**
0-5 cm	16.3	0-5 cm	30.1
5-25 cm	81.9	5-25 cm	59.2
25-35 cm	1.8	25-50 cm	10.7

4.3 Migration rate and migration depth of ^{137}Cs within the soil profiles sampled in 2008

The migration rates of ^{137}Cs in Knockandy Hill and Rora Moss were in accordance with each other and so were the migration depths, neither differed significantly from the other.

Table 4.5. Migration depths (cm) and migration rates (cm yr^{-1}) of ^{137}Cs deposition in Knockandy Hill and Rora Moss in 2008. Mean values and standard deviations. The migration depths were calculated as weighted mean values for the activity concentration in each profile

	Knockandy Hill	Rora Moss
Migration depth	8.2 ± 2.3	8.8 ± 5.5
Migration rate	0.36 ± 0.1	0.39 ± 0.2

Since the fallout, the radiocaesium activity concentration has moved down and reached an average depth of approximately 8 - 9 cm at both sampling site, see table 4.5.

4.4 Transfer of ^{137}Cs from soil to plants

The measured activity concentrations (Bq kg$^{-1}_{d.w.)}$ in the separate plants are shown in Appendix 5.

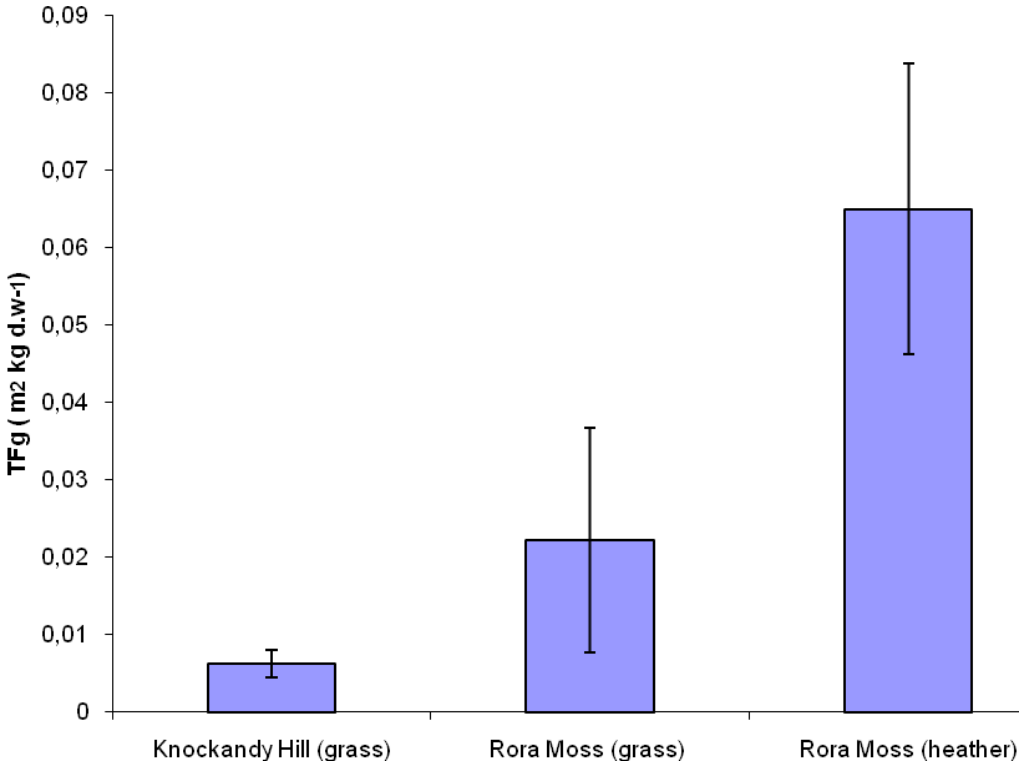

Figure 4.6. The transfer factors of radiocaesium (TFg, m^2 kg$^{-1}_{d.w}$) from soil to grass and heather for the sampled sites in 2008. Standard deviations. Transfer of ^{137}Cs to grass did not differ significantly between sites, but transfer to heather was significantly higher than that to grass at Rora Moss, $p<0.05$.

The transfer of ^{137}Cs from soil to heather in Rora Moss was significantly higher than the transfer to grass, see figure 4.6. The transfer from soil to grass did, however, not differ significantly between Rora Moss and Knockandy Hill. The mean soil to plant transfer factors were 0.009 m^2 $kg^{-1}_{d.w.}$ for Knockandy Hill, 0.022 m^2 $kg^{-1}_{d.w.}$ for grass from Rora Moss and 0.065 m^2 $kg^{-1}_{d.w.}$ for heather.

5. Discussion

5.1 Activity concentration of ^{137}Cs in the soil profiles

If using the levels of deposition estimated by Hart & Sly (1992) and Bar'ykhtar *et al*. (1998), of 0-3 000 or 600-3 000 Bq m^{-2}, the activity concentration in 1990 would be roughly 550-2700 Bq m^{-2}, 450-2200 in 1999 and 350-1800 in 2008 at all sites due to natural decay. This can be compared with 448 and 499 Bq m^{-2} that were detected in Knockandy Hill and Rora Moss in 2008. However, the Hart & Sly (1992) and Bar'ykhtar *et al*. (1998) values are approximates of large areas, while the values presented in this study are fairly precise.

5.1.1 The Knockandy Hill and Rora Moss sites

The differences in radiocaesium activity concentration in the upper five cm of the profiles between the sampling years 1990, 1999 and 2008 in both Knockandy Hill and Rora Moss are difficult to interpret, both of them having congruent trends. Firstly, the detected levels decrease but between 1999 and 2008 the levels seem to increase and get even higher than the detected levels in the 1990 sampling.

Generally, the difference in ^{137}Cs deposition at a sampling site can be due to several different factors such as; heterogenity of deposition, vegetation cover, sampling techniques, migration, topography and hydrological regime.

First of all, the deposition of ^{137}Cs can vary within meters at a site, especially at mosses where the micro topography and vegetation cover are quite rough, hence the fallout being unevenly distributed. Both Knockandy Hill and Rora Moss received the fallout as wet deposition. Due to the micro topography, the low laying pool complexes of the mosses gets higher levels of deposition. The more multi-layered vegetation is the more complicated will be the radioactivity pattern in the soils due to interception by vegetation. This might have led to almost horizontal transport, diluting the concentration at some parts, increasing them at others. The horizontal retranslocation can also be expected to be influenced by biological processes at the sites. The varying amounts of precipitation in the study area (see Appendix 1), the direction of the wind and possibility of trees sheltering from the wind might affect the deposition of the fallout in the micro environment. These circumstances mean that a consistent sampling strategy is of considerable importance. Otherwise it will be very difficult to interpret the results. The samplings in this study have been performed in different ways by many different people. It is also important that the exact positions of sampling are determined in a random way. Otherwise some samplers might prefer sampling on the grassy hillocks, while others might prefer the pool complexes. In the sites studied here the number of replicates varied from one to four individual samples per site between sampling occasions. Measuring the activity concentration in only one sample, does not seem to give reliable results.

Another aspect which needs to be considered in this respect is the vertical migration of ^{137}Cs. The samples from 1990 and 1999 are, as have been stated before, only taken down to a depth of five cm. In this case it is not known how much of the radiocaesium have

reached greater depths, though, it is likely to assume that some parts have, at least in the 1999 sampling. As can be seen in the migration diagrams in figures, 4.2 and 4.3 and also in table 4.4, it is evident that most of the radiocaesium is located deeper down in the profiles than those five cm presented in table 4.1, concerning the 2008 sampling.

5.1.2 The Loch Creran, Glencoe, Loch Rannoch, Drumochter Pass, Glen Feshie and Lecht sampling sites

The unsteady results from the sampling in Loch Creran, Glencoe, Loch Rannoch, Drumochter Pass, Glen Feshie and Lecht can, of course, also be explained by the factors mentioned above. Also here are the decreases in deposition levels not only explained by the decay. According to the data from table 4.1, the activity concentration of ^{137}Cs in 1999 at four out of eight sites, Glencoe, Loch Rannoch, Drumochter Pass and Lecht had increased during the study period, which is unrealistic. This might be due to either different sampling sites or extreme values in the single samples collected in 1990.

The only sampling sites showing reasonable results concerning decrease due to decay are Loch Creran and Glen Feshie, whose activity concentration levels had decreased about 10 % and 30 % respectively from 1990 to 1999.

5.2 The vertical distribution of ^{137}Cs in Knockandy Hill and Rora Moss

This study has shown a downward migration of radiocaesium since the fallout in 1986 in both the Knockandy Hill and the Rora Moss soil profile, which is in accordance with earlier studies conducted by e.g. Rosén *et al*. (1999), Arapis *et al*. (1997), Forsberg (2000) and Isaksson *et al.* (2001). In Knockandy Hill, the soil was sometimes impenetrable for the corer, due to impermeable roots and stones and many samples had to be discarded and new ones taken. In Rora Moss, all the soil cores were sampled down to a depth of 50.0 cm, but in Knockandy Hill no one was, the maximum sampling depth there varied between 27.5 - 35.0 cm. Probably not all of the radiocaesium was gathered in the Knockandy Hill profiles, but presumably at least 90 %. However, according to the measured samples, the elevated activity concentration had already ceased at a depth of approximately 25 cm, hence the shallower sampling may not matter.

The Knockandy Hill and Rora Moss profiles do not differ significantly from each other, either concerning the mean migration depths or the mean migration rates. Though, the relative distributions within the layers of the different soil profiles are slightly different. In Knockandy Hill the activity concentration in the bottommost layer is significantly lower than the activity concentration in the same layer at Rora Moss. This might be because of higher clay content in the upper parts of the Knockandy Hill profile, fixing the caesium and slowing down a downward migration. An indication of a higher mineral content is the higher bulk density of the Knockandy Hill samples, see Appendix 1. In the appendix it can also be seen that the organic content is much less in Knockandy Hill than in Rora Moss, 21.5 % versus 50.8 %. However, the actual clay content is not investigated at the Scottish sites.

From 10.0 cm depth on, another thinner corer is used. This sometimes leads to contamination, particularly of the layer at 10.0 - 12.5 cm depth and increased the activity

concentrations from the wider core pit above. The reason for the uneven distribution of caesium in the deeper parts of the profiles at both Rora Moss and Knockandy Hill is probably due to contamination from layers above during the sampling procedure, due to a less suitable sampling method. The Ultuna corer is best used in mineral soils, but it is not fitting as good at organic soils, since it does not cut off the organic matter in an appropriate way. There is no corer suitable for this kind of study in organic soils.

5.2.1. The importance of the soil fauna

In mineral soils the redistribution of caesium is sometimes influenced by the presence of earthworms (Jarvis *et al*. 2009). Müller-Lemans and van Dorp (1996) showed that material displacement is often caused by soil fauna, especially earthworms, since they live, breed and feed in and on organic matter. Depending on the clay content (Dumat *et al.* 1997), there is usually some caesium being bound to the organic material, which will increase or decrease the ^{137}Cs available for earthworm induced transport.

However, redistribution of ^{137}Cs within the soil profile caused by earthworms is unlikely to be the case in my study. The habitat of earthworms is the A, B and C-horizons of mineral soils near neutral in pH, not the H-horizon found in acid organic soils. No earthworms were seen at either of the sampling sites, in agreement with Müller-Lemans and van Dorp (1996). That is to say, the uneven distribution is not due to bioturbation.

5.2.2. Comparisons with migration studies at similar sites

Hille, Möjsjövik, Pålsjömossen and Åsenmossen are Swedish sampling sites, with similar soil types as the peat soils sampled in Scotland. They have been used in many Swedish studies concerning radiocaesium, conducted by *e.g*. Jansson (2004), Persson (2008) and Rosén *et al.* (1999), all of them using the same method as I did in this study. The mean migration rate (0.36 and 0.39 cm y^{-1}) and the mean migration depth (8.2 and 8.8 cm) of both Knockandy Hill and Rora Moss can be compared with the migration rates and migration depths estimated in the previous studies.

The last time Möjsjövik, a pasture sampling site on a fen-peat soil, was studied concerning migration was in 2007 (Persson 2008), the mean migration depth at that time was estimated to 6.1 cm. This is not significantly different from either Rora Moss or Knockandy Hill, neither is the mean migration rate, which was estimated to 0.3 cm yr^{-1}.

In the same study by Persson (2008) another fen-peat soil was studied namely Hille, utilized as a temporary grass land. Hille had a mean migration depth of 11.4 cm and a mean migration rate of 0.5 cm yr^{-1}. The migration depth does not differ significantly from either Knockandy Hill or Rora Moss, nor does the migration rate differ.

Åsenmossen, a bog, was sampled in 2003 by (Jansson 2004). Then, the ^{137}Cs had reached a mean migration depth of 17.1 cm, which is not significantly different from Rora Moss or Knockandy Hill. Though, when comparing Knockandy Hill with Åsenmossen, the p-value was close to 0.05 and considering the time difference of five years, the ^{137}Cs deposition in Åsenmossen will have reached a greater depth by now, hence there will probably be a significant difference between the migration depth of Knockandy Hill and

Åsenmossen. The mean migration rate at Åsenmossen was estimated to 1.0 cm yr^{-1}, which is significantly higher than that at Rora Moss, this could be because one of the single values of the migration rate at Rora Moss is as slow as 0.13 cm yr^{-1}, however the Åsenmossen migration rate is not different to Knockandy Hill.

In another study, by Rosén *et al.* (2009) on a raised bog, Pålsjömossen, a mean migration rate of ^{137}Cs of 0.8 cm per yr^{-1} was found on the outskirts of the bog. The mean migration depth was estimated to 14.9 cm, which is slightly different to the mean results of this study. Though, no significant differences were to be found neither between mean migration depth nor mean migration rate at Pålsjömossen and Rora Moss nor between Pålsjömossen and Knockandy Hill.

The trend seems to be that, the migration rate at Rora Moss might not differ from the Swedish sampling sites. However, at Knockandy Hill the migration rate is lower. This is probably due to higher clay content and the lower carbon content, as mentioned earlier.

In Arapis *et al*. (1997) studies were performed on five different soils and mean migration rates varying from 0.4 to 1.2 cm y^{-1} were found. The highest rate being a soddy podzoilc gley and the lowest being a peat march, which agrees with the results of this study.

In general terms, the migration in soil is a complex process. The mobility is closely connected with clay content, but all the discussed sampling sites contain little clay, which probably indicates a weak binding to the cation exchange capacity of organic matter instead. The differences in migration depth could possibly depend on the characteristics of the organic matter in the different soils. Other factors that influence the migration in soils in general are the possible application of countermeasures like plowing, but at these sites no measures to reduce the mobility of radiocaesium after the accident have been taken. Binding of ^{137}Cs to the living vegetation, hence an upward transportation within the plants can also be affecting the migration (Rosén *et al*. 2009). The climate affects the migration, the weather conditions varies from the west to the east coast of Scotland, higher amounts of precipitation comes in the west, closer to the Atlantic, see Appendix 1. Glencoe and Loch Creran have the highest amounts of annual precipitation, but it is Glen Feshie and Lecht that have the biggest decrease in activity concentration, looking at the 1990 and 1999 sampling. Hence, this factor alone does not determine the migration.

5.3 Transfer from soil to plants

Plants were collected at both Knockandy Hill and Rora Moss. The grasses and sedges were sampled separately in each sampling circle. At Knockandy Hill there were no heather growing.

5.3.1 The transfer of ^{137}Cs from soil to heather

The transfer from soil to heather was shown to be significantly higher than that for soil to grass in Rora Moss, which was also the case in studies conducted by *e.g.* Rosén *et al.* (2009). At Pålsjömossen, in Sweden, the transfer of ^{137}Cs from soil to different plants, including heather, have been studied at a couple of occasions (Rosén *et al.* 2009). In 1989 the transfer factor was estimated to 0.7 m^2 kg^{-1} and in 2004 it was estimated to 0.5 m^2

kg^{-1}, which is almost 10 times larger, hence significantly higher than the transfer factor for heather observed in Rora Moss (0.065 m^2 $kg^{-1}_{d.w.}$). This is probably due to the different environment of the different sites. The Pålsjömossen soil contains no measurable, structured minerals at all, hence the caesium ions are biologically available for uptake. However, both Knockandy Hill and Rora Moss have some mineral content and that how most of the ions will be bound to the clay particles and not available for uptake by plants.

Heather usually grows in areas with soils containing high contents of organic matter, which are also poor, sandy and acidic. Like other plants growing in nutrient-poor areas, also heather is vulnerable to atmospheric pollution, e.g. radioactive fallout. (Strandberg & Johansson 1999)

5.3.2 The transfer of ^{137}Cs from soil to grass

Assembling all grass and sedge species into one sample per sampling circle and call it all grass maybe was to simplify the method. Different species have different need of nutrients and varying root depths, hence they might have different caesium uptake.

The transfer of ^{137}Cs to grass did not differ significantly between Knockandy Hill (0.009 m^2 $kg^{-1}_{d.w.}$) and Rora Moss (0.022 m^2 $kg^{-1}_{d.w.}$) This is probably because it was more or less the same species, growing in environmental conditions resembling each other, even though the mineral content of the soils differed. Probably the plants absorbed their nutrients at depths in the soils that contained comparable activity concentration levels.

In year 2000, the transfer to grass was studied in Möjsjövik by (Persson 2008). The mean transfer factor was found to be 0.006 m^2 kg^{-1}, which is not significantly different from the mean transfer factor estimated neither in Knockandy Hill nor in Rora Moss. The difference between the mean values is almost a factor of ten, but the variation of the single values is too great to make the means differ significantly.

According to Broadley *et al*. (1999), there are no general factors explaining the species differences in caesium uptake. However, the same authors state that there definitely are differences. Differences in the transfer from soil to plant occur principally between plant taxa growing on the same soil, but also in the same plant taxa growing on different soils. Diversity in caesium uptake has also been found in mycorrhizal versus non-mycorrhizal plants and in fast growing plants versus slower growing ones, higher concentrations being concluded in the first-mentioned.

Both heather and grasses can have mycorrhiza. According to Rosén *et al*. (2009) the mycorrhiza of heather can be very effective in caesium uptake since it increases the root surface of heather at the same depth as where the most of the ^{137}Cs lingers. The mycorrhiza usually increases the nutrient uptake in plants and it seems like it increases the caesium uptake as well. The finer and thinner roots of the grasses probably penetrates the soil to greater depths than heather, hence the nutrient uptake will occur at depths were the caesium content is much lower. This could be an explanation to why the transfer is higher to heather than to grass. Another suggestion is the possible accumulation of

potassium in the plants as a preparation method in prospect of the winter, instead of the accumulation of potassium, the heather might by storing caesium because of their resemblance.

6. Conclusion

This study was conducted on two mosses, one containing more organic matter than the other. Neither of the mosses have been disturbed since the Chernobyl accident in 1986. The study has shown a slow downward migration of ^{137}Cs in both sites. The main parts of the radionuclides were found to be lingering in the upper 10 cm of the profiles at both sites, which is corresponding to other studies conducted on similar soils in Sweden.

Another aim of the project was to present the current state of plants growing on these ^{137}Cs contaminated soils. The plants investigated were heather and different grass and sedge species. It was found that there was still some uptake of ^{137}Cs in the plants, heather showing the highest transfer factor. Though, if repeating this study in the future it is advisable to collect the plants during the growing season to get more reliable results, not in the winter as have been done here.

It is of outmost importance to have a well thought-out approach for repeated sampling; to take many core samples, to have exact coordinates to be able to retrace old sampling sites and a thorough documentation.

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Appendix 1

Table A.2. National grid reference, carbon and nitrogen content, moisture, altitude and rainfall for the eight different sampling sites in 1999

Site	National Grid Reference	Bulk density dw (g/cm^3)	Carbon content %	Nitrogen content %	Field water content %	Altitude (m a sl)	Average annual rainfall (mm)
Loch Creran, 2:1	NN 972 459	0.11	49.37	1.74	86.11	155	1 600
Glencoe, 2:2	NN 285 535	0.09	48.56	1.51	91.70	310	2 600
Loch Rannoch, 2:3	NN 470 553	0.09	49.95	1.58	85.12	400	1 500
Drumochter Pass, 2:4	NN 725 725	0.15	49.11	1.86	86.50	450	1 200
Glen Feshie, 2:5	NN 861 ?78	0.14	48.62	1.36	81.13	480	1 300
Lecht, 2:6	NJ 254 124	0.14	49.65	1.06	88.70	680	1 100
Knockandy Hill, 2:7	NJ 557 313	0.22	21.51	0.89	57.48	350	950
Rora Moss, 2:8	NK 042 522	0.11	50.81	1.63	87.59	50	775

Appendix 2

Table A.3. The names of the sampled sites in transect 2, the sampling date, the sample dry weight (g), the ^{137}Cs activity concentration (Bq kg^{-1}) and the deposition (Bq m^{-2}) at the day of fallout

	Sampling date	Sample weight, dw (g)	Bq kg^{-1} sampling date	Bq kg^{-1} fallout date
Loch Creran				
single sample	1-9 -90	0.6	119.7	132.3
north	1-9 -99	13.6	76.8	104.4
south	1-9 -99	9.0	118.0	160.5
east	1-9 -99	10.4	137.0	186.3
west	1-9 -99	18.3	106.0	144.1
Glencoe				
single sample	1-9 -90	30.0	108.0	119.3
north	1-9 -99	1.4	246.8	335.6
south	1-9 -99	10.9	240.5	327.0
east	1-9 -99	10.7	241.0	327.7
west	1-9 -99	11.7	314.1	427.1
Loch Rannoch				
single sample	1-9 -90	24.7	469.0	518.3
north	1-9 -99	15.4	221.0	300.5
south	1-9 -99	14.3	533.0	724.8
east	1-9 -99	22.6	445.0	605.1
west	1-9 -99	10.1	677.4	921.2
Drumochter Pass				
single sample	1-9 -90	33.2	94.3	104.2
north	1-9 -99	33.0	164.0	223.0
south	1-9 -99	24.5	151.0	205.3
east	1-9 -99	22.2	99.7	135.6
west	1-9 -99	1.6	133.1	181.0
Glen Feshie				
single sample	1-9 -90	42.8	159.0	175.7
north	1-9 -99	21.8	77.1	104.8
south	1-9 -99	47.9	84.7	115.2
east	1-9 -99	11.7	146.0	198.5
west	1-9 -99	14.3	125.0	170.0
Lecht				
single sample	1-9 -90	42.1	145.0	160.2
north	1-9 -99	6.3	252.0	342.7
south	1-9 -99	12.1	292.0	397.1
east	1-9 -99	18.8	163.0	221.7
west	1-9 -99	20.5	188.0	255.7
Knockandy Hill				
single sample	1-9 -90	21.8	163.0	180.1
north	1-9 -99	31.5	190.0	258.4
south	1-9 -99	29.8	47.6	64.7
east	1-9 -99	33.0	87.6	119.1
west	1-9 -99	38.6	49.9	67.9

Appendix 2 (continued)

	Sampling date	Sample weight, dw (g)	Bq kg^{-1} sampling date	Bq kg^{-1} fallout date
Rora Moss				
single sample	1-9 -90	5.0	151.6	167.5
north	1-9 -99	18.0	42.6	57.9
south	1-9 -99	10.0	90.8	123.5
east	1-9 -99	8.7	80.8	109.8
west	1-9 -99	21.1	43.2	58.7

Appendix 3

Table A.1. The mean ^{137}Cs activity concentration (Bq m^{-2}) on the sampling dates at Knockandy Hill and Rora Moss in the different layers in 2008. Mean values with standard deviations

Depth (cm)	Knockandy Hill	%	Rora Moss	%
0 - 1	33 ± 16	1.2	50 ± 13	3.0
1 - 2	52 ± 20	1.9	76 ± 14	4.6
2 - 3	77 ± 25	2.8	122 ± 52	7.4
3 - 4	137 ± 61	5.0	125 ± 27	7.5
4 - 5	149 ± 95	5.4	126 ± 54	7.6
5 - 6	184 ± 91	6.7	145 ± 94	8.7
6 - 7	214 ± 112	7.8	127 ± 110	7.7
7 - 8	260 ± 168	9.4	109 ± 75	6.6
8 - 9	248 ± 105	9.0	64 ± 51	3.9
9 - 10	204 ± 15	7.4	67 ± 57	4.0
10 - 12.5	391 ± 92	14.2	106 ± 157	6.4
12.5 - 15	298 ± 90	10.8	109 ± 105	6.6
15 - 17.5	238 ± 191	8.6	90 ± 73	5.4
17.5 - 20	136 ± 99	4.9	80 ± 95	4.8
20 - 22.5	59 ± 54	2.1	45 ± 46	2.7
22.5 - 25	30 ± 42	1.1	38 ± 37	2.3
25 - 27.5	0 ± 0	0.0	43 ± 41	2.6
27.5 - 30	0 ± -	0.0	31 ± 23	1.9
30 - 32.5	50 ± -	1.8	18 ± 19	1.1
32.5 - 35	0 ± -	0.0	24 ± 21	1.4
35 - 37.5	±		22 ± 29	1.3
37.5 - 40	±		2 ± 3	0.1
40 - 42.5	±		16 ± 14	1.0
42.5 - 45	±		7 ± 9	0.4
45 - 47.5	±		0 ± 0	0.0
47.5 - 50	±		16 ± 27	1.0
Total	**2 760** ± 543	100.0	**1 658** ± 729	100.0

Appendix 4

Table A.4. Total weight and activity concentration of composite samples from each sampled circle (A, B and C) and depth at Knockandy Hill. Activity concentration calculated back to sampling date, the 12th of December 2008, n is the number of replicates in the circle

Depth (cm)	A total weight (g)	n	B total weight (g)	n	C total weight (g)	n	A Bq kg^{-1} sampling date	B Bq kg^{-1} sampling date	C Bq kg^{-1} sampling date
0 - 1	23.6	5	17.2	5	16.3	5	27.2	14.2	23.9
1 - 2	28.9	5	18.5	5	18.2	5	33.4	26.7	29.4
2 - 3	36.6	5	17.3	5	21.9	5	34.7	60.1	29.1
3 - 4	48.7	5	24.9	5	28.0	5	42.3	93.5	30.8
4 - 5	54.5	5	37.6	5	29.7	5	44.6	73.5	17.8
5 - 6	56.6	5	51.6	5	31.9	5	45.1	65.7	34.5
6 - 7	75.6	5	39.4	5	35.5	5	54.9	69.6	36.2
7 - 8	82.5	5	35.6	5	48.4	5	70.2	58.3	42.8
8 - 9	82.8	5	38.3	5	68.0	5	56.4	54.5	40.2
9 - 10	78.6	5	52.2	5	91.7	5	32.7	46.3	30.6
10 - 12.5	25.5	5	43.0	5	51.6	5	26.2	14.4	18.3
12.5 - 15	33.0	5	32.0	5	55.7	5	11.8	18.2	13.1
15 - 17.5	38.6	5	23.5	5	23.0	2	10.6	4.8	14.5
17.5 - 20	34.0	5	12.0	3	26.9	2	11.2	2.1	5.2
20 - 22.5	34.9	5	11.4	3	30.3	2	5.8	0.0	1.8
22.5 - 25	23.7	5	9.7	2			4.8	0.0	-
25 - 27.5	12.6	3	13.2	2			0.0	0.0	-
27.5 - 30	11.8	3					0.0	-	-
30 - 32.5	13.4	3					4.2	-	-
32.5 - 35	17.3	3					0.0	-	-

Appendix 4 (continued)

Table A.5. Total weight and activity concentration of composite samples from each sampled circle (A, B and C) and depth at Rora Moss. Activity concentration calculated back to sampling date, the 8th of December 2008, n is number of replicates in the circle

Depth (cm)	A total weight (g)	n	B total weight (g)	n	C total weight (g)	n	A Bq kg^{-1} sampling date	B Bq kg^{-1} sampling date	C Bq kg^{-1} sampling date
0 - 1	10.8	5	11.8	5	10.0	5	75.8	46.1	54.7
1 - 2	15.3	5	12.4	5	16.8	5	62.7	64.0	67.9
2 - 3	12.0	5	19.4	5	21.1	5	68.9	110.0	81.9
3 - 4	16.9	5	16.7	5	20.8	5	90.6	118.0	62.0
4 - 5	19.8	5	20.8	5	27.0	5	72.8	113.4	37.9
5 - 6	22.8	5	25.9	5	26.0	5	68.9	122.0	31.4
6 - 7	21.5	5	29.5	5	31.8	5	44.4	110.0	21.5
7 - 8	25.7	5	31.7	5	33.0	5	41.0	77.9	19.3
8 - 9	24.9	5	25.6	5	39.2	5	20.2	61.6	9.9
9 - 10	22.5	5	28.6	5	32.6	5	21.3	59.2	11.9
10 - 12.5	12.9	5	7.2	5	6.8	5	42.3	5.5	2.5
12.5 - 15	17.9	5	9.3	5	8.8	5	24.4	12.8	7.7
15 - 17.5	21.4	5	8.6	5	7.4	5	15.3	15.3	7.7
17.5 - 20	23.9	5	12.8	5	4.7	5	14.7	8.3	0.0
20 - 22.5	11.1	5	9.4	5	4.8	5	15.8	8.8	0.0
22.5 - 25	14.1	5	10.7	5	4.3	5	9.9	7.3	0.0
25 - 27.5	16.1	5	9.2	5	7.0	5	10.3	7.4	1.5
27.5 - 30	19.8	5	12.7	5	8.3	5	5.5	2.5	4.0
30 - 32.5	16.4	5	9.7	5	7.1	5	1.8	7.4	0.0
32.5 - 35	11.5	5	8.9	5	9.5	5	5.9	0.0	7.5
35 - 37.5	11.4	5	13.3	5	10.6	5	9.2	1.5	0.0
37.5 - 40	10.6	5	9.7	5	10.3	5	0.9	0.0	0.0
40 - 42.5	9.7	5	11.1	5	10.0	5	4.4	4.4	0.0
42.5 - 45	9.5	5	8.3	5	9.6	5	0.0	3.9	1.0
45 - 47.5	9.1	5	8.9	5	9.8	5	0.0	0.0	0.0
47.5 - 50	8.9	5	11.0	5	12.0	5	0.0	8.0	0.0

Appendix 5

Table A.6. The activity concentration in the plants (Bq kg^{-1} d.w.). in the sampling areas within the sampling circles (A, B and C) at Knockandy Hill (12th of December 2008) and Rora Moss (8th of December 2008). (-) indicate that no plants were found in the sampling area

	A	B	C
Knockany Hill. grass	0	15.0	0
	14.0	79.0	30.7
	0	-	0
	3.4	-	41.6
	0	-	-
Rora Moss. grass	0	64.8	8.4
	7.4	26.3	37.8
	39.2	105.0	23.4
	3.1	-	19.7
	9.1	-	30.1
Rora Moss. heather	104.0	122.0	33.6
	-	132.0	68.9
	-	115.0	108.0
	-	123.0	70.1
	-	-	-