# Ecological Half-Lives of Radioactive Elements in Semi-Natural Systems





## Nordic Nuclear Safety Research (NKS)

organizes joint four-year research programs involving some 300 Nordic scientists and dozens of central authorities, nuclear facilities and other concerned organizations in five countries. The aim is to produce practical, easy-to-use background material for decision makers and help achieve a better understanding of nuclear issues.

To that end, the results of the fifth four-year NKS program (1994 – 1997) are herewith presented in a series of final reports comprising reactor safety, waste management, radioecology, nuclear emergency preparedness and information issues. Each report summarizes one of the ten projects carried out during that period, including the administrative support and coordination project. A special Summary Report, with a brief résumé of all ten projects, is also published. Additional copies of the reports on the individual projects can be ordered free of charge from the NKS Secretariat.

The final reports – together with some technical reports and other material produced during the 1994 – 1997 period – have been collected on a CD-ROM, also available free of charge from the NKS Secretariat.

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# Ecological Half-Lives of Radioactive Elements in Semi-Natural Systems

Final Report of the Nordic Nuclear Safety Research Project EKO-2

Tone D. Bergan

October 2000

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## 1. This is NKS

NKS (Nordic Nuclear Safety Research) is a scientific cooperation program in nuclear safety, radiation protection and emergency preparedness. Its purpose is to carry out cost-effective Nordic projects, thus producing research results, exercises, information, manuals, recommendations, and other types of background material. This material is to serve decision-makers and other concerned staff members at authorities, research establishments and enterprises in the nuclear field.

The following major fields of research are presently dealt with: reactor safety, radioactive waste, radioecology, emergency preparedness and information issues. A total of nine projects have been carried out in the years 1994 - 1997.

Only projects that are of interest to end-users and financing organizations have been considered, and the results are intended to be practical, useful and directly applicable. The main financing organizations are:

- The Danish Emergency Management Agency
- The Finnish Ministry for Trade and Industry
- The Icelandic Radiation Protection Institute
- The Norwegian Radiation Protection Authority
- The Swedish Nuclear Power Inspectorate and the Swedish RadiationProtection Institute

Additional financial support has been given by the following organizations:

In Finland: Ministry of the Interior; Imatran Voima Oy (IVO); Teollisuuden Voima Oy (TVO)

In Norway: Ministry of the Environment

In Sweden: Swedish Rescue Services Board; Sydkraft AB; Vattenfall AB; Swedish Nuclear Fuel and Waste Management Co. (SKB); Nuclear Training and Safety Center (KSU)

To this should be added contributions in kind by several participating organizations.

NKS expresses its sincere thanks to all financing and participation organizations, the project managers and all participants for their support and dedicated work, without which the NKS program and this report would not have been possible.

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## Abstract

The recovery of Nordic seminatural ecosystems from radioactive fallout 8 - 11 years after the Chernobyl accident is studied. Ecological halflives of radiocesium in various ecosystems such as uncultivated pastures, mountain areas and uplands are important, since foodstuffs from these areas account for a considerable dose to man. Dominant types of foodstuffs are identified. Three types of problem areas have been chosen: sheep grazing on uncultivated pasture; mushrooms; and freshwater fish. By incorporating the new data on ecological half-lives into existing models, uncertainties in dose calculations may be decreased.

## Key words

Radioecology, radiocesium, radiostrontium, Nordic natural environment, sheep, mushroom, fish, doses to man



## 2. Summary in English

Foodstuffs from semi-natural areas, such as uncultivated pastures, mountain areas and uplands account for a considerable portion of intake of radiocesium and radiostrontium, and thus the dose to man. Products from intensive agricultural production show short half-lives, so as time after a radioactive fallout increases, foodstuffs such as mushroom, game and freshwater fish become dominant with respect to intake of radionuclides.

Within this EKO-2 project three problem areas have been chosen; sheep grazing on uncultivated pasture, the influence of mushroom and freshwater fish. The main aim has been to identify the contribution from semi-natural systems, by determining ecological half-lives for specific foodstuffs from these areas, and thus determining dose to man. By incorporating these half-lives into existing models, we can decrease the uncertainties in dose calculations. In a fallout situation it is possible to quickly develop a picture of possible consequences, and implement appropriate countermeasures.

In the ongoing projects we have produced or compiled data for 8-11 years after the Chernobyl accident. The time series have been very necessary for predicting ecological half-lives. Within this project period, we have demonstrated that the time dependent levels of radionuclides in foodstuffs from the selected areas are not well described by any «ecological half-life». As time increases, so does the half-lives.

The recovery of Nordic ecosystems from contamination by radiocesium originating from the Chernobyl accident is gradually slowing down, at the same time as areas vary widely in susceptibility and recovery rates. Accordingly, ecological half-lives are gradually increasing and cannot be treated as constants, over neither time nor space. Although it has not been easy to determine simple or general ecological half-lives, the projects have given us useful understanding of the mechanisms governing the transfer of radionuclides, and more knowledge about typical Nordic ecosystems.

#### The sheep-project

The soil - vegetation - sheep - system has been studied in five countries; Iceland, The Faroe Islands, Denmark, Sweden and Norway. Co-ordinated sampling started in 1990 and has been continued up until 1997. Large differences in transfer are found, and by studying the production intensitivity, biomass production, climatic conditions, the presence of mushrooms, intake of soil and experimental studies of stable elements in the soil, we have been able to explain some of the differences. Since soil represents an important reservoir for radionuclides in the terrestrial

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system, the soil characteristics has been the most important factor for the different transfer factors that we have observed in the different grazing areas.

## The forest project

Little attention has previously been paid to the consumption of food products from the forest system, such as wild berries, mushroom, freshwater fish and game. A questionnaire has therefore been performed, with focus on consumption of wild berries and mushrooms, with surveys performed in Denmark, Finland, Norway and Sweden. In Sweden up to 25 GBq of radiocesium yearly are transferred to man via mushroom.

Mushroom also plays an important role in the uptake of radiocesium by vegetation at higher levels, and is probably the cause for radiocesium being easily available for a long time. Most animals show strongly increasing levels of radiocesium when mushrooms are available in August-September, and roe deer is one of the largest consumers of mushroom. Up to 20-30% of the paunch content is mushroom in this period.

#### Limnic systems: Ecological half-lives in Nordic lakes

The significance of fish consumption as a contributor to the internal doses of <sup>137</sup>Cs to man was demonstrated in previous NKS-projects. Main aim of the project has been to investigate the processes and mechanisms leading to radiocesium being easily available for uptake in fish. A Nordic map has been developed, containing descriptions of fallout, limnic data (such as water quality, size, water transport), radiocesium levels in freshwater fish and water, as well as runoff from surrounding areas. Resuspension of sedimented radiocesium, along with runoff from catchment area, has shown to be important sources for biological uptake, and thus the dominating factor contributing to long ecological half-lives in freshwater fish.

Under the assumption that the future decrease will be mainly governed by physical decay, the infinite time-integrated activity can be approximated to 10 to 20 kBq/kg per year, which then would give a significant contribution to critical groups. It is thus important to follow the time development of <sup>137</sup>Cs in fish, and the controlling factors of critical catchments and lakes.

#### Conclusions

Semi-natural systems are becoming increasingly more important with time when it comes to transfer of radionuclides to man. Ecological half-lives are increasing with time.

Key words: Radioecology, radiocesium, radiostrontium, Nordic natural environment, sheep, mushroom, fish, doses to man

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## 3. Sammendrag på norsk

Matvarer fra semi-naturlige områder, som for eksempel udyrket beitemark, skogog fjellområder og utmark står for en anselig andel av det total inntaket av radiocesium og radiostrontium, og derved dose til mennesker. Matprodukter fra intensiv landbruk viser korte halveringstider, slik at når tiden etter et radioaktivt nedfall øker, er det matvarer som sopp, vilt og ferskvannsfisk som dominerer menneskers inntak av radionuklider.

I dette EKO-2 prosjektet ble tre problemområder valgt ut; sau som beiter i utmark, soppens betydning for totalinntaket og ferskvannsfisk. Hovedmålet med prosjektene har vært å identifisere bidraget fra semi-naturlige områder, ved å bestemme økologiske halveringstider for utvalgte matvarer fra disse områdene, og derved bestemme doser til menneske. Ved å benytte disse halveringstidene i eksisterende modeller, kan vi redusere usikkerheten i doseberegningene. I en nedfallssituasjon kan en det derved hurtig danne seg et bilde av mulige konsekvenser, og deretter iverksette relevante mottiltak.

I de pågående prosjektene har vi produsert eller samlet data fra 8-11 år etter Tsjernobyl-ulykken. Tidsseriene har vært svært nødvendige for å si noe om økologiske halveringstider. I løpet av prosjektperioden har vi demonstrert at den tidsavhengige konsentrasjonen av radionuklider i matvarer fra de utvalgte områdene ikke er godt beskrevet ved noen "økologisk halveringstid". Når tiden fra et nedfall øker, øker også halveringstiden. Restituering av nordiske økosystemer etter forurensning av radiocesium fra Tsjernobyl-ulykken går gradvis langsommere, samtidig som det er store variasjoner mellom ulike områder når det gjelder sårbarhet og evne til restitusjon. Tilsvarende er de økologiske halveringstidene gradvis økende, og kan ikke betraktes som konstanter verken med tid eller rom. Selv om det ikke har vært enkelt å bestemme generelle økologiske halveringstider, så har prosjektet gitt oss nyttig informasjon om de mekanismene som styrer overføring av radionuklider, og bedre kunnskap om nordiske økosystemer.

## Saue-prosjektet

Jord – vegetasjon – sau - systemet har vært undersøkt i fem land; Island, Færøyene, Danmark, Sverige og Norge. Koordinert prøveinnsamling ble startet i 1990, og har vært gjennomført inntil 1997. En har observert store forskjeller i overføring av radioaktivitet, og ved å undersøke produksjonsintensivitet, biomasseproduksjon, klimatiske forhold, mengden av sopp tilgjengelig, inntak av jord og eksperimentelle studier med stabile elementer i jord, har vi vært i stand til å forklare noen av de forskjellene som vi observerer. Siden jord representerer et viktig reservoar for radionuklider i det terrestre miljø, har jordegenskapene vært

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den viktigste faktoren for å forklare de store forskjellene vi ser i overføringsfaktorer i de ulike beiteområdene.

## Skogsprosjektet

Tidligere har en viet lite oppmerksomhet mot matvarer som for eksempel ville bær, sopp ferskvannsfisk og vilt som kommer fra skogssystemet. Et spørreskjema har derfor vært utarbeidet, med fokus på konsum av ville bær og sopp, og undersøkelser har vært gjennomført i Danmark, Finland, Norge og Sverige. I Sverige er opp til 25 GBq radiocesium overført årlig fra sopp til menneske.

Sopp spiller også en viktig rolle når det gjelder opptak av radiocesium hos høyerestående vegetasjon, og er sannsynligvis hovedårsaken til at radiocesium er tilgjengelig for planteopptak over lang tid. De fleste dyr viser også sterk økning i radiocesiumkonsentrasjon når sopp er tilgjengelig i august-september i beiteområdene. Rådyr er en av de største konsumentene når det gjelder sopp, i perioder kan 20-30% av vominnholdet bestå av sopp.

## Limniske systemer: Økologiske halveringstider i nordiske innsjøer

Betydningen av fiskekonsum som et viktig bidrag til interndosen fra radiocesium hos mennesker ble demonstrert i forrige NKS-periode. Hovedmålet med dette prosjektet var å undersøke de prosessene og mekanismene som fører til at radiocesium er lett tilgjengelig for opptak i ferskvannsfisk. Et nordisk kart har blitt utviklet, med informasjon om tidligere nedfall, limniske data (som for eksempel vannkvalitet, størrelse, vanntransport), radiocesiumkonsentrasjoner i fisk og vann, og avrenning fra tilliggende områder. Resuspensjon av sedimentert radiocesium, sammen med avrenning fra området har vist seg å være viktige kilder til biologisk opptak, og derved de dominerende faktorene som bidrar til lange økologiske halveringstider i ferskvannsfisk.

Dersom en forutsetter at fremtidig reduksjon i radiocesiumkonsentrasjoner vil i hovedsak være regulert av fysisk halveringstid, vil den tidsintegrerte konsentrasjonen estimeres til mellom 10 til 20 kBq/kg fisk per år. Dette vil gi et signifikant bidrag til dosen til kritiske grupper. Det vil derfor være viktig å følge utviklingen i ferskvannsfisk fremover, og de kontrollerende faktorene for spesielt utsatte avrenningsområder og innsjøer.

#### Konklusjon

Semi-naturlige områder blir mer og mer viktige med tiden, når en ser på overføring av radionuklider til mennesker. Økologiske halveringstider øker med tiden, og kan ikke betraktes som konstanter.

Nøkkelord: Radioøkologi, radiocesium, radiostrontium, Nordiske naturlige systemer, sau, sopp, fisk, doser til mennesker

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## 5. Introduction and aims of studies

## 5.1 Background

After the Chernobyl accident it became clear that the transfer of radionuclides via food to man could result in significant internal radiation doses to the Nordic population. In the long term the most significant internal doses were expected to be related to the contamination of especially sensitive Nordic environments leading to a high transfer of radiocesium to man. Some of the sensitive pathways have been known for a long time, such as the lichen-reindeer-man, whereas others were less studied (fresh water fish in oligothropic lakes, the influence of mushroom in upland pastures, among others).

## 5.2 Previous NKS-works

Radioecology has long traditions within the framework of NKS, and focus has been put on special Nordic conditions. The previous RAD-programme (from the period 1990-1993) is well documented in the book «Nordic Radioecology. The transfer of radionuclides through Nordic ecosystems to man» published by Elsevier.

## 5.3 Main tasks

The main task of EKO-2 has been to investigate the fate of radionuclides in typical Nordic semi-natural or natural ecosystems, and determine the transfer to man. Some money has been available for co-ordinated sampling and analysis, but most of the work has been to collate existing data and applying simplified models to the data.

## 5.4 Organisation of the work

This NKS period started with a planning period in spring 1994. Hanne Solheim Hansen, Agricultural University of Norway, led the preparatory work, and compiled ideas and project plans from the Nordic community of radioecologists into what became the project plan for EKO-2 1994-1997. The plans were presented to a reference group, and changes were made according to comments from the reference group. The project plan was carefully made, and we have been able to follow it without major changes through this four-year period.

The project period started in fall, 1994, and Tone D. Bergan, Institute for Energy Technology, Norway, took over as project leader. EKO-2 has been organised in three separate sub-projects. Markus Meili, University of Uppsala, Sweden, have functioned as a sub-project leader for the lake project, the other two have been lead by Tone D. Bergan directly. In total, there have been 35 project participants.

Progress reports have been presented every half-year, and more detailed annual reports have reported both scientific progress and administrative matters. There was a halfway evaluation of all the projects early in 1996, and for this evaluation technical reports from all sub-projects were presented.

The project participants have met once or twice every year, either at working meetings or seminars. All the project participants should be thanked for their enthusiasm and positive attitude to NKS work. Scientific results and ideas have been freely exchanged, and this Nordic forum has been very important for keeping radioecolgical competence alive, and securing new life. As Henning Dahlgaard expressed in «Nordic Radioecology»: «A nuclear preparedness plan without working scientific projects is like an air force without trained fighter pilots.»

## 5.5 Funding

Funding for the project period has come from the NKS, and approximately 75% of the work has been financed by the participating institutions, or via other research programmes. The exact amounts have not been estimated, since it is mainly manhours and free use of laboratory facilities.

Funding granted from NKS has in total been 5 215 000. All figures are in Danish kroner (DKK). In general the money have been spent according to budget.

| Project        | 1994      | 1995      | 1996      | 1997      |
|----------------|-----------|-----------|-----------|-----------|
| 2.1 Sheep      | 560.000   | 580.000   | 630.000   | 600.000   |
| 2.2 Forest     | 220.000   | 260.000   | 320.000   | 205.000   |
| 2.3 Lakes      | 120.000   | 210.000   | 330.000   | 300.000   |
| Project leader | 130.000   | 250.000   | 250.000   | 250.000   |
| Total          | 1.030.000 | 1.300.000 | 1.530.000 | 1.355.000 |

The sheep project is financed also by the Norwegian Research Council (NFR) (Norwegian part), Swedish Radiation Protection Institute (SSI) (Swedish part). The forest project is financed by the Finnish Radiation Protection Institute (STUK) (Finnish part) and the Directorate for Nature Management (NINA) (Norwegian part). Within the lake project SSI, STUK and NFR have financed most of the work.

# 6. The sheep project: Transfer of radiocesium and radiostrontium from soil to vegetation and sheep

Uplands are generally considered as land that is not cultivated, but they are frequently used as rangelands. Many types of soil are present, but poor, organic and acid soils are prevailing. The lack of cultivation secures cycling of radionuclides, and the role of microbiological activity seems to be important for this recycling of radionuclides. Standardisation of sampling and data presentation has been important, so that the results are comparable.

The majority of lamb production in the Nordic countries occurs on uncultivated pastures or in semi-natural or natural environments. The length of the grazing period varies from 4 to 10 months, as plant growth does not take place in sufficient quantities to sustain grazing for the full year in any of the countries. Lambs are born in April to May and slaughtered at the end of the grazing season in September to October.

The extent and economical importance of sheep farming varies considerably. According to government statistics, the summer sheep stock in each country is: Denmark 111 000, the Faroe Islands 70 000, Finland 107 000, Iceland 700 000, Norway 2 211 000, and Sweden 406 000. In Iceland, Norway and the Faroe Islands, the sheep is the most abundant domestic animal and sheep farming is of relatively greater importance than in the other Nordic countries. The annual per capita consumption of lamb meat in Finland (0.3 kg), Sweden (0.7 kg), and Denmark (0.8 kg) is low compared to Norway (5.2 kg), the Faroe Islands (10 kg), and Iceland (24 kg).

#### The production method's influence on ecological half-lives in sheep meat

As part of the yearly mapping of radioactivity levels, the number of grazing animals, grazing area, the length of the grazing period and the carcass weight have been recorded. Also, average precipitation and temperature during the grazing period is recorded. When needed, chemical analyses of the soil and vegetation (nutrition levels) have been included.

## Study of the effects of intake of fungi

Faeces samples were collected during the grazing period at all sites. Unfortunately, neither 1995 nor 1996 were good mushroom years, and the amount of fungi spores in faeces samples varies from 1-5 spores per sample. This did not enable us to estimate the intake of fungi by sheep. Results from 1997 showed a dramatically increase of spores in faeces, according to more mushroom available in the pastures.



Figure 6.1. Different factors studied within the sheep project: climate, grazing intensity, biomass production, ingestion of mushrooms and soil, and the relationship between stable cesium/radiocesium and stable strontium/radiostrontium.

## Study of the effects of ingestion of soil

The study started in 1995, using scandium analysis of faeces as an indicator of soil intake. The method proved to be successful, and analyses have been performed on faeces samples from all sites. The scandium method is very sensitive and results show that the amount of soil ingested is very small at all sites. In some cases the vegetation contains more soil than what is found in sheep faeces, indicating that the sheep are grazing selectively on cleaner plants.

#### Study of the effects of mobility of stable cesium and strontium in soil

Soil and vegetation samples from all sites have been analysed for stable cesium and strontium, and other trace elements (Ce, Co, Cr, Eu, Fe, Rb, Sc, Se, and Zn). Stable Cs and Sr transfer factors give important information to the equilibrium transfer factors at different sites and help identifying sites that are ecological sensitive to Cs. Especially the soil from Iceland shows high transfer of stable Cs and Sr. Tracer studies of <sup>134</sup>Cs and <sup>85</sup>Sr have been included to study the rate of fixation to irreversibly bound sites in soils.

## 6.1 Study areas and sampling procedures

## 6.1.1 Description of the geographical areas

Sites which showed longer than anticipated half-lives in lamb's meat were already identified in the previous NKS-period. Their geographical location is indicated on the map in Figure 6.2. The sites were not representative for the country as a whole, but would still exhibit typical national problems (e.g. the Icelandic sites), and all of them represented semi-natural pastures.

Table 6.1 Size and localisation of the sampling areas in the different countries.

| Country       | Code | Grazing area         | No. of | Latitude      | Longitude     |
|---------------|------|----------------------|--------|---------------|---------------|
|               |      | size km <sup>2</sup> | sites  |               |               |
| Denmark       | DEN  | 0.09                 | 1      | 55.3°N        | 8.7°E         |
| Faroe Islands | FAI  | 29                   | 9      | 62.0°N        | 7.0°W         |
| Finland       | FIN  | 0.02                 | 2      | 60.9°N        | 23.5°E        |
| Iceland       | ICE  | 0.12                 | 2      | 64.4°N/63.6°N | 21.4°W/20.6°E |
| Norway        | NOR  | 0.004                | 1      | 66.6°N        | 12.4°E        |
| Sweden        | SWE  | 10                   | 1      | 64.4°N        | 14.4°E        |



Figure 6.2. Map with the selected sampling sites in the sheep project.

Table 6.2 Altitudes, soil types and vegetation types of the grazing areas.

| Country | Environment     | Altitude | Soil types            | Vegetation types              |
|---------|-----------------|----------|-----------------------|-------------------------------|
| code    |                 | m.a.s.   |                       |                               |
| DEN     | Coastal         | 2        | Sandy                 | Permanent grassland           |
| FAI     | Coastal         | 50-240   | Peaty                 | Permanent grassland           |
| FIN     | Inland          | 110      | Clay                  | Natural pasture <sup>*)</sup> |
| ICE     | Coastal         | 20       | Peat, gravelly        | Lowland mire                  |
| NOR     | Coastal         | 10       | Peaty                 | Permanent grassland           |
| SWE     | Mountain forest | 580-770  | Peat, gravelly, sandy | Mountain moor, Betula forest, |
|         |                 |          | moraine               | permanent grassland           |

 $^{\ast)}$  50 % grass-growing old field, 50 % forest meadow

| Country code | Mean temperature   | Mean precipitation  |
|--------------|--------------------|---------------------|
|              | May-September (°C) | May-September (mm). |
| DEN          | 13.9               | 332                 |
| FAI          | 9.2                | 426                 |
| FIN          | 12.6               | 310                 |
| ICE          | 8.1                | 291                 |
| NOR          | 11.0               | 434                 |
| SWE          | 9.5                | 466                 |

*Table 6.3 Data on precipitation and temperature from weather stations adjacent to the respective grazing areas.* 

## 6.1.2 Soil Characteristics

Soil represents the major sink for radionuclides released into the terrestrial environment. In any soil-plant-animal system, soil and soil characteristics becomes important parameters when looking at transfer. The general soil characterisiticas are given in Table 6.4.

Table 6.4 Chemical data for the top soil layer (0-10 cm) of the grazing area. Mean  $\pm$  standard deviation for the sample sites

| Country | Organic matter | pH (H <sub>2</sub> O) | mg pe            | er 100 g dry s | oil               |
|---------|----------------|-----------------------|------------------|----------------|-------------------|
| code    | % w/w          |                       |                  |                |                   |
|         |                |                       | Ca <sub>AL</sub> | $K_{AL}$       | K <sub>HNO3</sub> |
| DEN     | $8.2\pm0.6$    | $5.1 \pm 0.7$         | $39 \pm 6$       | $21 \pm 5$     | -                 |
| FAI     | $51 \pm 21$    | $4.8 \pm 0.4$         | -                | $52 \pm 22$    | -                 |
| FIN     | 17             | 5.0                   | 87               | 22             | 155               |
| ICE     | 52.0           | 6.0                   | 50               | 7.6            | 25                |
| NOR     | $60 \pm 26$    | $5.1 \pm 0.8$         | $737 \pm 259$    | $34 \pm 21$    | $53 \pm 19$       |
| SWE     | $10.0\pm4.0$   | $4.6\pm0.3$           | $36 \pm 10$      | $12.1\pm4.5$   | $32 \pm 10$       |

#### 6.1.3 Chemical composition and nutritive value of the vegetation

Samples of vegetation from the study sites were taken in Sweden, Denmark and Norway for analyses of chemical composition and metabolizable energy (ME) content. The analyses were performed according to conventional methods referring to ruminants. The results indicate, when compared with standard values of grass of second cut, for the Swedish herbage a very low content of ME, 6.0 and 6.5 MJ per kg dry weight (d.w.), a low crude protein content, a relatively high content of crude fat, possibly due to cortex waxes, a low content of ash and a "normal" content of calcium, while the content of potassium was very low. The three

vegetation samples taken in Denmark and Norway had a "normal" content of ME, 9.2 and 10.0 MJ/kg d.w., respectively, a high crude protein content and a "normal" potassium content. The content of calcium was "normal" in the Norwegian samples, while the Danish samples had a low content, about 3 g/kg d.w. No vegetation samples were collected in Iceland or The Faroe Islands for nutritive analyses.

## 6.2 **Production intensity**

Both biomass (herbage) production and grazing intensity (stocking rate) was studied at the different sites. Herbage density was recorded at the cut- and sampling occasion each year. The herbage density was fairly similar from year to year.

The grazing intensity varied from site to site. Normally, low biomass production of the grazing area was compensated by a low grazing intensity. As the soil ingestion by animals showed to be low, grazing intensity seem to not be important when explaining the differences observed between the different sites.

## 6.3 The influence of fungi in the grazing area

## 6.3.1 Occurrence of mushrooms and their contents of <sup>137</sup>Cs

Mushrooms among which the most frequent represented species of *Russula*, *Cortinarius, Lactarius, Leccinium* and *Rozites* were collected each year, and <sup>137</sup>Cs concentrations within the range of 200 and 80 000 Bq/kg d.w. were detected. As shown in the forest project, the concentrations of <sup>137</sup>Cs in mushroom did not decrease during the study period.

## 6.3.2 Collection and analysis of spores in faeces

In deer it has been shown that fungi make up a large part of the diet during the mushroom season (Fraiture, 1992). Later it was shown that mushrooms are responsible for an increase in the concentration of radiocesium in meat from roe deer, this was done through studies of the occurrence of mushroom spores in faeces (Strandberg and Knudsen, 1994). Rafferty *et al.* (1994) found that among sheep grazing in the same area, those having access to areas with fruitbodies of species having ectomycorrhiza with trees, had the highest levels of radiocesium in the meat.

If mushrooms with a high concentration of radiocesium generally are eaten by ruminants both in household and nature, a reasonable countermeasure will be to relate the season of hunting or slaughter to a time of the year when mushrooms are sparse. From studies of the relation between the intake of mushrooms and the occurrence of spores in the faeces of sheep it can preliminary be said that one spore in 0.3 mg dry weight of sheep faeces equals a daily intake of 12 g fresh

mushrooms (Strandberg and Hansen in prep.) At the time being this is a rough estimate, and it is likely that further investigations may increase the information about the dependence upon the spore type i.e. thick-walled spores are more likely to pass intact through the gastro intestinal tract (GIT) than spores with thinner walls. Hence it may be necessary to operate with more conversion factors than the one used in this study i.e. the more the spore is adapted to pass through the GIT the smaller the intake of fresh mushrooms per spore found in the faeces is.

Sheep faeces from the four Nordic sampling sites were collected. The faeces were sampled during the mushroom season for the purpose of analyses for occurrence of mushroom-spores in the faeces. The faecal pellets were grinded gently to obtain a dry powdery material. A small sample (0.3 mg) of this material was soaked with distilled water and placed under the light microscope. All the samples were studied for occurrence and counting of mushroom spores at 20 times magnification. All types of spores found were studied more detailed at 40 times magnification for further identification. Identification was carried out according to Hansen & Knudsen (1995).

#### 6.3.3 Consumption of fungi by sheep

#### Iceland 1990 (Table 6.5)

Only few mushroom spores (0 - 1, on average 0.4 per specimen) were found in the samples from June and July 1990. In the samples from August and September 0 - 3, on average 1.7 spores per specimen were detected. This equals a daily intake of 4 g in June and July and 20 g in August and September. The spores were from typical grassland species of mushrooms, *Stropharia semiglobata* and probably *Stropharia coronilla*, *Psilocybe sp*. one species of *Entoloma* and probably *Clitocybe* and some *Coprinus spp*. Most of the spores are from species commonly found associated with dung. The amount of spores found in the faeces gives only a rough indication of the amount of fungi eaten by Icelandic sheep in the mushroom season.

The intake of mushrooms increases over the season from June to July. There was no difference between sheep and lamb as regards the amount and species ingested. The most abundant spores are from the genus *Stropharia* which have thick walled spores adapted to survive the passage through the GIT. There were no observations of spores from species known to concentrate radiocesium.

#### Norway 1995 (Table 6.6)

At the Norwegian sampling site, the daily intake was the highest in the beginning of the season with 28 g mushroom fresh weight. In July and August the intake decreased to between 8 and 12 gram of fresh weight per day. The species eaten were ordinary grassland mushrooms, many of them associated with dung from ruminants (Table 5). None of the spores found are from species known to concentrate radiocesium.

#### Sweden 1994 (Table 6.7)

On the locality Blomhöjden faeces were sampled twice during the mushroom season of 1994. Three samples were from the 25th of July and another three were from the 23rd of August. From these samples the number of spores detected were between 0 and 1. Both in July and August one out of three samples contained one spore. Both was non-pigmented spores, the genus could be *Marasmius* and the spore from August could belong to the species *Marasmius oreades*, which commonly grows on grassland. Contrary to expectation no spores from any species of *Boletus* or related species were found, although samples of mushrooms has demonstrated that these species are available in the forested areas at Blomhöjden. The spores represented in the faeces are not from species that could explain an increased amount of radiocesium in the faeces.

The number of spores found in the faeces of the Swedish sheep in 1994 indicates a consumption of mushrooms less than 10 g of fresh weight per day during July and August. Spot-tests from the material from Blomhöjden from 1995 and 1996 did not indicate signs of ingestion of mushrooms beyond what was observed in the 1994 material.

The relative high transfer of radiocesium to Swedish sheep at Blomhöjden should probably not be explained by an extraordinary high consumption of mushroom. Rather the explanation could be that the diet of the Swedish sheep contains a lot of herb species other than grass. If Ericaceous species e.g. bilberry, *Vaccinium myrtillus* and cowberry, *Vaccinium vitis-idaea* are included in the diet it may explain a higher level in the meat than would be expected if the diet was pure grass.

#### The Faroe Islands 1996 (Table 6.8)

From the Faroe Islands material sampled from 5 localities in June 1996 was analysed for occurrence of spores. Only few spores were found in the faeces. Two different species of *Agaricus*, the one probably *Agaricus campestris* and the other not identified. Then also a spore from the genus *Inocybe* was found. This genus forms ectomycorrhiza with different tree species and also some herbs. The daily intake of mushrooms ranged between 0 and 30 g, depending upon locality. None of the species identified is known to concentrate radiocesium.

| Location | Month | Spores<br>per specimen | Mean spores | SD   | Daily intake<br>(gram fresh weight) |
|----------|-------|------------------------|-------------|------|-------------------------------------|
| Iceland  | Jun   | 0                      |             |      |                                     |
| Iceland  | Jun   | 1                      | 0.33        | 0.58 | 4                                   |
| Iceland  | Jun   | 0                      |             |      |                                     |
| Iceland  | Jul   | 1                      |             |      |                                     |
| Iceland  | Jul   | 0                      |             |      |                                     |
| Iceland  | Jul   | 1                      | 0.50        | 0.58 | 6                                   |
| Iceland  | Jul   | 0                      |             |      |                                     |
| Iceland  | Aug   | 2                      |             |      |                                     |
| Iceland  | Aug   | 1                      |             |      |                                     |
| Iceland  | Aug   | 2                      | 1.60        | 0.55 | 19                                  |
| Iceland  | Aug   | 2                      |             |      |                                     |
| Iceland  | Aug   | 1                      |             |      |                                     |
| Iceland  | Sep   | 1                      |             |      |                                     |
| Iceland  | Sep   | 3                      |             |      |                                     |
| Iceland  | Sep   | 3                      | 1.80        | 1.30 | 21                                  |
| Iceland  | Sep   | 2                      |             |      |                                     |
| Iceland  | Sep   | 0                      |             |      |                                     |

Table 6.5. Results from Iceland 1990

Table 6.6 Results from Norway 1995

| Location | Month | spores<br>per specimen | Mean<br>spores | SD   | Daily intake<br>(gram fresh weight) |
|----------|-------|------------------------|----------------|------|-------------------------------------|
| Larvekra | Jul   | 2                      |                |      |                                     |
| Larvekra | Jul   | 0                      | 2.33           | 2.52 | 28                                  |
| Larvekra | Jul   | 5                      |                |      |                                     |
| Larvekra | Aug   | 1                      |                |      |                                     |
| Larvekra | Aug   | 1                      | 0.67           | 0.58 | 8                                   |
| Larvekra | Aug   | 0                      |                |      |                                     |
| Larvekra | Sep   | 2                      |                |      |                                     |
| Larvekra | Sep   | 1                      | 1.00           | 1.00 | 12                                  |
| Larvekra | Sep   | 0                      |                |      |                                     |

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| Location    | Month | spores<br>per specimen | Mean<br>spores | SD   | Daily intake<br>(gram fresh weight) |
|-------------|-------|------------------------|----------------|------|-------------------------------------|
| Blomh. 1994 | Jul   | 0                      |                |      |                                     |
| Blomh. 1994 | Jul   | 1                      | 0.33           | 0.57 | 4                                   |
| Blomh. 1994 | Jul   | 0                      |                |      |                                     |
| Blomh. 1994 | Aug   | 0                      |                |      |                                     |
| Blomh. 1994 | Aug   | 0                      | 0.33           | 0.57 | 4                                   |
| Blomh. 1994 | Aug   | 1                      |                |      |                                     |

Table 6.7 Results from Sweden 1994

Table 6.8 Results from the Faroe Islands 1996

| Location  | Month | spores       | Mean   | SD   | Daily intake        |
|-----------|-------|--------------|--------|------|---------------------|
|           |       | per specimen | spores |      | (gram fresh weight) |
| Hvalvik   | Jun   | 0            |        |      |                     |
| Hvalvik   | Jun   | 2            | 1.00   | 1.41 | 12                  |
| Bøur      | Jun   | 2            |        |      |                     |
| Bøur      | Jun   | 3            | 2.50   | 0.71 | 30                  |
| Velbastad | Jun   | 0            |        |      |                     |
| Velbastad | Jun   | 0            | 0.00   | 0.00 | 0                   |
| Hvalba    | Jun   | 1            |        |      |                     |
| Hvalba    | Jun   | 0            | 0.50   | 0.71 | 6                   |
| Sumba     | Jun   | 0            |        |      |                     |
| Sumba     | Jun   | 0            | 0.00   | 0    | 0                   |
| Total     |       | 8            | 0.80   | 1.14 | 9,6                 |

Table 6.9 List of species and genera recognised from spores found in faeces from the localities.

| Iceland   | Norway   | Sweden               | The Faroe Islands  |
|---|--|----------------------|--|
| 1990  | 1995   | 1994                 | 1996   |
| Stropharia<br>semiglobata                                       | Stropharia<br>semiglobata  | Marasmius<br>oreades | Stropharia sp.   |
| Stropharia sp.<br>Entoloma sp.<br>Clitocybe sp.<br>Coprinus sp. | Stropharia sp.<br>Psilocybe sp.<br>Omphalina sp.<br>Not identified 2 | Not identified 1     | Agaricus campestris<br>Agaricus sp.<br>Inocybe sp.<br>Not identified 1 |
| Not identified 3  | Not identified 2   |                      | Not identified 1   |

Consumption of mushrooms was not revealed as a major source of radiocesium to sheep at the localities examined through this investigation. However, a marked difference among years may exist, i.e. in years with a long lasting mushroom season sheep may learn to exploit this source of food. It must be pointed out that neither of the investigated years were good mushroom years, on the contrary, these years were especially poor mushroom years in general. In normal years, only few animals will spend time searching for mushrooms.

Probably the higher uptake rates observed in Sweden and on the Faroes should be explained by differences in the composition of the diet between the different countries i.e. localities. Consumption of ericaceous shrubs like *Calluna vulgaris*, Vaccinium myrtillus & Vaccinium vitis-idaea can explain the differences in transfer from soil to meat between the localities. Hence it is important to know the precise composition of the sheep diet at least in the period before they are slaughtered. In this context a few samples of grass does not satisfactorily represent the diet. Observations from the Faroes clearly demonstrates that sheep consumes large amounts of heather Calluna vulgaris. Faecal samples from sheep that consumes mushroom could be screened by an investigation of the radiocesium and radiopotassium content. In the case of a significant mushroom consumption these concentrations should be at least a factor two higher than in faeces collected from the same place when there is no mushrooms available. If there is an increased amount of radiocesium in the mushrooms consumed, the radiocesium content in the faeces will be increased. If the mushrooms consumed belongs to species with a low radiocesium content e.g. Agaricus spp., then the high concentration of potassium in mushrooms will be reflected by the <sup>40</sup>K concentration in the faeces.

## 6.4 Soil ingestion by animals

## 6.4.1 Literature survey

The main pathway of radiocesium to grazing lambs is by the consumption of vegetation, but also other pathways may be possible. As mentioned above consumption of fungi may not be excluded, even if in the present study only small numbers of fungi spores were found in the lamb faeces. Other radiocesium vectors may be ingested soil particles or drinking water. In connection to the EKO-2 project a literature review was therefore performed on the phenomenon soil ingestion by farm animals (Herlin & Andersson, 1996). A summary of the survey is given in the following. Herlin & Andersson (1996) give references in the original report.

Soil ingestion in farm animals occurs mainly under grazing conditions. It takes place either as inadvertent intake due to soil adhered to vegetation caused by rain splash, resuspension or wind erosion or as an active ingestion suggested being due to lack of essential mineral elements such as Cu, Co and Mn. Among domestic animals sheep and cattle have been more investigated in this field than horses and pigs. With the ingested soil different contaminants like heavy metals, radionuclides, chemicals and pesticides can be consumed. These elements are often found in high concentrations in the upper soil layers and are more or less transferred to meat and milk depending on, e.g. the adhesion of the specific element to the soil particles.

#### 6.4.2 Methods for measuring soil content in plants and faeces

Measuring methods are referred like soil ingestion estimates. A rough method for measuring the soil content is the determination of ash content (acid insoluble residue method, AIR-method) (Healy & Ludwig, 1965). The AIR is taken as a measure of soil ingested, and is related to it by the formula given by Healy & Ludwig (1965) and Fleming (1986):

AIR = (100 - S) X / 100 + Y / 100,

where S is the percentage of soil in faeces, X is the percentage AIR derived solely from plant material and Y is AIR in per cent of soil. The determination of ash content in plant material or faeces is, however, considered as a rough method, which only approximately estimates the soil content (Mayland *et al.*, 1975).

A soil element that is not incorporated by plants and not digested by the animal can be determined and thus, knowing the concentration of the element in the soil, the soil content of the matter (plant surface contamination or faeces) can be calculated. This procedure has been proven to the most convenient and accurate (Mayland *et al.*, 1975; Lie *et al.*, 1994). Several elements have been suggested and evaluated in order to determine soil concentration in plant material and faeces, such as titanium (Ti) by using spectrophotometri (Sherman & Kanehiro, 1965) and scandium (<sup>45</sup>Sc) by using neutron activation analysis (Oughton & Day, 1993; Lie *et al.*, 1994). The determination of <sup>46</sup>Sc by neutron activation analysis seems to be the most reliable.

Calculation of daily intake (in per cent of dry matter, DM, intake) of soil can be made by using the equation suggested by Thornton & Abrahams (1983):

Soil ingested, per cent of DM intake = $[(1 - D_h)Ti_f / Ti_s - D_hTi_f]$  100,

where  $D_h$  is the herbage digestibility,  $Ti_s$  the titanium concentration of DM in soil and  $Ti_f$  the titanium concentration of DM in faeces. Scandium content and other elements fulfilling the criteria of not being incorporated by plants or digested by the animal can also be used for the calculation.

Russell et al. (1985) presented another equation, which avoids estimating herbage digestibility. The fraction of intake of an element (F) coming from ingested soil can here be calculated:

 $F = (Ti_{f} E_{s}) / (E_{f} Ti_{s}),$ 

where  $E_f$  is concentration of an element in faeces and  $E_s$  the concentration of an element in soil.

## 6.4.3 Investigations on soil ingestion in animals

Results from investigations on ingested soil measured as faecal soil in different animal species kept on pasture or stable fed, respectively, are reviewed in the report. Factors that influence on the intake of soil are listed, such as animal species, available forage and root intake, season and soil type. The influence by animal species can be accounted by factors as different grazing behaviour: the way of separating the grazed plant parts from the rest of the plant, how close to the ground the grazing is performed and soil contamination of plants by treading due to the difference in claw size, live weight and amount of locomotion. Reports are referred to, demonstrating that sheep as being close grazers are more likely to ingest greater amounts of soil than cattle (Green & Dodd, 1988), e.g. as much as 30 % of the daily dry matter intake versus 18 %, respectively (Thorntorn & Abrahams, 1983). In another report a daily soil intake per sheep of about 0.7 kg is mentioned (Vaithiyanathan & Singh, 1994)

Soil in faeces of, e.g. sheep has been found to increase when the availability of feed decreases. Growth rates of the plants and stocking rate are therefore important factors in this context. The soil consumption is considered to be more dependent on intake of soil-contaminated roots that are pulled up than on soil dust on leaves and stems. The influence of season on soil consumption often reflects the availability of feed.

Low soil ingestion has been found when animals are kept on well-drained soils, which have a strong structure. In contrast, soils with a weak structure and poor drainage are associated with high pasture soil contamination (Healy, 1968).

## 6.4.4 Radioactivity transfer by soil intake

Reports are referred (Wilkins & Green, 1988; Riise et al., 1990; Green et al., 1995) demonstrating that <sup>137</sup>Cs in soil has a low bioavailability. In these extraction or in vitro experiments only a small portion of the total activity intake was found to be soluble in the rumen fluid. Radionuclides, such as <sup>137</sup>Cs were most readily extracted from organic soils (Cooke et al., 1995). Several studies have shown that clay minerals have binding effects on the <sup>137</sup>Cs transfer to animal products.

## 6.4.5 Soil ingestion by lambs in the present study

In the present Nordic study the soil ingestion by lambs was studied, to demonstrate possible radioactivity transfer to lamb meat (Oughton & Standring) Soil content in faeces was measured by determination of the content of scandium by neutron activation analysis according to Oughton & Day (1993). It can be concluded that soil ingestion by the lambs was low in all countries and was thus probably negligible as a pathway of activity transfer to lamb meat.

## 6.5 Mobility of stable Cesium and Strontium

Comparison of radionuclides with the stable elements can provide valuable information on the «equilibrium status» of the deposited radionuclide within the ecosystem, and can be a considerable aid in prediction of future transfer. As <sup>137</sup>Cs is transported down the soil profile, and with binding to the inaccessible sites in the clay matrix, the plan-to-soil concentration ratio for <sup>137</sup>Cs will be expected to approach that of stable Cs. Compared to <sup>137</sup>Cs, the stable Cs concentrations are relatively constant down the soil profile.

Soil, vegetation and lamb muscle samples were collected from the study sites in Norway, Sweden, Iceland, Denmark and Faroe Islands. Soils were air dried prior to extraction and tracer studies. Standard soil characteristics were measured in all soil samples included in tracer studies. Stable Cs and other trace element concentrations were determined by neutron activation analysis (NAA).

Sorption kinetics in soils were studied using controlled laboratory tracer ( $^{134}$ Cs and  $^{85}$ Sr) experiments. Particular emphasis was placed on the rates of change of radionuclide mobility, sorption mechanisms and speciation in soils. Desorption studies included the application of sequential extraction techniques. Aliquots of soil were removed as a function of time (up to 12-18 months) and extracted first with water and then with 1 M NH<sub>4</sub>Ac (soil pH).Extracts and residues were counted, and the rate of adsorption calculated as a function of time after addition.

Variations in <sup>137</sup>Cs transfer factors have been observed between the different Nordic sites (Hove et. al., 1994). This could be due to site-specific environmental parameters such as soil characteristics or agricultural practice, or it might reflect varying degrees of isotopic exchange - confounded by the fact that the sites have varying contributions from weapons and Chernobyl fallout.

Results from sorption studies showed that both <sup>134</sup>Cs and <sup>85</sup>Sr became rapidly associated with soil components. After 1 hour contact time with soils, more than 90% of the total activity was adsorbed to soils: less than 10 % was displaced with a water extraction. However, soil-bound <sup>85</sup>Sr was significantly more mobile than <sup>134</sup>Cs: between 60 and 100 % of the sorbed <sup>85</sup>Sr could be displaced from binding sites by a single NH<sub>4</sub>Ac extraction, compared to 4 to 35 % for <sup>134</sup>Cs. Both nuclides

showed a time-dependent increase in the degree of sorption to soils, the percentage extracted both by water and by  $NH_4Ac$  decreasing with contact time.

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Figure 6.3 A simplified model for radiocesium and radiostrontium in soil, and the availability for plant uptake.

Laboratory studies show that the mobility of <sup>134</sup>Cs tracers added to soils decreases with time after addition. Sequential extraction studies show that radiocesium is rapidly and strongly bound to soils. Use of a simple box model provides estimates of the long-term fixation rates of <0.0001 - 0.00059 d<sup>-1</sup>, equivalent to half-times of 3 to > 15 years. Rate constants for other sorption processes (e.g. ion-exchange and sorption to reversibly bound sites) are faster, with half-lives from hours to days.

Fixation rates are of the same order of magnitude as ecological half-lives in soilplant-sheep system, adding support to the assumption that radionuclide fixation in minerals will have a strong influence on ecological half-lives.

Decreases in the mobility of <sup>134</sup>Cs tracers does not follow simple: sorption and fixation rates decrease with time. This both supports and stresses the importance of observations that ecological half-lives need not be constant as a function of time. Of course, for modelling purposes, fixation rates are just one of the rate determining parameters that can influence ecological half-lives (transport down the soil profile and removal with produce and run-off are almost certainly also important).

Tracer studies suggest that the fraction of mobile Cs and Sr in soils can show considerable variation between the different sites, especially in the first 100 days after contact time. There was evidence that soils with reduced organic content often showed reduced mobility and faster fixation rates (e.g. Ribe).

As Sr is not fixed in these soils, the rate-determining parameters for ecological half-lives would be expected to be transport down the soil profile and removal with produce and run-off.

Analysis of stable Cs shows a large variation in trace element concentrations and soil-to-plant concentration ratios between the different sites. However, observed concentration ratios for stable Cs showed good correlation with <sup>137</sup>Cs concentration ratios, suggesting that differences between the locations were due to site-specific factors rather than source term or lack of equilibrium. Stable Cs analysis can give some indication of the potential vulnerability of ecosystems due to high transfer factors.

Plant-to sheep concentration ratios are similar, apart from the Faroe Islands. Although some Faroe Island vegetation samples showed evidence of higher levels of soil contamination as compared to other sites, up to 16 mg soil per g vegetation d.w., preliminary calculations suggest that this was not large enough to have a significant influence on stable Cs levels in vegetation

## 6.6 Concentrations of <sup>137</sup>Cs - Transfer Parameters

The following parameters are used for describing the radioactivity transfer in different steps of the chain soil - plant (vegetation) - lamb meat.

 $TF = Transfer factor for {}^{137}Cs from soil to plant, calculated as: Bq/kg dry weight plant per kBq/ m<sup>2</sup> soil.$ 

- $CF = Concentration factor for {}^{137}Cs$  from vegetation to lamb meat, calculated as: Bq/kg fresh weight meat per Bq/kg dry weight plant.
- $T_{ag}$  = Aggregated transfer factor for <sup>137</sup>Cs from soil to meat, calculated as: Bq/kg fresh weight meat per kBq/ m<sup>2</sup> soil.

The concentration of <sup>137</sup>Cs in lambs are based on live monitoring or meat samples after slaughtering. The effective ecological half-life of <sup>137</sup>Cs in pasture-sheep is calculated by taken the average activity levels measured by live monitoring in the period 8-12 weeks of grazing. An exponential regression curve was fitted to the data to estimate the effective ecological half-life of <sup>137</sup>Cs.

## 6.6.1 Concentration of <sup>137</sup>Cs in Soil and Vegetation. Radiocesium Transfer from Soil to Plant

Data on mean <sup>137</sup>Cs concentrations in soil for the different sites each year are summarised in Figure 6.4. In general, the concentration in soil showed a decrease from 1990 to 1997, but with great variation. A new Icelandic site was chosen in 1995, with a slightly higher deposition of <sup>137</sup>Cs. The variation illustrates the inhomogeneity within a grazing area, even when the same method for sampling and measurements is applied each year. The deposition varies from 2.9 kBq/m<sup>2</sup> at the Danish site, to 34 kBq/m<sup>2</sup> at the Norwegian site (average values). The origin of the deposition also varies, for the Icelandic, the Faroe Islands and Danish site, the global fallout from nuclear weapons tests is dominant, whereas for Norwegian and the Swedish site, Chernobyl fallout is dominant.



Figure 6.4  $^{137}$ Cs concentrations in soil at the different sites, and the variation with time. The concentration is expressed as  $kBq/m^2$ .

The <sup>137</sup>Cs concentration in vegetation is illustrated in Figure 6.5. The average <sup>137</sup>Cs concentration in the collected single plant species was about the same as that in the cut herbage.



Figure 6.5  $^{137}Cs$  concentrations in vegetation at the different sites, and the variation with time. The concentration is expressed as Bq/kg dry weight.

# 6.6.2 Concentration of <sup>137</sup>Cs in Lamb Meat. Radiocesium Transfer from Plant to Meat and from Soil to Meat.

The concentration of <sup>137</sup>Cs in lamb's meat obviously reflects the amount of deposited <sup>137</sup>Cs in each site. However, also the time dependent development varies from site to site. This is better reflected looking at the transfer at each site.



Figure 6.6<sup>137</sup>Cs concentrations in lamb's meat at the different sites, and the variation with time. The concentration is expressed as Bq/kg fresh weight.



Figure 6.7 Transfer of <sup>137</sup>Cs from soil to grass at the different study sites.

The transfer factor of <sup>137</sup>Cs from soil to grass (TF) for each year are shown in Figure 6.7, demonstrating great variations. At some sites the TF decreases, indicating that radiocesium will be less available by root uptake over time. The TF varies from 0.6 at the Danish site to 59 at the Swedish site (average values for 8 years).



*Figure 6.8 Transfer of*<sup>137</sup>*Cs from grass to meat at the different study sites.* 

The concentration factor (CF) from grass to lamb's meat varied from year to year. No evident time trend could be demonstrated, if any, there was a possible increase with time. The range for this transfer is considerably smaller (0.2 to 1.8) than the range for transfer from soil to plant. The range might indicate an increased bioavailability of <sup>137</sup>Cs in plant, but this conclusion may be incorrect if there are other pathways present of radioactivity transfer to lamb meat than through plant intake.

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Figure 6.9 Aggregated transfer of  $^{137}Cs$  from soil to meat at the different study sites.

The aggregated transfer factor  $(T_{ag})$  varies from 0.4 at the Danish site to 47 at the Swedish site. No evident time trend could be demonstrated, Figure 6.9.

## 6.7 Effective Ecological Half-Lives of <sup>137</sup>Cs

The effective ecological half-lives of <sup>137</sup>Cs during the period 1990-1997 in lamb's meat are summarised in Table 6.10. Calculations are made according to the observed values in each year.

The effective ecological half-life of <sup>137</sup>Cs was thus estimated to 19.3 years in the soil, 8.0 years in the vegetation and 20.9 years in the lamb herd. Corresponding values for the ecological half-life (based on data corrected for physical decay to August 1990) were 54.7, 11.0 and 66.6 years. The results show a long half-life for <sup>137</sup>Cs, especially in the soil and lamb herd. This indicates that radiocesium is becoming fixed in the soil, being less available for root uptake in the vegetation and finally that there must be another radiocesium pathway to the lambs, besides by the intake of vegetation.

| Country | T <sub>eff</sub> whole period | T <sub>eff</sub> 1990-1993 | T <sub>eff</sub> 1994-1997 |
|---------|-------------------------------|----------------------------|----------------------------|
| DEN     | 1.6                           | 0.9                        | 3.5                        |
| FAI     | 10.6                          | 1.7                        | 6.7                        |
| ICE     | 32.4                          | 32.4                       | - 10                       |
| NOR     | 6.8                           | 20.8                       | 10.3                       |
| SWE     | 20.9                          | 3.4                        | 18.0                       |

*Table 6.10 The effective ecological half-life of* <sup>137</sup>*Cs in lamb's meat during the period 1990-1997.* 

## 6.8 Modelling

A simple dynamic model was developed and used for the analysis of the data (Nielsen, 1994). The model incorporates an atmospheric compartment, a soil compartment, two grass compartments (one for the external plant and one for the internal plant), and two lamb compartments (one for the gastro-intestinal tract and one for the meat) representing a single animal. The atmosphere, soil and grass compartments are associated with a surface area of one square metre.

The model is based on first-order differential equations where the transfer of radiocesium between the model compartments is based on rate constants that determine the fractional transfer between the compartments for each time step. The model included a single surface soil compartment from which radiocesium was transferred partly to vegetation by root uptake and partly to deeper soil. The model structure was adjusted for the present work by including an additional soil compartment to be able to simulate two fractions of radiocesium in the surface soil, a mobile fraction and a fixed fraction. The modelling concept of separating radiocesium in the soil into two fractions is a step toward a more realistic simulation considering the well-known strong fixation of radiocesium to certain soil minerals. Greater detail can be incorporated into the model by including more than two fractions for radiocesium in surface soil, but it has been considered important to keep the model simple without more detail than justified from the basic data. The simulated transfer of radiocesium to vegetation by root uptake is possible from the mobile fraction, only, but exchange between the two soil fractions is included.

The model assumes an initial contamination of the surface soil from which a fraction is resuspended into the atmosphere compartment which is limited by an atmospheric mixing height of 1000 m. The simulation of the resuspension is based on a resuspension factor of  $10^{-9}$  /m. From the atmospheric compartment the activity is transferred to the ground through the application of a generic deposition velocity of 0.023 m/s representing both wet and dry processes. The deposition is split between a fraction of 30% intercepted by vegetation surfaces and the
remainder which is transferred to the mobile soil compartment. From the mobile soil compartment the bioavailable activity is reduced through the application of a removal process, e.g. run off or transfer to deeper soil layers. The model implementation of root uptake from soil to grass is based on the concentrationratio concept.

For simplicity, generic assumptions are made for annually occurring events. The growing season is assumed to start every year at day no. 100 and end at day no. 300. The lambs are assumed to be born at day no. 50, let out on pasture on day no. 120 and slaughtered at day no. 300. The model assumption of slaughtering the lambs at 8 months is at the high end compared with the field data which represent a range from 4 to 8 months.

The transfer of the activity in the lamb is simulated by two compartments only. The daily intake of radiocesium by the lamb is calculated from the current concentration in the grass and the amount eaten per day. From the meat compartment there is a transfer through other metabolic processes corresponding to a biological half life of three weeks.

Site-specific data on soil and biomass densities were collected. The values are shown in Table 6.11. Furthermore, information was collected on rates of lamb consumption covering average individuals and high-rate consumers (critical groups) in the countries. These rates are used for dose calculations and are shown in Table 6.12.

Table 6.11 Site-specific values for soil and grass densities at the study sites.

| Location   | Soil density                  | Grass density          |  |
|------------|-------------------------------|------------------------|--|
|            | $(\text{kg d.w.}/\text{m}^3)$ | $(\text{kg d.w./m}^2)$ |  |
| Ribe       | 1000                          | 0.18                   |  |
| Tjøtta     | 520                           | 0.15                   |  |
| Faroes     | 214                           | 0.10                   |  |
| Blomhöjden | 800                           | 0.15                   |  |

| Country | Average Individual | Critical Group |
|---------|--------------------|----------------|
|         | (kg/y)             | (kg/y)         |
| Denmark | 1.1                | 17             |
| Norway  | 6                  | 15 (*)         |
| Faroes  | 19                 | 25 (*)         |
| Sweden  | 0.5                | 7              |

*Table 6.12 Lamb consumption data used for dose calculations.* 

(\*) estimated by the authors

The data on radiocesium in soil, grass and lamb from the four sites were analysed with the model by adjusting parameters to obtain the best possible agreement between observations and predictions.

The results of the model calculations with the annual mean of the observations are illustrated in Fig. 6.10 which shows the model results and the observations for comparison for each of the four sites. The calculated effective half lives of radiocesium in the surface soil layer vary across the sites from a value of 10 y at Ribe to a value at the Faroes of 30 y which is equal to the physical half life of <sup>137</sup>Cs. These values are higher (typically a factor 2) than those that were used by Aarkrog (1994) for the effective ecological half lives for <sup>137</sup>Cs in Nordic lamb. The relative amounts of mobile radiocesium in soil at steady state ( $F_{mob}$ ) is showed in Table 6.13. The values are correlated with the amount of Chernobyl fallout ( $R^2 = 0.9$ ).

*Table 6.13 Values of*<sup>137</sup>*Cs deposition in 1990 and parameters derived from model calculations with the annual mean data.* 

| Location   | Deposition  | CR    | $F_{f}$ | T <sub>1/2, eff</sub> | $F_{mob}$ |
|------------|-------------|-------|---------|-----------------------|-----------|
|            | $(kBq/m^2)$ |       | (d/kg)  | (y)                   |           |
| Ribe       | 4.5         | 0.026 | 1.1     | 10                    | 0.035     |
| Tjøtta     | 43          | 0.97  | 0.74    | 18                    | 0.23      |
| Faroes     | 6.0         | 0.21  | 0.26    | 30                    | 0.027     |
| Blomhöjden | 17          | 1.2   | 0.86    | 23                    | 0.17      |

The values of the concentration ratios vary considerably (about a factor 50) across the sites. This variability is mainly due to the different soil characteristics between the sites. Mineral soils are known to retain radiocesium efficiently from plant uptake compared with organic soils, and the Tjøtta, Faroese and Blomhöjden sites are characterised by peaty soils in contrast to the Ribe site. However, if the radiocesium content in the grass is related to the mobile fraction in the soil, the concentration ratios become 0.7 for Ribe, 4.1 for Tjøtta, 7.9 for the Faroes and 7.1 for Blomhöjden. The total variation across the sites thus reduces to a factor of 10, still with the Ribe site at the low end, and with a significantly smaller variation across the sites with peaty soils. The lower transfer of radiocesium from soil to Faroese grass may thus be explained by a lower biological availability ( $F_{mob} \approx$ 0.03) than at the two other sites with peaty soil ( $F_{mob} \approx 0.2$  for Tjøtta and Blomhöjden).

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Figure 6.10 Comparison of model results and observations.

The lower transfer of radiocesium from grass to lamb in the Faroes compared to the other Nordic sites has been noted during the project, but no obvious explanation for the apparent difference has been found. However, reported values of the meat transfer coefficient from the literature for uptake of radiocesium in lamb demonstrate significant variability. Pre-Chernobyl values are reported in the range 0.08 - 0.5 d/kg (Ng et al., 1982), but post-Chernobyl field studies have found values at 0.8 d/kg (Howard et al., 1987) and at 1.6 d/kg (Beresford et al., 1989).

The estimates of doses to individuals from consumption of lamb are based on timeintegrated concentrations of radiocesium in lamb from 1990 to 2040. These values are used with the consumption data from Table 6.12 and a dose-per-unit factor of 1.3E-08 Sv/Bq (IAEA, 1996). The resulting dose estimates are shown in Table 6.14.

| лп | io from the study          | siles.            |                     |
|----|----------------------------|-------------------|---------------------|
|    | Location Dose from average |                   | Dose from high-rate |
|    |                            | consumption (µSv) | consumption (µSv)   |
|    | Ribe                       | 0.2               | 4                   |
|    | Tjøtta                     | 1300              | 3200                |
|    | Faroes                     | 110               | 140                 |
|    | Blomhöjden                 | 70                | 1000                |

Table 6.14 Estimated 50-y doses from  $^{137}$ Cs to individuals from consumption of lamb from the study sites.

The analysis of the data with the model has made it possible to identify similarities and differences between transfer processes across the sites. The effective ecological half lives for <sup>137</sup>Cs in the soil were found to vary from 10 y at the Danish site to 30 y at the Faroese site.

For the sites with low levels of  $^{137}$ Cs and consequently of Chernobyl fallout (Ribe and the Faroes, below 10 kBq/m<sup>2</sup>) low mobile fractions of radiocesium in the soil (about 3%) available for root uptake in the grass were found, while the other sites with higher levels of Chernobyl fallout (Tjøtta and Blomhöjden, some tens of kBq/m<sup>2</sup>) showed higher mobile fractions (about 20%).

The uptake of <sup>137</sup>Cs in grass was found to be more consistent at sites with peaty soils when the levels in the grass were related to the mobile fractions in the soil rather than to the total activities. The root uptake of radiocesium in grass from Ribe with mineral soil is a factor 10 lower than that from the site with peaty soils.

The transfer of <sup>137</sup>Cs from grass to lamb is found to be similar at the Danish, Norwegian and Swedish sites with a meat transfer coefficient,  $F_f$ , of about 0.8 d/kg. The transfer to lamb at the Faroese site is lower by about a factor 3. No clear explanation of this difference has been found. However, the  $F_f$  values compare well with corresponding data from the literature.

Doses have been calculated to individuals from the consumption of lamb from the four locations. Two categories of individuals were considered: average consumers and high-rate consumers, and doses were calculated for a 50-y period 1990-2040. The doses from average consumption of lamb range from 0.2  $\mu$ Sv in Ribe to 1 mSv in Tjøtta, and doses from high-rate consumption range from 4  $\mu$ Sv in Ribe to 3 mSv in Tjøtta.

This work has illustrated that reliable predictions of radiological consequences require long time series of field data, particularly when environmental levels of radioactivity are characterised by long ecological half lives.

## 6.9 Recommendations and future work

The study has shown that soil characterisitcs are the most important factors for transfer of <sup>137</sup>Cs to sheep. Climatic conditions, biomass production and soil ingestion by animals (indirectly grazing intensity) are of minor importance, and cannot explain the large differences we observe between the different study sites.

Studies of stable cesium and strontium gives an indication on wheter the fallout has reached equilibrium in soil, but it is expected that the radionuclides will be somewhat more mobile than the stable analogue.

The ecolgocial half-lives increases as time since fallout increases, indicating that a first-order exponential fit is not the best model. The Nordic lamb-model has been modified according to the collected data, and gives a good prediction of <sup>137</sup>Cs in lamb's meet at the different sites.

The long time series has been very important for this study, and should to some extent be continued. Also, it is important to investigate country wide differences, to combine this with soil characteristics and identify sensitive areas to radioactive fallout.

# 7. The forest project: Transfer of radiocesium via mushroom to roe deer and man

In the forest ecosystems fungi have a very important role both in the transfer of <sup>137</sup>Cs from soil to fruit bodies of fungi as well as in the soil-plant transfer and for both pathways further to animal and man. The radiocesium concentration in fungal fruit bodies is often more than 50 times higher than in plants growing in the same location, and the levels of radiocesium in fungi has tended to be stable or even increasing.

Questionnaires have been implemented in Sweden, Norway, Denmark and Finland, in Sweden, Norway and Denmark with focus on mushrooms, and a more thorough investigation in Finland on natural products for consumption. Unfortunately, non of the years during the project period were good mushroom years, and some of the results may not be representative for a good year.

Mushrooms also play an important role in the uptake of radiocesium by animals living in the forest. As a representative of these animals, roe deer was chosen, and samples of roe deer meat, rumen and grazing plant was collected during 1995 and 1996.



Figure 7.1 Shortly after an accident leading to deposition of radionuclides, agricultural products are most exposed (left part of the picture). Within the agricultural system there is a regime of practices leading to less uptake of radionuclides in food products, and as time since the accident increases, most of the intake of radiocesium comes via food produced in natural environments (right part of the picture).

# 7.1 Ecological half-lives in fungi, selected species

The evaluation of the half-lives in selected species of fungi was performed at selected sites. From 1990 to 1996 all fruit bodies of fungi growing at three sites at

the Swedish research area have been collected. Results show that the concentrations are not decreasing during the study period, even when the same mycelium is sampled.



Figure 7.2 The human consumption of mushrooms has been studied, as well as the intake of mushrooms by roe deer.

# 7.2 Concentrations of <sup>137</sup>Cs in roe deer – consumption of fungi

The <sup>137</sup>Cs concentration in roe deer show a very marked seasonal variation with peak concentrations during the mushroom season in August to October. During this period, often 20 to 30% of the roe deer rumen content is found to be mushrooms, so it is quite clear that roe deer consume large quantities of mushrooms.

In 1992 and 1993, the mean aggregated transfer factor, T<sub>ag</sub> for roe deer during August to October was  $0.093 \text{ m}^2/\text{kg}$  and 0.027 during the rest of the year. The annual harvest of roe deer in Sweden has been about 300,000 animals and assuming a mean meat weight of 10 kg per roe deer the annual meat production is 3 million kg. About one third of the harvested roe deer corresponding to one million kg meat is harvested during August to October. Assuming that our research area is representative for all roe deer ecosystems in Sweden, the mean roe deer in Sweden has a <sup>137</sup>Cs level of 930 Bq/kg in August to October and during the other part of the year 270. Of this 930 Bq/ kg, 660 is due to <sup>137</sup>Cs coming from mushrooms and the rest - 270 Bq/kg - coming from intake of "normal" fodder. The total potential transfer of <sup>137</sup>Cs to man by roe deer meat is 930 MBg during August to October and 540 MBg during the rest of the year, totally 1.470 MBg. This will mean that mushrooms were the vectors for 45 % of the total roe deer based transfer. Using the ICRP dose conversion factor of  $1.3 \cdot 10^{-8}$ , the corresponding potential dose commitment is 19 manSv of which 9 manSv is due to mushroom transfer.

One of the aims for the research was to study which species of fungi could be found in the rumen of roe deer. In September 1996 there were practically no fruit bodies of mushrooms in our sampling area. The mushroom occurrence in 1996 was, in general, extremely low and therefore we realised that it was not possible to obtain relevant information this year.

Ytterøya is an island located in the Trondheim Fjord. The total area is 28 km<sup>2</sup>, whereof 50% agricultural land, 25% productive pine forest and the rest less productive forest and some mire. Most probably the most populated roe deer areas in Norway are found on Ytterøya, with an annual hunt of 300 deer. Roe deer are found everywhere, with a change between cultural landscapes with good autumn/winter and early spring grazing areas.

Biological samples have been collected both in 1995 and 1996: 45 samples of muscle, 25 rumen or faeces samples and 30 samples of main grazing plant species. At 4 locations, gamma spectrometry has been performed *in situ*. Samples from animals have been collected in the period September - December, and plant species and in situ measurements are made in September 1996. The 4 locations were chosen based on fresh roe deer tracks. Mushrooms could not be found during the fieldwork, and both 1995 and 1996 were reported to be poor mushroom years by the local population.

Based on calibration, the present activity of  $^{137}$ Cs is calculated to 8 kBq/m<sup>2</sup> in uncultivated areas and 2 kBq/m<sup>2</sup> in grassland.

The meat samples from 1995 show an average concentration of  $^{137}$ Cs of 200 Bq/kg (number of samples, n=79). There is a large variation within the group ranging

from 0 to 1100 Bq/kg. Values related to time of hunting show apparently no trend connected to time of year. The variation most probably reflects the different grazing patterns. The ratio between meat and faeces samples is about 1:1. The rumen samples show lower values.

## 7.3 The use of natural products for consumption

We have performed simplified dietary studies in Denmark, Sweden, Finland and Norway. The goal of the study was to collect consumption data relevant for radiation dose assessments. Normally the food consumption often ignores the differences in radionuclide contents of wild food types. Also regional differences in consumption patterns are of importance for dose assessments, when the deposition densities of fallout radionuclides differ by regions. The direct pathway to man is that people pick mushrooms and since the <sup>137</sup>Cs activity concentrations in many species is very high quite large activities can be transferred this way.

Previous NKS-work (RAD-3) investigated the annual consumption of wild products, expressed in kg per year per capita.

Table 7.1 Previous estimates of annual consumption of some wild products, expressed in kg per year per capita. There are no figures for Iceland or the Faroe Islands.

| Produce  | DEN  | FIN | NOR  | SWE  |
|----------|------|-----|------|------|
| Mushroom | 0.05 | 1.4 | 0.24 | 2.8  |
| Berries  | 0    | 3.3 | 2.6  | 4.5  |
| Game     | 0.46 | 0.2 | 0.23 | 0.05 |

Table 7.2 Revised estimates of annual consumption of some wild products, expressed in kg per year per capita. There are no figures for Iceland or the Faroe Islands. The \* indicates no new estimate.

| Produce  | DEN | FIN | NOR | SWE |
|----------|-----|-----|-----|-----|
| Mushroom | 0.4 | 1.5 | 0.9 | 2.1 |
| Berries  | *   | 8.5 | 3.9 | *   |
| Game     | *   | 1.7 | *   | *   |

#### 7.3.1 Denmark

The main conclusions are that the Danes use their natural and semi-natural ecosystems to collect or shoot food products in an average amount of approx. 2 kg/year. Berries and game animals are the most important categories, whereas mushrooms and fresh water fish are less important. Besides fresh water fish, marine fish not in trade make up a part of the use of natural ecosystems that was not treated in the survey.

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A total of 30% did not use consumable products from nature at all; some of the rest buy products of Danish origin at restaurants or in shops. There was, however, a great individual variation, primarily determined by geography, education and employment. The total average intake of natural products was about 1% of the total consumption of food in Denmark. Hence it could be concluded that the value of natural and semi-natural ecosystems as a food source was of minor importance in Denmark.

#### 7.3.2 Sweden

In 1995, a questionnaire was sent to 1000 families in the municipality of Avesta in central parts of Sweden asking about their mushroom picking habits. Based on the answer of the first questionnaire a second was sent to those families who were picking mushrooms at least two times a year. They were asked to answer in more detail how much of various species of mushrooms they were picking either 1995 (a poor mushroom year) and also asked to try to estimate how much they were picking a normal mushroom year. To the later group a similar questionnaire was also sent in 1996.

59 % of the families who answered picked mushrooms more than two times per year and to them we sent a second questionnaire. The three most frequent species in the mushroom basket were *Cantharellus tubaeformis* (0.35 kg fresh weight (f.w.) per capita during 1995 and 0.84 kg a normal year), *Cantharellus cibarius* (0.17 kg in 1995 and 0.54 kg in a normal year) and *Boletus edulis* (0.04 kg in 1995 and 0.25 kg in a normal year). Additionally 0.19 kg of other species was picked in 1995 and 0.49 kg in a normal year.

Previously we had determined the aggregated transfer factor,  $TF_g$  for *C*. *tubaeformis* to be 0.94 m<sup>2</sup>/kg d.w, 0.39 for *C*. *cibarius* and 0.28 for *B*. *edulis*. The  $TF_g$  for the other species was usually higher and we used the value 1.0. From this values and the mean ground deposition in Sweden - 10000 Bq/m<sup>2</sup> - an estimation of the mean Swedish <sup>137</sup>Cs levels in *C*. *tubaeformis*, *C*. *cibarius*, *B*. *edulis* and the other mushrooms in Sweden could be done. In this estimation we assume that our research area is representative for all Sweden, which of course could be questioned. The estimated mean levels were for *C*. *tubaeformis* 752 Bq/kg f.w, for *C*. *cibarius* 312 and for *Boletus edulis* 224. For the other species the mean level was 800 Bq/kg.

Combining these values with the consumption data the estimated transfer of  $^{137}$ Cs to each person in Sweden was for *C. tubaeformis* 263 Bq in 1995 and 632 a normal year, for *C. cibarius* 53 Bq in 1995 and 168 a normal year, for *Boletus edulis* 9 Bq in 1995 and 56 Bq a normal year. The other species contribute with 152 Bq in 1995 and 392 a normal year. All together the estimated potential transfer to the mean Swede was 477 Bq in 1995 and 1248 a normal year corresponding to 6.0  $\mu$ Sv

in 1995 and 16  $\mu$ Sv a normal year. The corresponding potential collective dose in Sweden was 48 manSv in 1995 and 128 manSv in a normal year. These values correspond to the total activity in the mushroom basket, which is higher than the real intake due to reduction of <sup>137</sup>Cs levels during the food processing.

Table 7.3 The consumption (kg fresh weight per year per average Swede) and transfer factors ( $TF_g$ ) of the three most important species of fungi and the estimated potential transfer of <sup>137</sup>Cs (Bq/y) to average Swede.

| Species        | Consumption       |             | TFg            | Trans           | sfer to man |
|----------------|-------------------|-------------|----------------|-----------------|-------------|
|                | (kg fresh weight) |             | $(m^2/kg dry)$ | (Bq per person) |             |
|                |                   |             | weight)        |                 |             |
|                | 1995              | normal year |                | 1995            | normal year |
| C. tubaeformis | 0.35              | 0.84        | 0.94           | 263             | 632         |
| C. cibarius    | 0.17              | 0.54        | 0.39           | 53              | 168         |
| Boletus edulis | 0.04              | 0.25        | 0.28           | 9               | 56          |
| Other species  | 0.19              | 0.49        | 1.0            | 152             | 392         |
| Total          | 0.75              | 2.12        |                | 477             | 1248        |

#### 7.3.3 Finland

A mail survey was sent to 1500 randomly selected households. The country was divided into four regions: metropolitan region, western, eastern and northern Finland. Questions referred only to food, which was consumed at home.

The questionnaire was focused on the consumption of game, berries, reindeer meat, freshwater fish, wild mushrooms and some other wild food products of vegetative origin. The response rate of 59 % was reached after two reminders. Filled out questionnaires were returned from 884 households and the number of persons included in the responding households were 2360.

In our sample the percentage of hunters exceeded that in the whole population, which had to be taken into account. Hunters seem to have been more eager to return the questionnaire than other subgroups of population. The data was therefore analysed separately for hunters and non-hunters and thereafter the figures concerning the whole country were corrected by weighing.

For hunters the groups of berries, freshwater fish and game were all equally important with a mean consumption rate of 13 kg per year for a member of the household.

| food group                               | consumption per person (kg per year) |
|--|--------------------------------------|
| berries                                  | 8.3                                  |
| fresh water fish                         | 7.9                                  |
| mushroom                                 | 1.5                                  |
| game                                     | 1.2                                  |
| other wild products of vegetative origin | 0.4                                  |
| rein deer meat                           | 0.5                                  |

Table 7.4 Average consumption of wild products in Finland, 1995 survey.

Regional differences were quite obvious. Biggest consumption of mushrooms was in eastern Finland, with 2.2 kg per year and more than half of the mushrooms consumed were of *Lactarius* type. In this sub-region the mean consumption of wild berries was greatest, 13 kg per year, to which mountain cranberries contributed most. Freshwater fish was used most in eastern Finland in the large lake district. The consumption of game meat differed already between metropolitan regions 0.32 kg per year to northern Finland's 1.8 kg per year. The most important species was moose. The wild products of vegetative origin consist of sap, nettle, herbs and lichen. The consumption of sap was surprisingly high; 0.24 kg per year in the whole country, while people in eastern Finland consumed 0.44 kg per year. The use of reindeer meat was as was expected: biggest amounts in northern Finland and least in western and eastern Finland. The consumption pattern followed the availability of the product.

In the metropolitan region it is more common to buy wild food products than in the other regions. About half of the households used berries and fish outside the season, while berries were used in most of the households (91%) throughout the year. A good harvest year would increase the consumption of mushrooms by 60% and of berries 20% compared with the year 1995, which was not a good mushroom year.

Only 3.3% of population did not use any wild food products at home. In the group of non-hunters the most consuming households used a total of 72 kg per year of all wild products. In the group of hunters the same figure was 130 kg per year per user.

Consumption of mushrooms and berries was greater than findings of the previous studies. This was probably not a non-response error, as shown by the preliminary results of the telephone survey of the group of non-responders.

The study revealed regional differences in consumption rates. It is also important to consider the different consumption patterns in rural and densely populated areas and in sub-regions of the country.

#### 7.3.4 Norway

In Norway to surveys have been performed, one at Ytterøya in middle parts of Norway, and one in the Oslo area.

In the Oslo area 471 persons were asked to fill in a questionnaire, and 50% of these responded. The results show that 32-45 % pick mushroom, one to three times per year. The tendency is that people living in Oslo pick more often mushrooms compared to rural areas. Among those who pick mushroom, the annual consumption is 1.9 - 4.3 kg (fresh weight), depending on the occurrence of mushroom.

In the whole group as an average, annual consumption of mushroom is 0.9 kg. The most common species are: *Boletus edulis, Cantharellus cibarius, Albatrellus ovinus, Leccinum* species and *Suillus luteus*.

The group was also asked about their consumption of wild berries. Mushroom pickers tended to pick more berries than those that did not pick mushroom. In total the consumption of berries seems to be higher than earlier estimates.

| Group            | blueberries | cranberries | cloudberries | total |
|------------------|-------------|-------------|--------------|-------|
| Mushroom pickers | 1.8         | 1.3         | 1.3          | 5.9   |
| Non pickers      | 0.9         | 0.5         | 0.6          | 2.2   |
| Whole group      | 1.3         | 0.8         | 0.9          | 3.9   |

Table 7.5 The annual consumption of wild berries expressed as kg per year per capita. The total includes also wild raspberry.

At Ytterøya 173 families (close to 90% of the total population) responded, and of these 28 families (16%) pick mushroom. Especially two families had a large consumption of mushroom, and among those who picked mushroom 3 times per year or more, the average was  $10.4 \pm 12.8$  kg (fresh weight) per year. Excluding the two families, the average was  $4.4 \pm 3.6$  kg. Looking at the whole group, the annual consumption is between 0.7 - 1.7 kg per year.

## 7.4 Recommendations and future work

The forest project has shown that ecological half-lives in mushroom are long, and that this influences both <sup>137</sup>Cs concentrations in animal and man.

Unfortunately, there were no good mushroom years within the study period, it was therefore difficult to quantify the effect. Data on the use of natural products for consumption has been collected, and we now have better estimates on the intake by man. There is still great need for consumption data, for better estimates of doses to man, as few of the food products not commercially exchanged are covered in national statistics.

# 8. Limnic systems: Ecological half-lives in Nordic lakes

Main aims for the lake project have been to quantify the mobility of radionuclides in soils and sediments, and the biological transfer to fish in typical Nordic lakes. Much of the project has been based on a compilation of data and models covering large scales of time -10 years - and space -1 million km<sup>2</sup>.

The recovery of Nordic lake ecosystems from contamination by <sup>137</sup>Cs originating from the Chernobyl accident is gradually slowing down, at the same time as lakes vary widely in recovery rates and their sensibility to radioactive fallout. Accordingly, «ecological half-lives» are gradually increasing and cannot be treated as constants, over neither time nor space.

We have tried to quantify such «half-lives» in different types of landscapes in the Nordic countries, and assess the relative importance of various mechanisms distributing radioactivity in the system. In lakes, the initial phase of contamination and equilibration after fallout of <sup>137</sup>Cs lasts up to five years and appears to be largely ruled by biological processes. After that, <sup>137</sup>Cs activities in fish approach a «steady» state, with a slow decline that seems to be controlled by continuous secondary inputs of <sup>137</sup>Cs into the lakes and their food webs. The most important sources are the loss of <sup>137</sup>Cs from land to water and the recycling of <sup>137</sup>Cs from sediments.

<sup>90</sup>Sr is less frequently studied, but more mobile than <sup>137</sup>Cs, and has to some extent been included in our project. Due to the scarcity of data, comprehensive models have not been developed for <sup>90</sup>Sr.



Figure 8.1 Different factors studied within the limnic project; run off from catchment area, important limnic parameters, resuspension and relocation of sediments and the availability of radiocesium for biological uptake.

## 8.1 Geographical and temporal variation in transfer of <sup>137</sup>Cs

#### 8.1.1 Key parameters

The main objective of this part of the project was to compile data from the Nordic countries in order to describe the geographical variation in the transfer of <sup>137</sup>Cs from soils to water and freshwater fish and to present the results in Nordic maps. The resulting maps are based on field data and models that are derived from the other sub-programmes within EKO-2.

The first modelled maps that describe the geographical variation in the transfer of radiocesium from fallout to water and fish was based on Swedish data. Besides data on fallout and limnological characteristics these maps were derived from data on soil type and vegetation. In Sweden, an already existing database on <sup>137</sup>Cs concentration in fish and water was used for calibration of the model. Data on fallout and other key parameters was compiled from Finland and Norway and the model was then applied on these data but with some generalisations compared to Sweden.

When producing the Nordic maps, a particular problem has been the scarcity of available data on fish in Norway, where most freshwater systems are comparatively low in ionic strength and poor in nutrients and organic matter. In the latter respect, most Norwegian lakes differ from the forest lakes dominating in Sweden and Finland where wetlands and humic waters are common. To overcome this problem, a special effort was made to compile data from Norwegian lakes.

The transport of <sup>137</sup>Cs from the catchments to the lakes and the internal loading from the sediments, these factors are considered as crucial for the long-term temporal development of <sup>137</sup>Cs in water and fish. The loss of radiocesium during the first 10 years after Chernobyl fallout was quantified using a model that describes the loss from 3 different soil types (wet mire, dry mire and mineral soil) with strongly differing loss. The basics of this approach were given in the technical report EKO-2.3. The loss model gives a gradually changing map picture with smaller differences between different regions as the compartments with the highest loss rates (wet mires) are being emptied. However, still more than 10 years after Chernobyl the highest losses per area unit could be expected in areas with a high frequency of peat soils.

The output of this model combined to the ground deposition gives the input concentration to the lakes. The secondary input from lake sediments to water is not accounted for as a direct input source in the model, partly due to the problem that any variation in such input is due to factors describing lake morphometry and sediment characteristics. Due to the poor geographical coverage of such data, it is not possible to incorporate these factors in a model that should be applied on this geographical scale. However, from the relationship between soil types and lake

sediment characteristics and the structure of the model, it could be argued that also the quasi-equilibrium between sediment and water is at least partly described by the model. In this context it has to be stressed that even if the model has been evaluated on individual small catchments and lakes, it is attempted to describe the variation between larger geographical units.

#### 8.1.2 Lake susceptibility

Shortly after the Chernobyl accident (Håkanson et al., 1988) a map was presented that pointed out regions in Sweden with a high frequency of lakes that were especially sensitive to high levels of radiocesium in fish due to Chernobyl fallout. That early map was based on the empirical inverse relationship between ionic strength and <sup>137</sup>Cs activity in fish, and the geographical pattern was derived from the average ionic strength of lake waters. With increased knowledge about the processes behind that empirical relationship, it has been possible to separate different components and what geographical variation they may have.

The mean activity of <sup>137</sup>Cs in fish at a certain trophic level is then calculated from a bioaccumulation factor (= <sup>137</sup>Cs in fish (fresh weight): <sup>137</sup>Cs in water). This bioaccumulation factor is calculated from the concentration of potassium in lake water according to the relationship given by Rowan & Rasmussen (1994). Empirical data from more than 6000 Nordic lakes give a reasonably good geographical resolution. The model states that in the range of potassium concentrations in Nordic lake waters, the bioaccumulation factor to non-pescivore fish would be in the range 1000 to 7000 in most Nordic lakes and should then increase by about a factor of 2 for each trophic level.

The model has continuously been evaluated against empirical data from individual lakes and on a regional scale. There is generally a good agreement between empirical and predicted data in both geographical resolutions and the observed slower rate of decrease in fish during the mid-1990's is also indicated by the model. A problem for the future seems to be that the monitoring of <sup>137</sup>Cs in lake water has terminated in many freshwater systems during the last few years. This is unfortunate from a radioecological aspect since we will lack data for validation and testing of models.



Figure 8.2 Geographical variations in the bioaccumulation factor (<sup>137</sup>Cs in fish: <sup>137</sup>Cs in water) to non-pescivore fish as based on the potassium concentration in lake waters of Sweden, Norway and Finland.

The map presented in Figure 8.2 shows the regional differences in the transfer of <sup>137</sup>Cs from water to non-pescivore fish within Norway, Sweden and Finland. The key parameter is the concentration of potassium in lake water. Thus, the geographical differences in this bioaccumulation factor are governed by the geographical variation in potassium concentration of lake waters. From the map it could be observed that the northern parts of Finland and Sweden are generally having lakes with a high bioaccumulation factor, but the highest values are found in Norwegian lakes and in the northernmost parts of Sweden.

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The highest losses are found in areas with a high frequency of peat soils. Such conditions exist in the northern and interior parts of northern Finland and Sweden. At the time being and with this geographical resolution, the annual loss never comprises more than a few per mille of the initial fallout. When combining this with the map, we find that highly susceptible lakes towards <sup>137</sup>Cs fallout are most common in the northern parts of Finland and Sweden and within the Caledonian mountain range. These regions were only in part affected by the Chernobyl accident.

Figure 8.3 illustrates the resulting geographical variation of <sup>137</sup>Cs concentration in predatory fish during 1997 by accounting for the Chernobyl deposition of <sup>137</sup>Cs. As expected, the large-scale pattern is foremost governed by the fallout pattern. This is partly due to the minor fallout in much of the most susceptible areas in Finland. However, quite large deviations from what could be expected from the fallout pattern exist in the interior parts of northern and central Sweden. This is the Swedish region with the highest average concentration in freshwater fish despite an intermediate level of fallout.

In Finland the highest average concentration is found in the central parts of southern Finland where a high level of fallout is combined to medium high lake susceptibility. The lakes in the coastal regions and in the southern parts of Finland and Sweden are generally having lower concentrations in fish despite a high level of fallout. The Norwegian lakes are generally having even higher bioaccumulation factors than the lakes of Finland and Sweden. However, the expected quasi-equilibrium concentrations in lake water are lower and the dominating fish species (trout and char) are at a lower trophic level.

Thus, differences in lake susceptibility are affecting both the initial transfer from fallout to fish and the long-term transfer in terms of base-line level and ecological half-lives. Geographical variation in this respect is leading to a significant modification of the geographical distribution of concentration in freshwater fish as related to the distribution of <sup>137</sup>Cs deposition.



*Figure 8.3 Model predicted geographical variation in the mean activity of* <sup>137</sup>*Cs in Nordic Pike in Finland and Sweden and in Brown Trout in Norway during 1997.* 

## 8.1.3 Long time series of <sup>137</sup>Cs in fish

One model that have been developed within the frames of IAEA is the VAMPmodel, a model that predicts the <sup>137</sup>Cs-concentration in lake water and predatory fish using nine components meant to increase the predictive capacity of the model. The two components that we have put special emphasis on are the transport of <sup>137</sup>Cs from the catchments to the lakes and the internal loading from sediments, which both are crucial for the long-term temporal development in water and fish.

The model has continuously been evaluated against empirical data. A comparison between the predicted and observed transfer from Chernobyl fallout to non-pescivore fish in 41 Swedish lakes and lakes from 7 different regions in Finland during the 1990's gave a reasonably good agreement in both geographical resolutions. The model is also indicating the observed slower rate of decrease in fish during the mid-1990's.

The recovery of Nordic lake ecosystems from contamination by <sup>137</sup>Cs originating from the Chernobyl accident is gradually slowing down, at the same time as lakes vary widely in recovery rates and their sensibility to radioactive fallout. Accordingly, «ecological half-lives» are gradually increasing and cannot be treated as constants, over neither time nor space.

The present recovery rate is very slow and in several lakes the content of <sup>137</sup>Cs content has not decreased significantly during the last few years. This phenomenon can be identified for a wide range of fish and lake types. But nevertheless, both the fish type and lake characteristics are of importance for the timing of the shift from a faster exponential decrease to the quasi-equilibrium stage. Typically, carnivorous fish not only reach maximum contamination later than their prey, but the activity does also begin levelling out several years later. While small roach and perch show an almost constant contamination since 1992, pike have only done so the last few years. This difference between different trophic levels is clearly shown by Figure 8.4(A) where the temporal development of the average content of <sup>137</sup>Cs in pike in illustrated for some different types of lakes.



Figure 8.4 Average <sup>137</sup>Cs activity levels in pike, medium-sized perch and roach from Lake Päijänne, Finland, and in trout from Caledonian lakes (A). Long-term temporal variation of the <sup>137</sup>Cs activity levels in pike in Nordic lakes with different susceptibility towards radiocesium contamination. All values are decay corrected and normalised to a deposition of 30 000 Bg/m<sup>2</sup> (B).

The importance of the type of lake is illustrated with time series of <sup>137</sup>Cs in pike from different lakes. In lakes with long water retention time the establishment of the quasi-equilibrium is delayed and the contamination level in pike could be very high for a long time if the catchment soils also are having a high loss rate. However, the timing is also influenced by food web and fish population structures, which especially in large lakes can show high complexity. Shallow lakes, which rarely stratifies are more susceptible to resuspension of the lake sediments. This brings the water mass into frequent contact with contaminated sediment particles, and depending on the type of sediments this could lead to a slow decrease and a higher quasi-equilibrium level in such lakes. Lakes with a high input from the drainage area and short hydraulic residence time show a similar slow decrease during the present stage. The lowest maximum contamination and lowest quasi-equilibrium level are found in lakes where accumulation bottoms are dominating and the input from the catchment is low which in Nordic lakes generally is indicating a higher sediment retention and a relatively higher potassium concentration in lake water.

An important and interesting question for the future is, how accurate the observed geographical variation in lake sensitivity related to Chernobyl fallout would be in another, hypothetical fallout situation. It is not possible to definitely answer this

question, but the geographical variation is well described by fundamental factors that regulate the retention in soil, the turnover and retention in lakes and the transfer from water to fish. Thus, it also seems likely that the major geographical variation in the transfer of radionuclides to water and fish due to Chernobyl fallout will be of general validity, even if the amplitude of the transfer will depend on the timing and durability of the radioactive deposition.



Figure 8.5 Map showing the lakes where fieldwork has been performed to determine the influence of run-off from catchment areas.

It seems also clear that a large portion of the Nordic fresh water systems in an international perspective is susceptible to fallout of <sup>137</sup>Cs due to a combination of relatively low retention in soils, low sedimentation rates and low concentrations of potassium and other base cations in water. Accordingly, it seems of great interest to get the best possible description of the long-term development of radioactivity levels in water, sediment and aquatic biota in Nordic limnic systems.

# 8.2 The origin and dynamics of <sup>137</sup>Cs discharges

The discharge of <sup>137</sup>Cs and <sup>90</sup>Sr in catchments was studied in four Nordic regions (Figure 8.5). In Addition to the fieldwork, existing data on radionuclide discharge from the catchments was compiled. The aim of the study was

- to follow the long time development in stream water
- to investigate the influence of bogs on the development in streams
- and to investigate the significance of <sup>137</sup>Cs discharge on the development of activity concentration in lakes, by comparing the activity ratio between stream water in streams and lakes in typical Nordic environments.

Five different Nordic catchments were chosen (Table 8.1):

- two arable land catschments (Ängland and Hille south) discharging into the lake Hillesjön in central Sweden,
- one mainly subalpine (Lektorbekken) discharging into lake Øvre Heimdalsvann in central Norway,
- two in forests, the first (Puutteenophja) discharging into lake Päijänne in southern Finland and the second (Nyänget) discharging into the river Vindelälven in northern Sweden, located in the Svartberget experimental forest station in Vindeln, northern Sweden.

| Catchment         | Annual  | Catchment       | Fraction of bog           | Deposition,                          |
|-------------------|---------|-----------------|---------------------------|--------------------------------------|
|                   | run off | area            | in %                      | Chernobyl                            |
|                   | mm/year | km <sup>2</sup> |                           | kBq <sup>137</sup> Cs/m <sup>2</sup> |
| Lektorbekken (NO) |         | 23.6            | A: 0 ; B: ca 1.4; C: ca 1 | 130                                  |
| Nyänget (SE)      | 330     | 0.5             | A: 42; B:16               | 20                                   |
| Ängland (SE)      | 300     | 368             | A: 0; B: ca 2             | 100                                  |
| Hille syd (SE)    | 300     | 380             | 0                         | 100                                  |
| Päijänne (FI)     | Ca 200  | ca 0.86         | A: ca 20; B: ca 10 ; C: 4 | 80                                   |

Table 8.1. Some characteristics of the catchment areas studied.

Water was sampled from at least one stream and location in each area. In most of the catchments, these samples complete long time series that has been carried out since 1986 or earlier.

#### 8.2.1 Results from fieldwork

Among the pathways that might contribute to elevated concentrations of <sup>137</sup>Cs in lakes discharge via drainage water can be an important source. The main pathway for <sup>137</sup>Cs loss from land is via saturated surface runoff. Loss through groundwater has not been shown to give any significant contributions. An effective retention in the top organic and inorganic layers of mineral soils has been reported from numerous of investigations. Intercalation in clay lattice, effective incorporation

into the biosphere and chemical binding in abiotic organic compounds in humus has been suggested as explanations for the effective retention. Discharge areas, with shallow groundwater surfaces are often found in areas of thick organic soils. If they are in close vicinity to streams or lakes the saturated surface runoff from these areas is able to generate a transfer of radiocesium to streams. In the Nordic countries most lake systems include a fraction of bog or other water saturated discharge areas. Hence, areas with high contributions of these areas can give a significant secondary contribution to radiocesium in lakes. It is possible that the rapid initial decrease in fish that was observed a few years after the Chernobyl fallout event can level out in lakes surrounded by bogs as a result of <sup>137</sup>Cs discharge from land.

The observed remaining fraction of the deposited <sup>137</sup>Cs of Chernobyl origin (Bq/m<sup>2</sup>) in soil is in most cases significantly lower in bogs than in the surrounding mineral soils (Figure 8.6). This is, at least in one case, also true for the pre-Chernobyl fraction of <sup>137</sup>Cs (Nylén and Grip, 1997). The activity is typically 10-90% lower on the mire compared to surrounding areas.



Figure 8.6 The ratio between  $^{137}$ Cs in bogs and surrounding mineral soils ( $Bq/m^2 / Bq/m^2$ ) in Sweden some years after the Chernobyl fallout (Vindeln 3 to 6 years, subalpine and middle Sweden 10 years)

The processes behind these ratios might be a horizontal discharge of <sup>137</sup>Cs from bogs with high water levels to streams due to the higher frequency of saturated

surface runoff. In recharge areas raised bogs with relatively deep groundwater (southern part of the Nordic countries) a faster vertical migration of radiocesium into the bog may be possible.

A typical discharge area constitutes 1 to about 30% of the catchment with a spatial and temporal variation depending on topography, vegetation cover and climatic factors. It has been shown that the contribution to saturated surface discharge reaches its maximum of 20-30% during spring and its minimum 1% during summer and that the discharge of Cs follows this dynamic contribution of surface water (Nylén and Grip, 1997). The rest of the catchment only contributes to runoff with very low Cs-discharge, and hence the activity concentrations in the streams are proportional to the contribution of mires saturated by water in contact with the streams. Further the change in activity in the stream should to some extent reflect the change in the pool of <sup>137</sup>Cs in the discharge areas. Fractions of the mires that constantly undergoes saturated surface discharge and are in contact with draining streams should empty their pool faster than fractions that only are saturated during snow melt or heavy rainfall or loose its <sup>137</sup>Cs via infiltration during these events.

Catchments with high contributions of bogs with contact to streams generate higher activity concentrations in drainage water than catchments where mires are absent. This was shown in investigated catchments were the levels of <sup>137</sup>Cs after 10 years are more elevated in catchments with high fractions of bogs (Päijänne 4-20 %, Vindeln 16-42% bog, Figure 8.8). In Hille and Heimdalen the levels are about 100 times lower.



Figure 8.7 The relative activity in streams compared to the contribution of bogs.

The decrease in activity in stream water has not been uniform over time and at least two phases were observed in Northern Sweden. One rapid phase that lasted no longer than some weeks, generating an effective decrease in the remaining deposition in mires but not in mineral soils (Figure 8.7). This phase was driven by thaw of the 0.5 m snow cover generating saturated surface runoff on mires and probably also parts of the stream banks. The second phase was much slower in this catchment and still 11 years after the fallout event activity concentrations of the same levels as in the summer of 1986 can be found. In the two catchments discharging into the lake Hille the decrease is more rapid but seem to level out at a lower concentration after 5 years. The activity in the sub alpine stream Lektorbekken resembles the fate of the Hille streams and may together with them reflect washout from the stream banks.

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Figure 8.8 The time development of relative activity  $(^{137}Cs)$  in stream water from the studied catchments.

The influence of runoff on the concentration in stream water was mainly studied in the Nyänget catchment. The positive influence in Nyänget is not evident in the alpine and subalpine catchment. High water flow takes part mainly during the spring flood. The loss during these few days of high runoff constitutes the main fraction of the yearly loss in Vindeln, Heimdalen and probably also in the other catchments.



Figure 8.9 The influence of water flow on the relative activity in stream water.

The intensive investigation in the Nyänget catchment has shown that the initial loss of <sup>137</sup>Cs during the snowmelt of 1986 originated in the water saturated mire and that the amount was 44% of the original deposition. It has also been proved that the Nyänget mire has two fractions of different dynamics: the drier fraction with a loss of on average 2% per year and the wetter fraction that is constantly saturated by water and leaches 30% per year. This approach has been applied to several catchment areas, and fits well with the amount of dry and wet fractions of the mires.

#### 8.2.2 A simple model for the discharge of radiocesium from catchments

Different approaches has been used to model the loss of  $^{137}$ Cs from catchments to lakes including different parameters, such as environments, K<sub>d</sub>, through fall and retention from vegetation, average yearly runoff and seasonal changes in runoff. In the frame of this project the model is composed by five compartments (Figure 8.11). This is to our opinion the limit of reduction. The model does not take seasonal changes into consideration. The reason for constructing such a simple model is that it can easily be included in a GIS-model and generate examples of integrated yearly loss from different environments.



Figure 8.10 The reduced model generating yearly loss from catchment with different contributions of discharge areas.

| Compartment   | Symbol | Yearly loss Yearly loss |             | Ecological |
|---------------|--------|-------------------------|-------------|------------|
|               |        | t < 1 month             | t > 1 month | half life  |
| Mineral soil  | F1     | 0.0002                  | 0.0002      | >1000      |
| Discharge dry | F2     | 7.1                     | 0.02        | 30         |
| Discharge wet | F3     | 7.1                     | 0.3         | 2          |
| Stream        | F4     | 365                     | 365         | -          |

Table 8.2 Transfer coefficients for the GIS-model.

The model is composed of three terrestrial compartments. The transfer coefficients F1, F2 and F3 are taken from the observations in the Nyänget catchment, F3 also from the time development in Hille and Heimdalen (Figure 8.8). The recharge areas are discharging into the stream bank with a transfer of 0.0003 per year, which is the detection limit for the groundwater measurements in the Nyänget catchment. The drier fractions of mire are engaged in surface discharge mainly during spring with an annual loss of 0.02. The wet fraction of permanent surface saturated overland flow on mires as well as in organic fractions of the stream banks have a transfer to the stream of 0.3 of its inventory per year, and the stream it self a transfer to lakes of 365 per year. The compartments are in direct contact with the stream, which is a simplification of the real conditions.

#### 8.2.3 Conclusions

After an initial decrease in activity concentration of <sup>137</sup>Cs in streams lasting for some weeks after the Chernobyl accident, the rate in decrease will mainly be controlled by the fraction of mires in the catchments. Except for a trend at low flow rates no significant decrease in activity concentration has been observed since the summer of 1986 in catchments with high fractions of bogs. In catchments with only a minor contribution of bogs a significant decrease in activity was observed during the first years. This seems to level out after about 5 years.

The ratio of activity between stream water and lake water indicate that in areas rich in bogs the development in lake water will be given by loss from the bogs while in areas of mineral soil (Hille) resuspension of radiocesium from sediments will dominate.



Figure 8.11 The ratio between activity in streams and lakes.

According to measurements in stream water certain lake systems will generate activity in fish that will decrease with a rate that is close to 2% per year (physical decay).

# 8.3 Sediments as a secondary source of <sup>137</sup>Cs to fish

In most Nordic lakes, most of the radiocesium (>90%, >99%) is associated with sediments. It is thus likely that sediments are an important secondary source of <sup>137</sup>Cs to the lake environment. The question is, to what extent, and during how long time. Our aims were to determine the relative importance of sediments versus catchments as a secondary source of <sup>137</sup>Cs and to quantify long-term trends.

Fundamentally different processes may control the abiotic mobility of <sup>137</sup>Cs in sediments:

- Resuspension of particulate radiocesium, especially in shallow areas/shallow lakes
- Diffusion of dissolved radiocesium, especially in anoxic environments/deep lakes

Sediments can act as a secondary source of <sup>137</sup>Cs to organisms in several ways: Resuspension of sediments (physical remobilisation):

- equilibration with the dissolved phase in the water followed by direct uptake in biota
- ingestion of suspended particles by planktonic animals

Direct uptake from sediments (biological remobilisation):

- assimilation by rooted plants
- ingestion of contaminated sediments by benthic animals
- Diffusion from sediments into the water (chemical remobilisation)
- dissolution and diffusion into the water followed by direct uptake in biota

The mobility and accessibility of particulate <sup>137</sup>Cs in contaminated surface sediments is controlled in several ways:

- Dilution and burial by new sedimentation
- Vertical mixing of surface sediments by bioturbation or physical mixing through wind and wave action
- · Horizontal redistribution of surface sediments by resuspension and settling

In this project, various methods are applied in different lakes and streams:

- Horizontal and vertical (re-)distribution of <sup>137</sup>Cs in lake sediment
- Particulate <sup>137</sup>Cs in sediment traps (sedimenting material)
- Physical and chemical speciation of <sup>137</sup>Cs in different sediments and waters
- Diffusion studies in the field and in the laboratory

The mean inventory  $(Bq/m^2)$  varies widely among lakes. The mean inventory appears to be controlled by the water residence time, i.e. by the initial retention during the spring flushing in 1986.



Figure 8.12 Fate of radiocesium in a lake - some important mechanisms.

# 8.3.1 Relationship of <sup>137</sup>Cs inventories with water depth

Sediment studies show that in shallow lakes, the sediment inventories of <sup>137</sup>Cs are fairly evenly distributed across the lake. In deep lakes, however, the sediment inventory of <sup>137</sup>Cs is highly correlated with water depth, demonstrating a substantial net transport of sedimentary <sup>137</sup>Cs from littoral areas towards the deeper areas of the lake basin. Flux estimates suggest that much of the <sup>137</sup>Cs have been removed from the shallow areas within the first six years after contamination, while the mean inventory in deep areas has increased substantially.

In the profundal sediments, the amount of <sup>137</sup>Cs stored showed no obvious relationship with slope. In the sediments of the littoral zone, however, an increasing slope appears to result in a reduced amount of <sup>137</sup>Cs stored in the sediment: <sup>137</sup>Cs inventories on flat bottoms exceeded the inventories at the steep slopes about fivefold. Thus, flat bottoms seem to allow continuous sediment accumulation, even in the presence of resuspension.

## 8.3.2 Resuspension of <sup>137</sup>Cs

The focusing of <sup>137</sup>Cs can be explained by wind-driven resuspension and redeposition of surficial sediments. This is supported by the horizontal uniformity of <sup>137</sup>Cs activity concentrations in surficial sediment, irrespective of water depth, and by their similarity with the concentrations in settling material collected in sediment traps, even in epilimnetic traps deployed during thermal stratification.

Primarily in shallow near-shore sediments, mixing processes carry buried contaminated particles back to the surface, where they are frequently resuspended. Subsequently, they are distributed in the whole water mass and settle at random. This has resulted in a pronounced focusing of <sup>137</sup>Cs towards deeper lake areas.

The vertical and horizontal fluxes of <sup>137</sup>Cs caused by resuspension and focusing can be quantified from the undisturbed cores taken in the deepest part of some lakes. Such cores show a significant secondary deposition of <sup>137</sup>Cs during the years following the initial contamination. In at least some lakes, this secondary contamination of sediments is several times larger than the <sup>137</sup>Cs input from surrounding land and must be explained otherwise.

<sup>137</sup>Cs activities of settling material can be used to trace the source of the accumulating material. Mean activity concentrations of <sup>137</sup>Cs in surface sediments (0.5 cm) from different depth zones were all similar, suggesting an even deposition of the same material throughout the whole lake basin. Moreover, settling material collected in epilimnetic sediment traps was just as contaminated as the surficial sediments, even during the period of maximal thermal stratification. The contamination of settling material during the summer season, when inflow from the watershed is almost absent, strongly suggests that a large proportion of the settling material originate from resuspended sediments.

The fluxes of sediment-associated <sup>137</sup>Cs in different parts of the lake can be quantified from the losses of <sup>137</sup>Cs relative to a total gross deposition. The areal distribution of sedimentary <sup>137</sup>Cs in deep lakes is strongly correlated with water depth, and interpolated mean <sup>137</sup>Cs inventories can be derived for each depth. Horizontal and vertical annual fluxes of <sup>137</sup>Cs can be calculated by comparing the gross losses to the net losses or increases relative to the mean inventory or the
initial sedimentation of <sup>137</sup>Cs. Resuspension fluxes to the water column can be estimated as the gross losses from the sediment, assuming even distribution of deposition over the lake.

The procedure provides annual net rates of change at different depths, showing a transition from losses to accumulation with increasing water depth. Shallow areas have lost up to almost 90% of the initial <sup>137</sup>Cs input, while the inventory in the deepest area exceeds the lake mean by up to twofold. The water depth where the inventory of <sup>137</sup>Cs is equal to the average inventory in the lake (where gross losses are balanced by redeposition) often coincides with the mixing depth of the water column during summer stratification. The calculated annual gross losses can reach values up to 30% in the littoral zone, which indicates a substantial erosion and remobilisation of sedimentary matter. In the deep areas, substantial gross losses indicate that resuspension occurs even in deep waters, but the data also suggest that resuspension probability gradually decrease with depth. Accordingly, the gross loss rates of <sup>137</sup>Cs from sediments are strongly related to the intensity of water mixing. The strong relationship supports wind-driven resuspension to be the cause of the strong focusing of sedimentary <sup>137</sup>Cs.

### 8.3.3 A prognostic lake recovery model based on sediment resuspension

The horizontal and vertical distribution of sedimentary <sup>137</sup>Cs in lake sediments can be used to assess the long-term immobilisation of <sup>137</sup>Cs emitted by the nuclear accident in Chernobyl in 1986. Observed resuspension fluxes can be projected in order to make prognoses about the state of contamination in different parts of a lake ecosystem. Most biota lives in shallow areas within the warm water layer, which is mixed during the whole summer. In this zone, which is most exposed to wind-driven wave action, sediment resuspension results in a prolonged contamination of the water column. Accordingly, <sup>137</sup>Cs concentrations in fish decrease very slowly, especially in shallow lakes. At the same time, shallow areas in deep lakes are gradually cleaned by a net removal of contaminated surface sediments, which are deposited in biologically less active zones where total contamination is increasing.

A model was developed to assess the long-term immobilisation of <sup>137</sup>Cs and to make prognoses about the state of contamination in different parts of a lake ecosystem. The observed fluxes of sedimentary <sup>137</sup>Cs and their projection into the future indicate that the initial recovery of the sediments is limited to the shallowest area, where erosion results in an elimination half-time of about half a decade. Only later, decontamination progresses towards deeper zones, where contaminated particles are slowly buried.

The long-term removal of <sup>137</sup>Cs from the water column appears thus to be largely controlled by sediment resuspension. This process is likely to delay the recovery of many lakes from <sup>137</sup>Cs contamination. This suggests that the effects of the Chernobyl accident will remain a problem in Swedish lakes for several decades. On the other hand the long-term consequence of particle focusing is an eventual accumulation of <sup>137</sup>Cs in the least bioactive areas of the lake.

## 8.3.4 Diffusion of dissolved <sup>137</sup>Cs in and from lake sediments

Eight years after the Chernobyl fallout, the distribution of  $^{137}$ Cs observed in the sediment of a small lake suggests that most of the  $^{137}$ Cs entering the lake was rapidly deposited in the sediment. However, vertical profiles in undisturbed sediments show that  $^{137}$ Cs is not irreversibly bound to particles, but slowly remobilise during the following years. These profiles show that a large fraction of the  $^{137}$ Cs inventory is confined to a sediment layer of 0.5 cm. Below this peak layer, a tail of  $^{137}$ Cs is visible to a depth >0.1 m below the peak. Apparently, part of the radiocesium originating from the initial deposition has penetrated deeper into the sediment. Discontinuous gradients are evident on both sides around the peak, suggesting complex patterns of sorption and desorption.

The <sup>137</sup>Cs profile with both a distinct peak and long tails indicates a gradual desorption of <sup>137</sup>Cs from the original deposit and a vertical redistribution caused by diffusion in a matrix with different sorption properties. A three-phase model was developed that describes slowly desorbing binding sites in the initial deposit by an instantaneous equilibration changing slowly over time.

An explanation for the observed behaviour of rapid sorption in the water and subsequent slow desorption in the sediment can be found in the roughly hundred fold difference in K<sub>d</sub> between these environments. The solute and particulate partitioning of the initial inventory is constituted by the environmental condition prevailing in the lake water during the fall-out. Since the conditions in the sediment are different from the lake water, the equilibrium ratio of solute and particulate <sup>137</sup>Cs of the inventory may successively adjust to the surrounding partitioning ratio. Such a change in  $K_d$  can be caused by ammonia (NH<sub>4</sub><sup>+</sup>) originating from the decomposition of organic matter. Ammonia ions are directly competing with  $^{137}$ Cs for sorption sites, causing an inverse relationship between K<sub>d</sub> and NH<sub>4</sub><sup>+</sup> concentration, which usually increases with sediment depth. A hundredfold difference in K<sub>d</sub> between water and sediment would require a similar increase in  $NH_4^+$  concentration. This is not unlikely, since only 1‰ of the sediment nitrogen needs to be in the form of NH<sub>4</sub><sup>+</sup> to meet this condition. Equilibration of intact sediment cores from the same site indeed produced NH<sub>4</sub><sup>+</sup> concentrations in overlaying water that were thousand-fold higher than in the surface water of the lake. Accordingly, the vertical redistribution of <sup>137</sup>Cs in this sediment can be accounted for by ionic diffusion alone.

The slow desorption can be explained by an inert rather than an instantaneous equilibration with the dissolved phase. This would be in accordance with the <sup>137</sup>Cs sorption on clay minerals during short-term experiments, suggesting a slow binding with a half-saturation time of only one year, and a subsequent redistribution in minerals that was earlier concluded to be irreversible.

Our sediment profile rather suggests a continuous loss of <sup>137</sup>Cs from the inert to the dissolved phase, as indicated by long tails surrounding a sharp peak, and by fairly steep tail gradients being maintained after several years of equilibration in a natural environment. This chemical remobilisation may contribute to the slow recovery of lakes contaminated with <sup>137</sup>Cs, especially in lakes with anoxic bottom waters.



Figure 8.13 Amount of <sup>137</sup>Cs at different depths within a lake as a result of sediment resuspension and focusing alone. The figure shows values obtained from a simple model as a function of time after fallout. Note in particular the lowest curve representing the habitat of many fish or their diet, and the gradual slowing down of the relative decline ("half-life").

**8.3.5** Comparison of secondary inputs of <sup>137</sup>Cs: sediments versus soils In order to maintain observed concentrations of <sup>137</sup>Cs in lake waters in a typical shallow forest lake, soils must release 0.1-1% of their <sup>137</sup>Cs inventory every year. This is less than the observed loss from wetlands (>1%), but higher than the loss from upland soils (<0.03%). Consequently, at least in lakes with a catchment poor in wetlands, sediments are probably the main source of <sup>137</sup>Cs in lake waters and fish.

A preliminary evaluation of a study of stream sediments in a wetland-rich area indicates that these sediments contain ten times less <sup>137</sup>Cs than the surface sediments of a downstream lake, although water exchange is rather rapid in this lake. Moreover, the total inventory of <sup>137</sup>Cs in the sediment of this lake has not increased over time. This suggests that even in lakes with a significant supply of <sup>137</sup>Cs from land, the <sup>137</sup>Cs concentration on suspended and settling particles may be diluted rather than increased by stream input.

# 8.4 Bioavailability of particulate <sup>137</sup>Cs

Resuspended as well as deposited particles are taken up by planktonic or benthic invertebrates, which are likely to assimilate associated <sup>137</sup>Cs. Suspended particles are taken up by filtering zooplankton, whereby <sup>137</sup>Cs is likely to be transferred to fish. Moreover, benthic animals, which also constitute an important source of food to fish, are exposed to the <sup>137</sup>Cs at the sediment surface. Consequently, concentrations of <sup>137</sup>Cs in lacustrine food chains are likely to be controlled by the turnover and burial of contaminated sediment particles. Food chain transfer of both sedimentary and resuspended <sup>137</sup>Cs may thus substantially retard the natural decontamination of lacustrine fish communities.

**8.4.1 Influence of environmental factors on the bioconcentration of** <sup>137</sup>Cs Extensive laboratory studies were conducted to quantify the relative influence of various environmental factors on the bioconcentration factor of <sup>137</sup>Cs in freshwaters. For this purpose, <sup>137</sup>Cs uptake was studied in batch culture experiments using the unicellular planktonic green alga *Scenedesmus quadricauda* as a model organism. Of particular interest were the concentration of different cations (potassium, sodium, and calcium) and the metabolic activity (growth rate). Although little is known about the relative influence of different cations, in particular at the low concentrations common to Nordic freshwaters, it is a widespread assumption that the bioaccumulation of <sup>137</sup>Cs in freshwaters is largely regulated by the ionic strength in general and by the potassium concentration in particular. One aim was to test this assumption by studying both bioconcentration and uptake kinetics along combined gradients of major cations, thereby also changing total ionic strength. Even less is known about the influence of biotic factors on the bioaccumulation of <sup>137</sup>Cs, and about their importance relative to

chemical factors. Therefore, ionic gradients were combined with a simultaneous gradient in light intensity to manipulate algal growth rate without changing the chemical environment.

For each experimental combination, bioconcentration factors (BCF) were calculated as the concentration ratio between the organism and its abiotic environment. Here, intracellular concentrations of <sup>137</sup>Cs were related to the particulate algal carbon (POC) and to the concentration of dissolved <sup>137</sup>Cs, yielding a bioconcentration factor (BCF<sub>C</sub>) that is analogous to the widely used "solid/liquid" distribution coefficient related to organic carbon (K<sub>OC</sub>). This parameter is about 8 times higher than conventional BCF relating to fresh weight.

Variations in ionic composition and light intensity resulted in  $BCF_C$  ranging over four orders of magnitude (Figure 8.15). The potassium concentration (from 0 to almost 3000  $\mu$ M) showed to be by far the most important contributor to the variation and magnitude of BCF. However, it is evident that BCF is not always a simple linear inverse of potassium concentration, but that the relationship itself depends on potassium concentration. Furthermore, other variables showed a significant influence as well. Not only potassium but also sodium (40 versus almost 3000  $\mu$ M) showed a significant influence on BCF. On the other hand, a hundred fold variation of calcium concentration (5 and 500  $\mu$ M) showed no significant effect.

The BCF<sub>C</sub> in growing algae was found to be roughly an order of magnitude higher than in non-growing algae, which demonstrates the strong influence of metabolic activity on the bioconcentration of  $^{137}$ Cs, even at apparent equilibrium conditions.

Complex relationships among variables were observed. For example, the importance of potassium relative to sodium increases with metabolic activity (Figure 8.15). Furthermore, the magnitude of this influence is also dependent on the potassium concentration.

The experimental results suggest that both ionic concentrations and growth metabolism (metabolic rate) control bioconcentration factors of <sup>137</sup>Cs. At a given intensity of light, the major effector on biouptake of <sup>137</sup>Cs is the potassium concentration. The second most important contributor appears to be the metabolic activity, generating a 2-20 times lower BCF in darkness compared to light (9  $\mu$ E m<sup>-2</sup> s<sup>-1</sup>) along the potassium gradient. The third major abiotic factor is the sodium concentration. The bioconcentration factor is inversely related to the dissolved concentrations of these cations over a broad range of potassium concentrations. This can be explained by the constancy of potassium and sodium concentrations within organisms (homeostasis), irrespective of the concentrations in the external environment except at extreme conditions. Since both cations are nonisotopic carriers of <sup>137</sup>Cs, it appears that the concentration factor of <sup>137</sup>Cs is directly

proportional to the concentration factors of each of these nonisotopic analogues and can be quantified from a physiological model based on simple discrimination factors.

The experimentally derived bioconcentration factors are in good agreement with field data from freshwater ecosystems. It should be noted that the typical range of potassium concentrations found in Scandinavian freshwater ecosystems (10-100  $\mu$ M) is within the experimental range that generated the highest variability, with a pronounced inverse relationship of BCF with potassium concentration (Figure ð). This suggests that radioecological models based on this inverse relationship may be justified in Nordic freshwaters if available information is limited.

From an ecological viewpoint, it is often relevant to express the magnitude of biouptake as the fraction of biologically available <sup>137</sup>Cs that is present in the organic matrix. Our results suggest that in Nordic lake waters, this fraction is usually around 1-10%.



Figure 8.14 Influence of ionic concentrations and metabolic activity (light intensity) on the bioconcentration factor of <sup>137</sup>Cs a planktonic freshwater alga (Scenedesmus quadricauda). Bioconcentration factors ( $L kg^{-1}$ ) are related to algal organic carbon rather than fresh weight. Data from batch culture experiments (means and geometric SD, n=3 to 6) are compared with a physiological modelling approach based on ion regulation mechanisms.

### 8.4.2 Chemical mobility of <sup>137</sup>Cs in soils and sediment

A sequential extraction procedure was applied to study the chemical mobility of  $^{137}$ Cs in soils and sediments from two watersheds in central Sweden. Five extraction reagents were used sequentially, two of them representing mobile fractions (NH<sub>4</sub>Ac+NH<sub>2</sub>OH·HCl) and one (HNO<sub>3</sub>+Residual) representing strongly bound  $^{137}$ Cs. In most samples, the strongly bound fraction dominated and usually accounted for around 70% of total  $^{137}$ Cs, while the mobile fraction accounted for only about 20% of total  $^{137}$ Cs in many samples (Figure 8.16). However, the desorption experiment also showed a large difference in the chemical mobility of  $^{137}$ Cs between peaty and other samples. The mobile fraction of  $^{137}$ Cs was about 3 times larger in pure peat soils than in agricultural or forest soils, leaving only 5-35% in the strongly bound fraction. This is confirming evidence that the capacity of peat to bind  $^{137}$ Cs is low, and is in good agreement with the depleted inventories of  $^{137}$ Cs in the same mire as well as in peat areas studied elsewhere. It should also be noted, however, that more than 70 % of the initial  $^{137}$ Cs inventories is still left in the mire studied.

The differences in the amount of strongly bound caesium probably depend on the well-known ability of clay minerals to fix caesium, and on the abundance or rather scarcity of clay minerals. The extraction patterns indicate that most <sup>137</sup>Cs in highly organic peat soils is not bound to mineral particles, but rather to organic matter, possibly in association with iron oxyhydroxides. In contrast, podzolic forest soils showed a pattern similar to agricultural soils despite a dominance of organic matter. This suggests either that a minimal fraction of mineral particles is sufficient to dominate the binding of <sup>137</sup>Cs, or that reducing conditions common to saturated peat soils contribute to increase the mobility of <sup>137</sup>Cs.

In contrast to the humic lake Blacksåstjärn, sediments from another lake with agricultural influences (Loppesjön) unexpectedly showed a binding pattern similar to the one found in most soils, both in oxic and anoxic water strata. In contrast, aquatic systems downstream the mire studied (BL) showed systematic differences, with intermediate <sup>137</sup>Cs binding both in stream and lake sediments (Figure 8.16). This may indicate a downstream influence of peat on the mobility of <sup>137</sup>Cs, and a potential role of humic matter as a carrier of <sup>137</sup>Cs in systems where the abundance of clay minerals is low. However, the notable differences between sediments and soils as well as among sediments suggest that the organic content is not the only controlling factor. Interestingly, lake sediments differed more in the mobile fraction of <sup>137</sup>Cs than in the strongly bound fraction.



Figure 8.15 Mobile and strongly bound fractions of <sup>137</sup>Cs in surficial soil and sediment samples, based on sequential chemical extraction. The potential influence of humic matter is highlighted. The samples were collected at different locations in the catchments of lake Blacksåstjärn (BL) and Loppesjön (LO) in Hudiksvall, central Sweden. While both catchments are dominated by coniferous forest, Blacksåstjärn is a humic lake located downstream of the mire studied, in contrast to Loppesjön where agricultural soils constitute a significant influence. Organic matter content from left (soils) to right (sediments): 25-38%, 9-13%, 55-85%, 97-98%, 12-29%, 21-30%, 7-31% (usually loss on ignition, 2·orgC for lake sediments). Mobile <sup>137</sup>Cs is defined as the sum of the exchangeable (NH<sub>4</sub>Ac) and the easily reducible (NH<sub>2</sub>OH·HCl) fractions, and the strongly bound fraction refers to <sup>137</sup>Cs extracted in HNO<sub>3</sub> and in the residual.

### 8.5 Recommendations and future work

During this project period we have compiled available data, which were then used to calibrate a simple. radioecological model applicable to <sup>137</sup>Cs in Nordic lakes. The model is based on relationships between levels of radiocesium in fish, the occurrence mire in catchment areas, and the concentration of potassium in lake waters. The structure and parametrisation of the model were derived from extensive field observations including long time series and spatial distributions. The model was successfully transferred to a GIS platform to produce useful maps of regional susceptibility to fallout.

Studies of sediment show that redistribution of contaminated sediments within the lake is an important factor, and even if most radiocesium is strongly bound to the sediment, there is both an upwards and downwards diffusion of <sup>137</sup>Cs. In most cases, the sediment inventory of <sup>137</sup>Cs in the lake is lower than the inventory in the catchment area, and the inventory in the sediment has not increased over time which suggests an insignificant secondary net input. The concentration in lake waters over time may to a large extent be controlled by the extent (depth) of bioturbation determining the amount of "active" sediment available for sorption/desorption.

The relative importance of sediments versus soil runoff as sources of secondary <sup>137</sup>Cs input to lakes is not well resolved yet. The loss from sediments must be substantial in order to explain the <sup>137</sup>Cs concentrations observed in some lake waters. On the other hand, catchment input increases in importance with the relative extent of wetland areas, and may exceed the losses from sediments if wetland constitute more than a few percent of the catchment area. However, the critical fraction is extremely uncertain and may vary with hydrological and sediment characteristics of a lake. A better understanding would facilitate the planning of remedial measures in the case of a fallout.

The time series of <sup>137</sup>Cs in fish show that the decline of <sup>137</sup>Cs in fish has slowed down dramatically, indicating a steady source of <sup>137</sup>Cs available for biological uptake. At present, the «half-lives» of <sup>137</sup>Cs in fish are long and appear to increase with time. The same is the case for concentrations in surface waters, which implies that the bioavailability of <sup>137</sup>Cs today is largely controlled by processes controlling the supply of <sup>137</sup>Cs to the water column. This is in contrast to the initial years after the fallout when food web transfer of <sup>137</sup>Cs resulted in complex dynamics including negative half-lives.

Effort should also be put into continued monitoring of existing time series. It is suggested that activity time series should be described by other means than half-lives, which are based on the unrealistic assumption of first-order decline. It is evident that many "half-lives" are not constant numbers but increase over time and are often proportional to time, suggesting that power functions or even a simple inverse of time may be more appropriate than first-order exponential functions as a first estimate or modelling default to describe temporal changes. Valuable input on the long-term behaviour of <sup>137</sup>Cs in Nordic environments can be obtained from earlier time series since the bomb fallout.

Our study confirms that foodstuffs from natural environments increases in relative importance with regard to long-term doses of <sup>137</sup>Cs to man. Unfortunately, only limited data are available on <sup>90</sup>Sr in fresh water systems - more effort should be put into studies of <sup>90</sup>Sr, preferable along with a continuous comparison to <sup>137</sup>Cs.



Figure 8.16 Typical ecological "half-lives" of <sup>137</sup>Cs (years, decay-corrected) in Nordic watersheds and lake ecosystems during the initial years after the Chernobyl fallout in late April 1986 and in the near future: Runoff (left) from upland areas (upper) and wetland or discharge areas (lower), release from sediments (centre) through bioturbation and resuspension (upper) and diffusion (lower), and concentrations in water, small fish, and large predatory fish (right, from bottom to top). All numbers are derived from field data but should be regarded as rough estimates since they may vary by an order of magnitude among different ecosystems.

It is evident that many "half-lives" are not constant numbers but increase over time and are often proportional to time, suggesting that power functions or even a simple inverse of time may be more appropriate than first-order exponential functions as a first estimate or modelling default to describe temporal changes.

Table 8.3 Typical ecological "half-lives" of <sup>137</sup>Cs (years – decay-corrected) in Nordic watersheds and lake ecosystems during the initial years after the Chernobyl accident. Apparent half-lives: based on observed rates, assuming first order decay. Fast component: excluding the first month after fallout (initial phase); listed estimates relate to fallout around 1 May 1986 but may depend on the fallout season and year. Slow component: application and extrapolation of present rates.

| Period  |                      |                  | Jun-Jul1986           | 1987-89                | 1996-2016              |               |
|---------|----------------------|------------------|-----------------------|------------------------|------------------------|---------------|
| Time si | ince Chernobyl fall  | out              | 0.2 y                 | 2 у                    | 20 y                   |               |
| Phase   |                      |                  |                       | Early dynamic          | Late dynamic           | Quasi-steady  |
| Typica  | l rates of dominatin | g processes      |                       | Fast                   | Intermediate           | Slow          |
| Compa   | artment              | Parameter        | Process               | Typical T <sub>a</sub> | Typical T <sub>a</sub> | Typical Ta    |
|         |                      | (Inv. or Conc.)  | (intrinsic)           | (years)                | (years)                | (years)       |
|         |                      |                  |                       |                        |                        |               |
| LF      | Conifer trees        | Conc & Inventory | Litter & through-fall | ca 0.6                 | ca 2                   | ca 30         |
| PS      | Peat soils           | Inventory        | Runoff                | (<1?)                  | ca 3 ("wet")           | ca 30 ("dry") |
| OS      | Other soils          | Inventory        | Runoff                | >> 100?                | > 1000                 | >> 1000?      |
| SWP     | Stream water PS      | Concentration    | Runoff                | variable               | ca 1, variable         | > 100?        |
|         |                      |                  |                       | (0.0210?)              | (negative?)            |               |
| SWO     | Stream water OS      | Concentration    | Runoff                | < 0.2                  | ca 1                   | > 10          |
| SH      | Lake sediment        | Inventory hypol. | Desorption/Diffusion  | > 20                   | ca 200                 | ca 2000       |
| SE      | Lake sediment        | Inventory epil.  | Resuspension          | < 4                    | > 4                    | > 10          |
| SC      | Lake sediment        | Conc. Surface    | Sorption              | (negative?)            | = 2? = 5?              | > 10          |
| WC      | Lake water           | Concentration    | All above             | ca 0.3                 | ca 2                   | > 10          |
| F1      | Small fish           | Concentration    | Food chain transfer   | negative!              | ca 2                   | > 10          |
| F2      | Predatory fish       | Concentration    | Food chain transfer   | negative!              | ca 8                   | > 10          |

#### Sources, Support, Comments

#### Jun-Jul 1986

- PS T= 1 y: runoff conc. in peat-rich areas decline to half from 1986 to 1987, likely faster initially (Saxén 1994).
- OS slowest regional runoff rates in Finland <1 %/y in 1986 based on conc. (Saxén 1994) and runoff<0.5m/y and assuming the contribution from peat to be negligible.
- SWP see PS for minimum; around 11 y in stream at Sälgsjön (this report). T~
  0.1...10 y: seasonal effects or long-term trends? (Nylén & Grip 1997 and in this report)
  T~1.2. Trend in part sick Einsich protecter had (Sara(a 1004) with endpotential)

T<sup>-</sup>1-2: Trend in peat-rich Finnish watersheds (Saxén 1994), with unknown contribution from peat soils

- SWO Slowest runoff rates (Saxén 1994), with unknown contribution from peat soils
- SH maximum diffusive loss (Meili & Wörman 1997)
- SE loss from littoral sediments (cf. Konitzer & Meili 1995) and erosion model (Meili in this report and unpubl., cf. Weyhenmeyer et al. 1997), rapid removal (effect and rate of vertical sediment mixing) not considered
- SC increasing due to slow sorption kinetics? (e.g. Comans), constant? (removal from water due to burial due to vertical sediment mixing?), declining? (equilibrium with water?)
- WC Andersson & Meili 1994
- F1 Meili 1992
- F2 Andersson & Meili 1994, Saxén 1996

#### 1987-89

- PS annual loss from most water-saturated peat areas: T<sup>~</sup> 4 y (cf. Nylén in this report); decline of runoff conc.: T<sup>~</sup> 1 y in peat-rich areas including lakes (Saxén 1994), T<sup>~</sup> 3 y at Sälgsjön (Sundblad et al. 1991), T<sup>~</sup> 4 y at Svartberget (Nylén & Grip 1997).
- OS slowest regional runoff rates in Finland (Saxén 1994), Hillesjön and Øvre Heimdalsvann (in this report), all with unknown contribution from peaty soils
- SWP variable, negative: "doubling time" = 4 years? (this report) T~1-2: Trend in peat-rich Finnish watersheds (Saxén 1994), with unknown contribution from peat soils
- SWO T<sup>-1-2</sup>: Trend in peat-poor Finnish watersheds (Saxén 1994), with unknown contribution from peat soils
  - T>1000?: pasture and arable land in the Hille catchment (this report)
- SH cf. peak and rapid 10-fold decline of conc. in vertical profile (Meili & Wörman 1996)

- SE loss from littoral sediments (cf. Konitzer & Meili 1995) and erosion model (Meili in this report and unpubl., cf. Weyhenmeyer et al. 1997), rapid removal (effect and rate of vertical sediment mixing) not considered
- SC As Water?, slow sorption kinetics?,
- WC as Fish 1, decline of runoff concentrations in peat-rich areas (Saxén 1994), decline in invertebrates (Andersson & Broberg 1992), cf. vertical profile (Meili & Wörman 1996)
- F1 Andersson & Meili 1994, Saxén 1996
- F2 max conc., Andersson & Meili 1994, Saxén 1996

### 1996-2016

- LF the transfer of Cs-137 via litter fall reached a steady state of about 2% of the total inventory per year during 1991 to 1996 in a mature mixed coniferous forest (Nylén 1996 and unpublished).
- PS runoff rates from low-loss ("dry") wetlands (Andersson in this report)
- OS The ecological half-life is too long to be unfolded from the effective half-life. Upper limit of concentration in groundwater suggests a loss from recharge areas of <0.02%, implying a half-life >1000 years (Nylén & Grip 1997).
- SWP T<sup>-140</sup>? this slow component is not significant (this report)
- SWO T<sup>-6</sup>...16 y from pasture and arable land in the Hille catchment (this report)
- SH cf. outflow from lakes in peat-poor areas
- SE erosion model (Meili in this report and unpubl., cf. Weyhenmeyer et al. 1997), as sed. conc. (or slower due to burial)
- SC as Water
- WC as Fish 1
- F1 Meili et al. 1997 and in prep.
- F2 as Fish 1

# 9. Conclusions and recommendations

During this project period we have compiled available data from different seminatural ecosystems, focusing on areas where we observe long ecological "halflives". In the sheep project we also have made extensive studies, to describe the dominant factors resulting in higher than average transfer of radionuclides to animals.

Our studies of foodstuffs from the semi-natural environment show that the decline of <sup>137</sup>Cs has slowed down dramatically, indicating a steady source of <sup>137</sup>Cs available for biological uptake. At present, the «half-lives» of <sup>137</sup>Cs in these foodstuffs are long, and appear to increase with time. This is in contrast to generic models, which emphasises food from the agricultural system, which exhibits much shorter half-lives.

Effort should also be put into continued monitoring of existing time series. It is suggested that activity time series should be described by other means than half-lives, which are based on the unrealistic assumption of first-order decline. It is evident that many "half-lives" are not constant numbers but increase over time and are often proportional to time, suggesting that power functions or even a simple inverse of time may be more appropriate than first-order exponential functions as a first estimate or modelling default to describe temporal changes. Valuable input on the long-term behaviour of <sup>137</sup>Cs in Nordic environments can be obtained from earlier time series since the bomb fallout.

Our study confirms that foodstuffs from natural environments increases in relative importance with regard to long-term doses of <sup>137</sup>Cs to man. Unfortunately, only limited data are available on <sup>90</sup>Sr - more effort should be put into studies of <sup>90</sup>Sr, preferable along with a continuous comparison to <sup>137</sup>Cs.

#### The sheep-project

The study has shown that soil characterisitcs are the most important factors for transfer of <sup>137</sup>Cs to sheep. Climatic conditions, biomass production and soil ingestion by animals (indirectly grazing intensity) are of minor importance, and cannot explain the large differences we observe between the different study sites.

Studies of stable cesium and strontium gives an indication on wheter the fallout has reached equilibrium in soil, but it is expected that the radionuclides will be somewhat more mobile than the stable analogue.

The ecolgocial half-lives increases as time since fallout increases, indicating that a first-order exponential fit is not the best model. The Nordic lamb-model has been

modified according to the collected data, and gives a good prediction of <sup>137</sup>Cs in lamb's meet at the different sites.

The long time series has been very important for this study, and should to some extent be continued. Also, it is important to investigate country wide differences, to combine this with soil characteristics and identify sensitive areas to radioactive fallout.

#### The forest project

The forest project has shown that ecological half-lives in mushroom are long, and that this influences both <sup>137</sup>Cs concentrations in animal and man.

Unfortunately, there were no good mushroom years within the study period, it was therefore difficult to quantify the effect. Data on the use of natural products for consumption has been collected, and we now have better estimates on the intake by man. There is still great need for consumption data, for better estimates of doses to man, as few of the food products not commercially exchanged are covered in national statistics.

#### The limnic study

In the limnic study, collected data were used to calibrate a simple. radioecological model applicable to <sup>137</sup>Cs in Nordic lakes. The model is based on relationships between levels of radiocesium in fish, the occurrence mire in catchment areas, and the concentration of potassium in lake waters. The structure and parametrisation of the model were derived from extensive field observations including long time series and spatial distributions. The model was successfully transferred to a GIS platform to produce useful maps of regional susceptibility to fallout.

Studies of sediment show that redistribution of contaminated sediments within the lake is an important factor, and even if most radiocesium is strongly bound to the sediment, there is both an upwards and downwards diffusion of <sup>137</sup>Cs. In most cases, the sediment inventory of <sup>137</sup>Cs in the lake is lower than the inventory in the catchment area, and the inventory in the sediment has not increased over time, which suggests an insignificant secondary net input. The concentration in lake waters over time may to a large extent be controlled by the extent (depth) of bioturbation determining the amount of "active" sediment available for sorption/desorption.

The relative importance of sediments versus soil runoff as sources of secondary <sup>137</sup>Cs input to lakes is not well resolved yet. The loss from sediments must be substantial in order to explain the <sup>137</sup>Cs concentrations observed in some lake waters. On the other hand, catchment input increases in importance with the relative extent of wetland areas, and may exceed the losses from sediments if wetland constitute more than a few percent of the catchment area. However, the critical fraction is extremely uncertain and may vary with hydrological and sediment characteristics of a lake. A better understanding would facilitate the planning of remedial measures in the case of a fallout.

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#### Appendix II List of participants

# EKO 2.1 "The sheep project"

| Morten Strandberg     | DMU, Denmark                             |
|-----------------------|--|
| Sven P. Nielsen       | Risø, Denmark                            |
| Mette Øhlenschlæger   | SIS, Denmark                             |
| Hans Pauli Joensen    | Fróðskaparsetur Føroya, Faeroe Islands   |
| Trygvi Vestergaard    | Fróðskaparsetur Føroya, Faeroe Islands   |
| Elísabet Ólafsdóttir  | Geislavarnir Ríkisins, Iceland           |
| Sigurður Emil Pálsson | Geislavarnir Ríkisins, Iceland           |
| Jóhann Þórsson        | Rannsóknastofnun landbúnaðarins, Iceland |
| Ingar Amundsen        | NRPA, Norway                             |
| Knut Hove             | NLH, Norway                              |
| Deborah Oughton       | NLH, Norway                              |
| William Standring     | NLH, Norway                              |
| Inger Andersson       | SLU, Sweden                              |
| Hans Lönsjö           | SLU, Sweden                              |
| Klas Rosén            | SLU, Sweden                              |

# **EKO 2.2 "The forest project"** Morten Strandberg

| EKO 2.2 "The forest project" |               |
|------------------------------|---------------|
| Morten Strandberg            | DMU, Denmark  |
| Aino Rantavaara              | STUK, Finland |
| Eldar Gaare                  | NINA, Norway  |
| Karl Johan Johanson          | SLU, Sweden   |

# EKO 2.3 "Limnic ecosystems"

| Sub-project leader: Markus Meili | University of Uppsala, Sweden   |
|----------------------------------|---------------------------------|
| Timo Jaakkola                    | University of Helsinki, Finland |
| Ritva Saxén                      | STUK, Finland                   |
| Helge Bjørnstad                  | FFI, Norway                     |
| John Brittain                    | University of Oslo, Norway      |
| Torbjørn Forseth                 | NINA, Norway                    |
| Per Varskog                      | IFE, Norway                     |
| Tord Andersson                   | University of Umeå, Sweden      |
| Karin Aquilonius                 | Studsvik Ecosafe, Sweden        |
| Torbjörn Nylén                   | FOA, Sweden                     |
| Anders Broberg                   | University of Uppsala, Sweden   |
| Jonas Hagström                   | University of Uppsala, Sweden   |
| Kerstin Konitzer                 | University of Uppsala, Sweden   |
| Annica Oscarson                  | University of Uppsala, Sweden   |
| Marcus Sundbom                   | University of Uppsala, Sweden   |
|                                  |                                 |

# Appendix III Glossary and abbreviations

To reduce the risk of misinterpretation, the following list gives explanations of selected technical terms used:

- Aggregated transfer coefficient: The relationship between the ground deposition of e.g.<sup>137</sup>Cs and the content of this in a product, for instance milk:  $T_{ag}$  (or  $T_{agg}$ ) = Bq <sup>137</sup>Cs/kg milk / Bq<sup>137</sup>Cs/m<sup>2</sup> deposited.
- $\mathbf{B}_{\mathbf{f}}$ : Bioconcentration factor, se concentration factor.
- **bioavailability**: being available for biological uptake and turnover. Generally, only part of the total amount of an element in an environment is bioavailable.
- **Bq**: bequerel; the unit of activity for a radioactive element: 1/s, i.e. the number of decays per second.
- **concentration factor**: In aquatic ecology: CF = Bq/kg sample (dry or fresh weight) / Bq/kg water at assumed equilibrium. As no general convention exists, it is essential always to indicate dry or fresh weight basis. Because true equilibrium hardly ever occurs under natural conditions, the terms concentration radio (CR) or bioconcentration factor ( $B_f$ ) are in practice used synonymously.
- **critical group**: That group of the population assumed to receive the highest radiation doses from a given source.
- half-life ( $\mathbf{T}_{\frac{1}{2}}$ ): Time (e.g. in years) during which the original amount or concentration of an element is reduced to half by an assumed exponential reduction. The physical half-life ( $\mathbf{T}_{\frac{1}{2},p}$ ) is the characteristic time (e.g. in years) during which a radioactive element is reduced to half its original amount by physical decay ( $\mathbf{T}_{\frac{1}{2},p} = \ln 2/\lambda_p$ , where  $\lambda_p$  is the decay constant (per yr)). Provided the amount or concentration of the radionuclide in a component of the environment, e.g. a plant species, follows a single exponential reduction with time, the **effective half-life** ( $\mathbf{T}_{\frac{1}{2},eff}$ ) (synonyms: **observed half-life** ( $\mathbf{T}_{\frac{1}{2},obs}$ ), **effective ecological half-life** ( $\mathbf{T}_{\frac{1}{2},eco}$ ), **effective environmental half-life**), where  $\mathbf{T}_{\frac{1}{2},eff} = \ln 2/\lambda_{eff}$ , describes the decrease in the observed concentration of a radionuclide with time, including the effects of environmental and ecological processes as well as physical decay. The **biological half-life** ( $\mathbf{T}_{\frac{1}{2},b}$ ) in an organism is that part of the effective halflife attributed to biological excretion, i.e., it does not include the effect of physical decay. Likewise, the **ecological half-life** ( $\mathbf{T}_{\frac{1}{2},e}$ ) (synonym:

**environmental half-life**) is that part of the effective half-life attributed to ecological or environmental processes. In other words,  $T_{\frac{1}{2},b}$  and  $T_{\frac{1}{2},e}$ , are equivalent to the half-life of a stable element without any decrease due to physical decay. Biological and ecological half-lives may be calculated after making a correction for physical decay to one common time, or by applying the relations  $\lambda_{eff} = \lambda_p + \lambda_{b \text{ and }} \lambda_{eff} = \lambda p + \lambda_e$  or the equivalent relations  $(T_{\frac{1}{2},eff})^{-1} = (T_{\frac{1}{2},p})^{-1} + (T_{\frac{1}{2},b})$  and  $(T_{\frac{1}{2},eff})^{-1} = (T_{\frac{1}{2},p})^{-1}$ . Unfortunately, the above definitions are not universally followed.

- K<sub>d</sub>: A distribution coefficient defined as the quotient between element concentrations at assumed equilibrium in solid materials (e.g. sediment) and water (Bq/kg dry material /Bq/ kg water).
- **radiation dose:** Health physicists use an abundant nomenclature to define various aspects of the absorption of ionising radiation by the human body. Only the chief terms will be discussed here. The **effective dose equivalent** (Sv) is corrected for various differences in radiation quality and in the different sensitivity of various organs to radiation damage to give the same magnitude of effect per unit. The **effective dose commitment** or the **committed effective dose** describes the assumed accumulated effective dose integrated over 50 years, whereas the **dose rate** (Sv/yr) gives e.g. the annual dose at a given time. **Internal radiation doses** result from radiation emitted by radionuclides inside the body; **external doses** are caused by penetrating radiation from nuclides outside the body, e.g. on the soil. The doses can be expressed as **individual doses** (Sv) to single individuals in a population or as **collective doses** (manSv), i.e. the sum of all individual doses in a given population.
- radioecological sensitivity: (Bq/kg yr/Bq/m) (synonym: transfer coefficient from fallout to sample): is defined as the ratio of the infinite integral of concentrations in the sample to the integrated deposition. This is equivalent to UNSCEAR's transfer coefficient  $P_{23} = IC/IF_r$ , where IC is the integrated concentration of the radionuclide in the diet and IF<sub>r</sub> the integrated deposition of that radionuclide.

#### statistical terms:

standard deviation: S.D. = 
$$\sqrt{\frac{\sum_{i=1}^{n} (\bar{x} - x_i)^2}{n-1}}$$
  
standard error: S.E. =  $\sqrt{\frac{\sum_{i=1}^{n} (\bar{x} - x_i)^2}{n(n-1)}} = \frac{s}{\sqrt{n}}$ 

coefficient of variation: CV: relative standard deviation.

- Sv: sievert: the unit of dose equivalent = 1 J/kg.
- transfer factor from deposition to surface water:  $TF_w = C_w / D$ , where  $C_w$  is annual average concentration of <sup>137</sup>Cs in water (Bq/m<sup>3</sup>) and D is the average deposition of <sup>137</sup>Cs (Bq/m<sup>2</sup>) corrected annually for the physical decay.
- **transfer factor from deposition to fish**:  $TF_f = C_f /D$ , where  $C_f$  is the annual average concentration of <sup>137</sup>Cs in fish (Bq/kg fresh weight) and D is the average deposition of <sup>137</sup>Cs (Bq/m<sup>2</sup>) corrected annually for the physical decay.
- transfer coefficient from fallout to sample: See: radioecological sensitivity (Bq/kg yr/ Bq/m<sup>2</sup>).
- transfer factor from ground deposition  $(TF_g)$  or synonym transfer factor from soil to plants  $(T_f)$  is the ratio of the concentration in a product, e.g. in plant material, and the deposition on the ground surface; Bq/kg / Bq/m<sup>2</sup> = m<sup>2</sup>/kg. Weight basis, dry or fresh-weight, must be specified.
- **transfer coefficient to milk (F**<sub>m</sub>): The milk transfer coefficient (F<sub>m</sub> = Bq/l milk / Bq ingested/d) represents the fraction of the daily intake of a nuclide that is transferred to a litre of milk at equilibrium.

UNSCEAR: United Nations Committee on the Effects of Atomic Radiation.

### **Prefixes:**

| E | exa   | $10^{18}$  |
|---|-------|------------|
| Р | peta  | $10^{15}$  |
| Т | tera  | $10^{12}$  |
| G | giga  | $10^{9}$   |
| Μ | mega  | $10^{6}$   |
| Κ | kilo  | $10^{3}$   |
| Μ | milli | $10^{-3}$  |
| μ | micro | $10^{-6}$  |
| N | nano  | 10-9       |
| Р | pico  | $10^{-12}$ |
| F | femto | $10^{-15}$ |
| А | atto  | $10^{-18}$ |

# **Appendix IV Publications**

Nylén T (1996). "Uptake, turnover and transport of radiocesium in boreal forest ecosystems." Thesis. FOA-R-96-00242-4.3-SE.

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Koulikov, AO & Meili, M (1996). "*Modelling the dynamics of fish contamination by Chernobyl radiocaesium: an analytical solution based on potassium mass balance.*" In: Proceedings of the 9th International Congress on Radiation Protection (IRPA9), Vienna, 14-19 Apr. 96 (extended abstract, 3 pages).

Meili, M & Wörman, A (1996). "Desorption and diffusion of episodic pollutants in sediments: a 3-phase model applied to Chernobyl <sup>137</sup>Cs." Applied Geochemistry 11:311-316.

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Konitzer, K & Meili, M (1997). "*Redistribution of sedimentary*<sup>137</sup>*Cs in small Swedish lakes after the Chernobyl fallout 1986*." In: Proceedings of the International Seminar on Freshwater and Estuarine Radioecology, Lisbon, March 1994. Eds. Desmet G, Blust RJ, Comans RNJ, Fernandez JA, Hilton J & de Bettencourt A. pp. 167-172. Elsevier, Amsterdam ISBN 0-444-82533-9.

Koulikov, AO & Meili, M (1996). "*Physiological modelling of radiocaesium turnover in fish.*" In: Proceedings of the International Symposium on Ionising Radiation: Protection of the Natural Environment. Swedish Radiation Protection Institute, Stockholm, 20-24 May 1996. Swedish Radiation Protection Institute (SSI) and Atomic Energy Control Board (AECB) of Canada, ISBN 91-630-5106-0, pp. 100-105.

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Nikolova, I Johanson KJ & Dahlberg A. "*Radiocesium in fruit bodies and mycorrhizae in ectomycorrhizal fungi*." J. Environ. Radioactivity. In press

Dahlberg, A Nikolova, I & Johanson KJ. "Intraspecific variation of <sup>137</sup>Cs activity concentrations in sporocarp of Suillus variegatus in seven Swedish population." Mycological Res. In press.

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Strandberg M & Knudsen H "*Mushroom spores and* <sup>137</sup>Cs in faeces of the Roe Deer." J. Envir. Radioactivity 23.

Herlin AH & Andersson I (1996). "Soil ingestion in farm animals." Swedish University of Agricultural Sciences, ISSN 1104-7313 Report 105 1996.

Weyhenmeyer GA, Håkanson L & Meili M "A validated model for daily variations in the flux, origin and distribution of settling particles within lakes." Limnology & Oceanography (accepted).

Meili M & Wörman A "Modelling the desorption and diffusion of Chernobyl <sup>137</sup>Cs in sediments." Applied Geochemistry (in press).

# **REPORT 1** RADIOCESIUM IN THE FOODCHAIN OF LAMB IN THE FAROE ISLANDS. RESULTS FROM THE RAD-3 AND EKO-2 PROGRAMMES.

Hans Pauli Joensen, Trygvi Vestergaard & J. Zachariasen Institute of Natural Sciences, University of the Faroe Islands

#### ABSTRACT

The paper summarizes the results from an eight-year investigation of transfer of radiocesium from soil to grass and further to lamb. The investigation is part of RAD-3 (1990-93) and EKO-2 (1994-97), being inter-Nordic radioecology projects under the Nordic Committee for Nuclear Safety research, NKS.

Nine uncultivated pastures have been selected for the Faroese part of the projects. The sample program involves lamb meat lamb, mixed grass and soil from the uppermost 10cm. Lamb faeces have been included since 1995.

The deposition of radiocesium is now around 5 kBq/m<sup>2</sup>. About 60 Bq/kg(d.w.) is observed in mixed grass and around 12 Bq/kg(f.w.) in lamb meat. However, despite the limited geographical extent of the Faroe Islands, a significant variation is observed between pastures. Significant inter-annual variation is observed within the pastures as well.

Taking all stations together gives the following estimates of transfer in 1997: Grass/soil concentration ratio around 0.3, meat/soil concentration ratio around 0.05, and meat/grass concentration ratio around 0.5. The aggregated transfer factors are about  $15 \cdot 10^{-3} \text{ m}^2/\text{kg}$  and  $2.5 \cdot 10^{-3} \text{ m}^2/\text{kg}$  for grass/soil and meat/soil, respectively. These are low compared to other Nordic countries.

The meat/faeces and faeces/grass concentration ratios are found to be  $0.20 \pm 0.14$  and  $2.3 \pm 2.0$ , respectively. These are comparable to results found in a feeding experiment.

The effective ecological half-life,  $T_{1/2}$ , of radiocesium in mixed grass and lamb meat is estimated, assuming an exponential decay model. Data from single pastures give  $T_{1/2}$ (grass) and  $T_{1/2}$ (meat) to be in the range 2.0-2.8y and 4.0-12.6y, respectively.

# 1. Introduction

Fairly high consumption of lamb meat in the Faroe Islands has been a motivation for making radioecology a field of priority at University of the Faroe Islands. The annual

per capita consumption of lamb meat is 9.5 kg. There is an additional consumption of imported lamb meat (mainly from Iceland) of 9.6 kg per capita.

The Faroes have been contaminated by radioactive isotopes from the bomb tests in the 50'ies and 60'ies, and from the Chernobyl accident in 1986. Both sources can be registered. Results presented in this paper are from the Faroese part of the RAD-3 (1990-93) and EKO-2 (1994-97) programs under the Nordic Committee for Nuclear Safety Research, NKS.

# 2. Material and methods

For the eight-year period 1990-1997, soil and grass sampling took place every summer in nine uncultivated pastures in the Faroe Islands. Samples were taken from lamb at time of slaughter, typically in October. The sampling program includes faeces from lamb in the period 1995-97.

# 2.1 Soil

In the project period, some differences have been with respect to soil sampling technique. In 1990, two or three  $1m^2$  areas were randomly chosen in each pasture and divided into four  $0.25m^2$  microplots for soil and grass sampling. The soil cores - taken with a corer with 6.0cm inner diameter - were split up into an upper 2cm layer followed by 5cm layers. However, in the pastures Sandur and Sumba, samples were taken from four randomly chosen  $0.25m^2$  microplots, and the cores were split up into 1cm layers down to 5cm followed by two 2.5cm thick layers from 5 to 10cm depth. The participating laboratories agreed upon the latter technique in 1990 when only Sandur and Sumba remained to be sampled.

Four  $0.25 \text{ m}^2$  microplots were randomly chosen in each pasture for soil and grass sampling in the years 1991-97. A corer with inner diameter of 5.7 cm and length of 10 cm was used for the sampling. The cores were divided into an upper and a lower 5 cm layer except for 1992, when they were split up into 1cm layers down to 5 cm depth and 2.5 cm layers further down to 10 cm depth.

Every core disc was measured in 1990 when typically three cores were taken from each  $0.25 \text{ m}^2$  microplot. In other years, three cores were taken from the microplots, and measurement was carried out on mixed samples from each layer.

Soil samples were dried at room temperature before measurement. Some chemical analyses have been carried out on the soil.

### 2.2 Grass

The grass was cut from the 0.25  $\text{m}^2$  microplots before soil sampling. It was dried at  $105^{\circ}$ C before measurement, and the samples from each microplot were measured.

## 2.3 Lamb

The stock of sheep varied from one pasture to another, from 65 to 260. At slaughter, lamb meat (neck muscle) was collected according to an arrangement made with the farmers. Samples were taken from 30-40 lambs every year, typically in October when the lambs were about 6 months of age. The carcass weight was around 12-13 kg. Meat samples were used fresh for measurement, and samples have not been mixed.

### 2.4 Detector and software

The samples were measured using a lead shielded Germanium-detector, and the software OMNIGAM from EG&G Ortec was used for the spectral analyses.

### 2.5 Pasture data

Table 1 shows some data for the selected pastures. The pasture Funningur is only included in the RAD-3 project (1990-93). Pasture locations can be seen in Appendix H.

*Table 1. The stock of sheep, area and approximate height above sea level of pastures. Local names are included.* 

|                      | Bøur,<br>N.í Haga     | Velbastað<br>Lambhagi | Hvalvík,<br>Miðdalah. | Skáli,<br>Hegnið    | Funning.,<br>I. í Haga |
|----------------------|-----------------------|-----------------------|-----------------------|---------------------|------------------------|
| Area, m <sup>2</sup> | 3.16                  | 2.01                  | 5.75                  | 3.60                | 3.67                   |
| Height, m            | 100                   | 150                   | 50                    | 70                  | 100                    |
| Stock                | 128                   | 150                   | 260                   | 105                 | -                      |
|                      | Norðoyri,<br>Mið&Lýðh | Sandur<br>Skálsafjórð | Hvalba,<br>S.í Haga   | Sumba,<br>Skúvabøli |                        |
| Area, m <sup>2</sup> | 4.21                  | 4.32                  | 0.96                  | 1.45                |                        |
| Height, m            | 160                   | 240                   | 100                   | 200                 |                        |
| Stock                | 160                   | 200                   | 65                    | 100                 |                        |

# 2.6 Climate conditions

Local phenomena may affect the climate conditions in the pastures. There are no weather stations close to the pastures, but the tables below with data from the

Faroese capital, Tórshavn, may indicate the inter-annual variation in precipitation and temperature.

Table 2.Accumulated precipitation (mm) in Tórshavn, the capital of the Faroe Islands. Data for May-September (i.e. 5 months. Ref: The Danish Met. Institute).

| 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1961-81 |  |
|------|------|------|------|------|------|------|------|---------|--|
| 330  | 340  | 393  | 286  | 376  | 262  | 438  | 465  | 426     |  |

Table 3. Mean temperatures (Celsius) in Tórshavn, the capital of the Faroe Islands. Data for May-September (i.e. 5 months. Ref: The Danish Met. Institute).

| 1990 | 1991 | 1992 | 1993 | 1994 | 1995 | 1996 | 1997 | 1961-88 |
|------|------|------|------|------|------|------|------|---------|
| 9.9  | 10.2 | 9.8  | 8.3  | 8.8  | 9.1  | 9.4  | 9.6  | 9.2     |

# 3. Results

### **3.1** Chemical analyses of the soil

Chemical analyses of the top 10cm of the soil show pH to be generally less than 5.3. The observed ignition loss is typically in the range 50-70 %. The content of potassium is mostly in the range 300-600 mg/kg, but values around 1000 mg/kg are also observed. The concentration of sodium is similar to potassium, but higher values are observed in the southern end of the country, particularly in Hvalba where the content of sodium exceeds 1400 mg/kg.

### 3.2 Radiocesium in soil

Soil sampling took place every year in July except for 1990, when it was carried out in August. In the top 10 cm, typically 50-80 % of the radiocesium deposition is found to be in the uppermost 5 cm, with practically the same relative distribution every year.

There is a lot of inter-annual scattering in the data, and it is obviously not possible to make any simple exponential curve fitting, although a decreasing trend is observed for some pastures. A discernible geographical variation may be noticed, but no simple geographic correlation is found when the time variation is compared between pastures. Estimates of deposition and concentration of radiocesium in soil for the country as a whole can be found in Figs. 1 and 2 (all samples are considered as one pool).



Figure 1. Radiocesium deposition in the 0-10 cm soil layer. Annual average  $\pm 1$  standard error.



Figure 2. Radiocesium concentration in the 0-10 cm soil layer. Annual average  $\pm 1$  standard error.

The <sup>134</sup>Cs/<sup>137</sup>Cs ratio in the soil - documenting contamination from Chernobyl - is presented in Table 4 (-no data for Skáli and Sandur in 1991). <sup>134</sup>Cs is not detected in 1990 or after 1995.

|      | Bøur         | Velbastað    | Hvalvík | Skáli        | Funning | Norðoyri | Sandur       | Hvalba       | Sumba        |
|------|--------------|--------------|---------|--------------|---------|----------|--------------|--------------|--------------|
| 1991 | 0.045        | 0.056        | 0.036   | -            | 0.029   | 0.044    | -            | 0.050        | 0.065        |
| 1992 | 0.029        | 0.037        | 0.039   | 0.023        | 0.018   | 0.037    | 0.030        | 0.049        | 0.040        |
| 1993 | 0.027        | 0.024        | 0.028   | 0.017        | 0.018   | 0.024    | 0.020        | 0.035        | 0.034        |
| 1994 | 0.022        | 0.023        | 0.014   | 0.010        | -       | 0.023    | 0.013        | 0.026        | 0.025        |
| 1995 | 0.013<br>N=2 | 0.023<br>N=1 | N.D.    | 0.010<br>N=1 | -       | N.D.     | 0.011<br>N=1 | 0.021<br>N=3 | 0.018<br>N=4 |

Table 4. The  ${}^{134}Cs/{}^{137}Cs$  ratio in the 0-5 cm soil layer. (No data in 1990 and after 1995. N = number of samples). N.D. = Not Detected

### 3.3 Mixed grass

The concentration of <sup>137</sup>Cs (Bq/kg(d.w.)) in grass is presented in Figure 3, pooled for all the pastures. Hvalvík in the northern part of the Faroes has generally higher values than other pastures, while the lowest level is found in Hvalba in the southern end of the country. A decreasing trend is observed for some pastures, while others show an even increasing trend for the last 3 years of the project period.



*Figure 3. Radiocesium in mixed grass. Annual average*  $\pm 1$  *standard error.* 

Putting all the data into one pool shows a decrease from 1990 to 1994 followed by a level around 50-60 Bq/kg(d.w.).

The  ${}^{134}Cs/{}^{137}Cs$  ratio found in 1990 and 1991 is presented in Table 5.  ${}^{134}Cs$  was not detected in 1992, and in 1993 it was only observed in a sample from Hvalvík, having a  ${}^{134}Cs/{}^{137}Cs$  ratio of 0.051.  ${}^{134}Cs$  has not been detected in grass samples since 1993.
Table 5. Cs-134/Cs-137 ratio in mixed grass.

|      | Bøur  | Velbastað | Hvalvík | Skáli | Funningur | Norðoyri | Sandur | Sumba |
|------|-------|-----------|---------|-------|-----------|----------|--------|-------|
| 1990 | 0.188 | 0.172     | 0.080   | 0.120 | 0.032     | 0.112    | 0.139  | 0.171 |
| 1991 | -     | 0.041     | 0.059   | -     | 0.027     | -        | -      | 0.095 |

### 3.4 Lamb

In the eight-year project period, samples have been taken from 30-40 lambs every year. The time of slaughter is from late September to early November. The radiocesium concentration (Bq/kg(f.w.)) in the pastures and the country as a whole is shown in Fig. 4. The results are based on single lamb measurements.



*Figure 4. Radiocesium in lamb meat. Annual average*  $\pm 1$  *standard error.* 

The average  ${}^{134}$ Cs/ ${}^{137}$ Cs ratio in meat, based on single lamb measurements, was 0.117 in 1990 and 0.067 in 1991. The  ${}^{134}$ Cs isotope could be detected in only 5 lamb samples in 1992 (all from different pastures), giving the mean value 0.065.

# 4. Concentration ratios and transfer factors

All the results are for radiocesium. The observed concentration ratios are presented in Figs.5-9. The results are arithmetic means of data for the pastures together with minimum and maximum observed ratios.



Figure 5. Grass/soil concentration ratio\*1000. Mean, min and max..

For each  $0.25m^2$  microplot, grass/soil concentration ratio has been calculated from the concentration (Bq/kg(d.w)) in the 0-10cm soil layer and the concentration in grass. An estimate for the pasture is obtained by averaging these data, and an estimate for the country is obtained by averaging the results for the pastures.



Figure 6. Meat/soil concentration ratio\*1000. Mean, min and max.

The ratio between the concentration in each lamb and the mean concentration in the 0-10 cm soil layer is calculated for every pasture. This gives an indication of the variation with respect to animal. Averaging these ratios gives an estimate of the meat/soil concentration ratio for the particular pasture. An estimate for the whole country is obtained by averaging the pasture results.



Figure 7. Meat/grass concentration ratio\*1000. Mean, min and max.

The meat/grass concentration ratio is calculated for each pasture from single-lamb data and average values for concentration in grass. The ratio for the whole country is found as the arithmetic mean of pasture results.

The observed soil-to-grass  $(m^2/kg(d.w.))$  and soil-to-meat transfer factors  $(m^2/kg(f.w.))$  have been calculated in the same way as the concentration ratios, using the deposition  $(Bq/m^2)$  in the 0-10cm soil layer.



Figure 8. Grass/soil aggregated transfer factor  $(m^2/kg)*1000$ . Mean, min and max..



Figure 9. Meat/soil aggregated transfer factor  $(m^2/kg)$ \*1000. Mean, min and max

# 5. Half-life

Doing a linear regression analysis between time and the logarithm of radiocesium in grass and in lamb meat gives the results in Table 6. It has not been possible to distinguish between the half-life of Chernobyl- and fallout-cesium, since <sup>134</sup>Cs content is mainly below the detection limit.

Table 6. Effective ecological half-lives (years) assuming exponential decay.  $R^2$  from the linear regression between time and natural logarithm of activity is shown in parenthesis. No data if  $R^2 < 0.3$ .

|                  | Bøur            | Velbastað      | Hvalvík | Skáli   | Norðoyri | Sandur         | Hvalba         | Sumba          |
|------------------|-----------------|----------------|---------|---------|----------|----------------|----------------|----------------|
| Grass<br>1990-97 | - (0.008)       | 2.1<br>(0.559) | (0.297) | (0.055) | (0.168)  | 2.0<br>(0.597) | (0.198)        | 2.8<br>(0.651) |
| Meat<br>1990-97  | 12.6<br>(0.425) | 7.8<br>(0.386) | (0.053) | (0.032) | (0.193)  | - (0.047)      | 6.9<br>(0.847) | 4.0<br>(0.513) |

Using annual mean values for the *whole country* and doing a semi-logarithmic fit between time and radiocesium in grass gives the half-life  $T_{1/2}(\text{grass})= 5.3$  years (R<sup>2</sup>=0.508). Using the same procedure for meat gives R<sup>2</sup>=0.168, which is too poor for estimating half-life.

Fig.4 shows that the level of radiocesium in meat is shifted from 1993 to 1994. Doing the same semi-logarithmic fit for the sub-periods 1990-93 and 1994-97 gives  $T_{1/2}(meat)=2.1y$  (R<sup>2</sup>=0.940) and  $T_{1/2}(meat)=4.0y$  (R<sup>2</sup>=0.853), respectively. For grass we obtain  $T_{1/2}(grass)=1.6y$  (R<sup>2</sup>=0.993) for the years 1990-93, while no decreasing trend is observed for 1994-97 (see Fig.3).

The results indicate that the decay model is more complicated than simple exponential.

# 6. Faeces as an indicator of sampling representability

The most obvious problem in transfer factor calculations is to estimate the activity of the vegetation the sheep have actually eaten. Concentration of <sup>137</sup>Cs in faeces can in steady state conditions be used to estimate the concentration in the feed. <sup>137</sup>Cs in faeces is not just a left-over from ingested <sup>137</sup>Cs, since sheep excrete around 2/3 of <sup>137</sup>Cs excreted into the intestines (refs. 8, 12, 13, 15). Table 7 shows <sup>137</sup>Cs concentrations in faeces 1995 –1997 from Faroese sampling stations. Annual variation in <sup>137</sup>Cs-concentration in faeces is less than in vegetation.

Table 7. Cs-137 (Bq/kg(d.w) in lamb faeces from Faroese pastures 1995-97.

|      |      |           |         |       | 0        |        |        |       |
|------|------|-----------|---------|-------|----------|--------|--------|-------|
|      | Bøur | Velbastað | Hvalvík | Skáli | Norðoyri | Sandur | Hvalba | Sumba |
| 1995 | 32.6 | 59.2      | 61.5    | 120.9 | 98.2     | 111.0  | 23.2   | 69.0  |
| 1996 | 30.1 | 29.3      | 64.0    | 117.6 | 55.6     | 76.6   | 18.4   | 65.3  |
| 1997 | 49.2 | 32.8      | 125.7   | 113.7 | 37.2     | 56.6   | 18.5   | 55.3  |

The average meat/faeces concentration ratio based on data in Table 5.2 is  $0.20 \pm 0.14$  (stdev). This ratio is generally more constant than the corresponding meat/grass ratio. From a feeding experiment the value 0.24 should be expected for the ratio (see section below, Table 9). From the data in Table 8 it seems likely that the lamb samples from Hvalvík are non-representative.

Table 8. Meat/faeces <sup>137</sup>Cs-concentration ratio in Faroese pastures 1995-97.

| 10000 | 0. 110000 | Jacces e  | s concent |       | ano mi i uno | ese pasia |        | //.   |
|-------|-----------|-----------|-----------|-------|--------------|-----------|--------|-------|
|       | Bøur      | Velbastað | Hvalvík   | Skáli | Norðoyri     | Sandur    | Hvalba | Sumba |
| 1995  | 0.19      | 0.04      | 0.58      | 0.27  | 0.20         | 0.14      | 0.09   | 0.09  |
| 1996  | 0.26      | 0.06      | 0.58      | 0.38  | 0.13         | 0.26      | 0.11   | 0.05  |
| 1997  | 0.09      | 0.12      | 0.26      | 0.21  | 0.19         | 0.22      | 0.10   | 0.16  |

Table 9 shows faeces/grass concentration ratios for the sampling stations. Five of the eight stations have ratios consistently around the expected value of  $2.8 \pm 1.0$  (see section below Table 9). Two stations, Hvalvík (which also had an unusually high meat/faeces ratio) and Bøur had ratios less than half the others. Sandur had a ratio more than double of what should be expected. The average and standard deviation of the data in Table 9 is  $2.3 \pm 2.1$ . Excluding Bøur, Hvalvík and Sandur we obtain  $2.0 \pm 0.9$ . From the data in Table 5.3, <sup>137</sup>Cs- concentration in feed may probably be overestimated in Hvalvík and Bøur, and underestimated in Sandur.

| Table | 9. <i>Fuece</i> | es/grass  | Cs-concentration ratio in Farbese pastures 1995-97. |       |          |        |        |       |
|-------|-----------------|-----------|---|-------|----------|--------|--------|-------|
|       | Bøur            | Velbastað | Hvalvík   | Skáli | Norðoyri | Sandur | Hvalba | Sumba |
| 1995  | 1,43            | 2,32      | 0,38  | 1,53  | 1,54     | 4,89   | 1,56   | 1,68  |
| 1996  | 0,37            | 3,66      | 0,40  | 2,44  | 1,23     | 8,06   | 1,28   | 2,41  |
| 1997  | 0,66            | 1,59      | 0,64  | 1,17  | 0,54     | 7,45   | 3,25   | 3,43  |

Table 9. Faeces/grass <sup>137</sup>Cs-concentration ratio in Faroese pastures 1995-97.

A feeding experiment was carried out in order to get some insight into connections between faeces and feed concentrations. Twin male lambs were moved from a meadow with a known <sup>137</sup>Cs- activity in grass to another with about 1/3 of the concentration of the former. Two weeks later the lambs were moved to a stable and fed grass from the second field. Amount of feed, faeces and their <sup>137</sup>Cs- activity were measured. Digestibility in this period ranged from 70 to 84%, with a mean of 82%. The first fortnight, faeces/grass <sup>137</sup>Cs- concentration ratios ranged from 4.5 to 9.5. In this period the lambs excreted more <sup>137</sup>Cs through their faeces than they ingested. In the following 3 weeks concentration ratios dropped rapidly to  $2.8 \pm 1.0$ . Therefore, the faeces/grass concentration ratio is expected to be around this value under steady state conditions.

Meat/faeces <sup>137</sup>Cs- concentration ratio in the feeding experiment was 0.24. Only faeces concentrations from the steady state period were used in the calculation. Standard deviation for concentration in faeces in this period was around 30%.

# 7. Discussion

The paper summarizes the results from the Faroese part of the RAD-3 and EKO-2 projects. The RAD-3 program involved 9 uncultivated pastures, one of which had to be abandoned for practical reasons in the EKO-2 program.

Chemical soil parameters have been measured in addition to radiocesium. The soil type is peaty with pH below 5.3. The ignition loss is generally around 50-70%. The content of potassium and sodium is typically in the range 300-600 mg/kg.

The ratio between the deposition in the uppermost 5cm of the soil and in the 0-10 cm soil layer has a range from 0.5 to 0.8 every year. The highest Cs-134/Cs-137 ratios were found in the southern part of the country.

Physical data for the pastures are presented in Table 1. In most cases it was manageable to find the same spots for soil and grass sampling every year, although no fencing was done for the project, mainly because the same people took the samples every year. However, it was necessary to move the sample-taking site in Velbastað about 3km for the years 1994-97 because of cattle grazing, and this change may be an explaining factor for the increasing trend in observed deposition in

Velbastað these years. Such changes have not occurred in other pastures, and it seems difficult to give a satisfactory explanation of the inter-annual variation of radiocesium in soil. The main reason may possibly be related to differences in soil characteristics, but this has not been analysed as yet.

Considering all radiocesium soil data as one pool show unexpected high deposition levels (Bq/m<sup>2</sup>) in 1994 and 1995 (Fig.2). A decreasing trend is observed for concentration (Bq/kg) for the years 1994-1997, while more scattering and lower values are observed for 1990-93. There seems to be a shift to higher levels in both deposition and concentration from 1993 to 1994.

The radiocesium content in lamb meat presented in Fig.4 for the whole country do also shift to higher levels from 1993 to 1994. The rise comes mainly from Hvalvík, Skáli, Norðoyri and Sandur. It is well-known that Hvalvík and Norðoyri are places with quite high precipitation rates. A tempting explanation for the shift in content of radiocesium in soil and lamb meat would be that an extra input of <sup>137</sup>Cs should have entered the Faroes, but no source for it has been revealed to our knowledge.

For the country as a whole the radiocesium content in grass decreases from 1990 to 1993, where upon it comes to a level around 50-60 Bq/kg(d.w). However, this trend is not observed for all pastures, as an increasing trend in Hvalvík, Skáli, Norðoyri and Bøur can be observed for the years 1994-97.

Radiocesium in grass is observed to vary a lot from one  $0.25 \text{ m}^2$  microplot to another. This confirms the difficulty of taking representative samples, but may also reflect difference in botanical composition on the microplots.

The results of a correlation analysis are presented in Table 10. Annual mean values are used for the pastures. Correlating radiocesium in meat and grass for 1990-97 give positive correlation in all pastures, with the highest coefficient for Sumba (0.908) and the lowest for Bøur (0.261). Making the correlation for the years 1994-97 gives lower correlation, and it is found to be negative for Hvalvík, Skáli and Sandur, indicating non-representative sample taking. However, such a correlation analyses is not satisfactory in deciding whether or not the sample taking has been representative.

|                            | Bøur  | Velbastað | Hvalvík | Skáli  | Norðoyri | Sandur | Hvalba | Sumba  |
|----------------------------|-------|-----------|---------|--------|----------|--------|--------|--------|
| Meat &<br>Grass<br>1990-97 | 0.261 | 0.834     | 0.594   | 0.296  | 0.391    | 0.756  | 0.471  | 0.908  |
| Meat &<br>Grass<br>1994-97 | 0.252 | 0.462     | -0.934  | -0.863 | -0.026   | -0.273 | 0.157  | -0.023 |

Table 10. Correlation coefficients for radiocesium in meat (Bq/kg(f.w.)) and grass (Bq/kg(d.w.)) calculated for 1990-97 and 1994-97.

Faeces have been sampled simultaneously with grass and soil sampling in the years 1995-97. For the years 1995-97 the average ( $\pm 1$  standard deviation) for the meat/faeces concentration ratio is found be  $0.20 \pm 0.14$ , while the analogue faeces/grass concentration ratio is  $2.3 \pm 2.0$ . Excluding extreme values for three of the pastures (Bøur, Hvalvík and Sandur) result in a faeces/grass concentration ratio of  $2.0 \pm 0.9$ . From a feeding experiment the meat/faeces concentration ratio is estimated to  $0.24 \pm 0.07$ , while the faeces/grass concentration ratio be  $2.8 \pm 1.0$ . The sampling must therefore be considered as representative for most pastures.

The observed concentration ratios and aggregated transfer factors (m<sup>2</sup>/kg) vary significantly both geographically and within the pastures. For the country as a whole the average grass/soil and meat/soil concentration ratios tend to decrease from 1990 to 1994 and from 1990 to 1993, respectively. After this they tend to be a bit higher (Figs.1 and 2), slightly increasing in the first case and having a decreasing trend in the second case. The meat/grass concentration ratio tends to increase slightly from 1990 to 1994, while it is more scattering after 1994. The average ratio between the concentration (Bq/kg) of radiocesium in meat and grass is observed to be a factor 2-3 lower in the Faroe Islands than in Norway, having similar soil types as the Faroe Islands. A controlled feeding experiment (not discussed in the paper) with two male twin lamb confirmed the observed meat/grass ratios.

The aggregated transfer factor from soil to meat for the country as a whole has a decreasing trend from 1990 to 1993. After 1993 it shifts to a bit higher value, slightly decreasing until 1997 (Fig.5). The observed meat/soil transfer factor is observed to be lower than in Norway by a factor of 15-20.

The aggregated transfer factor from soil to grass for the country as whole decreases from 1990 to a minimum in 1994 followed by a slightly increasing trend until 1997 (Fig.5). It is observed to be a factor 3-10 lower than in Norway.

The variation with respect to animal is found to be high for both meat/soil concentration ratio and meat/soil aggregated transfer factor (Figs.5 and 6), as well as for meat/grass concentration ratio (Fig.4).

The effective ecological half-life,  $T_{1/2}$ , has been estimated on the assumption of simple exponential decay with time. However, data from most pastures do not fit to this model (Table 7). For grass,  $T_{1/2}$  is found to be 2.0-2.8 y for the pastures and 5.3 y when data from all pastures are considered as one pool. For lamb meat,  $T_{1/2}$  is between 4.0 y and 12.6 y for single-pastures, but the exponential decay model does not fit to the data when all pastures are considered as one pool. The decay model seems therefore to be more complicated than simple exponential.

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# **REPORT 2** Analysis of mushroom spores in sheep faeces

Morten Strandberg National Environmental Research Institute

# 1. Introduction

In deer it has been shown that fungi make up a large part of the diet during the mushroom season (Fraiture, 1992). Later it was shown that mushrooms are responsible for an increase in the concentration of radiocesium in meat from roe deer, this was done through studies of the occurrence of mushroom spores in faeces (Strandberg and Knudsen, 1994). Rafferty et al.(1994) found that among sheep grazing in the same area, those having access to areas with fruitbodies of species having ectomycorrhiza with trees, had the highest levels of radiocesium in the meat.

If mushrooms with a high concentration of radiocesium generally are eaten by ruminants both in household and nature, a reasonable countermeasure will be to relate the season of hunting or slaughter to a time of the year when mushrooms are sparse.

From studies of the relation between the intake of mushrooms and the occurrence of spores in the faeces of sheep it can preliminary be said that one spore in 0.3 mg dry weight of sheep faeces equals a daily intake of 12 g fresh mushrooms (Strandberg and Hansen in prep.) At the time being this is a rough estimate, and it is likely that further investigations may increase the information about the dependence upon the spore type i.e. thick-walled spores are more likely to pass intact through the gastro intestinal tract (GIT) than spores with thinner walls. Hence it may be necessary to operate with more conversion factors than the one used in this study i.e. the more the spore is adapted to pass through the GIT the smaller the intake of fresh mushrooms per spore found in the faeces is.

# 2. Material and methods

Sheep faeces from the four Nordic sampling sites were collected. The faeces were sampled during the mushroom season for the purpose of analyses for occurrence of mushroom-spores in the faeces. The faecal pellets were grinded gently to obtain a dry powdery material. A small sample (0.3 mg) of this material was soaked with distilled water and placed under the light microscope. Here all of the sample was studied for occurrence and counting of mushroom spores at 20 times magnification. All types of spores found was studied more detailed at 40 times magnification for further identification. Identification was carried out according to Hansen & Knudsen (1995).

## 2.1 Iceland 1990 (Table 1)

Only few mushroom spores (0 - 1, on average 0.4 per specimen) were found in the samples from June and July 1990. In the samples from August and September 0 - 3, on average 1.7 spores per specimen were detected. This equals an daily intake of 4 g in June and July and 20 g in August and September. The spores were from typical grassland species of mushrooms, *Stropharia semiglobata* and probably *Stropharia coronilla*, *Psilocybe sp*. one species of *Entoloma* and probably *Clitocybe* and some *Coprinus spp*. Most of the spores are from species commonly found associated with dung. The amount of spores found in the faeces gives only a rough indication of the amount of fungi eaten by Icelandic sheep in the mushroom season.

The intake of mushrooms increases over the season from June to July. There was no difference between sheep and lamb as regards the amount and species ingested.

The most abundant spores are from the genus *Stropharia* which have thick walled spores adapted to survive the passage through the GIT.

There were no observations of spores from species known to concentrate radiocesium.

## 2.2 Norway 1995 (Table 2)

At the Norwegian sampling site, the daily intake was the highest in the beginning of the season with 28 g mushroom fresh weight. In July and August the intake decreased to between 8 and 12 gram of fresh weight per day. The species eaten were ordinary grassland mushrooms, many of them associated with dung from ruminants (Table 5). None of the spores found are from species known to concentrate radiocesium.

## 2.3 Sweden 1994 (Table 3)

On the locality Blomhöjden faeces were sampled twice during the mushroom season of 1994. Three samples were from the 25th of July and another three were from the 23rd of August. From these samples the number of spores detected were between 0 and 1. Both in July and August one out of three samples contained one spore. Both was non-pigmented spores, the genus could be *Marasmius* and the spore from August could belong to the species *Marasmius oreades*, which commonly grows on grassland. Contrary to expectation no spores from any species of *Boletus* or related species were found, although samples of mushrooms has demonstrated that these species are available in the forested areas at Blomhöjden. The spores represented in the faeces are not from species that could explain an increased amount of radiocesium in the faeces.

The number of spores found in the faeces of the Swedish sheep in 1994 indicates a consumption of mushrooms less than 10 g of fresh weight per day during July and August.

Spot-tests from the material from Blomhöjden from 1995 and 1996 did not indicate signs of ingestion of mushrooms beyond what was observed in the 1994 material.

The relative high transfer of radiocesium to Swedish sheep at Blomhöjden should probably not be explained by an extraordinary high consumption of mushroom. Rather the explanation could be that the diet of the Swedish sheep contains a lot of herb species other than grass. If Ericaceous species e.g. bilberry, *Vaccinium myrtillus* and cowberry, *Vaccinium vitis-idaea* are included in the diet it may explain a higher level in the meat than would be expected if the diet was pure grass.

### 2.4 The Faroe Islands 1996 (Table 4)

From the Faroe Islands material sampled from 5 localities in June 1996 was analysed for occurrence of spores. Only few spores were found in the faeces. Two different species of *Agaricus*, the one probably *Agaricus campestris* and the other not identified. Then also a spore from the genus *Inocybe* was found. This genus forms ectomycorrhiza with different tree species and also some herbs. The daily intake of mushrooms ranged between 0 and 30 g, depending upon locality. None of the species identified is known to concentrate radiocesium.

| Location | Month | spores       | Mean   | SD   | Daily intake     |
|----------|-------|--------------|--------|------|------------------|
|          |       | per specimen | spores |      | (g fresh weight) |
| Iceland  | Jun   | 0            |        |      |                  |
| Iceland  | Jun   | 1            | 0.33   | 0.58 | 4                |
| Iceland  | Jun   | 0            |        |      |                  |
| Iceland  | Jul   | 1            |        |      |                  |
| Iceland  | Jul   | 0            |        |      |                  |
| Iceland  | Jul   | 1            | 0.50   | 0.58 | 6                |
| Iceland  | Jul   | 0            |        |      |                  |
| Iceland  | Aug   | 2            |        |      |                  |
| Iceland  | Aug   | 1            |        |      |                  |
| Iceland  | Aug   | 2            | 1.60   | 0.55 | 19               |
| Iceland  | Aug   | 2            |        |      |                  |
| Iceland  | Aug   | 1            |        |      |                  |
| Iceland  | Sep   | 1            |        |      |                  |
| Iceland  | Sep   | 3            |        |      |                  |
| Iceland  | Sep   | 3            | 1.80   | 1.30 | 21               |
| Iceland  | Sep   | 2            |        |      |                  |
| Iceland  | Sep   | 0            |        |      |                  |

Table 1. Results from Iceland 1990

Table 2. Results from Norway 1995

| Location         | Month | spores       | Mean   | SD   | Daily intake     |
|------------------|-------|--------------|--------|------|------------------|
|                  |       | per specimen | spores |      | (g fresh weight) |
| Larvekra, Tjøtta | Jul   | 2            |        |      |                  |
| Larvekra, Tjøtta | Jul   | 0            | 2.33   | 2.52 | 28               |
| Larvekra, Tjøtta | Jul   | 5            |        |      |                  |
| Larvekra, Tjøtta | Aug   | 1            |        |      |                  |
| Larvekra, Tjøtta | Aug   | 1            | 0.67   | 0.58 | 8                |
| Larvekra, Tjøtta | Aug   | 0            |        |      |                  |
| Larvekra, Tjøtta | Sep   | 2            |        |      |                  |
| Larvekra, Tjøtta | Sep   | 1            | 1.00   | 1.00 | 12               |
| Larvekra, Tjøtta | Sep   | 0            |        |      |                  |

Table 3. Results from Sweden 1994

| Table 3. Results from Sweden 1994 |   |   |   |   |  |  |  |  |  |  |
|-----------------------------------|---|---|---|---|--|--|--|--|--|--|
| Month                             | spores  | Mean  | SD  | Daily intake  |  |  |  |  |  |  |
|                                   | per specimen                                    | spores  |   | (g fresh weight)  |  |  |  |  |  |  |
| Jul                               | 0   |   |   |   |  |  |  |  |  |  |
| Jul                               | 1   | 0.33  | 0.57  | 4   |  |  |  |  |  |  |
| Jul                               | 0   |   |   |   |  |  |  |  |  |  |
| Aug                               | 0   |   |   |   |  |  |  |  |  |  |
| Aug                               | 0   | 0.33  | 0.57  | 4   |  |  |  |  |  |  |
| Aug                               | 1   |   |   |   |  |  |  |  |  |  |
|                                   | Month<br>Jul<br>Jul<br>Jul<br>Aug<br>Aug<br>Aug | Irom Sweden 1994Monthspores<br>per specimenJul0Jul1Jul0Aug0Aug0Aug1 | Irom Sweden 1994MonthsporesMeanper specimensporesJul0Jul10.33Jul0Aug0Aug0Aug1 | Item Sweden 1994MonthsporesMeanSDper specimensporesJul0Jul10.330.57Jul0Aug0Aug00.330.57Aug1 |  |  |  |  |  |  |

Table 4. Results from the Faroe Islands

| Location  | Month | spores       | Mean   | SD   | Daily intake     |
|-----------|-------|--------------|--------|------|------------------|
|           |       | per specimen | spores |      | (g fresh weight) |
| Hvalvik   | Jun   | 0            |        |      |                  |
| Hvalvik   | Jun   | 2            | 1.00   | 1.41 | 12               |
| Bøur      | Jun   | 2            |        |      |                  |
| Bøur      | Jun   | 3            | 2.50   | 0.71 | 30               |
| Velbastad | Jun   | 0            |        |      |                  |
| Velbastad | Jun   | 0            | 0.00   | 0.00 | 0                |
| Hvalba    | Jun   | 1            |        |      |                  |
| Hvalba    | Jun   | 0            | 0.50   | 0.71 | 6                |
| Sumba     | Jun   | 0            |        |      |                  |
| Sumba     | Jun   | 0            | 0.00   | 0    | 0                |
| Total     |       | 8            | 0.80   | 1.14 | 9,6              |

Table 5. List of species and genera so far recognised from spores found in faeces from the localities - those recognised to species should be regarded as likely to be this species.

| Iceland                   | Norway                    | Sweden               | The Faroe Islands   |
|---------------------------|---------------------------|----------------------|---------------------|
| 1990                      | 1995                      | 1994                 | 1996                |
| Stropharia<br>semiglobata | Stropharia<br>semiglobata | Marasmius<br>oreades | Stropharia sp.      |
| Stropharia sp.            | Stropharia sp.            | Not identified 1     | Agaricus campestris |
| Entoloma sp.              | Psilocybe sp.             |                      | Agaricus sp.        |
| Clitocybe sp.             | Omphalina sp.             |                      | Inocybe sp.         |
| Coprinus sp.              | Not identified 2          |                      | Not identified 1    |
| Not identified 3          |                           |                      |                     |

# 3. Conclusions

Consumption of mushrooms was not revealed as a major source of radiocesium to sheep at the localities examined through this investigation. However, a marked difference among years may exist, i.e. in years with a long lasting mushroom season sheep may learn to exploit this source of food. It must be pointed out that neither of the investigated years were good mushroom years, on the contrary, these years were especially poor mushroom years in general. In normal years, only few animals will spend time searching for mushrooms.

Probably the higher uptake rates observed in Sweden and on the Faroes should be explained by differences in the composition of the diet between the different countries i.e. localities. Consumption of ericaceous shrubs like *Calluna vulgaris*, Vaccinium myrtillus & Vaccinium vitis-idaea can explain the differences in transfer from soil to meat between the localities. Hence it is important to know the precise composition of the sheep diet at least in the period before they are slaughtered. In this context a few samples of grass does not satisfactorily represent the diet. Observations from the Faroes clearly demonstrates that sheep consumes large amounts of heather Calluna vulgaris. Faecal samples from sheep that consumes mushroom could be screened by an investigation of the radiocesium and radiopotassium content. In the case of a significant mushroom consumption these concentrations should be at least a factor two higher than in faeces collected from the same place when there is no mushrooms available. If there is an increased amount of radiocesium in the mushrooms consumed, the radiocesium content in the faeces will be increased. If the mushrooms consumed belongs to species with a low radiocesium content e.g. Agaricus spp., then the high concentration of potassium in mushrooms will be reflected by the <sup>40</sup>K concentration in the faeces.

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# **REPORT 3** MOBILITY OF CESIUM AND STRONTIUM IN SOILS

#### Deborah Oughton

Laboratory for Analytical Chemistry, Agricultural University of Norway

Soil represents the major sink for radionuclides released into the terrestrial environment. Hence, the mobility of any radionuclide and its subsequent transfer through food chains will be dependent largely on its interactions with soil components. In any soil-plant-animal system, time dependent changes in vegetation and animal activity levels can reflect a number of factors. For example, ecological half-lives can be influenced by the rate at which radionuclides are physically removed from soil rooting zones (i.e., run-off, transport down the soil profile) and by the rate at which radionuclides are transformed *chemically* to species that are less available for biological uptake (e.g., diffusion of radiocesium into the clay mineral lattice) (Fig. 1). Providing that environmental and agricultural parameters remain constant, long-term changes in soil-to-plant transfer factors usually reflect a change in chemical speciation of the radionuclide and/or depth distribution in the rooting zone. If radionuclides are deposited in an inert form (i.e., uranium oxide fuel particles) weathering to more mobile species may lead to an increase in transfer factors. Fixation of long-lived radionuclides to soil minerals leads to a reduction in transfer factors, and often represents the major ratedetermining parameter for ecological half-lives. It follows that variations in both transfer factors and ecological half-lives between different soil-plant-lamb ecosystems may reflect site-specific parameters (i.e., soil characteristics, vegetation type, climate conditions and agricultural practice) and/or the source term of fallout radionuclides.

It has been known for many years that the mobility of radiocesium is limited by interactions with clay minerals (Sawnhey, 1960; Shultz et al., 1960; Sawnhey, 1972; Evans et al, 1983). The highly selective sorption of cesium to clay minerals is thought to take place in the interlayer spaces between the frayed particle edges (Sawnhey, 1972). Non-expanded layer silicates such as illite and micas show a greater retention of cesium than the expanded layer silicates vermiculite and montmorillonite (Tamura and Jacobs, 1960). However, in recent years, interest has been directed towards the relationship between cesium mobility and the *kinetics* of interactions with soil components (Absalom et al., in press) and, in particular, on the reversibility of cesium sorption processes (Comans et al., 1991; Comans and Hockley, 1992). Laboratory tracer studies on illite (Comans et al., 1991), soils (Absalom et al, in press) and freshwater sediments (Evans et al., 1983) have indicated a slow fixation of radiocesium, with reaction times of months to years. It is clear that the kinetics of such sorption processes will influence both the mobility and the ecological half-life of radiocesium.



Figure 1. Major soil processes influencing ecological half-lives in a soil-plantsheep ecosystem. In addition to physical decay of the radionuclide, the ratedetermining parameters controlling the long-term decrease in activity concentrations in vegetation and lamb meat are fixation to soils and removal from the system with run-off and produce.

The work presented here had two aims: first, to study the kinetics of radiocesium and radiostrontium sorption in soils using laboratory tracer experiments, and to relate results to the environmental studies of ecological half-lives at the selected Nordic sites; and, second, to obtain information on the equilibrium or steady-state transfer of naturally occurring stable Cs isotopes at the different sites and compare these with the observed transfer factors for <sup>137</sup>Cs.

# 1. Materials and methods

### **1.1 Sample Preparation**

Soil, vegetation and lamb muscle samples have been received from Tjøtta, Norway (1995 and 1996); Blomhöjden, Sweden (1994, 1995 and 1996); Hestúr, Iceland (1994), Iceland (1996), Ribe, Denmark (1995) and Faroe Islands (1994 and 1995). Soils were air dried ( $<60^{\circ}$ C) prior to extraction and tracer studies. Standard soil

characteristics were measured in all soil samples included in tracer studies. Parameters include: pH, dry weight (105°C), organic content by loss on ignition (LOI), cation exchange capacity (CEC), major available macro elements, and base saturation (Øien and Krogstad, 1987). A summary of soil characteristics has already been presented in Paper 1. Stable Cs and other trace element concentrations were determined by neutron activation analysis (NAA). Full details of the NAA technique can be found in Oughton and Day (1993). Vegetation samples were dried and/or ashed, homogenised and lamb tissue samples were ashed (350 - 400°C) prior to determination of stable Cs and trace element concentration (NAA). All analyses, including tracer studies, were carried out in duplicate.

### **1.2 Kinetic Tracer Studies**

Sorption kinetics in soils were studied using controlled laboratory tracer (<sup>134</sup>Cs and <sup>85</sup>Sr) experiments. The hypothesis is that rates of fixation in soils will play a central role in determining the ecological half-life of radiocesium in an ecosystem. Thus laboratory experiments enable us to determine the extent to which soil parameters alone (as compared to environmental and agricultural parameters) might account for observed differences in ecological half-lives between the Nordic sites. Particular emphasis was placed on the rates of change of radionuclide mobility, sorption mechanisms and speciation in soils. Desorption studies included the application of sequential extraction techniques.

Tracer solutions ( $^{137}$ Cs and  $^{85}$ Sr) in artificial rainwater (pH 5.2) were added to airdried soils, and the mixture homogenised gently using a food mixer. The volume of water added was enough to ensure that the soil was evenly wet without being waterlogged. All samples were stored at 4°C in aerobic conditions. Aliquots of soil were removed as a function of time (up to 12-18 months) and extracted first with water and then with 1 M NH<sub>4</sub>Ac (soil pH). The solid (dry weight) to liquid ratio was ca. 1:20 and extraction time was 1 hour. Extracts and residues were counted, and the rate of adsorption calculated as a function of time after addition. In order to provide more detailed information on soil-radionuclide interaction, selected aliquots were subject to more thorough sequential extraction procedures, including extraction with 1M NH<sub>4</sub>Ac (pH 5) and dissolution with H<sub>2</sub>O<sub>2</sub> and 7M HNO<sub>3</sub>. Full details of sequential extraction methods can be found elsewhere (Oughton et al. 1992).

## **1.3** Stable Cs Transfer

Variations in <sup>137</sup>Cs transfer factors have been observed between the different Nordic sites (Hove et. al., 1994). This could be due to site-specific environmental parameters such as soil characteristics or agricultural practice, or it might reflect varying degrees of isotopic exchange --- confounded by the fact that the sites have

varying contributions from weapons and Chernobyl fallout. Deposited <sup>137</sup>Cs should act as a tracer for the naturally-occurring stable Cs analogue: isotopic exchange will occur between radiocesium and stable cesium. Thus stable Cs levels will reflect the «steady-state» distribution in the ecosystem, hence determination of stable Cs in the soil-plant-lamb systems can provide valuable information on site-specific variations in transfer factors. In particular, determination of stable Cs transfer factors could help to identify sites that are ecologically sensitive to Cs (i.e. high transfer factors). Transfer factors and concentration ratios are calculated using data from trace element analysis (NAA). In addition to measurements on whole samples, the distribution of stable analogues in sequential extracts has been compared to both <sup>137</sup>Cs and <sup>90</sup>Sr and the laboratory-added <sup>134</sup>Cs and <sup>85</sup>Sr tracers.

# 2. Results

### 2.1 Characterisation of soils

Available data on soil characteristics is given in Report 1. Soils from the Blomhöjden, Tjøtta and Faroe Island sites (LOI, pH, CEC) varied between the different sampling plots, whereas soils from the other sites were more homogeneous. Trace element analysis revealed that concentrations of trace elements in soils showed rather large variations between the different sites (Table 1). Soils showed either similar levels or enhancement of trace elements in the lower mineral layers as compared to upper layers. Zinc was the only element to show consistently higher levels in the organic layer.

## 2.2 Kinetic Tracer Studies

Results from sorption studies showed that both <sup>134</sup>Cs and <sup>85</sup>Sr became rapidly associated with soil components. After 1 hour contact time with soils, more than 90% of the total activity was adsorbed to soils: less than 10 % was displaced with a water extraction. However, soil-bound <sup>85</sup>Sr was significantly more mobile than <sup>134</sup>Cs: between 60 and 100 % of the sorbed <sup>85</sup>Sr could be displaced from binding sites by a single NH<sub>4</sub>Ac extraction, compared to 4 to 35 % for <sup>134</sup>Cs (Figs. 2 and 3). Both nuclides showed a time-dependent increase in the degree of sorption to soils, the percentage extracted both by water and by NH<sub>4</sub>Ac decreasing with contact time.

|                         | Ce   | Со                         | Cr                 | Cs   | Eu   | Fe mg/g               | Rb                | Sc                         | Se                                      | Sr                        | Zn                        |
|-------------------------|--|----------------------------|--------------------|--|--|-----------------------|-------------------|----------------------------|---|---------------------------|---------------------------|
| Tjøtta<br>n=12          | $60.1 \pm 8.0$<br>(27-104)                                       | 6.1 ± 1.0<br>(3.9-10)      | 96 ±13<br>(57-194) | $\begin{array}{c} 1.31 \pm 0.16 \\ (1.12 \text{-} 1.91) \end{array}$ | 1.04 ±0.09<br>(0.42-1.5)   | 22 ± 3<br>(14-38)     | 32 ± 3<br>(26-42) | 6.8 ± 1.0<br>(5.6-12)      | 1.3 ±0.5<br>(1.2-1.5)                   | $206 \pm 11$<br>(149-263) | 44.5 ± 4.5<br>(19-73)     |
| Hestúr<br>n=11          | 18.5 ±1.3<br>(10-25)   | 33 ± 5<br>(19-67)          | 73 ± 3<br>(49-85)  | $\begin{array}{c} 0.11 \pm 0.01 \\ (0.08 \text{-} 0.13) \end{array}$ | $\begin{array}{c} 1.00 \pm 0.07 \\ (0.63 \text{-} 1.4) \end{array}$  | 51 ± 8<br>(31-122)    | ND                | $16.6 \pm 0.8 \\ (10-20)$  | $1.4 \pm 0.1$<br>(1.0-1.7)              | $154 \pm 23$<br>(87-293)  | $176 \pm 10$<br>(127-215) |
| S. Armot<br>n=12        | $31.7 \pm 0.8$<br>(26-36)  | 33 ± 1<br>(30-40)          | 80 ± 1<br>(76-83)  | $\begin{array}{c} 0.27 \pm 0.01 \\ (0.23 \text{-} 0.32) \end{array}$ | $\begin{array}{c} 2.2 \pm 0.1 \\ (1.8 \text{-} 2.9) \end{array}$     | 68 ±2<br>(58-76)      | ND                | $22.9 \pm 0.8$<br>(20-30)  | ND                                      | $349 \pm 43$<br>(165-509) | 117 ± 1<br>(114-119)      |
| Blom.<br>n=19           | $\begin{array}{c} 15.3 \pm 0.8 \\ (9.5\text{-}20.6) \end{array}$ | 8.1 ± 1.3<br>(3.5-24)      | 98 ± 6<br>(59-152) | $\begin{array}{c} 1.25 \pm 0.07 \\ (0.8\text{-}2.0) \end{array}$     | $\begin{array}{c} 0.56 \pm 0.04 \\ (0.36\text{-}1.1) \end{array}$    | 23 ± 1<br>(14-35)     | 46 ± 3<br>(28-66) | $10.8 \pm 0.5$<br>(7-15)   | $0.9 \pm 0.3$<br>(0.7-1.3) <sup>a</sup> | 192 ± 6<br>(148-255)      | $126 \pm 13$<br>(81-161)  |
| Blom.<br>1995<br>n = 21 | $16.0 \pm 1.8$<br>(7.1-34)                                       | $6.9 \pm 0.9$<br>(2.0-20)  | 72 ±6<br>(21-150)  | 1.30 ±0.08<br>(0.9-2.2)  | $\begin{array}{c} 0.44 \pm 0.03 \\ (0.16 \text{-} 0.89) \end{array}$ | 23 ± 2<br>(9-42)      | 41 ±3<br>(21-67)  | 9.5 ± 0.5<br>(4-13)        |   | 112 ±7<br>(47-175)        | 61.2 ±8.6<br>(38-194)     |
| Ribe<br>n=12            | 25.3 ±7.7<br>(9.8-104)   | $1.2 \pm 0.1$<br>(1.0-1.7) | 42 ±14<br>(15-186) | $\begin{array}{c} 1.49 \pm 0.15 \\ (0.7 \text{-} 2.1) \end{array}$   | $\begin{array}{c} 0.23 \pm 0.01 \\ (0.16 \text{-} 0.30) \end{array}$ | 9.8 ± 3.1<br>(5.4-43) | 36 ± 2<br>(29-44) | $2.5 \pm 0.2$<br>(1.5-3.0) |   | 47 ± 5<br>(35-56)         | 18.0 ±1.3<br>(12-27)      |
| Ribe 1995<br>n=2        | 23 ±0.6  | 1.6<br>±0.005              | 47 ±4              | 2.2 ±0.04  | 0.40<br>±0.005   | 6.8<br>±0.006         | ND                | 1.8 ±0.2                   | 0.5                                     | 98                        | 33 ± 1                    |
| Faroe Is.<br>n=8        | 10.1<br>(6.2-18.3)   | 23.0<br>(9.2-35)           | 169<br>(69-449)    | 0.20<br>(0.07-0.36)  | 0.76<br>(0.52-1.2)   |                       | ND                | 13.6<br>(6.9-17.7)         | 3.3<br>(1.1-5.7)                        | 138<br>(75-254)           | 79<br>(40-130)            |

Table 1. Trace Element Concentrations ( $\mu g/g$ ) in Whole Soil Samples (NAA analysis). Mean  $\pm$  SEM (range). ND = Not detected

% Mobile

Sorption of Cs-134 to soils



Figure 2. Sorption of Cs-134 tracer to Nordic soils as a function of time after addition. Mobile fraction (%) is the sum of water and  $NH_4Ac$  extractable Cs-134.



Figure 3. Sorption of Sr-85 tracer to Nordic soils as a function of time after addition. Mobile fraction (%) is the sum of water and  $NH_4Ac$  extractable Sr-85

Results indicate clear differences in the degree of tracer sorption and the rate of fixation in soils. After 200 days contact time, <sup>134</sup>Cs showed the greatest mobility in Blomhöjden and Hestúr soils (30 % NH<sub>4</sub>Ac extractable) and the lowest mobility in Tjøtta -- less than 2 % extractable by NH<sub>4</sub>Ac (Table 2). For <sup>85</sup>Sr, Tjøtta soils showed rather low extractability into NH<sub>4</sub>Ac, about 60-70 %, the other soils giving between 80 and 100 % extractability into NH<sub>4</sub>Ac (Fig. 3). There was little indication of a correlation between Cs-134 mobility and soil characteristics, probably reflecting the complex relationship between mineral and clay type, competing cations and organic content on Cs binding in soils.

|          | pН                 | LOI  | CEC  | Base sat. | MF   |
|----------|--------------------|------|------|-----------|------|
|          | (H <sub>2</sub> O) | (%)  |      | (%)       | (%)  |
| Tjøtta 1 | 6.6                | 21.1 | 29.9 | 100       | <50% |
| Tjøtta 2 | 5.2                | 61.4 | 88.7 | 56.5      | <50% |
| Tjøtta 5 | 5.4                | 77.5 | 87.9 | 59.1      | 1%   |
| Hestúr 1 | 6.0                | 35.5 | 34.4 | 48.9      | 30%  |
| Hestúr 2 | 5.3                | 44.4 | 41.2 | 31.9      | 18%  |
| Ribe 1   | 4.9                | 10.5 | 18.4 | 22.0      | 8%   |
| Ribe 2   | 5.4                | 7.3  | 15.4 | 38.1      | 4%   |
| Blom 3   | 5.5                | 44.4 | 49.0 | 46.9      | 27%  |
| Blom 4a  | 5.9                | 28.7 | 23.1 | 49.5      | 26%  |
| Blom 5   | 4.5                | 6.3  | 17.0 | 12.7      | 5%   |
| Hvalvik  | 4.5                | 71.3 |      |           | 23%  |
| Hvalba   | 5.1                | 54.2 |      |           | 14%  |
| Sumba    | 5.2                | 65.1 |      |           | 29%  |
| St.Armot | 5.7                | 41.2 |      |           | 16%  |
| St.Armot | 6.1                | 33.2 |      |           | 19%  |

Table 2. Soil characteristics and Cs-134 mobility at 200 days contact time. MF: Mobile Fraction of 137Cs (NH<sub>4</sub>Ac extractable) after 400 days contact time.

Results of sequential extraction studies confirmed that <sup>134</sup>Cs was both rapidly and strongly adsorbed to all soils (Fig. 1.6.4). Comparisons of the tracer distribution in Tjøtta soils with that observed for fallout <sup>137</sup>Cs and <sup>90</sup>Sr, as well as stable analogues, indicate that the tracer studies are giving a relatively good picture of environmental processes (Figs 4 and 5). The higher percentages found in residue for the fallout radionuclides undoubtedly reflect the longer contact time and diffusion of Cs into the clay mineral lattice. Strontium isotopes showed similar distribution throughout the extraction fractions, highlighting that fixation and/or soil sorption processes are unlikely to have a strong influence on <sup>90</sup>Sr transfer and ecological half-lives in ecosystems.

### 2.3 Soil fixation rates: modelling

Data shows clearly that cesium fixation in soil does not follow simple first order kinetics: half-lives increase with contact time. Sorption and desorption studies indicated that sorption of both the tracers could be described by a two stage process, probably reflecting a rapid adsorption to exchangeable sites  $- T_{1/2}$  from minutes to hours, and a slower removal to less available binding sites  $- T_{1/2}$  from

days to months (Fig. 6).  $K_{dl}$  represents the distribution between water and labile compartments, and is used instead of  $K_d$  because  $K_d$  changes with time whereas  $K_{dl}$ is rather constant. The decrease in activity in water and NH<sub>4</sub>Ac was modelled using a simple box model (SB Modelmaker, 1994). The rate constants for longterm Cs fixation ranged from <0.0001 to 0.0006 d<sup>-1</sup> (Table 3) which is equivalent to ecological half-lives of between 3 and >15 years, and within the range seen for Cs-137 at the sites. Differences between laboratory and environmental data for the individual sites probably reflect contact time, run-off and removal in the environment and possible temperature and climatic effects. Nevertheless, results indicate that fixation undoubtedly plays a significant role in determining ecological half-lives.



# Cs-Isotopes in Tjøtta soil, 1995

*Figure 4. Sequential extraction of Cs-isotopes in Tjøtta soil, 1995.* <sup>134</sup>*Cs contact time 1 month;* <sup>137</sup>*Cs contact time, 9 years.* 



Sr-Isotopes in Tjøtta soil, 1995



<sup>90</sup>Sr contact time 9 years.

Figure 6. Simple box model for radiocesium sorption to soils.

| Site          | T <sup>1</sup> /2 (y) |
|---------------|-----------------------|
| Tjøtta        | > 15                  |
| Blomhöjden    | $9.4 \pm 1.7$         |
| Hestúr        | 7.1 ± 1.4             |
| Ribe          | $3.2\pm0.9$           |
| Faroe Islands | $4.8 \pm 2.1$         |
| Iceland II    | > 10                  |

Table 3. Long-term fixation rates for removal of Cs-134 tracer to «irreversibly» bound sites in Nordic soils.

### 2.4 Stable Cs Transfer in Soil-Plant-Sheep System

Results of trace element analysis of vegetation and lamb meat samples are given Tables 4 and 5. Concentration levels of trace elements in vegetation were generally less variable than in soils. Levels of Sc in vegetation were used to correct for soil contamination (Oughton and Day, 1993). Cs data was used to calculate soil-toplant and plant-to-lamb concentration ratios (Table 6). Plant-to-sheep concentration ratios were similar to averages presented for <sup>137</sup>Cs. Thus it seems reasonable to conclude that <sup>137</sup>Cs is more or less in equilibrium with stable Cs as far as plant to sheep transfer is concerned. Faroe Island sheep showed low plant-tomeat concentration ratios for both <sup>137</sup>Cs and stable Cs at certain sites (Table 7). Some Faroe Island vegetation had relatively high Sc concentrations (Table 4), up to 0.23 ug/g. From Sc concentrations in soil (Table 2), this is equivalent to 4 - 16 mg soil/g vegetation, which does not result in a significant correction for stable Cs levels in vegetation. Soil-to-plant concentration ratios for stable Cs showed good correlation with those observed for <sup>137</sup>Cs, confirming the stable Cs analysis can give useful information concerning the potential vulnerability of systems to radiocesium fallout due to high transfer factors.

|                       | Ce   | Со   | Cr             | Cs   | Eu pg/g                    | Fe                        | Rb                    | Sc  | Se                  | Sr                | Zn                     |
|-----------------------|--|--|----------------|--|----------------------------|---------------------------|-----------------------|---|---------------------|-------------------|------------------------|
| Tjøtta<br>n=8         | ND   | $0.12 \pm 0.06$  | ND             | $0.34 \pm 0.04$  | ND                         | 67 ± 11                   | 26 ± 6                | $0.004 \pm 0.001$   | ND                  | 26 ± 9            | 90 ± 13                |
| Hestúr<br>n=10        | $\begin{array}{c} 0.16 \pm 0.02 \\ (0.08 \text{-} 0.24) \end{array}$ | $\begin{array}{c} 0.7 \pm 0.1 \\ (0.5\text{-}1.4) \end{array}$       | ND             | $\begin{array}{c} 0.13 \pm 0.01 \\ (0.09 \text{-} 0.22) \end{array}$ | $5.9 \pm 0.7$<br>(2.6-9.3) | $412 \pm 63$<br>(131-744) | $11 \pm 2$<br>(4-20)  | $\begin{array}{c} 0.09 \pm 0.01 \\ (0.04 \text{-} 0.17) \end{array}$    | ND                  | 27 ± 1<br>(23-32) | 23 ± 3<br>(14-45)      |
| S.Ármót               |  |  |                |  |                            |                           |                       |   |                     |                   |                        |
| Blom.<br>1994<br>n=24 | ND   | $\begin{array}{c} 0.16 \pm 0.03 \\ (0.03 \text{-} 0.2) \end{array}$  | ND             | $\begin{array}{c} 0.13 \pm 0.02 \\ (0.03 \text{-} 0.57) \end{array}$ | ND                         | 48 ± 3<br>(30-97)         | 38 ± 3<br>(15-<br>66) | $\begin{array}{c} 0.006 \pm 0.001 \\ (0.002 \text{-} 0.07) \end{array}$ | ND                  | 23 ± 2<br>(10-48) | 83 ± 20<br>(19-351)    |
| Blom.<br>1995<br>n=5  | ND   | 0.11 ±0.05   | ND             | 0.13 ±0.04   | ND                         | 56 ± 10                   | 46 ±5                 | $0.007 \pm 0.002$   | ND                  | 17 ± 4            | 40 ± 9                 |
| Blom.<br>1996<br>n=9  | 0.078 ± 0.015  | $0.22 \pm 0.05$  | 0.49 ± 0.07    | $0.17\pm0.05$  | ND                         | 92 ± 13                   |                       | $0.016 \pm 0.05$  | ND                  | 17 ± 1            | 51 ± 7                 |
| Ribe<br>n=3           | $\begin{array}{c} 0.06 \pm 0.03 \\ (0.01 \text{-} 0.08) \end{array}$ | $\begin{array}{c} 0.19 \pm 0.03 \\ (0.15 \text{-} 0.26) \end{array}$ | 5.5<br>(1-15)  | 0.011±0.003<br>(0.004-0.017)   | $1.9 \pm 0.2$<br>(1.6-2)   |                           | 2.2 ±<br>0.3          | $\begin{array}{c} 0.12 \pm 0.004 \\ (0.04 \text{-} 0.16) \end{array}$   | ND                  |                   | 7.7 ± 1.2<br>(5.3-9.1) |
| Faroe Is.<br>Mean     | 0.29<br>(0.09-0.62)  | 0.32<br>(0.13-0.73)  | 2.4<br>(0.6-8) | 0.026<br>(0.004-0.069)   | 19<br>(3-24)               |                           | 4.6<br>(2-9)          | 0.12<br>(0.06-0.23)   | 0.34<br>(0.16-0.38) | 28.9<br>(19-34)   | 28.1<br>(18-44)        |

*Table 4. Trace Element Concentrations (\mu g/g) in Vegetation (NAA analysis). Mean*  $\pm$  *SEM (range). ND* = *Not Detected.* 

|                           | Co<br>(ng/g)             | Cr  | Cs  | Fe                  | Rb                         | Sc (ng/g)  | Se | Sr                     | Zn                    |
|---------------------------|--------------------------|---|---|---------------------|----------------------------|--|----|------------------------|-----------------------|
| Tjøtta, 1996<br>n=11      | 8.5 ± 1.4<br>(ND-20)     | 0.79 ± 0.07<br>(ND-1.4)                           | $\begin{array}{c} 0.32 \pm 0.03 \\ (0.12 \text{-} 0.69) \end{array}$          | 22 ± 3<br>(10-62)   | ND                         | 1.9 ± 0.3<br>(ND-5)  | ND | ND                     | 29 ± 2<br>(16-49)     |
| Hestúr, 1994<br>n=12      | $3.6 \pm 0.6$<br>(2-4.4) | ND  | 0.094 ±<br>0.006<br>(0.06-0.13)   | 11 ± 0.6<br>(9-16)  | $5.4 \pm 0.6$<br>(3.7-6.5) | $\begin{array}{c} 0.37 \pm 0.08 \\ (\text{ND-}0.9) \end{array}$  | ND | 1.2 ± 0.8<br>(ND- 2.0) | $30 \pm 2$<br>(19-43) |
| Stóra Ármót,<br>1995 n= 6 | 2.4±0.2<br>(1.8-3.4)     | $0.14 \pm 0.05$<br>(ND-0.39)                      | 0.0097<br>±0.0008<br>(0.23-0.32)  | 9.9 ± 0.4<br>(8-11) | ND                         | 0.16   | ND | ND                     | $32 \pm 2$<br>(23-40) |
| Blom. 1994,<br>n=7        | 5.6 ± 3.5<br>(1-60)      | 0.1 ± 0.01<br>(ND-1.5)                            | $0.092 \pm 0.005 \\ 2.1$  | 5.9±0.4<br>(4-9)    | 9.4±0.5<br>(7-13)          | 0.5  | ND | ND                     | 24 ± 1<br>(17-28)     |
| Blom. 1995<br>n = 13      | 4.7 ± 0.8<br>(1-9)       | 0.1 ± 0.01<br>(ND-0.16)                           | $\begin{array}{c} 0.16 \pm 0.02 \\ (0.9\text{-}2.2) \end{array}$              | 8 ±0.8<br>(3.5-12)  | ND                         | ND   | ND | ND                     | 34 ± 4<br>(15-54)     |
| Ribe, 1995<br>n=6         | 62 ±4<br>(51-77)         | 42 ±14<br>(15-186)                                | $\begin{array}{c} 0.0040 \pm \\ 0.0008 \\ (0.003 \text{-} 0.008) \end{array}$ |                     | $2.2 \pm 0.2 \\ (1.6-3.3)$ | $\begin{array}{c} 0.18 \pm 0.02 \\ (\text{ND-}0.24) \end{array}$ | ND | 1.6 ± 0.5              | 29 ± 1 (25-32)        |
| Faroe Is.<br>1994         | 3.3 ± 2.0                | $\begin{array}{c} 0.030 \pm \\ 0.012 \end{array}$ | $\begin{array}{c} 0.0085 \pm \\ 0.0044 \end{array}$                           | NA                  | NA                         | $0.42 \pm 0.21$  | NA | NA                     | 40 ± 3                |
| Faroe Is<br>1995          | 3.0 ± 1.2                | 0.62 ± 0.69                                       | 0.0058 ± 0.0041   | NA                  | NA                         | 0.48 ± 0.29  | NA | NA                     | 42 ± 5                |

Table 5. Trace Element Concentrations ( $\mu$ g/g, FW) in Lamb Muscle Samples (NAA analysis, ashed samples). Mean  $\pm$  SEM (range).

ND - Not Detected; NA - Not Analysed

|                   | Soil<br>DW   | Plant<br>DW  | Sheep<br>FW                     | CR Cs-133<br>soil-veg | CR Cs-133<br>veg-sheep | CR Cs-137<br>soil-veg | CR Cs-137<br>veg-sheep |
|-------------------|--|--|---------------------------------|-----------------------|------------------------|-----------------------|------------------------|
| Hestúr<br>1994    | $\begin{array}{c} 0.107 \pm 0.007 \\ (0.08 - 0.13) \end{array}$      | $\begin{array}{c} 0.127 \pm 0.015 \\ (0.084 - 0.22) \end{array}$         | $0.069 \pm 0.004$               | 1.19 ± 0.23           | 0.54 ± 0.09            | 1.04                  | 0.48                   |
| S.Ármót<br>1995   | $\begin{array}{c} 0.22 \pm 0.03 \\ (0.23 \text{-} 0.32) \end{array}$ | $\begin{array}{c} 0.017 \pm 0.003 \\ (0.012 \text{-} 0.027) \end{array}$ | $0.0097 \pm 0.0004$             | $0.077 \pm 0.018$     | 0.57 ± 0.11            | 0.069                 | -                      |
| Blom.<br>1994     | $\begin{array}{c} 1.25 \pm 0.07 \\ (0.83 - 2.0) \end{array}$         | $\begin{array}{c} 0.107 \pm 0.015 \\ (0.038 - 0.29) \end{array}$         | $0.092 \pm 0.012$<br>(0.48 ewe) | $0.086\pm0.017$       | $0.85 \pm 0.23$        | 0.23                  | 0.82                   |
| Blom.<br>1995     | 1.30 ± 0.08  | 0.13 ± 0.04  | 0.16 ± 0.02                     | $0.10 \pm 0.03$       | $1.2 \pm 0.8$          | 0.21                  | 0.88                   |
| Blom<br>1996      |  |  | 0.17 ± 0.05                     |                       |                        |                       | 0.82                   |
| Tjøtta 1995       | 1.31 ± 0.16  | $0.34 \pm 0.04$  | $0.32 \pm 0.03$                 | $0.25\pm0.05$         | $0.94 \pm 0.22$        | 0.36                  | 1.4                    |
| Faroe Is.<br>1994 | 0.20   | $0.017 \pm 0.004$  | $0.0085 \pm 0.0044$             | $0.085 \pm 0.015$     | $0.50 \pm 0.31$        | 0.18                  | 0.50                   |
| Faroe Is.<br>1995 | 0.20   | $0.026 \pm 0.008$  | $0.0058 \pm 0.0041$             | 0.13 ± 0.04           | 0.22 ± 0.39            | 0.18                  | 0.22                   |
| Ribe<br>1995      | 1.49 ± 0.15  | 0.0049   | 0.0040                          | 0.033                 | 0.81                   | 0.09                  | 0.89                   |

Table 6. Concentrations and concentration ratios (CR) of naturally-occurring stable cesium in soil-plant-sheep system. Mean $\pm SEM$  (Range). Units:  $\mu g/g$ .

| Site    | Stable Cs concentration ug/g |             |                 |                 | Stable Cs      |                      | Cs-137               |                |                     |                     |
|---------|------------------------------|-------------|-----------------|-----------------|----------------|----------------------|----------------------|----------------|---------------------|---------------------|
|         | Soil<br>DW                   | Plant<br>DW | Lamb 1994<br>FW | Lamb 1995<br>FW | CR<br>soil-veg | CR 1994<br>veg-sheep | CR 1995<br>veg-sheep | CR<br>soil-veg | CR 1994<br>veg-lamb | CR 1995<br>veg-lamb |
| Velb.   | 0.10                         | 0.069       | 0.0064          | 0.0024          | 0.60           | 0.093                | 0.035                | 0.104          | 0.48                | 0.11                |
| Skåli   | 0.22                         | 0.025       |                 | 0.0092          | 0.11           |                      | 0.37                 | 0.19           | 0.59                | 0.41                |
| Hvalik  | 0.17                         | 0.059       | 0.018           | 0.015           | 0.37           | 0.30                 | 0.25                 | 0.65           | 0.39                | 0.22                |
| Bøur    | 0.07                         | 0.0042      | 0.0040          | 0.0042          | 0.06           | 0.23                 | 1.0                  | 0.06           | 0.49                | 0.27                |
| Hvalba  | 0.18                         | 0.0063      | 0.0047          | 0.0025          | 0.035          | 0.74                 | 0.40                 | 0.055          | 0.26                | 0.14                |
| Nordov. | 0.23                         | 0.017       | 0.0091          | 0.0045          | 0.074          | 0.53                 | 0.26                 | 0.20           | 0.72                | 0.30                |
| Sandar  | 0.26                         | 0.0093      | 0.0107          | 0.0056          | 0.036          | 2.0                  | 0.60                 | 0.075          | 3.3                 | 0.70                |
| Sumba   | 0.36                         | 0.021       | 0.0069          | 0.0029          | 0.058          | 0.33                 | 0.14                 | 0.13           | 0.25                | 0.15                |

Table 7. Faroe Island: Soil, vegetation, lamb and concentration ratios for separate sites

# 3. Conclusions and Input to Model

- Laboratory studies show that the mobility of <sup>134</sup>Cs tracers added to soils decreases with time after addition. Sequential extraction studies show that radiocesium is rapidly and strongly bound to soils. Use of a simple box model provides estimates of the long-term fixation rates of  $<0.0001 0.00059 d^{-1}$ , equivalent to half-times of 3 to > 15 years. Rate constants for other sorption processes (e.g. ion-exchange and sorption to reversibly bound sites) are faster, with half-lives from hours to days
- Fixation rates are of the same order of magnitude as ecological half-lives in soil-plant-sheep system, adding support to the assumption that radionuclide fixation in minerals will have a strong influence on ecological half-lives.
- Decreases in the mobility of <sup>134</sup>Cs tracers does not follow simple: sorption and fixation rates decrease with time. This both supports and stresses the importance of observations that ecological half-lives need not be constant as a function of time. Of course, for modelling purposes, fixation rates are just one of the rate determining parameters that can influence ecological half-lives (transport down the soil profile and removal with produce and run-off are almost certainly also important).
- Tracer studies suggest that the fraction of mobile Cs and Sr in soils can show considerable variation between the different sites, especially in the first 100 days after contact time. There was evidence that soils with reduced organic content often showed reduced mobility and faster fixation rates (e.g. Ribe).
- As Sr is not fixed in these soils, the rate-determining parameters for ecological half-lives would be expected to be transport down the soil profile and removal with produce and run-off.
- Analysis of stable Cs shows a large variation in trace element concentrations and soil-to-plant concentration ratios between the different sites. However, observed concentration ratios for stable Cs showed good correlation with <sup>137</sup>Cs concentration ratios, suggesting that differences between the locations were due to site-specific factors rather than source term or lack of equilibrium. Stable Cs analysis can give some indication of the potential vulnerability of ecosystems due to high transfer factors.
- Plant-to sheep concentration ratios are similar, apart from the Faroe Islands. Although some Faroe Island vegetation samples showed evidence of higher levels of soil contamination as compared to other sites, up to 16 mg soil per g

vegetation d.w., preliminary calculations suggest that this was not large enough to have a significant influence on stable Cs levels in vegetation

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### **APPENDIX - SOIL ANALYSIS**

|          | pН                 | LOI  | Na  | Mg  | Ca   | K    | $H^+$ | CEC  | Base sat. |
|----------|--------------------|------|-----|-----|------|------|-------|------|-----------|
|          | (H <sub>2</sub> O) | (%)  |     |     |      |      |       |      | (%)       |
| Tjøtta 1 | 6.6                | 21.1 | 0.5 | 1.0 | 28.2 | 0.2  | 0     | 29.9 | 100       |
| Tjøtta 2 | 5.2                | 61.4 | 1.5 | 5.3 | 42.1 | 1.1  | 38.6  | 88.7 | 56.5      |
| Tjøtta 5 | 5.4                | 77.5 | 1.4 | 6.9 | 42.0 | 1.5  | 35.9  | 87.9 | 59.1      |
| Hestúr 1 | 6.0                | 35.5 | 1.1 | 7.5 | 7.0  | 1.1  | 17.6  | 34.4 | 48.9      |
| Hestúr 2 | 5.3                | 44.4 | 1.1 | 4.5 | 5.8  | 1.6  | 28.1  | 41.2 | 31.9      |
| Ribe 1   | 4.9                | 10.5 | 0.1 | 1.2 | 1.0  | 1.7  | 14.3  | 18.4 | 22.0      |
| Ribe 2   | 5.4                | 7.3  | 0.9 | 0.6 | 1.8  | 2.6  | 9.5   | 15.4 | 38.1      |
| Blom 3   | 5.5                | 44.4 | 0.4 | 2.1 | 5.5  | 15.0 | 26.0  | 49.0 | 46.9      |
| Blom 4a  | 5.9                | 28.7 | 0.2 | 1.2 | 2.2  | 7.8  | 11.7  | 23.1 | 49.5      |
| Blom 5   | 4.5                | 6.3  | 0.0 | 0.3 | 0.4  | 1.4  | 14.8  | 17.0 | 12.7      |
| Hvalvik  | 4.5                | 71.3 |     |     |      |      |       |      |           |
| Hvalba   | 5.1                | 34.2 |     |     |      |      |       |      |           |
| Sumba    | 5.2                | 52.1 |     |     |      |      |       |      |           |
| St.Armot | 5.7                | 41.2 |     |     |      |      |       |      |           |
| St.Armot | 6.1                | 33.2 |     |     |      |      |       |      |           |

Table A1: Soil Characteristics (Tracer Studies).

|               | pH water | pН                | LOI  | CEC      | Base sat % |
|---------------|----------|-------------------|------|----------|------------|
|               |          | CaCl <sub>2</sub> | %    | meq/100g |            |
| Tjøtta 1*     | 6.6      | -                 | 21.1 | 29.9     | 100        |
| Tjøtta 2*     | 5.2      | -                 | 61.4 | 88.7     | 56.5       |
| Tjøtta 5*     | 5.4      | -                 | 77.5 | 87.9     | 59.1       |
| Tjøtta 1      | 6.5      | -                 | 6.7  |          |            |
| Tjøtta 2      | 6.7      | -                 | 16.5 |          |            |
| Tjøtta 3      | 5.3      | -                 | 18.1 |          |            |
| Tjøtta 4      | 5.1      |                   | 29.4 |          |            |
| Tjøtta 5      | 5.7      |                   | 49.4 |          |            |
| Tjøtta 6      | 5.6      |                   | 90.9 |          |            |
| Hestúr 1      | 6.0      | 5.3               | 35.5 | 34.4     | 48.9       |
| Hestúr 2      | 5.3      | 4.8               | 44.4 | 41.2     | 31.8       |
| Blomhöjden 1  | 4.7      | -                 | 26.6 |          |            |
| Blomhöjden 2  | 5.5      | -                 | 10.8 |          |            |
| Blomhöjden 3  | 5.5      | -                 | 44.4 |          |            |
| Blomhöjden 4a | 5.9      | -                 | 28.7 |          |            |
| Blomhöjden 4b | 5.4      | -                 | 29.6 |          |            |
| Blomhöjden 4c | 4.3      | -                 | 37.7 |          |            |
| Blomhöjden 5  | 4.5      | -                 | 6.3  |          |            |
| Ribe 1        | 4.9      | -                 | 10.5 |          |            |
| Ribe 2        | 4.9      | -                 | 10.5 |          |            |
| Ribe 3        | 4.7      | -                 | 8.6  |          |            |
| Ribe 4        | 5.5      | -                 | 11.7 |          |            |
| Ribe 5        | 5.4      | -                 | 7.3  |          |            |
| Ribe 6        | 5.8      | -                 | 7.2  |          |            |

Table A2. LOI and pH (Other Soils)

\* 1993 samples
# **REPORT 4** Soil Ingestion - Scandium analysis

Deborah Oughton Laboratory for Analytical Chemistry, Agricultural University of Norway

# 1. Introduction

Soil ingestion is important for two main reasons: first, soil can act as a source of radionuclides to grazing animals; second, non-documented soil contamination of samples can lead to errors and uncertainties with respect to modelling the transfer of radionuclides in an ecosystem. Although radionuclides associated with soils are usually of low bioavailability for gut uptake, the relatively high levels in soils as compared to plants can lead to large variations in transfer factors. Soil contamination of vegetation and/or faeces by soil may lead to a underestimation of plant-to-animal transfer factors. Soil contamination of vegetation can also lead to overestimation of radionuclide root uptake, and soil-to-plant transfer factors.

Plant to animal concentration ratios are usually based on measurements of composite vegetation samples from the grazing area. Errors can arise if the collected sample does not reflect the animals' diet, either because of selective grazing of the animals -- different plant species often show wide variation in trace element and radionuclide concentration levels, because of soil contamination of the vegetation samples, or because the animals are ingesting large amounts of soil. For this reason, determining the level of radionuclides in the diet of grazing animals. From analysis of faeces one can both determine whether or not the animal has been ingesting soil, and by checking for deviations between the levels of trace elements and radionuclides in faeces from levels seen in the vegetation sources, provide information on the reliability of sheep diet estimates from vegetation samples.

Previous work has shown that neutron activation analysis (NAA) is a sensitive method for determining trace elements, including stable Cs, in a variety of samples and that the method also allows for a sensitive calculation of soil contamination in vegetation samples by measurement of the Sc content (Oughton and Day, 1993). Scandium is strongly bound within soil minerals, and soil to plant concentration ratios are of the order of 10,000.

To test the method, and provide data on soil ingestion for selected sites, NAA was carried out on a number of soil, vegetation and faeces samples from Blomhöjden and Ribe. Samples were dried or ashed, homogenised and weighed into small (0.5

ml) polyethylene vials. The vials were heat sealed and irradiated together with matrix matched standards. Activation products were counted using Ge-detectors.

In order to calculate the weight of soil per gram faeces one needs to assume that the animal is at equilibrium with its diet. Then the mass of soil per gram faeces can be calculated using the following equation:

mg soil/g faeces (dry weight) = {  $[Sc_{faeces}]_{calc} - [Sc_{faeces}]_{measured} } / [Sc_{soil}]$ 

 $[Sc_{faeces}]_{calc} \mbox{-} is the calculated concentration of Sc in the faeces based on the} \label{eq:scalc}$ 

concentration in vegetation

$$= [Sc_{veg}] * D * E_f$$

D = digestion factor, estimated at 2.5, which is equivalent to 60 % digestion of fodder.

 $E_f$  = fraction of total Sc excreted via the faeces. This is taken to be 1 for Sc, equivalent to 100 % excretion via the faeces.

The same equation can be used to compare calculated and measured concentrations of other trace elements in faeces. And for other elements having high soil-to-plant concentration ratios (e.g. Cr and Fe) an additional estimate of soil ingestion can be made. In this case, for all elements except for Cs,  $E_f = 1$  has been used. For Cs, an excretion partitioning of 2:1 for faces:urine has been assumed.

### 2. Results

Scandium is easily detected in the faeces samples, and the method is extremely sensitive. Results indicate that soil can be detected at levels of about 5-10 mg soil per g faeces/g which, assuming 60 % digestibility, is equivalent to about 2-4 g soil / kg vegetation, dry weight. This level is well below that which would expect to influence transfer factors or concentration ratios for radiocesium. In 1994, Blomhöjden faeces showed a *maximum* soil ingestion of about  $20 \pm 5$  mg soil/g faeces. In 1995, the levels were more variable, with most animals giving levels of less than 5 mg soil /g faeces, but one animal showing levels of between 60 and 135 mg/g. Ribe sheep showed very little evidence of soil ingestion. In fact sheep had a lower ingestion of soil than measured in the majority of vegetation samples. Two of the three Ribe vegetation samples analysed had high Sc concentrations, so calculations have been carried out with the one «clean» sample.

In Blomhöjden sheep, calculated and "expected" concentrations in faeces concentration differ for elements other than Sc/Fe/Cr. This can reflect a number of

factors including, non-equilibrium status of the animals and dietary sources not reflected by the sampled vegetation. Overestimation of digestibility (60 % is quite high) will result in the measured concentrations in faeces being *lower than expected* for all elements, as will underestimation of the fraction excreted in faeces for individual elements. For Blömhöjden sheep, the concentrations of elements in faeces are higher than expected -- even after correction for soil ingestion. For Cs, Sr and Zn an "extra intake" of *at least* between 50-100 % is needed; for Co calculated levels are "down" by 400%. Analysis of individual vegetation species from Blomhöjden suggests that this is not impossible.

NAA gives very promising results as a sensitive method for assessment of soil ingestion. The above calculations make a lot of assumptions, and hence there is room for both error and improvement (e.g., dry weight digestibility of the vegetation; faeces to urine excretion ratios, equilibrium status). Nevertheless, data is robust enough to give a rough estimate of soil ingestion and to provide evidence that sampled vegetation does not necessarily reflect the sheep diet.

| J                     |            |            |                  |             | DI       | 0            | a           | -           |
|-----------------------|------------|------------|------------------|-------------|----------|--------------|-------------|-------------|
|                       | Со         | Cr         | Cs               | Fe          | Rb       | Sc           | Sr          | Zn          |
|                       |            |            |                  |             |          |              |             |             |
| Soil                  | $8.7 \pm$  | $98\pm 6$  | $1.25 \pm$       | $22500 \pm$ | 46 ±     | $10.8\pm0.5$ | 185 ±       | $127 \pm 9$ |
| (n=19)                | 1.3        |            | 0.07             | 1400        | 3        |              | 12          |             |
| Veg                   | $0.16 \pm$ | N.D.       | 0.12 ±           | 47.7 ±      | 38 ±     | $0.006 \pm$  | $23 \pm 2$  | $83 \pm 20$ |
| (n=24)                | 0.03       |            | 0.02             | 3.3         | 3        | 0.001        |             |             |
| Faeces -              | 0.4        | 0          | 0.2 <sup>b</sup> | 120         | 95       | 0.015        | 58          | 200         |
| expected <sup>a</sup> |            |            |                  |             |          |              |             |             |
| Faeces -              | $2.7 \pm$  | 2.3 ±      | 0.39 ±           | 637 ±17     | $48 \pm$ | 0.17 ±       | $122 \pm 5$ | 330 ±       |
| measured              | 0.1        | 0.8        | 0.03             |             | 14       | 0.07         |             | 21          |
| (n=17)                |            |            |                  |             |          |              |             |             |
| mg soil/              | -          | $23 \pm 7$ | -                | $23 \pm 5$  | -        | $14 \pm 7$   | -           | -           |
| g faeces              |            |            |                  |             |          |              |             |             |

*Table 1. Trace element concentrations in faeces, vegetation and soil from Blomhöjden, Sweden, 1994 (ug/g \pm SEM)* 

<sup>a</sup> The amount in faeces calculated from vegetation concentration levels. This assumes that the animal is at equilibrium with respect to the stable element, that the dry weight digestibility of vegetation is 60%, and that 100% of the ingested element is excreted in faeces for all elements apart from <sup>b</sup>, Cs, where an excretion ratio of 2:1 faeces:urine has been used.

Table 2. Trace element concentrations in faeces, vegetation and soil fromBlomhöjden, Sweden, 1995 ( $ug/g \pm SEM$ )

|          | Co         | Cr              | Cs         | Fe          | Rb       | Sc             | Sr         | Zn          |
|----------|------------|-----------------|------------|-------------|----------|----------------|------------|-------------|
|          |            |                 |            |             |          |                |            |             |
| Soil     | $8.7 \pm$  | 98 ±6           | $1.25 \pm$ | $22500 \pm$ | $46 \pm$ | $10.8 \pm 0.5$ | $185 \pm$  | $127 \pm 9$ |
| (n=19)   | 1.3        |                 | 0.07       | 1400        | 3        |                | 12         |             |
| Veg      | $0.22 \pm$ | $0.5\pm0.02$    | 0.17 ±     | $92 \pm 2$  | N.D      | $0.016 \pm$    | $17 \pm 1$ | $51 \pm 7$  |
| (n=9)    | 0.05       |                 | 0.05       |             |          | 0.005          |            |             |
| Faeces - | 0.5        | 1.3             | 0.28       | 240         | N.D      | 0.044          | 42         | 127         |
| expected |            |                 |            |             |          |                |            |             |
| Faeces - | 1 ±        | $2 \pm 0.8$     | $0.8 \pm$  |             | N.D      | 0.04           | $96 \pm 5$ | 513 ±       |
| measured | 0.1        | 10 <sup>a</sup> | 0.03       |             |          | 0.7            |            | 14          |
| (n=22)   |            |                 |            |             |          |                |            |             |
| mg soil/ | -          | <17             | -          | <3          | -        | < 0.5          | -          | -           |
| g faeces |            | 89              |            | 135         |          | 65             |            |             |

a - one sample had high concentrations of Cr, Sc and Fe.

Table 3. Trace element concentrations in faeces, vegetation and soil from Ribe, Denmark 1995 ( $ug/g \pm SEM$ )

|          | Со    | Cr     | Cs     | Fe         | Rb   | Sc          | Sr         | Zn           |
|----------|-------|--------|--------|------------|------|-------------|------------|--------------|
|          |       |        |        |            |      |             |            |              |
| Soil     | 1.2 ± | 42 ±14 | 1.49 ± | $9800 \pm$ | 36 ± | $2.5\pm0.2$ | $47 \pm 5$ | $18 \pm 1.3$ |
| (n=12)   | 0.1   |        | 0.15   | 3100       | 2    |             |            |              |
| Veg      | 0.18  | 0.5    | 0.004  | 17         | 2.1  | 0.004       |            | 5.3          |
| (n=1)    |       |        |        |            |      |             |            |              |
| Faeces - | 0.44  | 1.25   | 0.011  | 44         | 5.3  | 0.01        | -          | 13.3         |
| expected |       |        | b      |            |      |             |            |              |
| Faeces - | 0.05  | 0.55   | 0.026  | 77         | 4.0  | 0.02        | 14.5       | 11.6         |
| measured |       |        |        |            |      |             |            |              |
| (n=3)    |       |        |        |            |      |             |            |              |
| mg soil/ | -     | -      | -      | 3-8        | -    | 4-8         | -          | -            |
| g faeces |       |        |        |            |      |             |            |              |

|                               | Co           | Cr     | Cs          | Fe             | Rb        | Sc        | Sr     | Zn       |
|-------------------------------|--------------|--------|-------------|----------------|-----------|-----------|--------|----------|
| Soil<br>(n=12)                | 1.6 ±<br>0.1 | 47 ±14 | 1.49 ± 0.15 | 9800 ±<br>3100 | 36 ±<br>2 | 2.5 ± 0.2 | 47 ± 5 | 18 ± 1.3 |
| Veg<br>(n=1)                  | 0.18         | 0.5    | 0.004       | 17             | 2.1       | 0.004     |        | 5.3      |
| Faeces -<br>expected          | 0.44         | 1.25   | 0.011       | 44             | 5.3       | 0.01      | -      | 13.3     |
| Faeces -<br>measured<br>(n=4) | 0.08         | 1.3    | 0.03        | 170            | N.D       | 0.05      | 8      |          |
| mg soil/<br>g faeces          | -            | < 10   | -           | ~20            | -         | ~20       | -      | -        |

Table 4. Trace element concentrations in faeces, vegetation and soil from Ribe, Denmark 1996 (ug/g  $\pm$  SEM)

## **REPORT 5** Transfer of Radiocesium from Soil to Lamb in Nordic Countries

Sven P. Nielsen<sup>1</sup> and Mette Øhlenschlæger<sup>2</sup>

- 1. Risø National Laboratory, DK-4000 Roskilde, Denmark
- 2. National Institute of Radiation Protection, Frederikssundsvej 378, DK-2700 Brønshøj, Denmark

#### ABSTRACT

Data on <sup>137</sup>Cs in soil, grass and lamb have been collected at four locations in Denmark, Norway, the Faroe Islands and Sweden for the period 1990-1996. The data have been analysed with a simple radioecological model in order to identify similarities and differences between transfer processes across the sites. The effective ecological half lives of <sup>137</sup>Cs in soil at the sites range from 10 y at the Danish site to 30 y at the Faroese site. The sites with low levels of Chernobyl fallout (Danish and Faroese sites) show low mobile fractions of <sup>137</sup>Cs in the soil (about 3%) while the other sites with higher levels of Chernobyl fallout show higher mobile fractions (about 20%). Improved consistency is found for the uptake of radiocesium in grass from peaty soils when the uptake is related to the mobile fractions of radiocesium in the soil rather than the total activity. The uptake in grass from mineral soil is found to be lower than that from peaty soils by a factor 10. The transfer of <sup>137</sup>Cs from grass to lamb is found to be similar at three sites with a meat transfer coefficient of about 0.8 d kg<sup>-1</sup>, but about a factor 3 lower at the Faroese site. Doses to individuals from consumption of <sup>137</sup>Cs in lamb from the study sites have been estimated for the 50-y period 1990-2040. For high-rate consumers the doses are estimated to range from 4 µSv to 3 mSv.

### 1. Introduction

Studies on the transfer of radiocesium through the soil-grass-lamb foodchain in semi-natural ecosystems have been carried out in the Nordic countries in a coordinated manner since 1990. The studies were carried out under the NKS (Nordic Committee for Nuclear Safety Research) programme, during 1990-1993 in the RAD-3 project, and during 1994-1997 in the EKO-2 project. These studies have included sites in the Faroe Islands, Norway, Sweden and Denmark and have provided data covering 1990 to 1996. The amount of Chernobyl fallout from 1986 varies significantly as do the soil characteristics across the sites. The data illustrate the long-term trends of the radiocesium transfer which are quite different between the locations. These trends are now well documented due to the continuation of the projects, which permit improved estimates to be made of the long-term radiological consequences. Furthermore, the data illustrate the similarities and differences across the sites with respect to the transfer processes. The objective of

this work is to carry out an analysis of the data obtained using a simple radioecological model in order to improve the understanding of the transfer processes involved and to permit reliable estimates to be made of the radiation doses to humans from consumption of lamb from Nordic environments contaminated with radiocesium.

## 2. Materials and methods

The results from the RAD-3 lamb project are reported by Hove et al. (1994) with a detailed description of sampling sites, soil characteristics and sampling procedures. Therefore, these are mentioned briefly here only. The field data used for the present work cover the period 1990-1996.



Figure 1. Data from the four study sites on <sup>137</sup>Cs in soil, grass and lamb showing annual mean values and associated standard errors of the means.

#### 2.1 Study Sites

The Danish data are from Vester Vedsted near Ribe in Jutland (55.3°N 8.7°E) and represent a grazing area of about 0.1 km<sup>2</sup>. The vegetation is permanent grassland on sandy soil at an altitude of 2 m above sea level. The Norwegian data are from Tjøtta located in Nordland (66.6°N 12.4°E) with a small grazing area of 0.004 km<sup>2</sup>. The vegetation is permanent grassland on peaty soil at an altitude of 10 m above sea level. The data from the Faroe Islands are collected from six islands (62.0°N 7.0°W) and represent a grazing area of about 30 km<sup>2</sup>. The vegetation is permanent grassland on peaty soil at altitudes of 50-240 m above sea level. The Swedish data

are from Blomhöjden in Jämtland (64.4°N 14.4°E) and represent a grazing area of 10 km<sup>2</sup>. The vegetation is mountain moor, *Betula* forest and permanent grassland on peat, gravelly sandy moraine at altitudes of 580-770 m above sea level.

### 2.2 Data Collection

Samples of soil, vegetation and lamb have been collected annually from the study sites and analysed for radiocesium content by gamma spectrometry. The soil samples were collected to a depth of 10 cm, dried, analysed and the results reported on an area basis. Vegetation was sampled, dried and the results reported on a dry weight basis. Samples of lamb were collected after slaughter and analysed either fresh or after ashing; the results were reported on a fresh weight basis. Recently, the laboratories producing these results participated in an intercomparison exercise on radionuclides in sediment samples (Nielsen, 1996). The analytical performance of the participants was evaluated for <sup>137</sup>Cs against a target standard deviation of 5%. All four laboratories met this performance criterion.

The data for the four sites are available as time series from 1990 to 1996 of annual mean concentrations of  $^{137}$ Cs in soil, grass and lamb including standard errors of the means as shown in Fig .1.

### 2.3 Model

A simple dynamic model was developed and used for the analysis of the data from the period 1990-1993 (Nielsen, 1994). The model incorporates an atmospheric compartment, a soil compartment, two grass compartments (one for the external plant and one for the internal plant), and two lamb compartments (one for the gastro-intestinal tract and one for the meat) representing a single animal. The atmosphere, soil and grass compartments are associated with a surface area of one square metre.

The model is based on first-order differential equations where the transfer of radiocesium between the model compartments is based on rate constants that determine the fractional transfer between the compartments for each time step. The model included a single surface soil compartment from which radiocesium was transferred partly to vegetation by root uptake and partly to deeper soil. The model structure was adjusted for the present work by including an additional soil compartment to be able to simulate two fractions of radiocesium in the surface soil, a mobile fraction and a fixed fraction. The modelling concept of separating radiocesium in the soil into two fractions is a step toward a more realistic simulation considering the well-known strong fixation of radiocesium to certain soil minerals. Greater detail can be incorporated into the model by including more than two fractions for radiocesium in surface soil, but it has been considered

important to keep the model simple without more detail than justified from the basic data. The simulated transfer of radiocesium to vegetation by root uptake is possible from the mobile fraction, only, but exchange between the two soil fractions is included. The schematic structure of the adjusted model is shown in Fig. 2 where the arrows indicate the transfers between the model compartments.

The model assumes an initial contamination of the surface soil from which a fraction  $(k_{21})$  is resuspended into the atmosphere compartment which is limited by an atmospheric mixing height of 1000 m. The simulation of the resuspension is based on a resuspension factor of  $10^{-9}$  m<sup>-1</sup>. From the atmospheric compartment the activity is transferred to the ground through the application of a generic deposition velocity of 0.023 m s<sup>-1</sup> representing both wet and dry processes. The deposition is split between a fraction of 30% (d<sub>2</sub>) intercepted by vegetation surfaces and the remainder (d<sub>1</sub>) which is transferred to the mobile soil compartment. From the mobile soil compartment the bioavailable activity is reduced through the application of a removal process (k<sub>20</sub>), e.g. run off or transfer to deeper soil layers. The model implementation of root uptake from soil to grass (k<sub>25</sub> and k<sub>52</sub>) is based on the concentration-ratio concept. The activity on external surfaces of the grass is transferred to the soil at a rate (k<sub>42</sub>) corresponding to a weathering half life of two weeks.

For simplicity, generic assumptions are made for annually occurring events. The growing season is assumed to start every year at day no. 100 and end at day no. 300. The lambs are assumed to be born at day no. 50, let out on pasture on day no. 120 and slaughtered at day no. 300. The model assumption of slaughtering the lambs at 8 months is at the high end compared with the field data which represent a range from 4 to 8 months.



Figure 2. Schematic model structure

The transfer of the activity in the lamb is simulated by two compartments only. The daily intake of radiocesium by the lamb is calculated from the current concentration in the grass and the amount eaten per day. This intake is supplied each day to the compartment of the gastro-intestinal tract from where there is a transfer through faecal and urine excretion ( $k_{68}$ ) and to the meat compartment ( $k_{67}$ ). From the meat compartment there is a transfer through other metabolic processes ( $k_{78}$ ) corresponding to a biological half life of three weeks. Faecal excretion is included at a rate of 70% of the total excretion, the remainder by urine excretion. The modelling of faecal excretion is included for the purpose of permitting additional field data to be compared with model predictions.

The daily intake of grass by the lamb is assumed to increase linearly with time from 0.5 kg d<sup>-1</sup> at an age of 3 months to 1.4 kg d<sup>-1</sup> at an age of 12 months (Haugsgaard Sørensen, 1990). This is expressed as I = 0.2 + 0.1 ·M, where I is the daily intake of grass in kg dry weight and M the age of the lamb in months. Correspondingly, the carcass weight of the lamb is assumed to increase linearly with time. From field data the following relation was obtained, where m<sub>f</sub> is the carcass weight in kg and M the age of the lamb in months at slaughter:  $m_f = 2.6 + 2.4 \cdot M$ .

The rate constants  $k_{67}$ ,  $k_{68}$  and  $k_{78}$  determine the dynamics of the transfer of radiocesium in the lamb. They are interrelated in the following way.

$$k_{67} = k_{68} \frac{F_f \cdot m_f \cdot k_{78}}{1 - F_f \cdot m_f \cdot k_{78}}$$

Here  $F_f$  is the meat-transfer coefficient (d kg<sup>-1</sup>) and m<sub>f</sub> the carcass weight.  $F_f$  is calculated by dividing the observed ratio between the radiocesium concentration in the lamb and grass (OR<sub>lamb/grass</sub>) with the daily intake. With this procedure, the transfer coefficient becomes dependent on the age of the animal with values that agree with observations (Galer et al. 1991). The rate constant k<sub>78</sub> determines the biological half life for radiocesium in the lamb. The rate constant k<sub>68</sub> is chosen arbitrarily at a value of 1 d<sup>-1</sup> to ensure a rapid turnover in the gastro-intestinal tract.

### 2.4 Long-term characteristics for radiocesium in soil and grass

The radiocesium activity in the soil is far greater than that in the grass and air, so for the purpose of the soil system we may neglect root uptake and resuspension. We define that the system has reached a steady state when there is a constant ratio between the activities in the two soil compartments ( $S_m$  and  $S_f$ ). This gives the following:

$$S_m = \alpha S_f \Rightarrow \frac{dS_m}{dt} = \alpha \frac{dS_f}{dt}$$
$$S_m = \frac{\alpha}{1+\alpha} S$$
$$\frac{dS_m}{dt} = -(\lambda_{fys} + k_{20} + k_{23})S_m + k_{32}S_f$$
$$\frac{dS_f}{dt} = -(\lambda_{fys} + k_{32})S_f + k_{23}S_m$$

By combining these equations we obtain an expression for the ratio  $\alpha$ :

$$\alpha = \frac{k_{32} - k_{20} - k_{23} + \sqrt{(k_{20} + k_{23} - k_{32})^2 + 4k_{23}k_{32}}}{2k_{23}}$$

The total activity S in the two soil compartments is reduced by physical decay and runoff, and by introducing the ratio  $\alpha$  into the differential equation for S we obtain:

$$\frac{dS}{dt} = -\lambda_{fys}S - k_{20}S_m = -\lambda_{fys}S - k_{20}\frac{\alpha}{1+\alpha}S = -(\lambda_{fys} + k_{20}\frac{\alpha}{1+\alpha})S$$

This gives the following expressions for the effective half life and corresponding rate constant for radiocesium in the soil:

$$T_{v_{2,eff}} = \frac{\ln 2}{k_{eff}}$$
$$k_{eff} = \lambda_{fys} + k_{20} \frac{\alpha}{1+\alpha}$$

The mobile fraction,  $F_{mob}$ , for the steady-state condition is determined by the ratio  $S_m/S$  which is derived from the above as:

$$F_{mob} = \frac{\alpha}{1+\alpha}$$

Root uptake is implemented in the model by the exchange of radiocesium between the mobile soil compartment  $(S_m)$  and the internal-grass compartment (P) through the rate constants  $k_{25}$  and  $k_{52}$ . For steady-state conditions the transfer from the soil to the plant is equal to the transfer from the plant to the soil:

$$k_{25}S_m = k_{52}P \Longrightarrow k_{25} = k_{52}\frac{P}{S_m}.$$

Furthermore, we assume that the steady-state conditions for the soil system apply as described above. The soil system is characterised by a unit area, a depth d, a dry-weight density of  $\rho$ , and a dry-weight biomass density of y for the grass. The concentration ratio CR is defined as the equilibrium ratio between the dry-weight concentrations of radioactivity between plant and soil:

$$CR = \frac{P}{y} \frac{d \cdot \rho}{S_m + S_f} = \frac{d \cdot \rho}{y} \frac{P}{S}.$$

and by combining with the above we get

$$k_{25} = k_{52} \frac{y \cdot CR}{d \cdot \rho} \frac{1+\alpha}{\alpha}.$$

The rate constant  $k_{25}$  is thus given from the model parameters and the rate constant  $k_{52}$ . The latter is set arbitrarily at 0.1 d<sup>-1</sup> to ensure a rapid equilibrium.

Site-specific data on soil and biomass densities were collected. The values are shown in Table 1. Furthermore, information was collected on rates of lamb consumption covering average individuals and high-rate consumers (critical groups) in the countries. These rates are used for dose calculations and are shown in Table 2.

| Table | 1. | Site- | specific | values | for | soil      | and | grass | densities | at the | study | sites. |
|-------|----|-------|----------|--------|-----|-----------|-----|-------|-----------|--------|-------|--------|
|       |    | ~     | ~r       |        | ,   | ~ ~ ~ ~ ~ |     | 0     |           |        | ~~~~~ | ~      |

| Location   | Soil density              | Grass density             |
|------------|---------------------------|---------------------------|
|            | $(\text{kg d.w. m}^{-3})$ | $(\text{kg d.w. m}^{-2})$ |
| Ribe       | 1000                      | 0.18                      |
| Tjøtta     | 520                       | 0.15                      |
| Faroes     | 214                       | 0.10                      |
| Blomhöjden | 800                       | 0.15                      |

|--|

| Country | Average Individual   | Critical Group       |
|---------|----------------------|----------------------|
|         | $(\text{kg y}^{-1})$ | $(\text{kg y}^{-1})$ |
| Denmark | 1.1                  | 17                   |
| Norway  | 6                    | 15 (*)               |
| Faroes  | 19                   | 25 (*)               |
| Sweden  | 0.5                  | 7                    |

(\*) estimated by the authors

The model is implemented as a Fortran code in the Time Zero modelling system (Kirchner, 1989).

#### 2.5 Data analysis

The data on radiocesium in soil, grass and lamb from the four sites were analysed with the model by adjusting parameters to obtain the best possible agreement between observations and predictions. The procedure used least squares fitting for all three types of data (soil, grass and lamb) simultaneously. The parameters investigated include the three rate constants for the soil system  $k_{23}$ ,  $k_{32}$  and  $k_{20}$ , the concentration ratio CR that determines the root uptake, and the observed ratio  $OR_{lamb/grass}$  that determines the meat-transfer coefficient  $F_f$ . Furthermore, initial values at the start of the simulations in 1990 for the deposition of <sup>137</sup>Cs in the soil and for the partitioning between the two soil fractions were optimised in the calculations.

The parameter optimisation procedure was carried out first on the annual mean values from the observations for each of the four sites. In order to illustrate the effect of the reported uncertainties on the observations, 10 additional sub-sets of data for each site were sampled from normal distributions based on the values of the means and the associated standard errors. The model optimisation procedure was then repeated on each of these additional data sets to provide 10 additional values for each site for each of the parameters mentioned above. The variability of the parameter values thus illustrate the combined effect of the reported uncertainties of the observed values and the robustness of the model for these

parameters. For practical reasons the number of additional sub-sets of data for each of the four sites was limited to 10.

### 3. Results and discussion

The results of the model calculations with the annual mean of the observations are illustrated in Fig. 3 which shows the model results and the observations for comparison for each of the four sites. The corresponding results of the soil parameter values are given in Table 3. The calculated effective half lives of radiocesium in the surface soil layer vary across the sites from a value of 10 y at Ribe to a value at the Faroes of 30 y which is equal to the physical half life of <sup>137</sup>Cs. These values are higher (typically a factor 2) than those that were used by

Aarkrog (1994) for the effective ecological half lives for <sup>137</sup>Cs in Nordic lamb. The relative amounts of mobile radiocesium in soil at steady state ( $F_{mob}$ ) is also showed in Table 3. The values are correlated with the amount of Chernobyl fallout ( $R^2 = 0.9$ ).

*Table 3. Values of*<sup>137</sup>*Cs deposition in 1990 and soil parameters derived from model calculations with the annual mean data.* 

| Location   | Deposition     | k <sub>20</sub> | k <sub>23</sub> | k <sub>32</sub> | T <sub>1/2, eff</sub> | F <sub>mob</sub> |
|------------|----------------|-----------------|-----------------|-----------------|-----------------------|------------------|
|            | $(kBq m^{-2})$ | $(y^{-1})$      | $(y^{-1})$      | $(y^{-1})$      | (y)                   |                  |
| Ribe       | 4.5            | 1.2             | 0.60            | 0.066           | 10                    | 0.035            |
| Tjøtta     | 43             | 0.070           | 0.051           | 0.032           | 18                    | 0.23             |
| Faroes     | 6.0            | 1.9E-06         | 1.2             | 0.032           | 30                    | 0.027            |
| Blomhöjden | 17             | 0.040           | 0.044           | 0.016           | 23                    | 0.17             |

The variabilities of the effective half lives for radiocesium in soil found from the calculations on the additional data sets described above are illustrated in Fig. 4. In the graph the label 'Mean' refers to the parameter values obtained from the model calculations on the annual mean of the observations, and the vertical bars range from the minimum to the maximum values of the parameters from the 10 additional data sets. The ranges show some overlap for the different sites.

The variabilities of the mobile fractions of radiocesium in surface soil,  $F_{mob}$ , are illustrated in Fig. 5 and indicate significant different values between the sites: about 0.03 for Ribe and the Faroes, and about 0.2 at Tjötta and Blomhöjden.

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Figure 3. Comparison of model results and observations.

The resulting parameters for the transfers of radiocesium from soil to grass (CR) and the transfers from grass to lamb ( $F_f$ ) are shown in Table 4 and the calculated variabilities in the Figs. 6 and 7. The variabilities indicate significant different concentration ratios between the sites with Ribe showing the lowest value of 0.03, followed by the Faroes showing a value of 0.2, and Tjøtta and Blomhöjden at a value of about 1.

Table 4. Estimated parameters for the transfer of radiocesium from soil to grass (*CR*) and from grass to lamb ( $F_i$ ).

| (ent) and from grass to tame (1 j). |       |               |  |  |  |  |  |
|-------------------------------------|-------|---------------|--|--|--|--|--|
| Location                            | CR    | $F_{f}$       |  |  |  |  |  |
|                                     |       | $(d kg^{-1})$ |  |  |  |  |  |
| Ribe                                | 0.026 | 1.1           |  |  |  |  |  |
| Tjøtta                              | 0.97  | 0.74          |  |  |  |  |  |
| Faroes                              | 0.21  | 0.26          |  |  |  |  |  |
| Blomhöjden                          | 1.2   | 0.86          |  |  |  |  |  |



*Figure 4. Effective half lives for radiocesium in soil estimated from the observations.* 



Figure 5. Estimated fractions of mobile radiocesium in surface soil,  $F_{mob}$ .



*Figure 6. Concentration ratios for the transfer of radiocesium from soil to grass.* 

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Figure 7. Estimated meat transfer coefficients ( $F_{fr}$  d kg<sup>1</sup>) for the transfer of radiocesium from grass to lamb.

The values of the concentration ratios vary considerably (about a factor 50) across the sites. This variability is mainly due to the different soil characteristics between the sites. Mineral soils are known to retain radiocesium efficiently from plant uptake compared with organic soils, and the Tjøtta, Faroese and Blomhöjden sites are characterised by peaty soils in contrast to the Ribe site. However, if the radiocesium content in the grass is related to the mobile fraction in the soil, the concentration ratios become 0.7 for Ribe, 4.1 for Tjøtta, 7.9 for the Faroes and 7.1 for Blomhöjden. The total variation across the sites thus reduces to a factor of 10, still with the Ribe site at the low end, and with a significantly smaller variation across the sites with peaty soils. The lower transfer of radiocesium from soil to Faroese grass may thus be explained by a lower biological availability ( $F_{mob} \approx$ 0.03) than at the two other sites with peaty soil ( $F_{mob} \approx 0.2$  for Tjøtta and Blomhöjden).

The results for the meat transfer coefficients shown in Fig. 7 indicate agreement between the values from Ribe, Tjøtta and Blomhöjden at around 0.8 d kg<sup>-1</sup>, but the

Faroese value at 0.3 d kg<sup>-1</sup> seems to be significantly lower. The lower transfer of radiocesium from grass to lamb in the Faroes compared to the other Nordic sites has been noted during the project, but no obvious explanation for the apparent difference has been found. The meat transfer coefficient is known to decrease with age, but according to the information collected, the age of the Faroese lambs at slaughter is not significantly greater than that at the other sites. However, reported values of the meat transfer coefficient from the literature for uptake of radiocesium in lamb demonstrate significant variability. Pre-Chernobyl values are reported in the range 0.08 - 0.5 d kg<sup>-1</sup> (Ng et al., 1982), but post-Chernobyl field studies have found values at 0.8 d kg<sup>-1</sup> (Howard et al., 1987) and at 1.6 d kg<sup>-1</sup> (Beresford et al., 1989).

#### 3.1 Doses

The estimates of doses to individuals from consumption of lamb are based on timeintegrated concentrations of radiocesium in lamb from 1990 to 2040. These values are used with the consumption data from Table 2 and a dose-per-unit factor of  $1.3E-08 \text{ Sv Bq}^{-1}$  (IAEA, 1996). The resulting dose estimates are shown in Table 5 and in Fig. 8 together with the variabilities based on the reported uncertainties of the field data.

Table 5. Estimated 50-y doses from  $^{137}$ Cs to individuals from consumption of lamb from the study sites.

| Location   | Dose from average consumption | Dose from high-rate consumption |
|------------|-------------------------------|---------------------------------|
|            | (µSv)                         | (µSv)                           |
| Ribe       | 0.2                           | 4                               |
| Tjøtta     | 1300                          | 3200                            |
| Faroes     | 110                           | 140                             |
| Blomhöjden | 70                            | 1000                            |



*Figure 8. Ranges of doses estimated to be received by individuals in the period 1990-2040 from consumption of*<sup>137</sup>*Cs in lamb from the four study sites.* 

## 4. Conclusions

A generic modelling system has been applied to field data covering 1990-1996 on the transfer of radiocesium through the soil-grass-lamb foodchain at locations in Ribe (Denmark), Tjøtta (Norway), the Faroe Islands and Blomhöjden (Sweden). The Danish site is characterised by sandy mineral soil and the others by peaty soils. The analysis of the data with the model has made it possible to identify similarities and differences between transfer processes across the sites.

The effective ecological half lives for  $^{137}$ Cs in the soil were found to vary from 10 y at the Danish site to 30 y at the Faroese site.

For the sites with low levels of <sup>137</sup>Cs and consequently of Chernobyl fallout (Ribe and the Faroes, below ten kBq m<sup>-2</sup>) low mobile fractions of radiocesium in the soil (about 3%) available for root uptake in the grass were found, while the other sites with higher levels of Chernobyl fallout (Tjøtta and Blomhöjden, some tens of kBq m<sup>-2</sup>) showed higher mobile fractions (about 20%).

The uptake of <sup>137</sup>Cs in grass was found to be more consistent at sites with peaty soils when the levels in the grass were related to the mobile fractions in the soil

rather than to the total activities. The root uptake of radiocesium in grass from Ribe with mineral soil is a factor 10 lower than that from the site with peaty soils.

The transfer of <sup>137</sup>Cs from grass to lamb is found to be similar at the Danish, Norwegian and Swedish sites with a meat transfer coefficient,  $F_f$ , of about 0.8 d kg<sup>-1</sup>. The transfer to lamb at the Faroese site is lower by about a factor 3. No clear explanation of this difference has been found. However, the  $F_f$  values compare well with corresponding data from the literature.

Doses have been calculated to individuals from the consumption of lamb from the four locations. Two categories of individuals were considered: average consumers and high-rate consumers, and doses were calculated for a 50-y period 1990-2040. The doses from average consumption of lamb range from 0.2  $\mu$ Sv in Ribe to 1 mSv in Tjøtta, and doses from high-rate consumption range from 4  $\mu$ Sv in Ribe to 3 mSv in Tjøtta.

This work has illustrated that reliable predictions of radiological consequences require long time series of field data, particularly when environmental levels of radioactivity are characterised by long ecological half lives.

### 5. Acknowledgement

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# **REPORT 6** Consumption survey in Finland

A. Rantavaara and M.-L. Markkula Finnish Centre for Radiation and Nuclear Safety

## 1. Consumption survey

A mail survey was sent to 1500 randomly selected households (Markkula & Rantavaara, 1997). The country was divided into four regions: Metropolitan region, Western, Eastern and Northern Finland (Fig. 1). Questions referred only to food which was consumed at home.

The questionnaire was focused on the consumption of game, berries, reindeer meat, freshwater fish, wild mushrooms and some other wild food products of vegetative origin. The response rate of 59 % was reached after two reminders. Filled out questionnaires were returned from 884 households and the number of persons included in the responding households was 2360.

In our sample the percentage of hunters exceeded that in the whole population, which had to be taken into account. Hunters seem to have been more eager to return the questionnaire than other subgroups of population. The data was therefore analysed separately for hunters and non-hunters and thereafter the figures concerning the whole country were corrected by weighing.



*Figure 1. The four regions and their population. Stars represent municipalities from which the responses came (one or more).* 

For hunters the groups of berries, freshwater fish and game were all equally important with a mean consumption rate of 13 kg per year for a member of the household.

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*Figure 2. Average individual consumption of mushrooms by species and by regions (kg*  $y^{-1}$ ).

Regional differences in amounts of consumed mushrooms were quite obvious (Fig. 2). Consumption was biggest in Eastern Finland, 2.2 kg per year, and more than half of the mushrooms consumed were of *Lactarius* type. In Northern Finland the mean consumption of wild berries was greatest, 13 kg per year, to which mountain cranberry contributed most. Freshwater fish was used most in Eastern Finland in the large lake district. The consumption of game meat differed between the Metropolitan region's 0.32 kg per year to Northern Finland's 1.8 kg per year. The most important species was moose. The wild products of vegetative origin consist of sap, nettle, herbs and lichen. The consumption of sap was surprisingly high; 0.24 kg per year. The use of reindeer meat was as expected: biggest in Northern Finland and least in Western and Eastern Finland. The consumption pattern followed the availability of the product. Berries and freshwater fish are the most consumed types of wild foods (Table 1).

| 1 ood type                               | (kg per year) |
|--|---------------|
| Berries                                  | 8.3           |
| Freshwater fish                          | 7.9           |
| Mushroom                                 | 1.5           |
| Game                                     | 1.2           |
| Other wild products of vegetative origin | 0.4           |
| Reindeer meat                            | 0.5           |
|  |               |

 Table 1. Annual average per capita consumption of different foods of wild origin.

 Food type
 Consumption per person

In the Metropolitan region it is more common to buy wild food products than in the other regions. About half of the households did not use mushrooms and fish outside the season, while berries were used in most of the households (91%) throughout the year. A good harvest year would increase the consumption of mushrooms by 60% and of berries 20% compared with the year 1995, which was not a good mushroom year.

Only 3.3% of population did not use any wild food products at home. In the group of non-hunters the most consuming ten percent of the households used a total of 5.7 kg per year of wild mushrooms. In the group of hunters the same figure was 5.2 kg per year per user.

In those households who use wild mushrooms at home not all children seem to eat them (Table 2). On average the families with children used 0.9 kg mushrooms per year, whereas among all other households the figure was significantly more, 2 kg  $y^{-1}$ . In the households of hunters the children eat mushrooms more often than in other households. Most mushrooms per person were consumed in households of one person, 2.8 kg  $y^{-1}$ .

Table 2. Percentage of adults and children, who are using mushrooms in the households of hunters and non-hunters.

|             | Adults | Children (under 15y) |
|-------------|--------|----------------------|
| Non-hunters | 64 %   | 40 %                 |
| Hunters     | 69 %   | 55 %                 |

Consumption of mushrooms and berries was greater than in previous studies (Household survey, 1990 & Kleemola et al., 1994). The telephone interview for ten percent of the non-responders indicates that they are not non-users. They had adopted a critical attitude to their ability to estimate the amounts of wild products used. The non-responders used mushrooms 0.5 kg, while responders used 1.5 kg. Our response-weighed estimate for lowest limit for consumption mean is 1.1 kg *per capita*. Obviously 1.5 kg *per capita* slightly overestimates the consumption of wild mushrooms.

The study revealed regional differences in consumption rates. It is also important to consider the different consumption patterns in rural and densely populated areas and in sub-regions of the country. Use of mushrooms is about the same in sparsely populated areas and in town areas. Game, berries and freshwater fish are used significantly more in countryside than in towns.

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# 1.1 Intake of <sup>137</sup>Cs through mushrooms

The annual mean intakes of <sup>137</sup>Cs were calculated using Finnish data for the transfer of radiocesium from fallout to mushrooms (Rantavaara, 1990), the survey of nation-wide ground contamination by <sup>137</sup>Cs (Arvela et al., 1990), population densities throughout the country (Statistical Yearbook, 1995), data for radiocesium losses during cooking of mushrooms (Rantavaara, 1989) and regional consumption rates for mushrooms (Markkula & Rantavaara, 1997). Collective intakes in Western Finland exceeded those in all other regions threefold (Fig. 3). Number of consumers, the level of accumulated <sup>137</sup>Cs fallout in the area and the species used in the area explain the difference, but not the *per capita* consumption rates which were almost equal to the nation-wide mean. (Fig. 2). It is worth noticing that the country was divided into sub-regions without consideration of fallout distribution. Western Finland includes of more municipalities above the average fallout than other sub-regions, in the Northern Finland the average fallout was negligible, most municipalities in Eastern and metropolitan area were below average fallout.



*Figure 3. Average individual doses through mushrooms by regions (* $\mu$  *Sv y*<sup>-1</sup>*).* 

The effect of cooking methods on the intake mostly follows the contribution of species to be parboiled to the consumption of mushrooms. Our study confirmed the use of traditional methods of processing mushrooms. The *Lactarius* mushrooms are almost always and the *Russula* mushrooms often either parboiled or soaked in water and/or salted. These treatments remove most radiocesium from mushrooms, and thereby change the intake of <sup>137</sup>Cs significantly compared to mushrooms which are cooked as such and therefore contain all the original radiocesium in the edible fraction. The species *Craterellus c*. and *Cantharellus tubaeformis* are sometimes dried for storing. Soaking in boiled water before cooking removes as much as 85 % of their radiocesium, assuming that the water is rejected (Rantavaara, 1989).

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The individual radiation doses received through the <sup>137</sup>Cs in mushrooms varied by sub-regions following the patterns of both consumption and regional fallout (Fig. 3). The collective annual intake of both mushrooms and their <sup>137</sup>Cs reflects also the differences in the size of population of sub-regions (Fig. 4).



*Figure 4. Annual consumption of mushrooms and their contribution to dietary intake of*<sup>137</sup>*Cs in Finland..* 

Compared to Sweden the consumption rates for mushrooms were somewhat higher. Species differed also, when *Lactarius* and *Russula* mushrooms were not mentioned among the most consumed species in Sweden at all. The importance of parboiling and soaking of mushrooms may therefore not essentially decrease the intake of radiocesium there.

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## **REPORT 7** Ecological half-lives in fungi, selected species

Karl Johan Johanson Swedish University of Agriculture

# 1. Introduction

One of the most pronounced problems when trying to determine the effective ecological half-lives in fungi growing in a specific forest is the large variation which appears. The variation between species could be at least two order of magnitude. Even within the same species the coefficient of variations could be more than 50 % within the same forest. These variations could be due to variation in nutrient conditions within the soil. Even within very small areas, about 400 m<sup>2</sup> and the same species, we found a very high variation, a mean <sup>137</sup>Cs activity concentration among 25 fruitbodies of 64.3 kBq per kg d.w. and a standard deviation of 31.9. The coefficient of variation was thus nearly 50 %. Even in clusters of fruitbodies of mushrooms grown and collected within a few dm<sup>2</sup> the variation can be very high although it seems quite clear that they are feed by the same mycelium.

In order to estimate the half-lives of <sup>137</sup>Cs in fungi we have used the results from three specific sites where we collected many fruitbodies of *Suillus variegatus* in 1994 to 1997. This period of 3 years is very short and it may of course be questioned if it possible to obtain a good estimate of the effective ecological half-lives. It is thus very important to stress that the estimation of the half-life will be rough. However, the advantage of this study is that it is based on quite a large number of determination of <sup>137</sup>Cs levels in individual fruitbodies. In other sets of samples, for example from three other sites (20x20 meters) where we have collected all fruitbodies, we have only a rather small number of fruitbodies from each species which will give larger uncertainty in the annual mean of <sup>137</sup>Cs levels.

## 2. Material and methods

During the mushroom seasons 1994 to 1997 we collected all fruitbodies of *Suillus variegatus* growing at 4 sites each of about 400 m<sup>2</sup>. In 1995 and 1996, only few fruitbodies of *Suillus variegatus* were found in these sites. A part of each fruitbody was used for determination of the <sup>137</sup>Cs activity and another part for in vitro culture and determination of somatic incompatibility in order to determine if the fruitbodies belong to the same individual or not. By using this method it was possible to obtain a map of each site with the distribution of the various individuals of *Suillus variegatus*. Usually there were about 2-4 individuals of *Suillus variegatus* producing fruitbodies at each site. Our hypothesis before the experiment was that the variation should be less within fruitbodies from the same

individual compared to fruitbodies from two different individuals, but this seems not to be the case. The variation within an individual is as large as between individuals. On the other hand the variation between the sites is much higher than within a site. For example the mean <sup>137</sup>Cs levels in fruitbodies at six sites varied between 45.9 to 119 kBq per kg d.w. This is probably due to nutrient states at the different sites.

In 1995 and 1996, only few fruitbodies were found in the 4 sites but during 1997 which was a good mushroom year we collected all fruitbodies of *Suillus variegatus* at these 4 sites and the production was high.

Each fruitbody was cut and transferred to plastic vials and the  $^{137}$ Cs activity was determined using HPGe detectors. The counting times were chosen so the standard deviations due to the random process of decay were less than 5 %. After  $^{137}$ Cs determination the samples were dried at 55°C. The activity concentration was expressed as Bq per kg of dry material (d.w.).

#### **Results and discussion**

In table 1 a comparison between the results from 1994 and 1997 is presented. As can be seen there is no significant decrease or increase during these three years and no consistent trend to decrease. From the results we draw the conclusion that the effective ecological half-life of <sup>137</sup>Cs in *Suillus variegatus* is very long and we suggest that the physical half-life of <sup>137</sup>Cs - 30 years - should be used until longer investigation periods have been carried out. It can also be seen that, although the number of samples is very high, decrease in <sup>137</sup>Cs levels of a few per cent per year will not be possible to observe. It seems also reasonable to assume similar long effective ecological half-lives in other species of at least mycorrizal fungi.

The effective ecological half-life of 30 years has also been suggested when evaluated by the <sup>137</sup>Cs levels in moose harvested during the October to December hunting period in Sweden. As can be seen in Figure 1, there has been no decrease of <sup>137</sup>Cs levels in moose muscles during the period 1986 to 1997. The reduction in levels during 1995 and 1996 depends on the lack of mushrooms in the forests in Heby commune during September. Similar as for *Suillus variegatus* an expected decrease due to the decay of <sup>137</sup>Cs should be expected but could not be seen in the results. Therefore the effective ecological half-lives could be even longer than 30 years.
| site       | year | no. of samples | kBq per kg |      |
|------------|------|----------------|------------|------|
|            |      | n              | mean       | SD   |
| Site A94   | 1994 | 36             | 69.2       | 24.6 |
|            | 1997 | 56             | 79.9       | 31.0 |
| Stalbo 3-K | 1994 | 38             | 43.1       | 28.3 |
|            | 1997 | 110            | 44.8       | 15.2 |
| Kyrkvägen  | 1994 | 20             | 74.2       | 28.6 |
|            | 1997 | 7              | 64.8       | 32.0 |
| Stalbo 2-K | 1994 | 17             | 81.4       | 33.1 |
|            | 1997 | 19             | 80.6       | 28.3 |

Table 1. The <sup>137</sup>Cs activity concentrations in fruitbodies of Suillus variegatus collected at four sites in 1994 and 1997.

In order to illustrate the variation in more details, the results from individual days at site Stalbo 3-K is shown in Table 2. As can be seen the variation is quite large which illustrate the difficulties to calculate an exact half-life for  $^{137}$ Cs in *Suillus variegatus*.



*Figure 1. The annual mean*<sup>137</sup>*Cs activity concentrations in moose harvested during the autumn hunting period in Heby commune.* 

## 3. Conclusions

The effective ecological half-life for  ${}^{137}$ Cs found in mushrooms is probably similar as the physical half-life of  ${}^{137}$ Cs and could even be longer.

The variation in <sup>137</sup>Cs levels in fruitbodies of fungi is very large even when feed from the same mycelium and therefore there is a need of large number of samples from the same sites in order to obtain relevant results.

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| date number of samples |    | kBq per kg d.w. |      |  |
|------------------------|----|-----------------|------|--|
| Stalbo 3-K             | n  | mean            | SD   |  |
| 940831                 | 7  | 29.8            | 12.7 |  |
| 940908                 | 14 | 28.1            | 7.5  |  |
| 940922                 | 8  | 73.4            | 35.9 |  |
| 940929                 | 7  | 50.1            | 25.2 |  |
| 970910                 | 21 | 48.7            | 16.1 |  |
| 970919                 | 30 | 32.8            | 14.7 |  |
| 970926                 | 44 | 56.7            | 16.3 |  |
| 971002                 | 15 | 28.3            | 7.6  |  |

Table 2. The mean<sup>137</sup>Cs activity concentrations in fruitbodies of Suillus variegatus collected at different days in 1994 and 1997.

## **REPORT 8** RADIOECOLOGICAL HALFLIFE OF <sup>137</sup>CS IN DANISH ROZITES CAPERATUS.

Morten Strandberg

National Environmental Research Institute

#### ABSTRACT

Samples of Rozites caperatus from Denmark were analysed for radiocesium. The main objective was to determine the effective half-life of <sup>137</sup>Cs in an important representative of the ectomycorrhizal basidiomycetes.

An effective half-life with a mean of 2.3 years was determined for Chernobyl derived <sup>137</sup>Cs for the period from 1991 to 1994. The corresponding ecological half-life was 2.5 years. Thereafter the curve is flattening and it is likely that the effective half-life is getting closer to the physical. For the period from 1991 to 1997 the effective half-life for <sup>137</sup>Cs for Chernobyl derived cesium is 10 years. For fallout derived <sup>137</sup>Cs no reliable effective or ecological half-lives could be calculated, but an effective half-life close to the physical is most probable. It is likely that the Cs-concentration in the ectomycorrhizal mushrooms begins to increase soon after deposition of radiocesium to the ecosystem. Those with their mycelia in the surface displays the highest concentration in the beginning. A theory for Rozites is presented predicting that a maximum concentration is reached after 3 to 4 years after deposition followed by a rather fast decrease where after a more stable period is reached. The theory is based on measurements of fallout and Chernobyl derived cesium in Rozites from one mycelia from 1991 to 1997.

### 1. Introduction

Species of mushrooms, especially those forming ectomycorrhiza with forest trees, are among the most radiocesium contaminated foodstuffs known. Their ability to concentrate radiocesium were known already before the Chernobyl accident in 1986 (Grueter 1971, Haselwandter 1977, Haselwandter 1978). However it was first after the severe contamination of great upland and forest areas in 1986 their importance was realised by the great majority of radioecologists. Since it was established that edible mushrooms are responsible for an unexpected high transfer of radiocesium to man, either directly through consumption of fruitbodies or indirectly through meat from ruminants feeding on mushrooms (Hove et al., 1990, Karlén et al., 1991, Strandberg & Knudsen 1994), they have been the subject of a large number of investigations e.g. Horyna (1991), Tsvetnova & Shcheglov (1994).

*Rozites caperatus* ((L.:Fr.) P. Karst.) is among the mushroom species with the highest uptake of radiocesium from soil. This is confirmed in a great number of investigations from all over Europe e.g. Mascanzoni (1987), Haselwandter et al.,

(1988), Bakken & Olsen (1990), Gulden & Amundsen (1994). Until today higher radiocesium concentrations, in Danish material, have only been measured in Hygrophorus hypothejus ((Fr.:Fr) Fr.) (Strandberg in prep.) and in one species of Cortinarius, Strandberg (1992). Most international, if not all, investigations have been concentrated on comparative studies of a larger number of mushroom species. Few authors have dealt with the time perspective. Elstner et al. (1987) and (1989) presents results from Germany from 1986 and 1987. Haselwandter (1978) and Haselwandter et al. (1988) compared the uptake of radiocesium in 7 species before (1974) and after the Chernobyl accident in 1986. It was found that the pattern of uptake was the same in both years and that it was species related. Heinrich (1992) found that radiocesium uptake in mushrooms generally was in the order symbiotic forest mushrooms > saprophytic forest mushrooms > meadow land mushrooms. In Norwegian mushrooms Olsen (1994) suggest a biological half-life of Chernobyl derived <sup>137</sup>Cs close to the physical, because no constant decrease was observed. However other observations from Norway reported by Gulden and Amundsen (1994) showed a decrease in some species, but an increase in others. Some species neither decreased nor increased during the period from 1989 - 1993 e.g. Rozites caperatus.

In common to all investigations cited are that they have no exact assessments of time dependent changes, such as determinations of ecological or effective half-life of radiocesium in any mushroom species. In this context ecological and effective half-life should be apprehended as defined by Strandberg & Strandgaard (1995). It might be so that the lack of a clear decrease has lead some authors to conclude that the effective half-life is equal to the physical. As fungi are among the products from natural ecosystems with a high potential for transfer of radiocesium to man, effective half-lives for radiocesium in fungi are valuable to know. The present study presents an assessment of the effective half-life of Chernobyl derived <sup>137</sup>Cs in one mushroom species, *Rozites caperatus*.

The use of mushrooms as bioindicators presents both advantages (Giovani et al., 1990 and Mietelski et al., 1994) and disadvantages, compared to lichen and moss species. The main difference is that mushrooms monitors a combination of parameters such as availability, deposition and depth penetration, while mosses and lichens simply gives an indication of the deposition pattern. The performance of lichens and mosses as bioindicators of radiocesium as well as other airborne contaminants has been assessed in several investigations e.g. Mattson & Lidén (1975), Eckl et al. (1986), Guillitte et al. (1990b), Crête et al. (1992) and Sloof & Wolterbeck (1992).

## 2. Material & methods

### 2.1 Samples

The material consists of 25 samples of *Rozites caperatus* obtained from 1 mycelia in Tisvilde Hegn in North Zealand, Denmark.

### 2.2 Site description

Tisvilde Hegn (56°03'N 12°06'E) is a near shore forest on sandy ground. The sampling site is an area with Scots pine (*Pinus silvestris*) on old deposits of shifting sand. The podzol soil is mainly sand covered by an approximately 5 cm thick organic layer. Mosses, such as *Pleurozium schreberi*, *Scleropodium purum* and *Hylocomium splendens*, dominates the forest floor. Local importance have lichens (especially *Cladina sp.*), herbs and dwarfbushes e.g. *Linnaea borealis*, *Fragaria vesca*, *Empetrum nigrum* and *Vaccinium vitis-idaea*. The clay content of the soil is low, between 0 and 2 % and pH varies between 4 and 5.

### 2.3 Sampling and treatment

Fruitbodies of *Rozites caperatus* was sampled from one distinct mycelia in Tisvilde Hegn. The limitation of mycelias of *Rozites caperatus* is most often easy (Strandberg & Rald, 1995), because they grow in well defined small groups.

Mushroom samples were cleaned from soil and litter and the base was cut off. They were brought to the laboratory where the mass was determined. Finally they were dried at 50°C for 48 hours and ground, before the dry weight mass was determined.

 $^{134}$ Cs results are not presented, but they were used for the calculation of the Chernobyl derived proportion of  $^{137}$ Cs in soil and fruitbodies. This calculation was carried out using a ratio of  $^{134}$ Cs to  $^{137}$ Cs of 0.54 (Aarkrog et al., 1988) on the 26th of April 1986.

## 3. Results & Discussion

## 3.1 Effective halflife of <sup>137</sup>Cs in distinct mycelias

Measurements and the decay curve is presented in fig 1. The effective half-life of <sup>137</sup>Cs in *Rozites caperatus* from 1991 - 1994 was 2.3 years for <sup>137</sup>Cs from Chernobyl and from 1991 to 1997 it was 9.6 years.  $T_{1/2eco}$  can be calculated from equation I.

(I)  $T_{1/2eco}^{-1} = T_{1/2eff}^{-1} - T_{1/2phy}^{-1}$ 



Figure 1. Regression line and concentrations of <sup>137</sup>Cs in Rozites caperatus from 1991 - 1997 at a distinct mycelia in Tisvilde Hegn.

Randa et al. (1990) observed an increase of the radiocesium content in *Boletus badius* from 1986 to 1988 from 14.8 in 1986 over 25.4 in 1987 to 38.3 kBq/kg D.W. in 1988. This increase is explained by the continued penetration of radiocesium to layers where it is available to *Boletus badius*.

Heinrich (1992) also observed an increase in most species from 1986 to 1989. In Norway Gulden & Amundsen (1994) reports results from Lierne on *Rozites caperatus* from 1989 to 1993. During this five year period no clear trend was observed, the material presented is apparently not from defined mycelias and very few replicates are included. However in species such as *Suillus variegatus* and *Amanita fulva* there may have been a decrease since 1990 and 1989 respectively. In other species the picture is more confused and in some cases an increase have taken place during the period e.g. in *Cortinarius armillatus* and *Tricholoma portentosa* (Fig 2).

4



Figure 2. The development of radiocesium concentrations in Tricholoma portentosa in which the concentration of radiocesium has increased from 1991 to 1997.

In a semi-logarithmic presentation of the mean concentrations of Chernobyl derived <sup>137</sup>Cs it is clearly demonstrated that a change in the uptake pattern takes place over the mushroom seasons from 1994 to 1995. The decrease ceases and a more constant rate of uptake now dominates (Fig. 3)



*Figure 3. The break of the curve around 1994. From that year the uptake of Chernobyl derived radiocesium in Rozites caperatus decreases with a long half-life.* 

From own observations and those cited it is possible, though with some uncertainty, to predict a general picture of mushroom <sup>137</sup>Cs-contamination over time after an accident (Fig. 4).

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Figure 4. Indications of the concentrations of the of origin different fractions of radiocesium in the fruitbodies of Rozites caperatus. C = Chernobyl, F = Fallout and T = Total. Lines indicates the fate according to the theory presented, dots represents actual measurements as presented in figure 1. For further explanation see text below.

- An initial phase where some saprophytes (especially *Clitocybe* sp. (Guillitte *et al.*, 1990a) are highly contaminated, but the radiocesium concentration start to decrease after the first or second season after deposition because radiocesium penetrates the litter layer and consequently becomes unavailable to the superficial mycelia of most saprophytes. In this initial phase most symbiotic (ectomycorrhizal) fungi displays an increase as radiocesium availability is enhanced by penetration to the layers where most species in this group have their mycelia. This early phase lasts from a few years (say 2 5) up to several years, depending on the depth of the mycelia, and the velocity of the depth penetration of radiocesium through the upper soil layers, which again is a function of at least vegetation, soil and climate.
- The intermediate phase, where the levels in the symbiotic mushrooms seems to be more or less constant, but with variations in both directions because of differences in availability of radiocesium between seasons. The supply of

radiocesium from layers above the mycelia is more or less equal to the removal by decay and biogeochemical processes. Levels in saprophytes decrease because the penetration of radiocesium keeps on moving cesium away from the litter layer. The intermediate phase may last from a few to several years.

• Finally the terminating phase where radiocesium levels decreases as a result of the joint forces of biogeochemical processes and physical decay of radiocesium. This phase is characterised by the efficient half-life of radiocesium, which now can be derived from measurements of decreasing levels of radiocesium in fruitbodies. The efficient half-life will probably increase towards the physical half-life during the terminating phase because loss and supply due to biogeochemical processes becomes more equal, hence it is mainly the physical decay that determines the rate of decrease. In the present study it is possible that this stage have been reached for fallout cesium from the early sixties. Thirty years, therefore, may consequently be the maximum time needed for  $^{137}$ Cs to reach equilibrium. In other ecosystems and climates the time needed can be longer or shorter depending on the turnover rates. The saprophytes are at this stage at a very low contamination level, because the main source of radiocesium to the litter layer are falling leaves and other remnants of the different forest plants contaminated through root uptake.

### 4. Conclusions

The effective half-life of Chernobyl derived <sup>137</sup>Cs in fruitbodies of *Rozites caperatus* was calculated to 2.3 years for the period from 1991 to 1994 - hereafter the effective half-life has been increasing. The effective half-life for <sup>137</sup>Cs from fallout in the same fruitbodies could not be determined, but definitely it was longer and it is probably close to the physical half-life of 30 years.

In most years but 1994 Chernobyl cesium were taken up more readily than fallout cesium, but in 1994 there was no clear difference in rate of uptake from soil.

### 5. Acknowledgements

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## **REPORT 9** DISTRIBUTION OF CHERNOBYL RADIOCESIUM IN ECOSYSTEM COMPONENTS IN NORWAY

Eldar Gaare Norwegian Institute for Nature Research

## 1. Aims

Transfer of <sup>137</sup>Cs by fungi and roe deer from range to man on the island Ytterøya in the Trondheims Fjord

### 2. Study area

Ytterøya, 63°45'N; 11°O, is an island in the Trondheims Fjord. Its closest distance to the mainland is 1,5 km. Communications to the Levanger municipality of which it is a part since 1964, is by a car ferry.

The total area is 28 km<sup>2</sup>, whereof 50% agricultural land, 25% productive spruce forest, and the rest less productive forest and some mire. The bedrock is sandstone and conglomerate which develops medium-rich to rich soils. Most agricultural land is situated on marine deposits. The highest peak is Sandsetheia, 211 m a s l. Oak (Quercus petraea) reproduce on a SV-exposed hillside. The summer climate is rather dry, total precipitation is about 750 mm annually. The snow cover lasts seldom more than a few weeks. The islands isolation show off in the mammalian fauna, roe deer is the only larger one, hare and fox is absent. The varied landscape with large transition areas between cultivated and forested land make a favourable roe deer habitat. This and the absence of predators make the roe deer population perhaps the most dense in Norway. The annual hunt is organised in 8 separate ranges A-H, and the 200-300 deer shot is the predominant mortality factor

Only about 500 of the 17000 inhabitants of Levanger municipality are residents on the island. The adult (>18+ years) population was in September 1997, 195 males and 191 females, the rest is children (<18 years). The island is know as an outdoor area with opportunities for some sea fishing, excellent roe deer hunting and mushroom picking. Therefore, in the summer time about 150 tourists, many with summer houses comes in addition.

### 3. Materials and methods

Ytterøya utmarkslag (the local ground owner organisation) organised the collection of 77 samples of roe deer muscle during the hunt 25.09 to 23.12.1995. By piece of bad luck all rumen and most faeces samples were lost. A similar sampling in 1997, complete with rumen content and faeces is not yet finished. Members of the NINA staff collected pasture plants in 1996 and measured by a 3" NaI equipped scintillator. At 4 field locations, gamma spectrometry has been performed *in situ* by a 3" NaI detector connected to a Canberra series 10. This was for 300s carried without collimator 1 m above the ground, covering systematically a homogenous plant community. The pulse score in the interval 517-723 keV has been calibrated as area activity, 1puls/min = 9,26 Bq/m<sup>2</sup>, (Gaare 1997)

Mushrooms could not be found during the fieldwork, and both 1995 and 1996 were reported to be poor mushroom years by the local population. Sampling of mushrooms from 1997, which is reported a good year, has not yet been processed.

All biological samples are dried at 70°C prior to counting. Muscle samples contain 25% dry matter and may be calculated to fresh matter for comparison.

The local population's use of range growing fungi was studied by a questionnaire given to all households in October 1997. The questionnaire followed closely a similar study in Sweden, (Johansson 19xx). A group of 15 years old pupils and their teacher at the local high school assisted in this. The same group picked fungi for analysis in addition to out own sampling.

### 4. Results

Based on the calibration, the present activity of  $^{137}$ Cs is calculated to 8 kBq/m<sup>2</sup> in uncultivated areas and 2 kBq/m<sup>2</sup> in grassland, Table 1. Chernobyl radiocesium was measured from helicopter by NGU (Norges geologiske undersøkelser), 1988. Their map show 0-100 pulses/s in cultivated land, 250-300 in forested land and upland and estimate that 300 pulses/s is 5-20 kBq/m<sup>2</sup>.

Table 2 show mean of concentrations in muscle from animals shot in separate hunt fields. Even if roe deer are territorial they do not always graze in the field were they are shot. Even if this is true we see that areas with high fallout have plants and animals with more radiocesium.

A few animals were shot in April 1997, in field B, E and H. Overall concentration in dry muscle,  $229 \pm 152$  Bq/kg (n=3), was not different from samples from autumn 1995. But the ratio between muscle concentration to rumen content or faeces was > 1 indicating animals with decreasing radiocesium load.

| Place                              | Hunt<br>field | <sup>137</sup> Cs activity Bq/m <sup>2</sup> |        |   | NGU<br>pulses/s |
|------------------------------------|---------------|--|--------|---|-----------------|
|                                    |               | Mean   | St.dev | n | _               |
| Tangen, old spruce forest          | А             | 8 300  | 500    | 4 | 100             |
| Solås, cultivated meadow           | А             | 3 000  | 700    | 3 | <100            |
| Solås, clear cut in spruce forest  | А             | 8 600  | 700    | 2 | 100             |
| Indre Berg, cultivated old meadow  | G             | 6 500  | 200    | 2 | 100             |
| Indre Berg, billberry birch forest | G             | 13 400                                       | 1 300  | 5 | 300             |
| Myr gård, old road in mixed forest | Н             | 5 900  | 200    | 2 | 200             |

Table 1 Area activity of  $^{137}$ Cs calculated from pulses in 517-723 keV range in a gamma spectrum measured in the field at Ytterøya, September 1996 (1 pulse/min = 9,26 Bq/m<sup>2</sup>). Stations chosen are known as frequently used roe deer habitats.

Roe deer graze mainly in the field layer and in the summer herbs, graminids and a few sedge species are most important.

The radiocesium content in more common food plants is shown in Table 2. In the autumn and winter *Vaccinium myrtillus* is known elsewhere as a very important component of the diet. Signs of severe grazing were observed to be very common. Botanical analysis of rumen samples collected 1997, will quantify its importance in transfer of radiocesium. Like other plants its <sup>137</sup>Cs concentration vary with the area fallout and reach a maximum in field G, 741 Bq/kg.

The meat samples from 1995, Figure 1 show an average concentration of  $^{137}$ Cs of 210±236 Bq/kg (n=77). There is a large variation ranging from beyond detection up to 1 100 Bq/kg. There is no significant change with date of felling. The variation between animals most probably reflects the different grazing habits. This again is connected to social status in a group. The ratio between meat and faeces samples was higher <1 indicating animals with increasing radiocesium load. Animals shot in hunt fields with high loads, table 1 show higher muscle and faeces concentrations, Table 3.

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Cs-137 in roe deer muscle and faeces, Ytterøya 1995

Figure 1. Concentration of radiocesium in 78 roe deer muscles and 11 faeces samples from Ytterøya in a poor fungi autumn, 1995

| Sample | Hunt field | Mean       | St.dev.    | Number |
|--------|------------|------------|------------|--------|
| type   |            | Bq/kg d.w. | Bq/kg d.w. |        |
| Faeces | В          | 621        | 346        | 5      |
| Faeces | E          | 954        | 317        | 6      |
| Muscle | А          | 103        | 59         | 13     |
| Muscle | В          | 82         | 85         | 10     |
| Muscle | С          | 190        | 88         | 8      |
| Muscle | E          | 319        | 277        | 17     |
| Muscle | F          | 202        | 208        | 9      |
| Muscle | G          | 187        | 169        | 12     |

*Table 3. Concentrations of* <sup>137</sup>*Cs in roe deer samples shot in separate hunting areas.* 

## 5. Use of fungi in Ytterøya households

The questionnaire was given to nearly all households. Of the ca 500 residents, 437 belong to 173 households who responded. Of the latter, 90 persons (21%) belong to the 28 households (16%) that eat mushrooms. The age distribution of all responders and those of them that use fungi is shown in Table 4. There is a small tendency that fungi are more used in families with younger persons.

*Table 4. Demography of persons in 173 responding households, and of those in 28 households that eats fungi* 

| nousenetus mai | nousenerus inur eurs jungt    |                            |  |  |  |  |  |  |
|----------------|-------------------------------|----------------------------|--|--|--|--|--|--|
| Age class      | Number in responding families | Number in families picking |  |  |  |  |  |  |
|                |                               | fungi                      |  |  |  |  |  |  |
| >60            | 104                           | 14                         |  |  |  |  |  |  |
| 40-60          | 108                           | 29                         |  |  |  |  |  |  |
| 25-39          | 84                            | 11                         |  |  |  |  |  |  |
| 18-24          | 37                            | 7                          |  |  |  |  |  |  |
| <18            | 106                           | 29                         |  |  |  |  |  |  |
| Total          | 439                           | 90                         |  |  |  |  |  |  |

16% of the 28 families who pick fungi, 5.8% does so 1 time annually, 4.6% 2 times, and 5.8% 3 times or more. The amount picked by those families picking 3 times or more (8 of 9 families have given this information) is on average  $10.4 \pm 12.8$ kg. The high figure is due primarily to the 2 families, if they are omitted the average is  $4.4 \pm 3.6$ kg annually. Only a few species are popular, even in a good fungi year, table 5.

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| Species                          | Amount  | St.dev. |
|----------------------------------|---------|---------|
|                                  | kg f.w. |         |
| Cantharellus cibarius            | 5.8     | 7.5     |
| Cantharellus tubaeformis         | 2.3     | 1.5     |
| Boletus edulis                   | 7.3     | 6.6     |
| Agaricus spp.                    | 5.0     | 1.7     |
| Lactarius spp                    | 1.0     | 0.4     |
| Hydnum repandum                  | 0.8     | 0.4     |
| Albatrellus ovinus               | 1.0     | 0.3     |
| Other species (often cf Bovista) | 2.7     | 1.7     |
| Annual average                   | 10.4    | 12.8    |

Table 5. Species and amount, given as kg fresh weight of peeled specimens of fungi picked in 1997, a good fungi year. The category «Other species» is by some respondents noted as cf Bovista spp.

## 6. Some measurement of moose muscles, 1992

Doses to man stems from different sources. In many parts of Norway, as is shown in Sweden, moose hunting today contributes significantly to the meat supply in many hunters families. Table 6 show the concentration of radiocesium in (dry) moose meat collected in 1991, and measured at NINA laboratories in 1992. In addition some measurements from 1990 are cited from Skurdal 1991. <sup>137</sup>Cs to dry weight are calculated from the original which was given as total caesium on a fresh weight basis. A dry weight of fresh meat is assumed 25%, and the relation <sup>137</sup>Cs: <sup>134</sup>Cs as 1,86:1 in 1986.

There are large variations within a municipality, but animals contamination seem to be correlated to the fallout levels measured in the municipalities. A material from Buskerud county stems from a period 1986-1987, Klute 1988 and is difficult to compare with and has not been included here.

| Municipality   | Mean       | St.dev     | n  | Year | Source         |
|----------------|------------|------------|----|------|----------------|
|                | Bq/kg d.w. | Bq/kg d.w. |    |      |                |
| Hedmark        |            |            |    |      |                |
| Åsnes          | 100        | 0          | 9  | 1992 | Gaare original |
| Nord-Fron      | 700        | 500        | 7  | 1992 | Gaare original |
| Gausdal        | 200        | 100        | 4  | 1992 | Gaare original |
| Nordre Land    | 500        | 300        | 7  | 1992 | Gaare original |
| Oppland        |            |            |    |      |                |
| Ramnes         | 300        | 100        | 2  | 1992 | Gaare original |
| Lardal         | 400        | 300        | 16 | 1992 | Gaare original |
| Vestre Slidre  | 710        | 450        | 4  | 1990 | Skurdal 1991   |
| Øystre Slidre  | 350        | 175        | 12 | 1990 | Skurdal 1991   |
| Vang           | 710        | 270        | 2  | 1990 | Skurdal 1991   |
| Aust-Agder     |            |            |    |      |                |
| Lillesand      | 390        | 170        | 16 | 1992 | Gaare original |
| Birkenes       | 560        | 310        | 23 | 1992 | Gaare original |
| Nord-Trøndelag |            |            |    |      |                |
| Meråker        | 730        | 450        | 3  | 1992 | Gaare original |
| Tydal          | 290        |            | 1  | 1990 | Skurdal 1991   |
| Stjørdal       | 600        | 290        | 11 | 1992 | Gaare original |
| Levanger       | 590        | 110        | 2  | 1992 | Gaare original |
| Inderøy        | 160        |            | 1  | 1992 | Gaare original |
| Troms          |            |            |    |      |                |
| Bardu          | 110        | 30         | 4  | 1992 | Gaare original |
| Nordland       |            |            |    |      |                |
| Hattfjelldal   | 120        | 20         | 3  | 1992 | Gaare original |

Table 6. Measurement of  $^{137}Cs$  concentrations in dry moose muscles in Norway. Data from Skurdal 1992 are recalculated to facilitate comparison. All samples are collected from ordinary hunt in September-October. A majority are males >1.5 years, but all age and sex classes are represented.

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## **REPORT 10** VARIATION OF <sup>137</sup>CS CONCENTRATIONS AMONG FISH IN TWO DIFFERENT LAKES OVER 10 YEARS FOLLOWING THE CHERNOBYL FALLOUT

E. Andersson, M. Meili<sup>\*</sup>, M. Sundbom & M. Östlund Institute of Limnology, Uppsala University and <sup>\*</sup>Institute of Earth Sciences, Uppsala University

## 1. Introduction

This study is based on unique time series of data that were collected regularly, usually several times per year, using consistent methods, and covering all abundant fish. <sup>137</sup>Cs in fish was monitored in a shallow and in a stratified Swedish lake over 10 years following the Chernobyl fallout in 1986. The data set includes >2700 fish individuals from 6 species and covers different size classes and seasonal variations.

These data were evaluated with the following focus:

- (1) long-term trends in different fish
- (2) influences of the fish ecology and physiology
- (3) influences of lake characteristics
- (4) relationships with current radiation protection guidelines

A nonlinear regression model were developed as tool for comparing time series of different fish classes.

### 2. Methods

Fish were collected in south-eastern Sweden, about 100-150 km NW of Stockholm, in the two lakes Flatsjön (N 60°29', E 17°10') and Siggeforasjön (N 59°52', E 17°03'). Both lakes are small oligotrophic forest lakes but differ in physical and chemical properties (see Broberg, Malmgren & Jansson, 1995). Flatsjön is a shallow monomictic lake, where all sediments are located at depths exposed to resuspension during the whole ice-free period, whereas Siggeforasjön is dimictic, being similar in size but almost three times deeper. Flatsjön has a high content of humic substances, about twice as much as mesohumic Siggeforasjön. Based on fish catch statistics, perch and roach are the most abundant fish species in both lakes, followed by bream and pike.

The lakes are located in an area where large amounts of fallout radionuclides were deposited after the Chernobyl nuclear accident on April 26, 1986, with <sup>137</sup>Cs deposition reaching a density up to 200 kBq m<sup>-2</sup>. Flatsjön received about 70 kBq m<sup>-2</sup>, and Siggeforasjön about 30 kBq m<sup>-2</sup>. The <sup>137</sup>Cs concentration in profundal surface sediment was in 1990 around 27 kBq g<sup>-1</sup> d.w. in Flatsjön and 12 kBq g<sup>-1</sup> d.w. in Siggeforasjön (Broberg, et al., 1995).

Fish were collected up to six times yearly during the period from May 1986 to June 1996. Gill nets with variable mesh sizes (10-75 mm) were used. The length, weight and sex of the fish was determined and thereafter muscle tissue was sampled and analyzed for radiocesium. Samples from large individuals were always analyzed individually, while those from small fish of the same species and size (within 1 cm of length) in many cases were pooled in order to improve measurements.

Activity concentrations of <sup>137</sup>Cs were quantified in wet samples using hyper-pure Ge-detectors (ORTEC, PGT) for most samples and a Gamma Counting System (Intertechnique Model 4000) equipped with a 3" NaI well detector for the smallest samples. Both equipments were regularly calibrated using blanks and dry standards of highly contaminated lichens collected after the Chernobyl fallout. All measurements were corrected for background radiation and sample geometry. Counting times were adjusted to obtain a coefficient of variation below 5 % (usually 1%). In samples below 100 Bq a limit of 10% was applied instead.

Fish from all species that were caught regularly and in sufficient numbers were included in the statistical analyses, which were applied to each lake independently. The fish from abundant species were divided into size classes (Table 1), with body length limits of each class based on empirical and statistical criteria of maturation, diet shifts, and available number of samples.

Before comparative and statistical analyses activity concentrations were logtransformed to equalize variance among fish types and over time. Activity concentrations were also corrected for radioactive decay back to May 1, 1986, the approximate time for the initial Chernobyl fallout. All values refer to muscle tissue and are expressed on a wet-weight basis.

| Species | Size Class | Numl | per of | Mean No. of |       | Median |         | Median |         |
|---------|------------|------|--------|-------------|-------|--------|---------|--------|---------|
|         |            | Sam  | ples   | Fish        | n per | Lengt  | th (cm) | Weig   | ght (g) |
|         |            |      |        | Sar         | nple  |        |         |        |         |
|         |            | Flat | Sigge  | Flat        | Sigge | Flat   | Sigge   | Flat   | Sigge   |
| Perch   | 5–13 cm    | 162  | 86     | 3.0         | 3.7   | 9.6    | 9.7     | 8.5    | 8.4     |
|         | 13-19 cm   | 140  | 121    | 1.4         | 1.8   | 16.8   | 17.2    | 56.8   | 54.1    |
|         | 19–25 cm   | 172  | 72     | 1.0         | 1.0   | 22.0   | 21.3    | 135.0  | 109.1   |
|         | 25-40 cm   | 110  | 48     | 1.0         | 1.0   | 27.8   | 29.7    | 291.2  | 300.0   |
| Roach   | 5-13 cm    | 102  | 64     | 4.5         | 5.1   | 10.6   | 10.8    | 10.1   | 11.0    |
|         | 13-19 cm   | 112  | 86     | 1.7         | 2.0   | 15.6   | 15.9    | 36.4   | 36.5    |
|         | 19-25 cm   | 67   | 67     | 1.0         | 1.1   | 20.6   | 20.6    | 93.1   | 84.3    |
| Pike    | 25-45 cm   | 115  | 37     | 1.0         | 1.0   | 39.2   | 34.5    | 354.0  | 218.9   |
|         | 45-100 cm  | 59   | 33     | 1.0         | 1.0   | 52.8   | 58.4    | 891.7  | 1134.0  |
| Bream   | 5–13 cm    | 67   | 62     | 2.0         | 1.8   | 13.7   | 16.2    | 26.5   | 36.8    |
|         | 19-25 cm   |      | 81     |             | 1.1   |        | 21.7    |        | 93.7    |
|         | 25-32 cm   |      | 87     |             | 1.0   |        | 29.0    |        | 233.7   |
|         | 19-32 cm   | 62   |        | 1.0         |       | 23.9   |         | 163.7  |         |
|         | 32-48 cm   | 55   | 34     | 1.0         | 1.0   | 39.2   | 34.1    | 633.9  | 430.1   |
| Ruffe   | 5-15 cm    | 25   | 41     | 2.8         | 3.0   | 7.9    | 8.8     | 6.4    | 7.2     |
| Bleak   | 8-16 cm    |      | 40     |             | 3.0   |        | 13.2    |        | 13.5    |
| Rudd    | 8-28 cm    | 43   | 71     | 1.2         | 1.5   | 22.6   | 18.7    | 170.9  | 83.3    |

Table 1. Summary of data on the studied fish species and size classes, the number of samples of fish muscle, the average number of equally sized individuals in each muscle sample, the median total length (from nose to tip of tail fin) and median fresh weight of each fish type.

To describe the temporal development of <sup>137</sup>Cs concentrations in each fish type, and to derive characteristic parameters for comparisons among fish types, a simple nonlinear model was applied to the data (Meili & Sundbom, in prep.). The basic formulation of the model is

$$C = \frac{a_3(e^{-\frac{1}{a_1}} - e^{-\frac{1}{a_2}})}{(a_1 - a_2)} + a_4,$$

where C is the <sup>137</sup>Cs concentration in fish muscle, *t* the time elapsed since fallout, and  $a_1$  to  $a_4$  are parameters. After reparametrisation and rearrangement (omitted here) of the model we have the following four intelligible parameters: (1) maximum activity concentration, (2) the time to reach maximum activity concentration, (3) the initial rate of decline, (4) an asymptotic baseline concentration. The advantages of the reparametrisation are that the new parameters

are easy to understand and that confidence limits for these key parameters can be estimated directly. The model was fitted by least-square methods to time series of log-transformed <sup>137</sup>Cs measurements. In the case of pooled samples, least squares were weighted with the number of fish in each vial. Parameter values were determined by minimizing the residual sum of squares using iterative methods (Gauss-Newton and Newton-Raphson). The approximate standard errors and, when possible, 95% likelihood confidence limits of each parameter were also iteratively calculated. Ordinary linear time scales were transformed into physiological time scales to account for the seasonal fluctuation of metabolic rates in dimictic lakes (Meili, in prep.).

### 3. Results and discussion

After a relatively fast decline following the maximum concentrations of <sup>137</sup>Cs in the late eighties, the rate of decline of <sup>137</sup>Cs in fish has decreased substantially during the past half decade, indicating that a change in the processes controlling the recovery of aquatic ecosystems has occurred. Correspondingly, ecological half-lives were not constant but increased continuously over time and are approaching infinity. The <sup>137</sup>Cs dynamics during the initial years as well as the leveling off during the following years were related to the type of fish. Due to limited space, time series for all fish types are not displayed. However, a few examples of time series for typical fish have been included for illustrating the general contamination pattern and variation over time, as well as the descriptive power of the nonlinear model introduced in this study (Fig 1). As an alternative to simply plotting all fish versus time, the key parameters of the model have been estimated and displayed for all types of fish in both lakes (Fig 2).

Both rise and decline of <sup>137</sup>Cs concentrations were related to fish species and trophic level (Fig. 1 & 2). The time needed for reaching maximum concentrations increased exponentially with trophic level. In piscivorous fish, the largest individuals reached their highest concentrations after an initial equilibration phase that lasted as long as 1.5 years in perch and 3 years in pike. Since the specific growth rate is higher in pike, the difference in the delay cannot be explained by differences in metabolic turnover, but indicates that pike occupies a higher trophic level, although both fish are piscivorous. This can be attributed to a larger prey size in pike, i.e. an indirect trophic-level effect. Also during the initial fast decline phase, i.e. before leveling off, differences between fish was obvious. While <sup>137</sup>Cs in small and rapidly growing fish at low trophic level decreased steeply and leveled off already in summer 1991 (Fig. 1A & B), concentrations in the largest predators appeared not starting leveling off until 1996 Fig 1D.

The estimated quasi-equilibrium that is approached during the leveling off level (the base parameter), appeared not to differ among fish on lower trophic levels but increased steeply with the degree of piscivory, i.e. for trophic levels above two (Fig. 2). But interestingly, for pike that is occupying the highest trophic level the base parameters were lower than for the largest size classes of perch. Also young perch had higher levels than all other non-piscivorous fish, and similar levels as medium-size pike despite a much lower trophic level. That perch is highly susceptible to radiocesium contamination is a well-known phenomenon, but no acceptable explanation has yet been given. Neither in this study could this be fully explained but our results indicate that physiological rather than dietary factors were most important.



Figure 1. <sup>137</sup>Cs concentration (Bq kg<sup>-1</sup>) in fish muscle during the period 1986-1996 in two Swedish lakes. The lines show a nonlinear regression model fitted to the data. The independent variable of the model was a physiological time scale which reflects the annual temperature dependent cycles of metabolic rates. Siggeforasjön: A) Small perch, 5-13 cm and B) roach, 13-19 cm. Flatsjön: C) Piscivorous perch 19-25 cm and D) Pike, 25-45 cm.

Also when comparing the observed leveling off concentrations within species over their whole size range, significant differences emerge. For pike the leveling off concentrations increased steeply with body size, while in the bottom dwelling bream concentrations seemed independent of size. Also for perch and roach the concentrations increased with size, but for roach the trend levels off at approximately 12 cm length. The increase of <sup>137</sup>Cs concentrations with size in

perch is sigmoid, with the steepest slope coinciding with the transition to piscivory. This pattern is the same as earlier reported for mercury, and reflects the biomagnification of Cs along food chains. The relationship between the leveling off concentrations and size can be attributed partly to a gradual ontogenetic diet shift toward higher trophic levels, partly to a decreasing growth efficiency with age.

Significant variation between sampling occasions was observed in several species, typically with a maximum in spring and a minimum in autumn. A seasonal variation in <sup>137</sup>Cs has previously been observed in North American populations and could successfully be described by a bioenergetic model (Rowan, Rasmussen & Chant, 1997). In order to distinguish seasonal patterns induced by annual cycles in ecology (e.g. diet) or physiology (e.g. reproduction) from temperature effects on metabolic kinetics, a temperature-corrected physiological time scale was developed to replace the linear time scale. Both the observations and the model graphs are plotted on a linear time scale, which explains the model's undulating appearance (Fig 1). Typically changes in <sup>137</sup>Cs concentrations are fastest, i.e. slopes are steepest, during summer. In support of the existence of a seasonal pattern the use of a physiological time scale did in most cases clearly improve the R<sup>2</sup> of the model. The impact of annual cycles in ecology and physiology is discussed elsewhere (Sundbom et al., in prep.).

While the species and trophic level of the fish had implications for the timing of both the maximum and leveling off (i.e. the width of the curves' "hump"), lake type was of importance for the levels of maxima (fallout corrected), especially in relation to the leveling off concentrations (height of humps). In Flatsjön the ratios between the model maxima and the corresponding leveling off levels were along the whole trophic gradient lower than in Siggeforasjön (Fig 2) The most apparent difference between these lakes are their morphologies (Table 1). Therefore it is believed that the low max/base-ratios in non-stratified Flatsjön result from frequent resuspension events. During the first months after fallout, when lakes were highly contaminated, suspended particles would bind <sup>137</sup>Cs and thereby reduce biouptake in plankton which subsequently lead to comparatively lower concentrations in fish. During later phases when total <sup>137</sup>Cs in the water is substantially lower resuspension might have the opposite effect by transferring radiocesium from surface sediment into the water column . The significance of this latter pathway, as is the bioavailability of Cs bound to sediment particles, is poorly known. One important conclusion, however, is that even if geochemical and physical processes differ between lakes, their response of the fish community is concordant, i.e. the contamination pattern at one trophic level relative to that of

another trophic level is maintained. This is illustrated by the striking similarity in fish community contamination patterns between Flatsjön and Siggeforasjön (compare the columns in Fig. 2). Absolute values differ due to differences in fallout, morphology etc. This inertia of the food web implies that its cesium dynamics may be partly uncoupled from other processes and would, if properly described and quantified, simplify the task by cross system modelers.



Figure 2. The relationship between some parameters of the nonlinear model and the trophic level of the fish. The trophic level indices are based on the relative contribution of different food sources: 1 = herbivores, 2 = invertebrate feeders, 3 = piscivores. The encircled dots are perch, other high trophic level dots are pike and all dots to the left of perch are the different cyprinids and Ruffe. A) The time in days from the pulse contamination until maximum levels were reached. B) Estimated long-term equilibration levels (assuming no physical decay). C) Calculated number of years from the pulse until concentrations return to the radiation protection guideline of 1500 Bq kg<sup>-1</sup> w.w. Physical decay is here accounted for. D) Ratio between maximum and long-term leveling off levels of <sup>137</sup>Cs in fish.

The nonlinear model could describe the contamination of fish over time very well (fig 1). In other words most of the variation in <sup>137</sup>Cs among years and taxa and size classes could be accounted for by simple empirical functions. Using the model for prediction of how many years it will take to reach the radiation protection guideline of 1500 Bq kg<sup>-1</sup> w.w., shows that this time, consequently, also is a function of trophic level with Such functions are not only useful for predictive purposes, but also facilitate comparisons among and within systems. The parameters of these functions could be related to the ecology (trophic level) and physiology (growth rate) of the fish, and are probably influenced by geochemical processes in the watershed and sediments. Our evaluation indicates dramatic changes over time in the relative importance of different processes.

### 4. Acknowledgment

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## **REPORT 11 Results of the study on surface runoff from a catchment** to Lake Päijänne, Asikkalanselkä

Timo Jaakkola, \*Ritva Saxén & \*Aino Rantavaara University of Helsinki and \*Finnish Centre for Radiation and Nuclear Safety

## 1. Introduction

Some small sub-catchments discharging through small rivers into Asikkalanselkä, the southern part of Lake Päijänne, were chosen to study the transfer of long lived radionuclides <sup>137</sup>Cs and <sup>90</sup>Sr from catchment to the lake. Results on <sup>137</sup>Cs and <sup>90</sup>Sr obtained in the study for one small catchment in 1996 are given.

The average runoff in Finland is about 310 mm/y. Spring runoff, which is formed of snow melt and precipitation, is on the average 100-120 mm in southern Finland, where the share of spring runoff about the annual runoff is 40 - 50%. Spring runoff is about 80% about the average maximum of the water equivalent of snow cover. Runoff is lowest in the lake area of southern Finland, 5 - 7  $l/s \cdot km^2$ , where evaporation is about 2/3 of the precipitation.

## 2. Material and methods

The area of the catchment was about  $0.86 \text{ km}^2$ . The small river studied begins from a bog and flows through areas of mostly forest, discharging finally into lake Päijänne, Asikkalanselkä. Distribution of the land use of the catchment was determined and is as follows:

| Puutteenpohja | distribution (in %) |
|---------------|---------------------|
| Forest        | 84.6                |
| Bog           | 4.0                 |
| Field         | 1.3                 |
| Meadow        | 10.1                |
| Lake          | -                   |

Table 1. Distribution of the land use of the Puutteenpohja catchment.

### 2.1 Soil

Soil profiles of about 20 cm from five different points along the flow of the river were taken and analysed for radiocesium, some samples also for <sup>90</sup>Sr. Short description of the five soil sampling points at Puutteenpohja is as follows:

Point 1: Forest and open coastal area. Point 2: Forest Point 3: Forest Point 4: Bog with sphagnum Point 5: Old field, ditched, birches

#### 2.2 Water

Water samples were taken from the small river near the soil sampling points 1, 3 and 4. Sampling points of soil and water and location of the catchment with respect to lake Päijänne, Asikkalanselkä, are given in Fig.1.

### 3. Results

#### 3.1 Soil

Total amounts of <sup>137</sup>Cs in soil samples at Puutteenpohja varied from 48 800 (point 2) to 71 600 (point 4) Bq/m<sup>2</sup>. In the sampling points, characterised by forest (points 2 and 3), maximum amounts of <sup>137</sup>Cs were found at the depth of 3-7 cm. At the point of old field, the top of <sup>137</sup>Cs was just at the surface layer of the ground (point 5). In the sampling point which is characterised by bog area, the peak of <sup>137</sup>Cs was found deeper, at the depth of 8-12 cm. At the point nearest the lake the peak of <sup>137</sup>Cs was broader than at the other stations studied (Fig.2).

Total amounts of <sup>90</sup>Sr were 1400 and 3200 Bq/m<sup>2</sup> at points 1 and 5 at Puutteenpohja catchment, respectively. Vertical distribution of <sup>90</sup>Sr in soil profile at point 1 shows higher mobility of <sup>90</sup>Sr compared to <sup>137</sup>Cs. At point 5 the peaks of both <sup>90</sup>Sr and <sup>137</sup>Cs are at the uppermost three centimetres (Fig.3).

### 3.2 Water

<sup>137</sup>Cs in water of the river at Puutteenpohja catchment was highest near the soil sampling point characterised by bog with sphagnum (point 4), 310 Bq/m<sup>3</sup>, and decreased when approaching the lake Päijänne (Table I). At the point nearest to the lake <sup>137</sup>Cs content was about the same as in water of Asikkalanselkä (sampling point A3), 69 Bq/m<sup>3</sup>. <sup>90</sup>Sr in water was also highest near the soil sampling station 4, characterised by wet area. At the two other points <sup>90</sup>Sr was somewhat more than one half of the value at point 4 (Fig. 4).

|         | Wa                                     | ter                                   | Soil                       |                                 |  |
|---------|--|---------------------------------------|----------------------------|---------------------------------|--|
|         | <sup>137</sup> Cs [Bq/m <sup>3</sup> ] | <sup>90</sup> Sr [Bq/m <sup>3</sup> ] | 137Cs [Bq/m <sup>2</sup> ] | $^{90}$ Sr [Bq/m <sup>2</sup> ] |  |
| Point 1 | 69                                     | 8.0                                   | 53 453                     | 1 369                           |  |
| Point 2 |  |                                       | 48 832                     |                                 |  |
| Point 3 | 220                                    | 7.3                                   | 66 014                     |                                 |  |
| Point 4 | 310                                    | 13                                    | 71 555                     |                                 |  |
| Point 5 |  |                                       | 68 936                     | 3223                            |  |

Table 2. <sup>137</sup>Cs and <sup>90</sup>Sr in water of the small river (Puutteenpohja) discharging into lake Päijänne, and at five soil sampling points (total amounts) 17.06.1996.

### 4. Discussion

Results of <sup>137</sup>Cs in water of the river from Puutteenpohja show that more <sup>137</sup>Cs is leached into water from wet area than from other type of catchment. <sup>137</sup>Cs was almost five times higher at the near bog point than at the sampling point near the river outlet. During the river flow <sup>137</sup>Cs has decreased gradually by binding into sinking material and transferring with it into the bottom of the river. In the nearest point to the outlet the <sup>137</sup>Cs contents of water is almost the same as in the water of Päijänne, Asikkalanselkä.

Also strontium is leached more from wet areas than from other types of catchments. The slighter decrease of  ${}^{90}$ Sr during the river flow is due to that its affinity to suspended matter of water and transfer with that into the sediment is less than that of  ${}^{137}$ Cs. Contents of  ${}^{90}$ Sr at the point near outlet (point 1) was about 60% of that at the point near the river start, while the corresponding figure for  ${}^{137}$ Cs was about 20%.

The ratio <sup>137</sup>Cs in water of the river / <sup>137</sup>Cs in soil at the catchment showed that transfer from soil to water ten years after the deposition is more than three times higher in wet soil than other type of soil. Transfer of <sup>90</sup>Sr is almost five fold compared to that of <sup>137</sup>Cs at the river outlet. (Values for <sup>90</sup>Sr in soil at wet area are missing, so the transfer there cannot be calculated).

Our results give information only at one time point ten years after deposition. Therefore, we can see neither effects of seasonal variations on leached amounts nor their other changes since deposition. The role of radionuclides leached from catchment seems to be rather small from the recipient lake's point of view ten years after the deposition, especially in this case, where the recipient lake is very large and the river very small. The situation may be quite different in case of a smaller lake and a larger river even ten years after deposition and has certainly been higher during shorter period after deposition even in this case.



Figure 1. Catchment of the river studied (Puutteenpohja) with estimated distribution of the area to forest, bog, field and meadow. M = Forest, 84.6 %, S = Bog, 4%, P = Field, 1.3%, N = Meadow, 10.1%. Sampling points 1-5 are also given. Soil samples were taken from all five points, water samples from the river close to the soil sampling points 1, 3 and 4.


Figure 2. Vertical distribution of  $^{137}$ Cs in soil at five sampling points in Puutteenpohja catchment 17.06.1996 (Bq/m<sup>2</sup>).



Figure 3. Vertical distribution of  $^{90}$ Sr in soil at two sampling points in Puutteenpohja catchment 17.06.1996 ( $Bq/m^2$ ).



*Figure 4.* <sup>137</sup>*Cs and* <sup>90</sup>*Sr in water of the river flowing from a small catchment of Puutteenpohja to lake Päijänne on 17.06.1996.* 

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