



Nordisk kernesikkerhedsforskning
Norrænar kjarnöryggisrannsóknir
Pohjoismainen ydinturvallisuustutkimus
Nordisk kjernesikkerhetsforskning
Nordisk kärnsäkerhetsforskning
Nordic nuclear safety research

NKS-35

ISBN 87-7893-086-3

Summaries of studies carried out in the NKS/BOK-2 project

Technical report

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December 2002

Abstract

Summaries of studies carried out in the NKS/BOK-2 project, *Radiological and Environmental Consequences*. The structure of the project as such is described in NKS-64, *Radiological and Environmental Consequences – Final Report of the Nordic Nuclear Safety Research Project BOK-2*. That report also includes compilations based on the summaries presented in this report. The project was carried out 1998-2001 with participants from all the Nordic countries. Representatives from the Baltic States were also invited to some of the meetings and seminars. The project consisted of work on terrestrial and marine radioecology and had a broad scope in order to enable participation of research groups with various fields of interest. The topics included improving assessment of old and recent fallout, use of radionuclides as tracers in Nordic marine areas, improving assessment of internal doses and use of mass spectrometry in radioecology. This report is a compilation of summaries from each research group, 32 papers in all, and gives references to papers published in scientific journals. Some of the studies have been described previously, at least to some degree, in NKS-70, *Proceedings of the 8th Nordic Seminar on Radioecology, 25-28 February 2001, Rovaniemi, Finland*.

Key words

Radioecology, radioactive fallout, sheep, radiocaesium, radiostrontium, technetium, seaweed, oceanic tracers, mass spectrometry, internal doses, doses to man

NKS-35
ISBN 87-7893-086-3

Pitney Bowes Management Services Denmark A/S, 2003

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Introduction

This report is a compilation of 32 summary papers, each written by a research group participating in the NKS/BOK-2 project, *Radiological and Environmental Consequences*. Most of this work has been described in papers published in scientific journals or conference proceedings. Reference to these papers can be found in the summaries. A general summary of the results obtained in the BOK-2 project can be found in the report NKS-64, *Radiological and Environmental Consequences – Final Report of the Nordic Nuclear Safety Research Project BOK-2*. That report also gives more information about the structure of the project work. The introductory information on the BOK-2 project that follows in this section is similar to what is presented as summary in NKS-64.

The NKS/BOK-2 project, *Radiological and Environmental Consequences, 1998-2001*, focuses on radioecology in the Nordic countries and areas of interest to them. The NKS has in previous programme periods included work on radioecology, and many basic questions concerning assessment of risk due to environmental radioactivity have been answered. One may then ask, why should one continue work in this field? This question, by itself, reflects the problem. Why should talented young people enter a field where many important questions have already been answered and the interest of society in nuclear methods is decreasing? In fact modern society still has pressing questions. The questions have changed, but answers are still urgently required and it seems there are fewer competent persons able to provide the answers.

The changed nature of questions asked by society can be seen from more requirements for studies to prove that there are no significant health risks from environmental radioactivity and for tracer studies to help understand processes and predict development of very low environmental concentrations. Risk and health related assessments are, however, still important. They form the backbone of emergency response plans. The threat scenarios change from time to time and thus new assessments are called for.

An important aim of the BOK-2 project was thus to provide a stimulating environment and to encourage contacts and co-operation between young and experienced researchers, between scientists in different fields (within and outside traditional radioecology) and between scientists within the Nordic countries and neighbouring regions. This was done through meetings, seminars and dissemination of information, including use of the Internet. The Nordic network within radioecology is important for national authorities and for new people in the field and for making it possible to start close co-operation quickly between countries, e.g. if needed because of a nuclear accident.

When organising the project work within BOK-2, a choice had to be made between having a project with a broad range of activities and many participants, and narrowing the focus, obtaining more depth in a few fields, at the expense of limiting the number of participants. No one ideal solution can be found, what might suit one country can be inappropriate for another. The conclusion was to have a broad range of activities, build on existing project work in each country as much as possible and try to build-up a large network with many participants. Some of the research groups that were involved in the core activities of BOK-2 were also willing to contribute with work in other fields, without seeking extra financial support from the NKS. This meant that the final report would become less focused. But this was considered a small price to pay for the positive effect of strengthening the networking aspect of the project and making it a more interesting forum (e.g. for young scientists).

A note on possible problems with colour / greyscale in illustrations

Please note that some of the papers contain illustrations, originally in colour or greyscale, which may not have been reproduced well in the printed version of the report. It is however available from the NKS as a PDF file, with the original colour/greyscale illustrations. Selected illustrations or papers can be viewed and printed from there.

Overview of structure of the BOK-2 project

In accordance with the suggestions of the NKS programme group, it was decided to structure the BOK-2 project as follows:

BOK-2.1 Important Nordic food chains

BOK-2.1.1 Radioecological vulnerability

BOK-2.1.2 Internal doses

BOK-2.2 Radioactive tracers in Nordic sea areas

BOK-2.2.1 Sea water transport

BOK-2.2.2 Biological and biogeochemical processes

BOK-2.3 Applications of ICP-MS for measuring radionuclides

BOK-2.4 Methodology for defining exemption levels of radionuclides in timber.

BOK-2.1 Important Nordic food chains

In BOK-2.1.1 the main emphasis was on using old fallout data to improve methods of estimating the effects of radionuclide deposition. This was done by using traditional UNSCEAR models on a combined data set of fallout and Chernobyl data, and by using precipitation data to predict deposition.

The aim with BOK-2.1.2, Internal doses, was to improve methods for dose calculations based on dietary methods (indirect method) and whole-body counting (direct method). It has e.g. involved two courses with practical exercises, calibration and intercalibration of equipment and preparation of a handbook for use in emergency situations.

BOK-2.2 Radioactive tracers in Nordic sea areas

BOK-2.2.1 focused mainly on radioactive tracers in Nordic waters, Tc-99, Cs-137 and to a lesser degree, I-129. Particular use was made of the Tc-99 peak in release from Sellafield in 1995. This release has been followed through the Danish straits into the Baltic Sea (with Cs-137 moving in the opposite direction) and along the Norwegian coast into the Arctic Ocean. At the end of the project period, no significant increase of Tc-99 had been observed at the Faroe Islands, but indications of increased concentrations in seaweed were found at the northern coast of Iceland.

The BOK-2.2.2 part focused on processes in the Baltic Sea. Main emphasis was on evaluating existing sediment data, comparing it with recent data, improving the coverage of sampling in the Gulf of Bothnia and improving the knowledge on the role of sedimentation in losses of radionuclides from the water column to the seabed. The last part of this study was to investigate the role of river discharges from Finland into the Baltic Sea.

BOK-2.3 Applications of ICP-MS for measuring radionuclides

BOK-2.3 was introduced late in the project period in order to meet increasing interest in investigating the applications of mass spectrometry for measuring long-lived radionuclides. It involved experimental studies and a training course, including an opportunity for work on own samples.

BOK-2.4 Methodology for defining exemption levels of radionuclides in timber

BOK-2.4 was also introduced late in the project period. It involved a study on methodology for defining exemption levels for radionuclides in timber.

The BOK-2 studies – A list of summary papers

BOK-2.1, Important Nordic food chains

BOK-2.1.1 – Radioecological vulnerability

Radioecological vulnerability or sensitivity to radioactive fallout was one of the core themes in the BOK-2 project. This was dealt with in the following six papers, all which also were extracting new information from old data sets.

- **Re-evaluation of Danish Fallout Data**, p. 6
Sven P. Nielsen and Mette Øhlenschläger
- **Dietary Cs-137 and Sr-90 in Finland in 1960-2000**, p. 10
Aino Rantavaara, Ritva Saxén and Eila Kostiainen
- **Modelling long-term variation of radioactivity in foodstuffs in the Faroe Islands**, p. 19
Hans Pauli Joensen
- **Radioactive fallout in Norway from atmospheric nuclear weapons tests**, p. 25
Tone D. Bergan
- **Estimating Cs-137 fallout inventories in Iceland from precipitation data**, p. 33
*Sigurður Emil Pálsson, Ólafur Arnalds, Magnús Á. Sigurgeirsson, Kjartan Guðnason
Brenda J. Howard, Simon M. Wright, Þórunn Pálsdóttir*
- **Radiocaesium (Cs-137) fallout in Iceland and its behaviour in Sub-Arctic volcanic soils**, p. 40
Magnús Á Sigurgeirsson, Ólafur Arnalds, Sigurður Emil Pálsson and Kjartan Guðnason

BOK-2.1.1 - Long term trends and roots of variability in semi-natural ecosystems, Cs-137 in lamb meat

This was a low-key continuation of a series of NKS studies started in 1990. In some cases the studies had been continued in order to obtain a better estimate of long term trends, in other cases the study developed into investigating roots of variability masking the long term trends.

- **Transfer of Cs-137 from soil to plants and lamb meat in Iceland**, p. 45
Sigurður Emil Pálsson, Jóhann Þórsson, Magnús Á. Sigurgeirsson, Eiríkur Þorkelsson and Kjartan Guðnason
- **Radiocaesium in Soil, Grass and Lamb at Ribe**, p. 52
Sven P. Nielsen
- **Cs-137 in the food-chain of lamb in the Faroe Islands. Measurements in the period 1990-2000**, p. 55
Hans Pauli Joensen
- **Transfer of Cs-137 from soil to plants and sheep at Tjøtta, Norway, 1988-1998**, p. 61
Ingar Amundsen

BOK-2.1.1 - Additional work contributed by research groups

- **Cs-137 concentrations in mushrooms collected in 1989-2000 at different forest stands in Kivalo experimental area**, p. 65
Kristina Rissanen, Jarkko Ylipietä and Yrjö Norokorpi
- **Sr-90 in cow's and goat's milk in Norway**, p. 70
Astrid Liland, Elisabeth Strålberg, Trine Kolstad, Arne Sæbø
- **A Study of Plutonium in different fresh water systems**, p. 77
Mats Eriksson, Elis Holm, Per Roos, Henning Dahlgaard
- **Field Sampling, Preparation Procedure and Plutonium Analyses of Large Freshwater Samples**, p. 89
Elisabeth Strålberg, Trygve O. Bjerk, Kristin Østmo and John. E. Brittain
- **Plutonium in coniferous forests**, p. 93
Aino Rantavaara and Eila Kostiainen

BOK-2.1.2 Internal doses

There are 2 papers in BOK-2 Technical Report describing this work (one is actually an edition of a handbook produced within the project):

- **Assessment of internal doses, p. 96**
Tua Rahola, Rolf Falk, Mats Isaksson, Lavrans Skuterud and Göran Ågren
- **Mätning av radioaktiva ämnen i människa i beredskapssituationer. En metodhandbok vid jod och cesium-kontaminering (in Swedish), p. 107**
Tua Rahola, Rolf Falk, Mats Isaksson

BOK-2.2 Radioactive tracers in Nordic sea areas

The following 12 papers in the BOK-2 Technical Report summarise the results from studies within the project on the marine environment.

- **Timing of fluxes in the Nordic Seas - overview, p. 122**
Svend Aage Malmberg
- **Long term study of Tc-99 in brown seaweed from the Swedish west coast, p. 125**
P. Lindahl, T. Gäfvert, P. Roos, S. Mattsson, B. Erlandsson and E. Holm
- **Radiative Tracers in Nordic Waters, Tc-99, Cs-137 and I-129, p. 129**
Henning Dahlgard, Xiaolin Hou, Sven P. Nielsen
- **Outflow of radiocaesium from the Baltic Sea detected in brown algae along the southern Norwegian coast, p. 134**
Elisabeth Strålberg and Gordon C. Christensen
- **Tc-99 in Norwegian Marine Environments, p. 140**
Anne Kathrine Kolstad, Justin Brown and Anne Liv Rudjord; Elisabeth Strålberg, and Gordon C. Christensen; Deborah H. Oughton
- **Cs-137 and Tc-99 in the ocean and at the shores around Iceland related to timing of fluxes in the Nordic Seas, p. 152**
Elisabet Dolinda Ólafsdóttir, Magnús Á Sigurgeirsson, Þórhallur Ingi Halldórsson, Svend Aage Malmberg, Sigurður Emil Pálsson, Kjartan Guðnason
- **Cs-137 and Tc-99 in seawater and seaweed around the Faroe Islands, p. 161**
H. P. Joensen, T. Vestergaard, J. Zachariasen and M. Mortensen
- **A sediment trap experiment to study seasonal variations in vertical fluxes in the Baltic sea, p. 165**
Per Roos, Örjan Gustafsson and Per Andersson
- **Use of Tc-99 as tracer for Sellafield discharges in the Baltic Sea: Technetium-99 in biota samples collected along the Finnish coast in 1999, p. 170**
Erkki Ilus, Vesa-Pekka Vartti, Tarja K. Ikäheimonen, Jukka Mattila and Seppo Klemola
- **Radionuclide processes in the Baltic Sea and its catchment and the adjacent areas: Discharge of Cs-137 and Sr-90 by Finnish rivers to the Baltic Sea in 1986-1996, p. 172**
Ritva Saxén and Erkki Ilus
- **Old data on Sr-90, Cs-137 and Pu-239,240 in Baltic Sea sediments prior to the Chernobyl accident (1966-1980), p. 173**
Erkki Ilus and Jukka Mattila
- **Cs-137 in the sediments of the Gulf of Bothnia: additional data on total inventory in sediments, p. 175**
Jukka Mattila and Erkki Ilus

BOK-2.3 Applications of ICP-MS for measuring radionuclides

The aim with work within this sub-project was to introduce possibilities (and limitations) of inductively coupled plasma mass spectrometry (ICP-MS) to Nordic participants. The work involved a training course and experimental studies.

- **Comparison of two ICP-MS systems for measuring Tc-99 in large volume samples, p. 178**
Miranda J. Keith-Roach, Stefan Stürup, Deborah H. Oughton and Henning Dahlgaard
- **Test of using ICP-MS for fast determination of Tc-99 in Icelandic coastal waters, p. 189**
Pórhallur Ingi Halldórsson, Niels Óskarsson, Magnús Á Sigurgeirsson, Kjartan Guðnason, Sigurður Emil Pálsson

BOK-2.4 Methodology for defining exemption levels of radionuclides in timber

Timber industry is very important in many Nordic areas. The NKS Board therefore approved that a study could be included in BOK-2 on the methodology for defining exemption levels for radionuclides in timber

- **Methodology for Defining Exemption Levels of Radionuclides in Timber and Wood Products, p. 196**
Elis Holm

Appendix: Related paper from the EKO-2 project

During the process of publishing and printing the final report of the EKO-2 project, one of the project papers was left out by mistake. It was to have been included in NKS(97)FR5 *Ecological half-lives of radioactive elements in semi-natural systems*. This missing paper is included in the compilation report published now, since it is relevant for other work being reported and the missing paper has been referred to in other papers. This was decided in consultation with the leader of the former EKO-2 project. It was also decided to place this paper in an appendix to the report, since it has not been updated since being written in 1997 and its style is different from the other papers.

- **Transfer of radiocaesium from soil to vegetation and to grazing lambs in Iceland – Icelandic part of the NKS/EKO-2.1 project, p. 206**
Sigurður Emil Pálsson, Jóhann Þórsson and Elísabet D. Ólafsdóttir

Re-evaluation of Danish Fallout Data

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Introduction

Data on environmental radioactivity in Denmark have been collected at Risø National Laboratory since the late 1950's covering abiotic samples, vegetation, foodstuffs and humans (Aarkrog et al., 1995, and previous reports). This collection has generated time series of data that illustrate the transfer of anthropogenic radioactivity through foodchains to the Danish population from a variety of sources including fallout from atmospheric nuclear weapons testing and from the Chernobyl accident, and discharges from European nuclear reprocessing facilities. The information obtained from the analysis of these data is useful in connection with predicting radiological consequences from potential nuclear accidents involving radioactive contamination of the Danish environment and for identifying effective countermeasures.

The radioecological sensitivity (Aarkrog, 1979) is a useful concept for characterising the transfer of radioactive contamination to different environmental compartments including man. The time-integrated dose rate to man is the relevant end point of a radiological assessment. Therefore the radioecological sensitivity is defined as the time-integrated concentration in an environmental sample from a unit ground deposition (e.g. Bq y kg⁻¹ per Bq m⁻²).

The data on environmental radioactivity are analysed with different radioecological models in order to obtain numerical values of radioecological sensitivities and to provide a comparison between models.

Fallout Data and Models

The data used here comprise observed levels in Denmark of the radionuclides ⁹⁰Sr and ¹³⁷Cs in precipitation, grass, cow milk, beef and diet. The data are given for Jutland, the western part of the country, and for the Islands, the eastern part. Basic references for the data are the time series of annual fallout shown in Fig. 1 for ⁹⁰Sr and ¹³⁷Cs in Jutland. Figure 2 shows the time series for ⁹⁰Sr and ¹³⁷Cs in milk from Jutland.

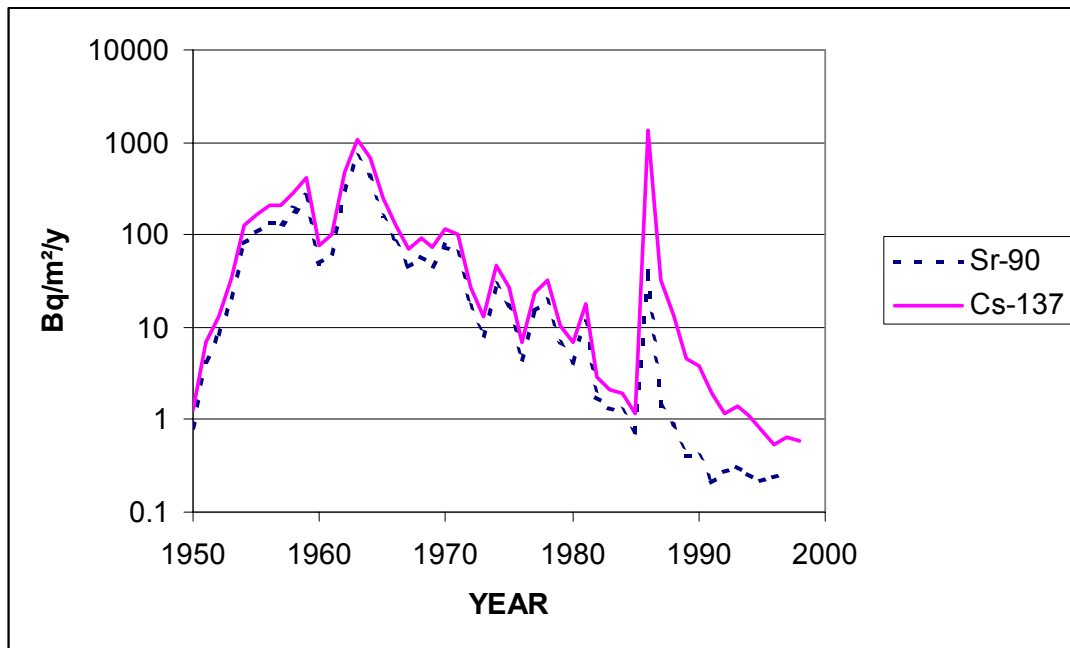


Fig. 1. Annual fallout (Bq m^{-2}) of ^{90}Sr and ^{137}Cs in Jutland.

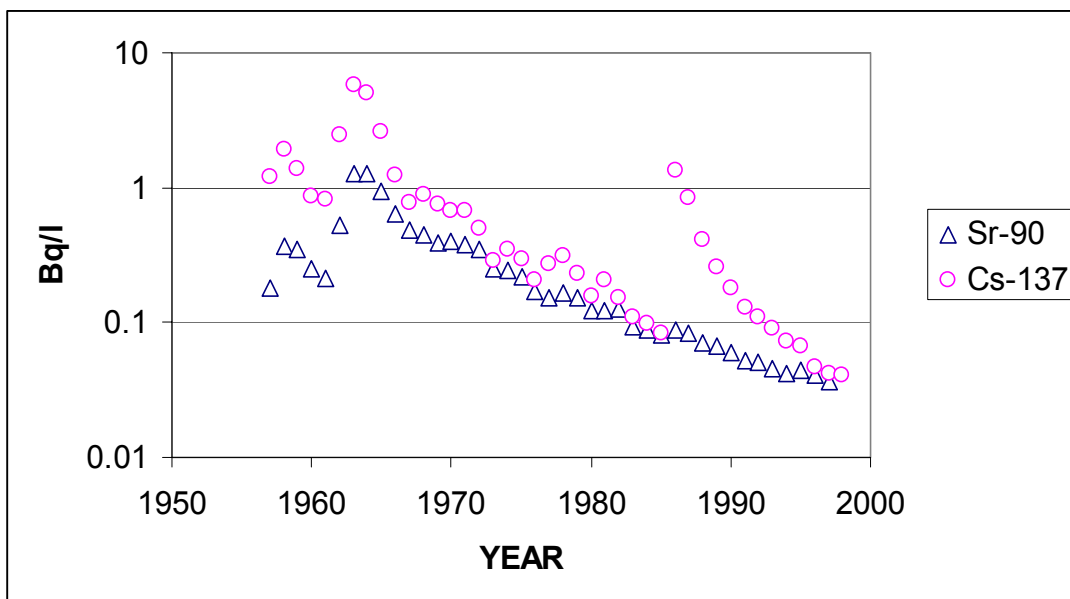


Fig. 2. Annual average concentrations (Bq l^{-1}) of ^{90}Sr and ^{137}Cs in milk from Jutland.

Mathematical models accounting for the transfer of deposited radioactivity to the human diet and further to man were developed in the 1960's and adopted by UNSCEAR (e.g. UNSCEAR, 1972; Aarkrog, 1979). The derivation of these models involves the determination of parameters of transfer functions by least-squares fitting of diet and deposit data. This type of models may be applied for any environmental compartment for which data are available. Radioecological sensitivities are calculated from the parameters of the transfer functions. The derivation of these models may be carried out from linear regression analyses of the data by spreadsheet (e.g. Excel) calculations.

More recent radioecological models are dynamic and mechanistic in the sense that they incorporate explicit descriptions of individual transfer processes, e.g. deposition, resuspension, exchange between soluble and insoluble phases, root uptake, weathering, uptake and excretion by animals. These models are more complex and require more computing power. Two such models were used here: ECOSYS (Müller and Pröhl, 1993) and FARMLAND (Brown and Simmonds, 1995). ECOSYS was available in an Excel version and FARMLAND in a version imbedded in a software package (PC-CREAM, 1997). Neither of these versions were well suited for the purpose of making comparisons with the above mentioned data since the models were intended for other purposes. But it was possible from both model versions to extract the transfer of ^{90}Sr and ^{137}Cs to grass, milk and beef from a unit ground deposition and fold this transfer with the annual deposition data from Denmark (cf. Fig. 1).

Radioecological Sensitivities

Transfer functions of the UNSCEAR model concept were fitted to the data, generally using all available data. In a few cases, however, data on ^{137}Cs were omitted from the fitting for the years 1986 and 1987 due to the different environmental transfer found in these years from fresh Chernobyl fallout compared to fallout from nuclear weapons testing. The prediction models are shown in Tables 1 and 2.

Table 1. UNSCEAR prediction models for ^{90}Sr in grass, milk, beef and diet.

	Jutland	Islands
Grass (mBq/kg fw)	$20 d_i + 13 d_{i-1} + 0.73 A_{28}$	$14 d_i + 28 d_{i-1} + 0.45 A_{28}$
Milk (mBq/l)	$1.2 d_i + 0.8 d_{i-1} + 0.21 A_5 + 0.013 A_{28}$	$0.9 d_i + 0.7 d_{i-1} + 0.14 A_5 + 0.015 A_{28}$
Beef (mBq/kg)	$0.45 d_i + 0.078 d_{i-2} + 0.027 A_{10}$	
Diet (mBq/d/cap)	$2.1 d_i + 1.8 d_{i-1} + 0.22 A_{10}$	$2.2 d_i + 1.8 d_{i-1} + 0.22 A_{10}$

Table 2. UNSCEAR prediction models for ^{137}Cs in grass, milk, beef and diet.

	Jutland	Islands
Grass (mBq/kg fw)	$21 d_i + 0.9 d_{i-1} + 0.15 A_{30}$	$14 d_i + 1.1 d_{i-1} + 0.74 A_5$
Milk (mBq/l)	$5.3 d_i + 2.1 d_{i-1} + 0.12 A_2 + 0.032 A_{30}$	$2.6 d_i + 1.6 d_{i-1} + 0.017 A_{30}$
Beef (mBq/kg)	$32 d_i + 0.16 A_{30}$	
Diet (mBq/d/cap)	$5.4 d_i + 4.3 d_{i-1} + 2.4 d_{i-2} + 0.083 A_{10}$	$4.7 d_i + 4.3 d_{i-1} + 2.2 d_{i-2} + 0.11 A_{10}$

Table 3. Radioecological sensitivities from UNSCEAR prediction models.

Sensitivity	Sr-90		Cs-137	
	Jutland	Islands	Jutland	Islands
Grass (mBq y/kg fw)	63 ± 18	60 ± 4	28 ± 2	20 ± 2
Milk (mBq y/l)	3.9 ± 0.5	3.2 ± 0.6	9.1 ± 1.4	4.9 ± 0.2
Beef (mBq y/kg)	0.9 ± 0.1		38 ± 3	
Diet (Bq/cap)	2.6 ± 0.1	2.6 ± 0.1	4.9 ± 1.0	4.6 ± 0.9

The radioecological sensitivities calculated from these prediction models are shown in Table 3 giving the time-integrated transfer from a deposition of 1 Bq m^{-2} . The sensitivities found from the ECOSYS and FARMLAND models are shown in Table 4.

Table 4. Radioecological sensitivities based on an application of the ECOSYS and FARMLAND models to Danish fallout data.

Sensitivity	Cs-137, Islands	
	ECOSYS	FARMLAND
Grass (mBq y/kg fw)	130	28
Milk (mBq y/l)	18	5.5
Beef (mBq y/kg)	47	28

Conclusions

Traditional radioecological models of the UNSCEAR type have been applied to time series of Danish data on ^{90}Sr and ^{137}Cs in grass, milk, beef and diet covering four decades. The models fit the data well for ^{90}Sr for the entire period, but for ^{137}Cs the models generally perform somewhat poorer from 1986 and onwards. The reason for this is the different transfer through the environment particularly in 1986 and 1987 of Chernobyl radiocaesium compared to that from nuclear weapons testing. This difference is caused mainly by different seasonal modes of deposition. But overall, the models reproduce the data quite well. The radioecological sensitivity of the Danish diet for the transfer of ^{90}Sr and ^{137}Cs through Danish foodchains is found to represent an average individual intake of 3 Bq ^{90}Sr and 5 Bq ^{137}Cs for a ground deposition of 1 Bq m^{-2} when no countermeasures are applied.

Dynamic radioecological models do not reproduce the fallout data better than the traditional models. But the general features of the dynamic models make them suited for prediction of radiological consequences of routine and accidental releases in areas where limited radioecological data are available.

Acknowledgement

The study was financially supported by Nordic Nuclear Safety Research.

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Dietary ^{137}Cs and ^{90}Sr in Finland in 1960 – 2000

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Introduction

Estimation of ingestion doses from environmental strontium 90 (^{90}Sr) and caesium 137 (^{137}Cs) through measured concentrations in Finnish foodstuffs has been one important aim of the regular foodstuff surveillance at STUK for decades. In northern Europe milk contributes significantly to the collective dose from these radionuclides due to its considerable consumption as a drink and its use in cooking. Foods from seminatural food chains, like freshwater fish, are important long-term sources of ^{137}Cs in the whole of Fennoscandia.

The aim of this study was to compile the historical data, estimate dietary intake of caesium 137 and derive time-integrated ingestion doses through agricultural produce and freshwater fish during 1960-2000. The measurements of different foodstuffs carried out at STUK do not fully represent the whole period since the beginning of environmental contamination from atmospheric nuclear weapon testing. Accuracy of dose estimation was improved with simple calculation models both by improved weighing the regional measurements and through completing the time series.

Material and Methods

Sampling and analyses

Long-lived radionuclides ^{90}Sr and ^{137}Cs in deposition and in domestic foodstuffs, particularly in milk have been regularly analysed at STUK during the last four decades, some of the sample types since the early 1960's. These data form the basis for estimation of radiation doses from deposition of atmospheric nuclear weapons tests carried out until the late 1970's and from the Chernobyl accident in 1986. Environmental gamma radiation surveys carried out in 1986-1987 essentially improved the data for surface densities of deposited radionuclides after the Chernobyl nuclear accident (Arvela et al. 1990).

The sampling and radionuclide determinations were carried out in accordance with the procedures described in the reports of the series SFL-A, STL-A and STUK-A by several authors (1966-2000). The sampling networks were often nationwide, organised in co-operation with authorities, industry and private persons. The radionuclide determinations were carried out with quality assured radiochemical and gamma spectrometric methods since the early phases of environmental surveillance of artificial radioactivity.

Deposited ^{137}Cs and ^{90}Sr

Measurements of dry plus wet deposition. Deposited ^{90}Sr and ^{137}Cs were monitored on monthly basis since 1961 at seventeen stations in Finland. Data files of annually and monthly deposited ^{137}Cs and ^{90}Sr in Finland were compiled (Fig. 1). The annual average depositions of these nuclides in Finland up to 1985 are the averages of the annually deposited activities at those seventeen stations. Since 1986 the figures for one station, Vaasa, were chosen for the assessments, because the value for ^{137}Cs deposition at Vaasa is close to the countrywide average value determined by measurements of external gamma radiation after the Chernobyl accident.

Environmental gamma radiation surveys. Important addition to the estimation of surface densities of ^{137}Cs activities in Finland has been the fallout survey carried out in 1986-1987 (Arvela et al. 1990). For regional milk and fish activities the transfer factor approach based on this survey and measurements of foodstuffs from the same or corresponding area have been used, and parameter values for simple calculation models derived.

Milk

The regular analysis of long-lived radioactivity in milk has been nationwide since 1960. The method of deriving nation-wide, production weighted means for monthly activity concentrations has been described in research reports of STUK in 1966 – 1992. The sampling areas constituting the network for nationwide sampling covered roughly one third (or 30 – 40%) of the milk production, and represented different conditions for environmental transfer of ^{137}Cs and ^{90}Sr . The differences in activities accumulated in soils, and the varying types of agricultural soils have caused variation in transfer of radionuclides to milk. These variations have been considered in assessment of nationwide means (Figures 2 and 3), which have been used for estimation of average radiation doses received through consumption of milk.

Agricultural produce other than milk

Cereals. Databases on regional and nationwide concentrations of ^{137}Cs and ^{90}Sr in cereals (rye, wheat, oats and barley) since the 1960's were available. The activity concentrations were used to assess the regional and soil type dependent differences in transfer of radionuclides to cereals. Combined with regional production weighted deposition of ^{137}Cs the nationwide means were derived (Kostiainen and Rantavaara, 2001).

Meat. Database on regional and nationwide radiocaesium concentrations of pork and beef were compiled for 1976 - 2000. The measurements were regional until 1985, and were extended to cover the most of beef and pork production in 1986. Thereafter the sampling programme was gradually reduced to include one large delivery area of beef cattle and pork in the late 1990's.

Vegetables and fruit. In 1975 – 1985 the sampling of vegetables and fruit was regional; it became nationwide during the harvest season of 1986. Thereafter the programme was gradually reduced to represent the mostly used products from the main production area.

Freshwater fish

The sampling and analysing programme carried out annually at STUK gave values of ^{137}Cs in fish from various lakes since the main fishing season of 1986. Countrywide annual averages of ^{137}Cs in fish, weighted for catches, were calculated after the Chernobyl accident (Fig. 4). The calculation was based on the results of the surveillance programme and the fish statistics (STUK-A61, STUK-A77, STUK-A94 and STUK-A129). In the pre-Chernobyl period ^{137}Cs in freshwater fishes in Finnish lakes were studied at STUK in 1982 (STL-A 47).

Values for ^{137}Cs activity concentrations in fishes for the years 1956-1985 were estimated using the method in which activity concentrations of surface water were first calculated with a model based on values of annual and accumulated deposition (Rantavaara and Saxén 1985). Transfer coefficients from water to fish were then used to estimate activity concentrations in fishes. Countrywide activity concentrations for ^{137}Cs in freshwater fishes in Finland in 1956-1998 are given in Fig. 4. Comparison of the values estimated by the model and via transfer coefficients with the analysed post-Chernobyl values are given in Fig. 5.

Results and Discussion

Dietary intake of ^{137}Cs and ^{90}Sr

For all food items the annual production data were used for estimation of nationwide means of activity concentrations in different produce. Food preparation losses, both mass reduction in cleaning before cooking, and fractionation of radio-caesium between edible parts and the rest of raw material were considered in intake estimation (Rajama & Rantavaara 1982; Rantavaara 1990). Mean consumption rates for different food items were derived from agricultural statistics or dietary surveys. They were used to calculate the dietary intake from nationwide annual mean concentrations of ^{137}Cs and ^{90}Sr in foodstuffs.

Consumption of freshwater fishes varies annually and in various fishery areas as do the activity concentrations of ^{137}Cs in fishes. Regional variations and consumption of various types of fishes have been taken into account in calculating of the countrywide averages for 1986-1994. Countrywide values for 1995-1998 were estimated on the basis of the results obtained in the analyses in those years and the decrease rate of the countrywide values for the previous years. The consumption of freshwater fish was assumed to be 4 kg per year when estimating average long-term radiation doses.

Ingestion doses from ^{137}Cs and ^{90}Sr

Using the conversion factor of $1.3 \times 10^{-8} \text{ Sv Bq}^{-1}$ for ^{137}Cs and $2.8 \times 10^{-8} \text{ Sv Bq}^{-1}$ for ^{90}Sr the effective committed doses for adults through ingestion were calculated from the dietary intakes (International Atomic Energy Agency, 1996; Table 1). The intake data for ^{137}Cs (Fig. 6) and activity concentrations of ^{90}Sr in milk (Fig. 2) and used for estimation of intake through milk correspond to 1.4 mSv dose for adults in the 41 years. For ^{90}Sr the non-milk foodstuffs may have contributed with almost a double intake compared to milk. Doses from other foodstuffs than agricultural products were estimated with simple assumptions, which indicate some 0.5 mSv additional dose contribution from ^{137}Cs in food types not assessed in this study. In all, our study suggests that adult Finns have on average received through the long-lived nuclides ^{90}Sr and ^{137}Cs in foodstuffs an individual internal dose slightly exceeding 2 mSv in 1960-2000.

Table 1. Committed effective doses received by adult Finns through different types of food in 1960 – 2000.

Food type	^{137}Cs mSv	^{90}Sr mSv
Domestic animal products	0.6	0.1
Vegetable foods of agricultural origin	0.3	0.1 ^a
Freshwater fish	0.3	0.01 ^a
Marine fish, wild forest products, reindeer meat and miscellaneous types of foods	0.5 ^a	0.02 ^a
All foods and drink	1.8	0.3

^a) Doses estimated roughly with a simple approach.

Conclusions

Domestic animal products dominated dietary intake of ^{137}Cs in 1960 – 2000. Freshwater fish and vegetable products of agricultural origin were almost equal contributors of ^{137}Cs to the diet. Wild foods received from forests, assessed using a simple approach, were also non-negligible sources of ^{137}Cs . Marine and brackish-water fishes, although of low activity concentration of ^{137}Cs compared to freshwater fishes, added to the ingestion dose through a higher consumption rate.

Sources of dietary ^{137}Cs during the period of atmospheric nuclear fallout from weapon tests differed from the post-Chernobyl time. Agricultural produce then seemed to dominate the ^{137}Cs received via the Finnish diet more than after the Chernobyl accident (Fig. 6). Subsequent growth periods of considerable stratospheric fallout made the pathways from vegetation to human diet important in the 1960's.

Strontium 90 is mobile in soils for long periods of time. However, it has not been a significant dietary nuclide for Finns during the history of environmental radioactive contamination (Table 1). In the years of maximum deposition rates in early 1960's the doses exceeded more than tenfold the doses in the following decades, and also the doses received after the Chernobyl accident (Rajama and Rantavaara 1982). The wild foods from forests are not likely to contribute to ingestion of ^{90}Sr essentially more than similar types of agricultural and garden products replacing wild foods in the diets of non-pickers and non-hunters.

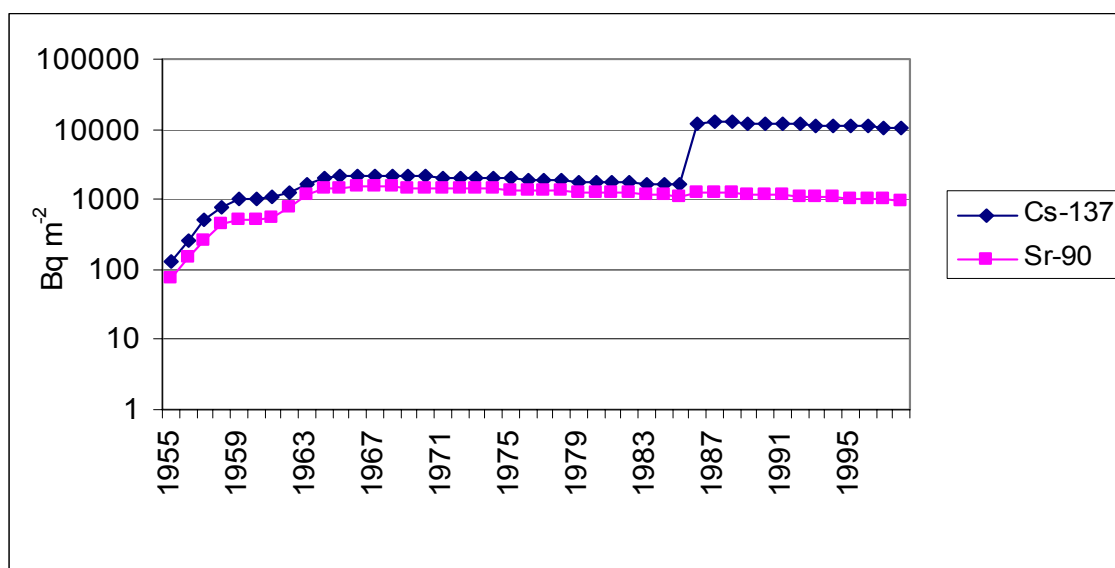


Figure 1. Estimated deposition of ^{90}Sr and ^{137}Cs accumulated in Finland in 1955-1998. In 1960-1985 the values were based on averages of the 17 sampling stations and after that on values in Vaasa (see text). For 1955-1959 data collected in St. Petersburg were used.

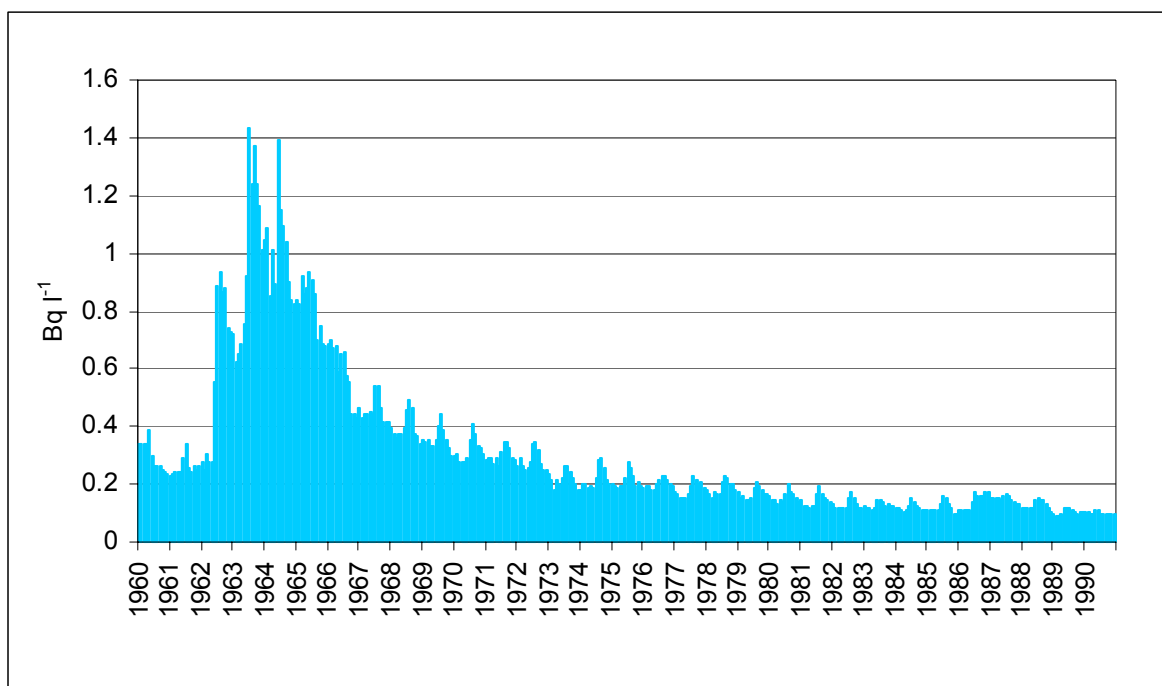


Figure 2. Nation-wide production-weighted monthly means of ^{90}Sr concentrations in Finnish milk since January 1960.

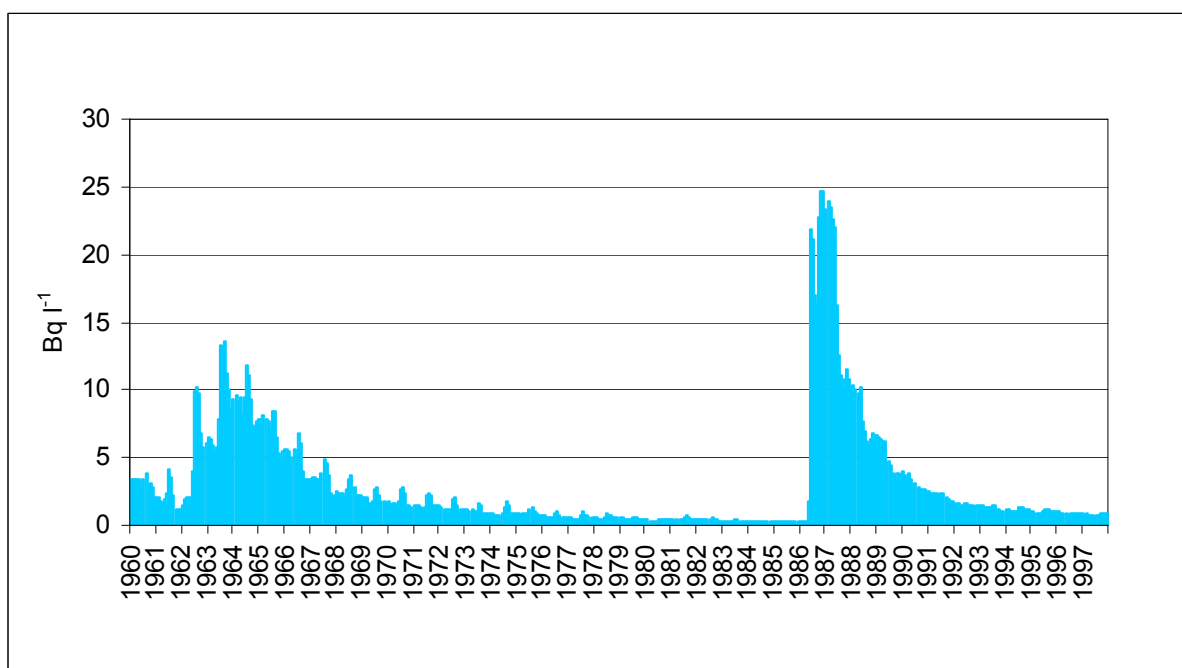


Figure 3. Nation-wide production-weighted monthly means of ^{137}Cs concentrations in Finnish milk since January 1960.

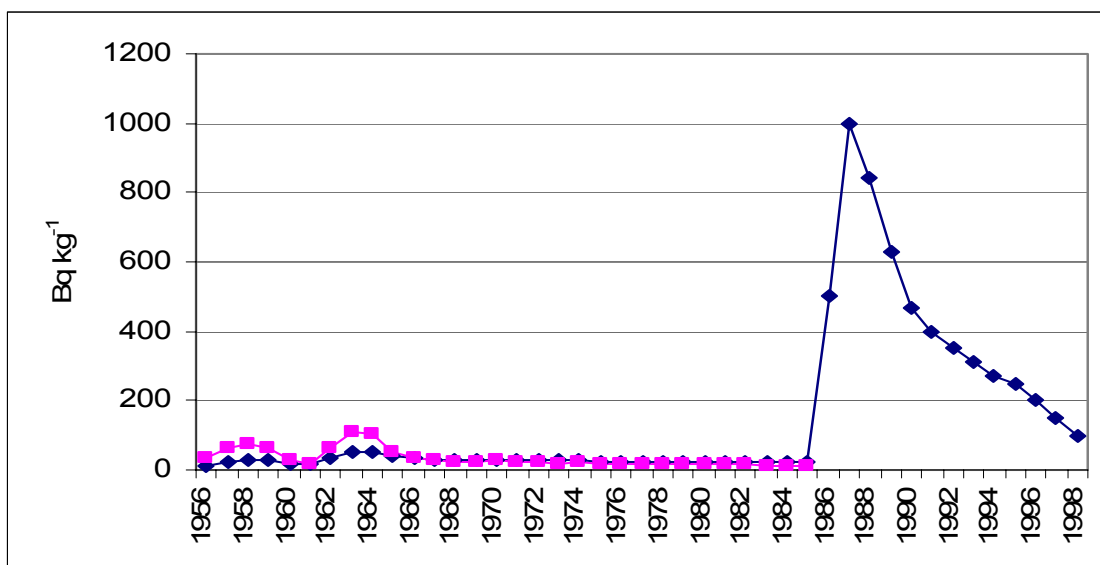


Figure 4. ^{137}Cs in freshwater fishes (Bq kg $^{-1}$ fresh weight) in Finland in 1956-1998. Values for 1956-1985 are estimated with a model using two sets of parameters (Rantavaara and Saxén 1985). The higher estimates for early years of atmospheric fallout were chosen for intake calculation. Values since 1986 are based on nationwide sample measurements.

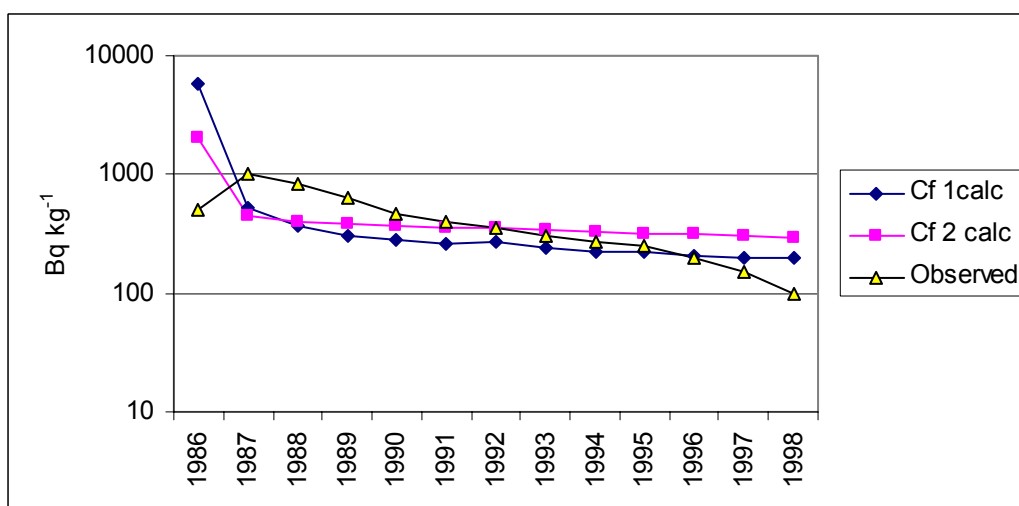


Figure 5. Comparison of activity concentrations of ^{137}Cs in fish, observed in 1986-1998, with values calculated with the model. Model parameters were based on pre-Chernobyl data.

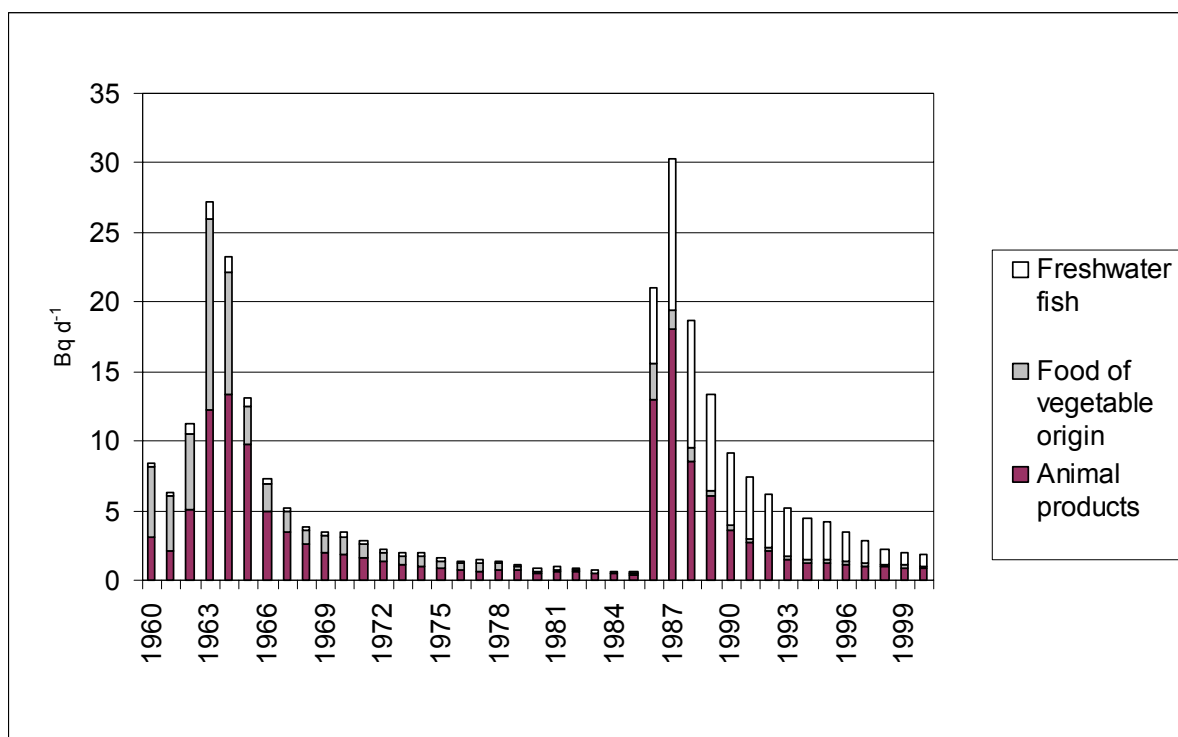


Fig. 6. Dietary ¹³⁷Cs from agricultural products and freshwater fish in Finland since 1960.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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Modelling long-term variation of radioactivity in foodstuffs in the Faroe Islands

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Introduction

Measurements of environmental radioactivity have been carried out on selected samples from the Faroe Islands since the beginning of the 1960's, with emphasis on marine and terrestrial foodstuffs. During the years, the work has been carried out in a co-operation between Risø National Laboratory in Roskilde, Denmark, and various Faroese institutions. The University of the Faroe Islands has been the Faroese partner since the beginning of the 1990's.

The paper presents results from modeling long-term variation of radioactivity in cow milk, lamb meat, drinking water and total diet in the Faroe Islands. The composition of the total diet in this context is according to estimates referred to by Aarkrog *et al.* (1963) and Aarkrog (1979). Model estimates are given for the effective ecological halflives of ^{137}Cs and ^{90}Sr in the foodstuffs and of the radioecological sensitivity, defined as time-integrated radionuclide concentration in an environmental sample from a unit ground deposition (Aarkrog, 1979).

Material and Methods

Sampling and data

The available data for the study are summarized in Table 1. The data have been obtained from annual Risø-reports (e.g. Aarkrog *et al.*, 1997) and from personal communication with

Table 1. Available sample data for the modeling

Samples	Fallout rates (3 localities)	Cow milk (3 localities)	Lamb meat	Total diet	Drinking water
Isotopes	^{137}Cs	^{137}Cs	^{137}Cs	^{137}Cs	^{90}Sr
Time period	1962-96	1962-96	1962-93	1962-93	1964-93

Sven P. Nielsen at Risø. The measured fallout rates are presented in Fig. 1, showing maximum values in the early 1960's and a pronounced signal from the Chernobyl accident in 1986. Precipitation and cow milk have been sampled at three different locations: Klaksvík in the north of the country, the capital Tórshavn in the central part and Tvøroyri in the south. The Faroese drinking water is obtained from surface water, and the samples have been collected as tap water in Tórshavn. Lamb meat and total diet derive from countrywide samplings. It should be noted that Faroese total diet does not only contain locally produced foods. All fruits and cereals are imported, and other foods have been imported to various degrees during the years, mainly from Denmark. Faroese lamb meat is mostly used for local specialties, and lamb meat for other purposes is imported, mainly from Iceland and New Zealand.

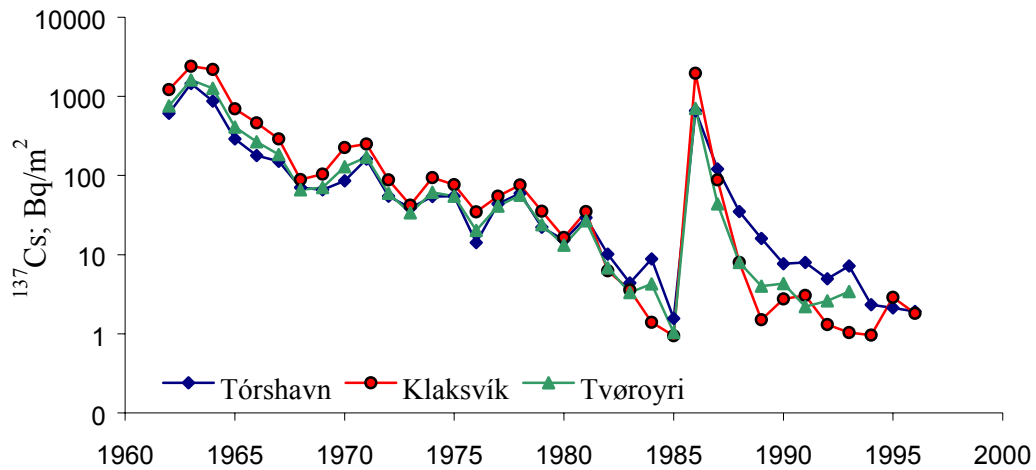


Figure 1. Fallout rates in the Faroes 1962-1996 measured at three locations.

Analyses

The radioactivity in a given sample has been related to the fallout rates in regression models based on the following equation (UNSCEAR, 1977; Aarkrog, 1979):

$$C_i = b_1 \cdot d_i + b_2 \cdot d_{i-1} + b_3 \cdot \sum_{k=1}^{k=\infty} d_{i-k} \cdot e^{-\lambda \cdot k} \quad (1)$$

where C_i is the concentration of a given radionuclide in a sample from year (i), and d_i and d_{i-1} are the deposition rates (kBq/m^2) in the years (i) and (i-1), respectively. Index $i=1$ represents the year 1950, taken as the first year with radioactive fallout. The sum on the right hand side represents the accumulated deposition from all preceding years, assuming an effective radiological halflife of T years, corresponding to a decay factor $\lambda = \ln(2)/T$. The estimated ecological halflife of a radionuclide in a given foodstuff is the value of T that gives the best fit between model and observations. It is presumed that all fallout is deposited by the start of a given year. This will underestimate the accumulated deposition, but it is immaterial for this study of long-lived radionuclides.

The observed deposition rates at the respective locations have been used for modelling ^{137}Cs concentration in cow milk, and for modelling ^{90}Sr concentration in drinking water from Tórshavn. An average deposition rate for the Faroe Islands has been calculated from precipitation rates and activities in precipitation at Klaksvík, Tórshavn and Tvøroyri. This average is used for modelling ^{137}Cs concentrations in lamb meat and in total diet.

The model-estimated radioecological sensitivity, S , is calculated as follows (Aarkrog, 1979; AMAP, 1998):

$$S = b_1 + b_2 + b_3 \cdot e^{-\lambda} / (1 - e^{-\lambda}) \quad (2)$$

where the parameters are as given in equation (1). The radioecological sensitivity estimates the transfer coefficient of a given radionuclide from fallout to the considered foodstuff.

Results and Discussion

The results are presented in Table 2 and Figs. 2-7. The model is found to represent the observations fairly well with high values for the square regression coefficient, R^2 , in all cases except for lamb meat. The relative low R^2 in the case of lamb meat may partly be explained by the fact that the meat samples have been collected countrywide from a few animals each year, as it is well-known that the ^{137}Cs concentration in lamb meat varies significantly between animals even from a single pasture (Joensen, 1999).

The model has been run in two steps in the case of cow milk: with all available data taken into account in the regression and with only data before 1986 taken into account. The model results after 1985 in parts (b) of Figs. 4-6 are therefore predictions of the ^{137}Cs activity in the milk after the Chernobyl accident. The R^2 is found to be slightly higher when only pre-Chernobyl observations are taken into account in the regression analyses.

The effective radioecological halflife of ^{137}Cs is estimated to 3-5 years for cow milk, 5.5 years for lamb meat and 4.5 years for total diet, and 5.5 years for ^{90}Sr in drinking water from Tórshavn (Table 2).

Table 2. Coefficients in the model when $b_4=0$, with p-values from t-test of the model coefficients given in brackets. R^2 is the square regression coefficient. S represents model-calculated sensitivities, and T is the estimated effective ecological half-life in years.

	b_1	b_2	b_3	T	R^2	Sens. S
^{90}Sr in drink. water; Tórshavn 1964-93	0.0205 (0.486)	0.0397 (0.018)	0.0114 (<0.001)	5.5	0.908	0.145 (Bq/l)y per kBq/m ²
^{137}Cs in total diet. Faroes 1962-93	0.0120 (<0.001)	0.0055 (0.001)	0.0031 (<0.001)	4.5	0.914	13.3 Bq/cap per Bq/m ²
^{137}Cs in lamb meat; Faroes 1962-93	0.1487 (<0.001)	0.0138 (0.594)	0.0168 (0.003)	5.5	0.675	288.0 (Bq/kg ww)y per kBq/m ²
^{137}Cs in milk from Klaksvík 1962-96	0.0075 (<0.001)	0.0086 (<0.001)	0.0040 (<0.001)	3.0	0.903	31.3 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tórshavn 1962-96	0.0093 (<0.001)	0.0171 (<0.001)	0.0041 (<0.001)	3.0	0.954	42.0 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tvøroyri 1964-93	0.0112 (<0.001)	0.0186 (<0.001)	0.0055 (<0.001)	4.5	0.982	62.6 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Klaksvík 1962-85	0.0080 (<0.001)	0.0111 (<0.001)	0.0030 (<0.001)	4.0	0.968	35.2 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tórshavn 1962-85	0.0094 (<0.001)	0.0189 (<0.001)	0.0036 (<0.001)	3.5	0.977	44.7 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tvøroyri 1964-85	0.0073 (0.062)	0.0222 (<0.001)	0.0050 (<0.001)	5.0	0.986	63.3 (Bq/l)y per kBq/m ²

The results for cow milk express a geographical variation of the radioecological sensitivity, with the highest value in the south (Tvøroyri). The reason for the geographical variation may partly derive from differences in the soil characteristics at the localities. This could, however, not be tested in the present study because of lack of data.

The calculated sensitivities are high compared to other countries (UNSCEAR, 1977; Aarkrog, 1979; Nielsen and Øhlenslæger, 1999). UNSCEAR (1977; p.143) reports sensitivities for milk in different countries around the world with the unit $\text{pCi}(\text{gK})^{-1}\text{y}$ per mCi km^{-2} . The highest value of 27.51 is reported for the Faroe Islands followed by 15.48 for Norway. The lowest value of 3.23 was reported for Denmark.

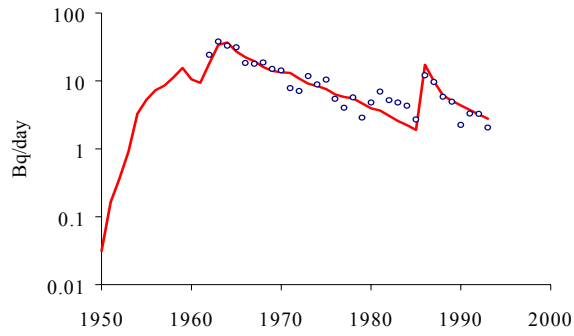


Figure 2. Observations (o) and model results (—) of ^{137}Cs in total Faroese diet. $R^2=0.914$.

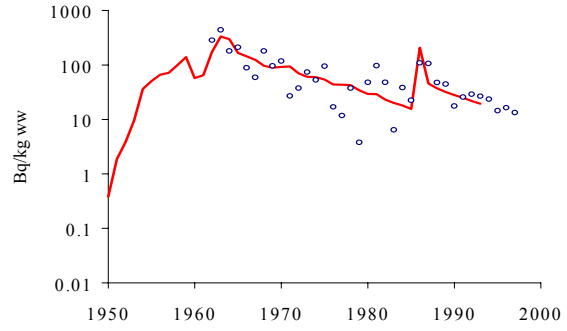


Figure 3. Observations (o) and model results (—) of ^{137}Cs in Faroese lamb meat. $R^2=0.675$

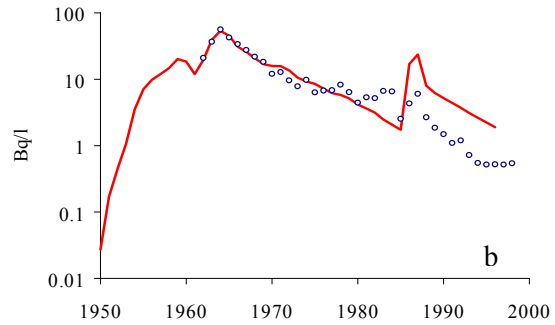
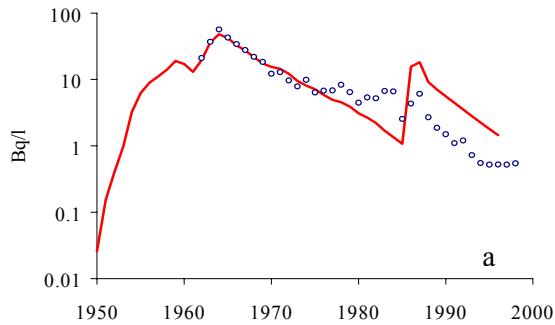


Figure 4. Observations (o) and model results (—) of ^{137}Cs in cow milk from Klaksvik. Regression: (a) 1962-96; $R^2=0.903$. (b) 1962-85; $R^2=0.968$.

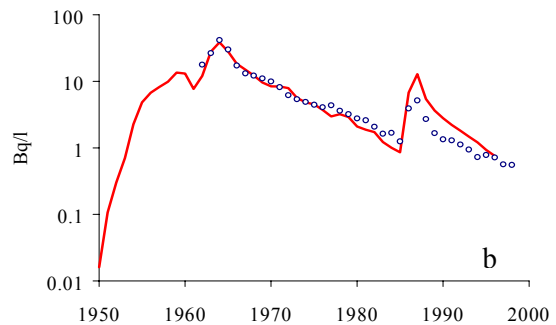
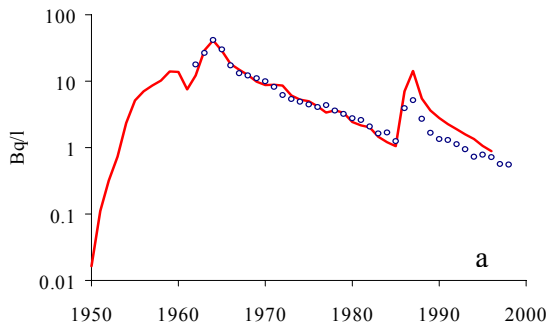


Figure 5. Observations (o) and model results (—) of ^{137}Cs in cow milk from Tórshavn. Regression: (a) 1962-96; $R^2=0.954$. (b) 1962-85; $R^2=0.977$.

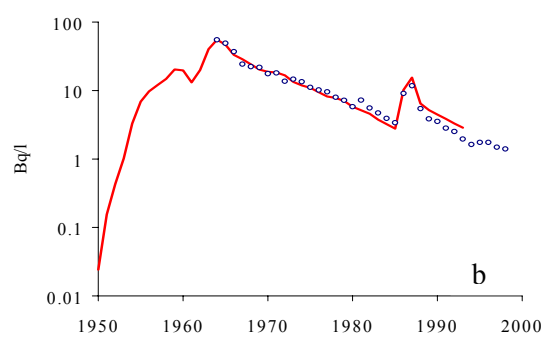
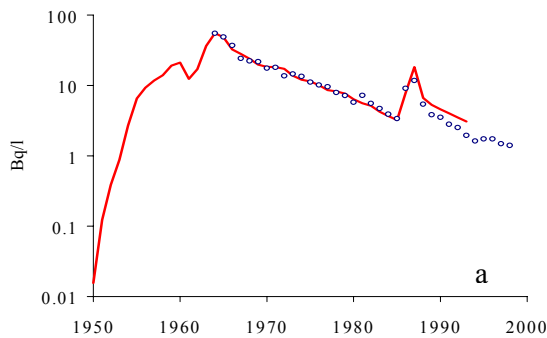


Figure 6. Observations (o) and model results (—) of ^{137}Cs in cow milk from Tvøroyri. Regression: (a) 1964-93; $R^2=0.982$. (b) 1964-85; $R^2=0.986$.

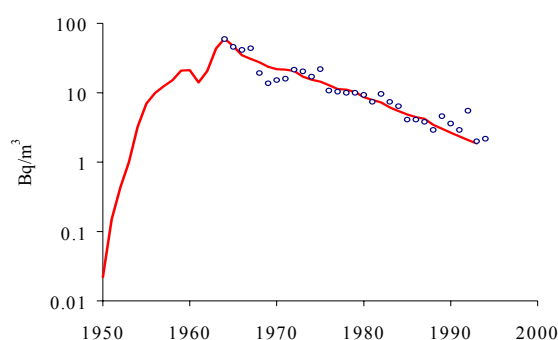


Figure 7. Observations (o) and model results (—) of ^{90}Sr in drinking water from Tórshavn. $R^2=0.908$.

Conclusions

The long-term variation of radioactivity in the selected foodstuffs is reproduced fairly well by the proposed regression model. The sensitivities in the Faroe Island are found to be high compared to other countries, indicating relatively high individual doses from ingestion of the foodstuffs in the Faroe Islands. Integrated transfer factors, like the sensitivity, provide a comprehensive assessment of transfer over the long term of ^{137}Cs and ^{90}Sr from deposition to foodstuffs.

The effective radioecological halflives of ^{137}Cs and ^{90}Sr in the foodstuffs are estimated to 2.0-5.5 years.

For a further study, more updated data sets should be available, and other sample types should be included, e.g. other foodstuffs, soil and grass. The present study shows that further modeling along the same line is worthwhile.

Acknowledgements

Thanks are extended to the Nordic Nuclear Safety Research (NKS) for financial support in the context of BOK 2.1. Thanks are also due to colleagues for valuable discussions, in particular to Sven P. Nielsen, Risø National Laboratory, Denmark, for advises and data delivery.

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Radioactive fallout in Norway from atmospheric nuclear weapons tests.

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Abstract

Historical data on radioactivity in air and precipitation samples have been collected and analysed from study sites in Norway. The purpose of the study was to investigate the correlation between air concentration, precipitation and deposition, and identify areas with high deposition. Areas with high precipitation have been compared with monitoring stations in other countries. The base data contains measurements of total beta in air and precipitation on a daily basis for the period 1956-1982. Radioactive fallout correlated strongly with annual precipitation which varies from 280-4200 mm per year in Norway. The deposition of ^{137}Cs was calculated to be $3.23 \pm 1.20 \text{ kBq/m}^2$ per 1000 mm annual precipitation for the period 1955-1975. Also, the relationship between total beta and ^{137}Cs has been investigated, in order to estimate the age of fallout. The age of fallout in Norway ranges from 3-9 months during the test periods, which is considerably shorter than the global average, where the mean residence time for debris in the lower stratosphere is estimated to be 1.3 years. There is no evidence of local fallout from tests on Novaya Zemlya reaching Norwegian areas.

Introduction

Norway received considerable radioactive fallout from the atmospheric nuclear weapons tests in the 1950s and early 1960s (Storebø, 1958, Hvinden & Lillegraven, 1961b). This was due to several factors including its geographical location (e.g. the rapid depletion of radionuclides in the polar stratosphere in 1959 following the 1958 Soviet tests on Novaya Zemlya) and high precipitation in coastal areas.

Since the first nuclear bomb was detonated in 1945, intensive testing activity occurred in the periods 1952-54, 1957-58 and 1961-62. A moratorium was effective in 1959, and largely observed in 1960 as well. Most countries signed a partial test-ban treaty, restricting nuclear tests in space, the atmosphere or under water, effective from 1963, but the last atmospheric explosion was performed as late as October 1980 by China.

UNSCEAR (2000) has recently re-evaluated the exposure of the public from atmospheric testing of nuclear weapons, as new information has become available on the numbers and yields of nuclear tests. The improved estimates of the production of each radionuclide in individual tests has led to a better agreement between the global atmospheric transport model (610 PBq of deposited ^{90}Sr) and the results of ^{90}Sr deposition from global monitoring networks (604 PBq).

As of January 1999, there have been 2532 nuclear detonations all over the world, (UNSCEAR, 2000). According to UNSCEAR, the official number of atmospheric tests has increased from 522 to 543. The total yield is estimated to have been 440 Mt, 189 Mt of which was fission yield. Deposited fission yield is estimated to have been 160 Mt (as opposed to the previous estimate of 189 Mt). The most intensive test period was from September 1961 to December 1962, where 57% of the total detonation power was detonated. The largest test, 50

Mt by USSR in 1961 is corrected to a fission yield of only 3% and a fusion yield of 97%. About 85% of the total fallout was deposited in the period 1950-1965.

The British Atomic Energy Research Establishment, AERE, and the US Naval Research Laboratory started systematically monitoring of the atmosphere for fission products in 1955 and 1957 respectively. Tromsø (69°40'N) and Bodø (67°17'N) were included as monitoring stations in the AERE-network since 1957 for rainwater, and Tromsø for air from 1977. Figure 1 illustrates the deposition of ^{137}Cs calculated for the 60°-70°N latitude using the UNSCEAR global fallout model, (UNSCEAR 2000), compared with measurements of deposited ^{137}Cs in Bodø and Tromsø. Estimated deposition in the years 1957-1963 is calculated from actual measurements of ^{90}Sr deposition using a ratio of 1.5 for $^{137}\text{Cs}/^{90}\text{Sr}$.

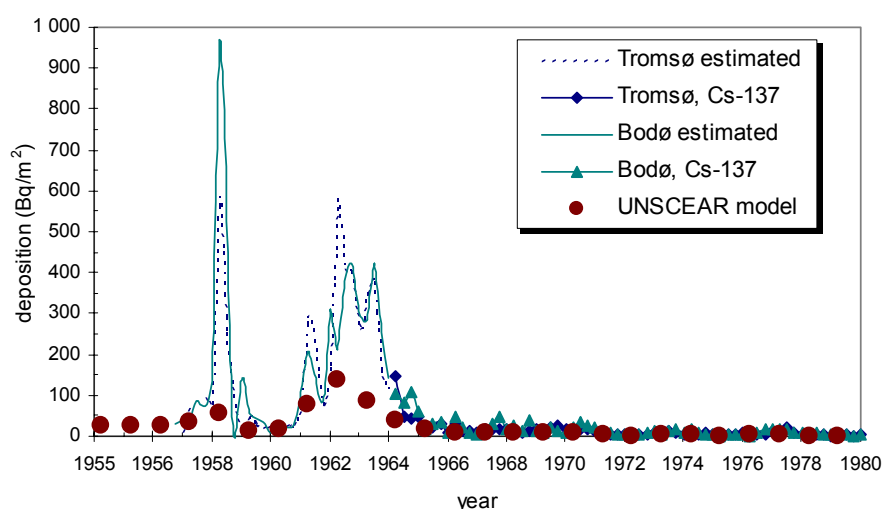


Figure 1. Deposition of ^{137}Cs calculated for the 60°-70°N latitude (UNSCEAR 2000), compared with measurements of ^{137}Cs deposition in Bodø (67°17'N) and Tromsø (69°40'N). Deposition of ^{137}Cs for the period 1957 to 1964 is calculated from measurements of ^{90}Sr deposition using a ratio of 1.5 for $^{137}\text{Cs}/^{90}\text{Sr}$.

Of special interest for Norway, are the nuclear tests conducted on Novaya Zemlya. Out of the 219 atmospheric tests performed in former Soviet Union, Matushchenko (1998) reports a total of 88 atmospheric, 2 surface water and 3 underwater tests on Novaya Zemlya. Among the atmospheric tests, the world's largest detonation of 58 Mt occurred, in October 1961. There has been speculation as to whether Norway received local fallout from any of the tests performed on Novaya Zemlya, especially in connection with the surface or under water tests in 1955, 1957 and 1961.

A peak in late spring and a minimum in winter have characterized the temporal distribution of the fallout during a typical year. The spring peak can be explained by an enhanced transport from the stratosphere, into which the main component of the debris from atmospheric nuclear tests is injected, down through the tropopause. Average residence time of debris in the atmosphere is about 1.3 years. Mean residence time of aerosols in the lower stratosphere ranges from 3-12 months in the polar regions, and 8-24 months in the equatorial regions.

In response to the nuclear tests, Norway quickly established a monitoring system, and from 1956 onwards the fallout situation was being monitored through daily measurements of radioactivity in samples of air, snow and drinking water, and monthly measurements of sea water, fish, milk, foodstuff, and occasionally animal and human tissue. In this study, data on total radioactivity in air and precipitation on a daily basis from 1956 to 1982 have been collated. The main purpose of the study was to investigate geographical differences in radioactive fallout across Norway, and to investigate if e.g. the number of days with precipitation was more important to the total deposited radioactivity than the amount of precipitation (expressed in mm).

Available information

The most extensive monitoring programme was conducted by The Norwegian Defence Research Establishment, FFI, located at Kjeller. FFI monitored radioactivity in air and precipitation at a number of sites across Norway. A total of 11-13 stations were operational in the period 1956-1984.

At every station, there was also a container for collecting snow, rain and/or settling dust. The containers were emptied every 10-14 days. If there had been no precipitation, the container was flushed with distilled water. Data on total beta fallout (combined dry and wet) was expressed as Bq (originally Ci) per sampling period, and measurement data from laboratory protocols and internal reports have been collated. At the end of every month, the air filters were bulked, and measured by gamma spectrometry for ^{137}Cs . The precipitation samples were also bulked and counted for ^{137}Cs .

Precipitation data for all stations are shown in Table 1. There are large differences in annual precipitation, especially along the coast where there are strong gradients within short distances. This may also result in large local variations in deposited radioactivity.

Table 1. Precipitation data for the monitoring stations (Norwegian Meteorological Institute)

Station	Average annual precipitation (mm/year) in 1957-1982	Range
Kjevik	1279	925 – 1653
Sola	1139	800 – 1733
Bergen	2059	1419 – 3068
Finse*	1030	n.a.
Gardermoen	867	635 – 1119
Ålesund	1270	930 – 1813
Røros	481	355 – 611
Værnes	846	617 – 1287
Bodø	1031	670 – 1529
Tromsø	1007	695 – 1548
Vadsø	516	315 – 675

*) Data for 1969-1982.

Current analysis

Data from the monitoring stations at Bergen, Gardermoen, (both situated at 60°N), Tromsø (69°N) and Vadsø (70°N) have been chosen as examples of variation in fallout. Bergen is

situated at the coast on the western side of the mountain range, whereas Gardermoen is an inland, lowland site to the east of the mountain range. Vadsø is situated approximately 800 km to the southwest of the most important test site of the former Soviet Union, Novaya Zemlya.

To compare fallout in different areas, the definition of a washout ratio, R , is useful. Washout ratios can be expressed as (in function of mass) (Engelmann, 1971):

$$R = \left(\frac{k}{\chi} \right)_m \quad (1)$$

where k is activity in fallout expressed as Bq/g water, and χ is the activity concentration in air, expressed as Bq/g air. Engelmann (1971) has compared different washout ratios, which vary between 200 and 4000 for different radionuclides, amounts of precipitation, and time periods. Washout ratios decrease with increasing precipitation. Engelmann also found that the highest washout ratios on the American continent occurred in the areas with the highest number of days with precipitation each year (Seattle). The Norwegian data has therefore been analysed both with respect to amount of precipitation (mm) and number of days with precipitation each year.

Results and discussion

The annual average concentration of total beta and ^{137}Cs in air is shown in Figure 2. The average is calculated based on data from all 13 Norwegian stations. As can be seen, there is roughly a 1 y delay from maximum beta concentration in air to maximum ^{137}Cs concentration in air. This is due to the large proportion of short-lived radionuclides in the fallout. Short-lived radionuclides, such as ^{140}Ba , ^{91}Y , ^{95}Zr , ^{103}Ru and ^{131}I , all contributing to the total beta fallout, exhibit maximum deposition rates in 1962. For the more long-lived radionuclides (e.g. ^{137}Cs , ^{90}Sr and ^{144}Ce), the maximum deposition rates are in 1963 (UNSCEAR 2000).

The predominant wind direction over Scandinavia is from the west throughout the year, and the annual precipitation at Bergen is about twice that of Gardermoen. Radioactivity in air at Bergen was generally higher than at Gardermoen, mainly because air from the west was washed out during passage over the western side of the mountains. It is only in periods with peak concentrations that air concentrations at Gardermoen are higher than those at Bergen. The seasonal variations in the air activity ratio may then be connected with the variations in the precipitation ratio and in the wind variability.

Table 2 shows the ratio between different stations and the average of all stations, for total beta in air and precipitation, calculated as monthly average of total beta in air (Bq/m^3) at that specific station compared with the average for all stations (Bq/m^3) and monthly fallout, both dry and wet (Bq/mm) at that specific station compared with the average for all stations (Bq/mm). The ratio is calculated as the average for every month from 1957 to 1982. As the numbers illustrate, the air concentration decreases from south to north, and the stations at the west coast, Bergen and Sola, show significantly higher air concentrations than the average of all stations. The ratio is rather constant, whether considering the periods 1957-1965 or 1957-1982.

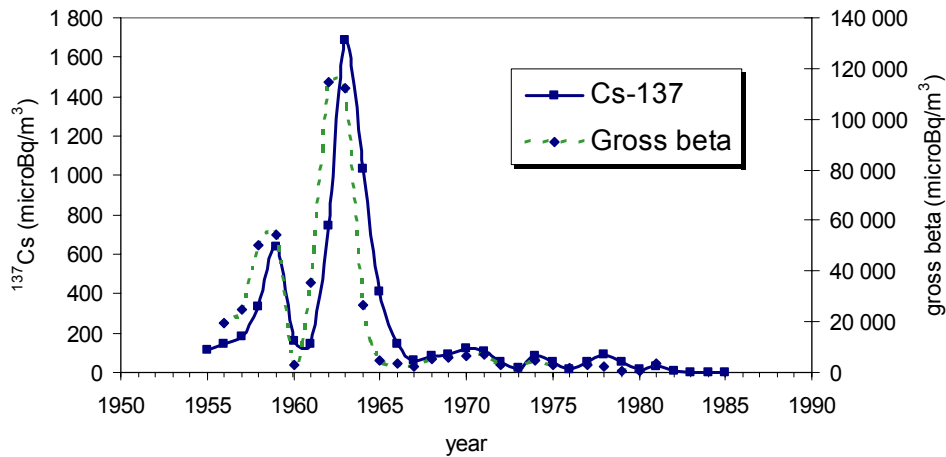


Figure 2. Total beta (right axis) and ^{137}Cs (left axis) in air, country average each year during the period 1955-1985.

Table 2. Ratio between each station and the average of all stations. The ratio is calculated for total beta in air, as monthly average of total beta in air (Bq/m^3) at that specific station compared with the average for all stations (Bq/m^3) and for total beta in fallout as monthly fallout, both dry and wet (Bq/mm) at that specific station compared with the average for all stations (Bq/mm). The ratio is calculated as the mean value ($\pm 95\%$ confidence interval of the average for every month from 1957 to 1982).

Station	Ratio in air	Ratio in fallout	Total number of days with precipitation 1957-69	Average days with precipitation per year
Sola	1.08 ± 0.10	0.6 ± 0.5	2879	221
Bergen	1.13 ± 0.12	2.3 ± 0.3	3019	232
Ålesund	0.99 ± 0.06	0.7 ± 0.1	3297	253
Værnes	0.93 ± 0.09	0.6 ± 0.1	2564	197
Røros	0.89 ± 0.08	0.6 ± 0.1	2320	192
Bodø	0.91 ± 0.10	1.1 ± 0.1	3320	255
Tromsø	0.84 ± 0.09	1.2 ± 0.2	2958	228
Vadsø	0.82 ± 0.05	0.5 ± 0.1	2915	224
Gardermoen	0.96 ± 0.16	1.1 ± 0.1	2320	178
Kjeller	1.23 ± 0.36	1.2 ± 0.1	2320	178

Vadsø is strategically located for detecting possible near zone fallout from Novaya Zemlya. There has been some speculation as to whether Norway received local fallout from any of the tests performed on Novaya Zemlya. Unfortunately, sampling at Vadsø has been irregular, and the data set is not as consistent as, for instance, Tromsø and Gardermoen. The air sampling was more regular during intense testing periods on Novaya Zemlya and air filters were changed daily. Any local fallout, originating from tests on Novaya Zemlya, should be evident in these air filters. As can be seen from Table 2, no such events are evident, as the S.E. is small for every month in the period of interest, from 1958 to 1962.

The ratios between monthly averages of total beta in air (mBq/m^2) and monthly fallout (Bq/m^2 per mm) for all stations are shown in Figure 3. Also the accumulated total beta fallout

(not corrected for decay) is compared with total precipitation for the period 1957-1969, Figure 4. The data in Figure 4 are best fitted to a linear regression ($R^2=0.74$), resulting in no dry deposition at all. Even with most of the radioactivity associated with particles with a low dry collection efficiency, an absence of dry deposition is not very likely. If one assumes some dry deposition, the data must be fitted to an exponential curve ($R^2=0.55$), which gives a total dry fallout of 40,000 Bq/m² (total beta).

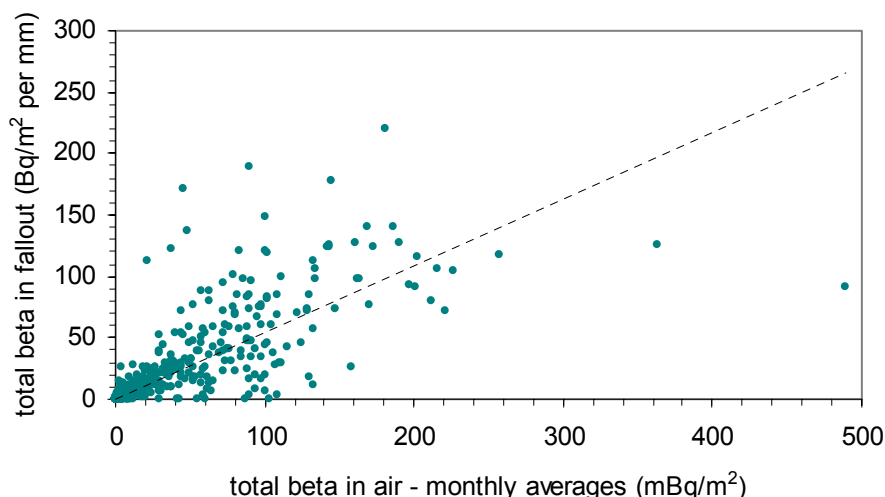


Figure 3. Correlation between total beta in air, monthly average expressed in mBq/m², and total beta in monthly fallout (both dry and wet) expressed as Bq/m² per mm precipitation. Data from Bergen, Gardermoen, Tromsø, and Vadsø.

On a global scale, the deposition of ⁹⁰Sr and ¹³⁷Cs shows a maximum at the temperate latitudes and a minimum at the poles and equator (UNSCEAR, 1982). Most tests were performed in the Northern Hemisphere, and the mean integrated fallout in the 60-70°N latitude band can be calculated from UNSCEAR (2000) to be 2.7 kBq/m² for ¹³⁷Cs and 1.8 kBq/m² for ⁹⁰Sr. Although describing the global pattern of deposition, the model does not in detail describe regional differences.

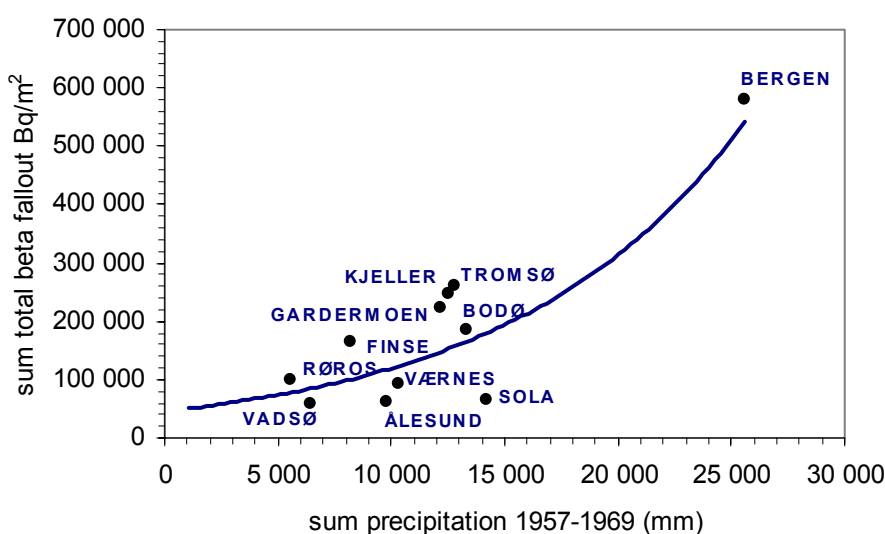


Figure 4. Correlation between total beta fallout (not decay corrected) and the total precipitation in the period 1957-1969.

When the annual deposition of ^{137}Cs calculated for the 60°-70°N latitude using the UNSCEAR global fallout model is compared with measurements of deposited ^{137}Cs in Bodø and Tromsø (Fig. 1), it becomes evident that the global model does not take into account the relatively rapid deposition of radionuclides in the northern hemisphere originating from the Soviet tests in 1958. The deposited ^{137}Cs in 1958 is also accompanied by high levels of ^{131}I . The UNSCEAR model significantly underestimates the annual deposition in Norway.

According to the global fallout model, one should in Norway expect a decrease in deposition of long-lived radionuclides from south to north. This conclusion is not valid for the 13 national monitoring stations (from 58°N to 70°N), as the deposition is strongly influenced by precipitation. Looking at Table 2, the trend in fallout (dry + wet) ratio has no south to north decreasing trend. Bergen has significantly higher fallout compared to the average; this is also the station with the highest annual precipitation, whereas both Ålesund and Bodø have more days of rain per year. Figure 4 also illustrates this, in that when the sum of total beta fallout for the period 1957-1969 is compared with the sum of precipitation for the same period, Bergen stands out as the station with the highest fallout.

Many authors have reported that the rates of deposition of long-lived nuclides in global fallout are correlated with precipitation patterns and rates (Hvinden & Lillegraven, 1961a & b, Livens et al., 1992, Blagoeva & Zikovsky, 1995, Bradley, 1970). A ratio of 3.7 kBq/m² of ^{137}Cs per 1000 mm annual precipitation measured in 1977 has been reported by Cawse and Horril (1986) from direct measurements of deposition in the UK. The AMAP-study, Wright et al. (1999) have estimated the cumulative deposition for the period 1955-85 to 3.69 ± 0.97 kBq/m² of ^{137}Cs per 1000 mm precipitation. Norwegian soil samples show that the highest depositions occur in areas with high precipitation (Lillegraven & Hvinden, 1982), Figure 5. The deposition, based on the soil samples reported by Lillegraven & Hvinden, is calculated to 3.23 ± 1.20 kBq/m² of ^{137}Cs per 1000 mm precipitation for soil samples collected after 1966, representing the cumulative deposition from 1955 up to sampling time. This is somewhat lower compared with previous estimates, but within the range of uncertainty.

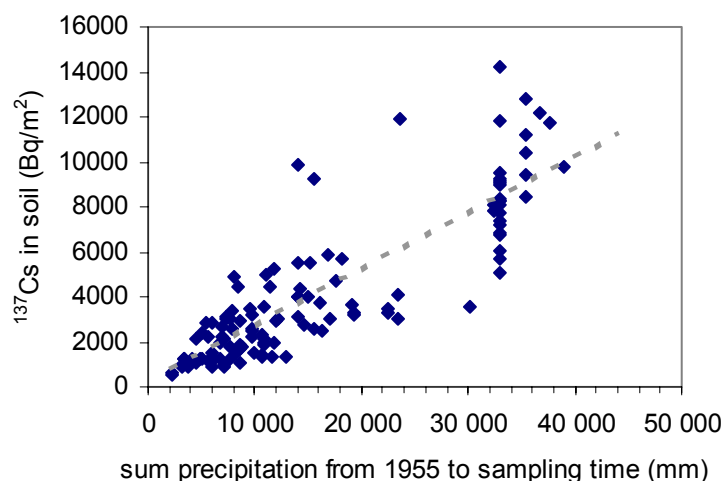


Figure 5. Correlation between ^{137}Cs in soil samples and total precipitation in the period 1955-up to sampling time. Soil sample results (1960-1979) from Lillegraven & Hvinden 1982.

Conclusions

Norway did, in most periods, receive more fallout than predicted by the UNSCEAR's global model. Especially in 1958-59 there is a rapid washout of radioactivity, with higher levels of e.g. ^{137}Cs , ^{90}Sr and ^{131}I than calculated by the model. The fallout is correlated to the amount of precipitation and concentration in air, and the deposited radioactivity is proportional to monthly precipitation. There is no decreasing trend in fallout from the south to north of Norway, as the fallout is well correlated to precipitation. The cumulative deposition for 1955-1975, based on soil samples is $3.23 \pm 1.20 \text{ kBq/m}^2$ of ^{137}Cs per 1000 mm annual precipitation. This calculated deposition is somewhat lower than previously reported (Wright et al., 1999, Cawse & Horril, 1986), but within the range of uncertainty.

There is no strong evidence that Norway received local fallout from the tests on Novaya Zemlya, as neither Vadsø nor Tromsø monitoring stations show higher air or fallout concentrations than other Norwegian stations. The only possible indication on local fallout is that Vadsø exhibits different washout factors, compared to the other stations. To investigate this question further, more nuclide-specific data (e.g. for ^{137}Cs , ^{131}I and ^{90}Sr) is needed, to date the fallout more correctly and correlate the ^{137}Cs in air and deposition to total beta measurements.

Acknowledgement

The Norwegian Research Council and the Norwegian Defence Research Establishment have financed the collection and analysis of data. Also the NKS/BOK-2 project has contributed to the work.

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Estimating Cs-137 Fallout Inventories in Iceland from Precipitation Data

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1. INTRODUCTION

This paper is based on a paper published in *Radioprotection Colloque*, **37**, C1, 1223-1228 [10]. The updated version given here contains updated estimates and some other improvements.

Iceland was identified in the Arctic Monitoring and Assessment Programme (AMAP) as one of the Arctic areas which received the most global fallout from atmospheric nuclear weapons tests, due to relatively high precipitation rates compared with much of the Arctic and sub arctic [1]. Cs-137 in the Icelandic terrestrial ecosystem almost entirely originates from the nuclear weapons tests carried out in the atmosphere until the early sixties. Fallout was greatest in mid nineteen sixties. Additional fallout from the accident at the Chernobyl Nuclear Power Plant was relatively small [2].

Measurements of fallout from nuclear weapons tests in soil, vegetation and agricultural products started in Iceland over 40 years ago [2]. Considerable variability was present in the results, even between adjacent sites, probably due to the mountainous terrain, changing strong winds and highly variable levels of precipitation. This variability has been especially noticeable in the case of soil samples but measurements of contamination in many areas are sparse, in part due to the difficult, remote and inaccessible terrain of much of the country. The important role of uncultivated rangelands in Icelandic agriculture (for instance in sheep farming) makes it desirable to be able to estimate deposition for many remote areas, as a basis for subsequent estimation of current and future transfer into agricultural products.

The study gave preliminary information on the spatial variation in ¹³⁷Cs deposition in Iceland, especially in areas used for agriculture. The objectives of the study were (i) to measure the spatial variation of radiocaesium inventories in soils in Iceland and (ii) to compare the results with different approaches to predicting ¹³⁷Cs contents in soil. This quantification is a necessary first step in an evaluation of vulnerability to radiocaesium deposition in Iceland. It is anticipated that Icelandic soils could be highly vulnerable to radiocaesium due to their volcanic nature and consequent lack of illitic minerals [3], as has been suggested by initial chemical studies on the properties of soils in the Nordic countries [4].

2. SITE DESCRIPTION

Soils in Iceland are unique amongst soils of Northern Europe. The terrain is mountainous and receives highly variable levels of precipitation. Volcanic eruptions are frequent, and volcanic ash deposits are widespread. In addition, there are vast unstable sandy areas near glacial

margins and along floodplains of glacial rivers. Soils that form in materials that are rich in volcanic ash are called Andosols. These soils have low cohesion but can absorb large quantities of water (>100% on dry weight basis). This high water holding capacity intensifies freezing effects that result in solifluction, landslides, needle ice formation, and the formation of hummocks. The very uneven surface of the rangeland areas and sparse vegetation can make it very difficult to obtain representative deposition estimates with soil sampling.

3. MATERIAL AND METHODS

3.1 Precipitation and fallout data

Measurements of the fallout radioactivity in precipitation have been carried out regularly in Iceland for a station close to Reykjavík (Rjúpnahæð, code in this paper: RJU) since 1958 [2]. These measurements were conducted by the United Kingdom Atomic Energy Authority in Harwell since April 1959. The precipitation data used in this study come from the Icelandic Meteorological Office and are regularly reported in its journal [5]. The journal also gives values from the United Kingdom Atomic Energy Authority in Harwell for ^{137}Cs activity concentration in precipitation from April 1959 to (and including) 1982. Therefore, this site was used as the base station for deriving the quarterly ratio of ^{137}Cs fallout to precipitation in Iceland, using the AMAP methodology outlined by Wright et al. [1]. The data from the base station, RJU, shows that 83% of the decay corrected deposition of ^{137}Cs in the period 1960-1982 occurred during the first eight years 1960-1967. The emphasis in this study was therefore placed on meteorological stations that had been in operation during these eight years; deposition for these years was estimated based on precipitation data for this period. These results were subsequently scaled up to the period 1960-1982, assuming that in all cases 83% of the deposition had occurred during the first eight years used for calculations. Fallout data from before 1960 was not taken into account in the initial study [11]. Later, as a part of an ongoing study, data from England was used for extrapolating the Icelandic data to cover the time period from the late fifties to 1986. This produced a precipitation ratio constant in a very good agreement with a constant calculated in the same way for a site in England and the agreement with measured concentration in soil was also very good on average (ratio of 0.98).

3.2 Soil sampling

Measurements of radioactive fallout in soils have been carried out by the Icelandic Radiation Protection Institute (IRPI) since 1989. In autumn 2000, IRPI started systematic analysis of radiocaesium in Icelandic soils. Soil samples from 14 sampling sites were collected in 2000. The sampling sites were deliberately located close to meteorological measurement stations so that good representative data was available on precipitation for each sampling site, and the exact location of each site was recorded using a GPS instrument (Figure 1). The sites were selected so that they would span a range of precipitation rates as well as representing different areas and soil types. Soils were sampled with 17 and 19 mm diameter sampling probes to a depth of up to 30 cm. Twenty cores were collected at each site at 1 metre intervals along a 20 m long transect and bulked. A more detailed description of the procedure adapted can be found in a report on sampling procedures in use in the Nordic countries [6].

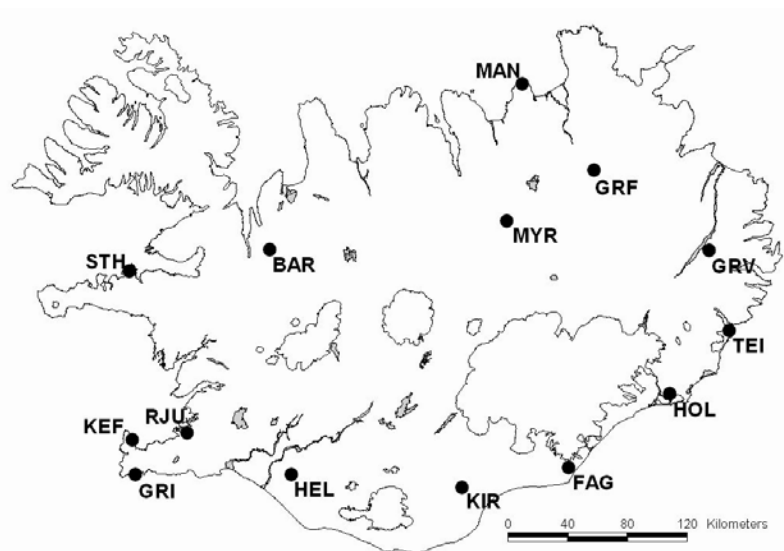


Figure 1. Soil sampling sites used in the initial study. Each site is identified with a 3 letter identification code, cf figure 2.

3.3 Radiocaesium measurements

The activity concentration of ^{137}Cs was measured using HPGe spectrometry and estimated on an activity per unit area basis (kBq m^{-2}).

3.4 Cs-137 deposition estimates

Various authors have linked ^{137}Cs deposition to precipitation [1,7,8]. The results are often normalised and presented as amount of deposition per unit area per 1000 mm annual precipitation. Using annual precipitation rates to predict deposition is based upon the assumption that the deposition is proportional to the average precipitation rate. This means that estimates of deposition can be made having one reference site where deposition is known, deposition at other sites can then be estimated by multiplying the reference deposition by the ratio of annual average precipitation rate at each site and dividing by the precipitation rate at the reference site. The time corrected accumulative deposition estimate for the reference site can be based on measurements of ^{137}Cs activity concentration in rainwater and the corresponding amount of precipitation, and it can also e.g. be based on measurements of amount per unit area of ^{137}Cs in soil. Sometimes text on this topic can become meaningless if only the reference *amount* (e.g. “per 1000 mm precipitation”) is specified, not the reference *rate* (e.g. “per 1000 mm annual precipitation”). Likewise if it is not clear what is the time reference for the deposition estimate.

3.5 Estimating annual precipitation, selection of time interval used for averaging

The average annual precipitation rate can be calculated in various ways. In areas with highly variable precipitation rates, the value of the calculated average depends on the method used for the averaging. The following methods were used in this study for calculating the average annual precipitation rates:

- a) **Averaging over 30 years (1931-1960, 1961-1990).** The 30 year standard reference time period, 1961-1990 is often used for reference meteorological data, especially when there

is a need to calculate average values for meteorological parameters. The 30 year period, 1931-1960 has previously also been widely used as a reference period.

- b) **Averaging over 8 years (1960-1967).** The same method as (a) apart from that the annual precipitation estimate is based on the 1960-1967 average. This is the time period in which most of the deposition of ^{137}Cs fallout occurred. Calculating an average based on this period should therefore give a better correlation between deposition and precipitation rates.
- c) **Averaging with respect to radiological effects.** In (b) the ^{137}Cs activity concentration in precipitation was used to help define the most suitable time window for averaging the precipitation rate. This method can be extended further by calculating the precipitation rate as a weighted average and using the ^{137}Cs activity concentration as the weighting function. No restrictions need to be made concerning the length of the time interval and this method automatically ensures that the calculated average is mainly affected by the precipitation rates in the periods which contributed most to the deposition. This method should be expected to give best results for correlation between precipitation rates and deposition. A more detailed mathematical description of the method can be found in a paper from the *International Conference on Radioactivity in the Environment, 1-5 September 2002, Monaco* [10].

The AMAP method. The method used successfully in the AMAP study [1] can be formulated as follows. The deposition at each site is estimated by assuming that the ^{137}Cs activity concentration in precipitation was the same at all sites during any given time period. Thus the deposition at each site can be estimated by measuring the activity concentration of ^{137}Cs in precipitation at one reference site and then estimating the deposition at other sites by summing the product of precipitation (in m) at the site and ^{137}Cs activity concentration in precipitation (in Bq m^{-3}) at the reference site for time periods of interest. The AMAP method can be shown [10] to be equivalent to method (c) described above, averaging with respect to radiological effects.

4. RESULTS

4.1 Effects of methods of averaging

Table 1. Effect of method of averaging annual precipitation at the Rjúpnahæð reference site

Method used for averaging	Average annual precipitation (mm y^{-1})
Average of the years 1931-1960	1026
Average of the years 1961-1990	984
Average of the years 1960-1967	870
Average with respect to radiological effects (method c)	802

Table 1 shows clearly how sensitive the calculated annual average precipitation can be to the time period used as the basis for the calculation. The average annual precipitation at the Rjúpnahæð reference site is e.g. 28% higher if it is calculated as a simple average of the years 1931-1960 than if the averaging is based on the radiological effects (method c).

4.2 Cs-137 inventories - correlation between measured and predicted values

The measured ^{137}Cs content per unit area of soil varied from 900-4700 Bq m^{-2} , with higher deposition in the south of Iceland which receives more precipitation. There are various ways of estimating the correlation between predicted and measured values for ^{137}Cs deposition. The method used in the comprehensive AMAP study [1] was to force the regression line to go through the origin and calculate correlation coefficients on that basis. This approach will give

a considerably higher value for the correlation coefficient than for an unbound regression line, but it can be justified on the basis that the assumption being tested is that deposition is directly proportional to precipitation. This same approach was used in this study. A comparison of measured and predicted values for each site is shown in Figure 2. In this figure the predicted estimates have been improved from the first study by extrapolation, making use of UK data when outside the scope of the Icelandic data (description in a paper to be published). This gave a very good correspondence between the predicted and measured values.

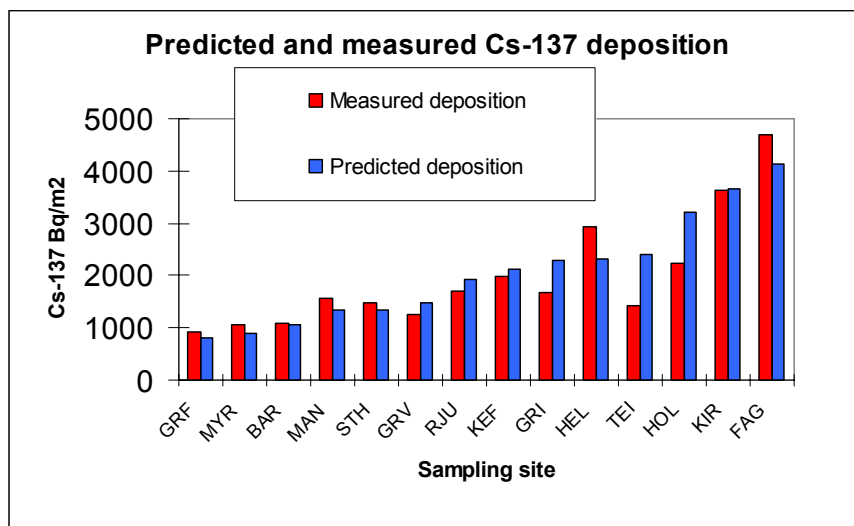


Figure 2. Comparison of predicted ^{137}Cs deposition based on precipitation data and measured values at 14 sites close to meteorological stations in Iceland. The annual precipitation was calculated by averaging with respect to radiological effects (method c).

The correlation between measured and predicted values (using method c) for ^{137}Cs deposition was good for all sampling sites ($r^2 = 0.96$) (Figure 2) and the ratio of measured to predicted values was also very good, 0.98. The predicted values are also highly correlated with the annual average precipitation at each site, based on the years 1960-1967 ($r^2 = 0.96$) (method b). Using the 30 year standard reference period 1961-1990 gave a poorer correlation ($r^2 = 0.93$) (method a) as expected.

5. DISCUSSION

Two methods of fallout prediction were considered. Firstly, a simple ratio approach (corresponding to methods (a) and (b) in section 3.5) was used based on published values for the amount of ^{137}Cs deposited per 1000 mm annual precipitation of 10 pCi per cm^2 per 1000 mm annual precipitation for samples collected in 1977 (equivalent to 3.7 kBq m^{-2}) by Cawse and Horrill [7] for sampling conducted in 1977 in the UK. Tone Bergan has reported [8] a corresponding estimate of 3.23 kBq m^{-2} per 1000 mm annual precipitation in Norway.

An alternative approach was to use a model predicting deposition using precipitation data, in a similar manner as that used previously within the Arctic Monitoring and Assessment Programme (AMAP) [1]. This corresponds to method (c) as described in section 3.5. In the AMAP analysis, the annual ratio between ^{137}Cs fallout and precipitation was derived for a base station (Tromsø) and the prediction spread over the Arctic using a precipitation data set with a resolution of $0.5^\circ \times 0.5^\circ$. Wright et al. [1] gave an estimate of $3.69 \pm 0.97 \text{ kBq m}^{-2}$ per 1000 mm annual precipitation for the end of 1985. The values given above are quoted for different years. To be able to compare the values, we have decay

corrected them to the end of 2000 when the Icelandic samples were taken. The results can be seen in Table 2 and vary from 1.8 to 2.6.

Table 2. Comparison of estimates of normalised deposition to precipitation ratios, time corrected to end of 2000

Source of data	kBq m ⁻² per 1000 mm annual precipitation decay corrected to end of 2000
Wright et al (AMAP)	2.6
Cawse and Horrill	2.1
Tone Bergan	1.8
This study, predicted estimate	2.5
This study, measured in soil	2.4

It should be noted that the time corrected value depends on the reference time for which the data are valid. This is not always clear in papers on this topic and some numbers in the table above could change for this reason.

The results of this study agree very well with the first study and reasonably well with the second one, taking into account the stated uncertainties of those estimates. Using exactly the same method to process the data sets might even reduce further the difference between the estimates.

The comparison of predicted and measured values for Iceland was much stronger than that originally reported by Wright et al [1]. In the AMAP study, data were analysed for 50 samples from Greenland, Norway and Russia from the period 1961-1985. A line through the origin was fitted to the data set using least squares regression giving an r^2 value of 0.51 [1] based on a coarse precipitation data set, disparate sources of measured ¹³⁷Cs deposition using different sampling methodologies. In Iceland, a comparison of predicted and measured values gives a corresponding r^2 value of 0.96. This same correlation was obtained, both when the AMAP method was used (method c) and when the prediction was based on average annual precipitation for the years 1960-1967 (method b). Some of the improved correlation compared with the AMAP study is probably due to the proximity of meteorological stations, where precipitation has been measured in a consistent manner. Furthermore, the soil sampling was conducted by the same team, with a consistent, rigorous methodology over a short period of time. However, the strength of the correlation is surprising considering that dry deposition is not accounted for in the method, although because of the high precipitation rate in much of Iceland dry deposition may not be expected to be an important contributor to total ¹³⁷Cs deposition. In addition, lateral transport by erosion would be expected to occur in some Icelandic areas [9]. Consistent methodology of soil sampling and compiling precipitation data can be crucial for successful application of precipitation based deposition estimates

The prediction based on the average annual precipitation for the years 1960-1967 (method b) gave a higher correlation with measured data than expected. The probable explanation for this is the highly correlated precipitation pattern between these stations, due to their proximity. This can be seen, for example, if the precipitation pattern as a function of time for all the stations is plotted. The method could thus be expected to provide significantly poorer results than the AMAP method, when applied to a larger geographical area with a less correlated precipitation pattern.

Overall, the AMAP methodology has been successful and has the advantage that it is possible to predict the ¹³⁷Cs deposition at any location in any year after 1960, even though fallout prior to this year was occurring but at lower levels than those prevailing in the nineteen sixties. This study will be followed up, investigating sites near and further from meteorological stations, including some sites where erosion might occur. The next step will

be to use this methodology to create a map of ^{137}Cs deposition across Iceland and to test the predictions.

ACKNOWLEDGEMENTS

The study is carried out in co-operation between the Icelandic Radiation Protection Institute, the Agricultural Research Institute and the Icelandic Meteorological Office. This study is a part of the NKS/BOK-2 project and has received support from the NKS through the project. It has also received financial support from the Icelandic Research Council. Co-operation with CEH occurred under the International Union of Radioecology task force on Arctic and Antarctic ecosystems.

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RADIOCAESIUM (CS-137) FALLOUT IN ICELAND AND ITS BEHAVIOUR IN SUBARCTIC VOLCANIC SOILS

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Introduction

The Cs-137 in the Icelandic terrestrial ecosystem almost entirely originates from the nuclear weapons tests carried out in the atmosphere until the early sixties, peaking in 1963. Insignificant radioactive fallout occurred in Iceland following the Chernobyl accident in 1986. Measurements of the fallout radioactivity in precipitation and the atmosphere have been carried out regularly in Iceland since 1958 (Pálsson 1996). Measurements of radioactive fallout in soils have been carried out by the Icelandic Radiation Protection Institute (IRPI) since 1989. In the autumn of 2000 IRPI started systematic analysis of radiocaesium in Icelandic soils. The main objectives of the study are to investigate the spatial variation of radiocaesium concentration in Iceland, make estimates of the retention of Cs-137 in major soil types and study the behaviour of radiocaesium in volcanic soils (i.e. Andosols).

The ability of soils to exchange cations (CEC), including Cs, is highly dependant on its physical and chemical properties, e.g. organic carbon content, clay content, type of clay and pH (Shoji *et al.* 1993). Soils in Iceland are unique among soils of Northern Europe. Iceland is a volcanic island situated in the North Atlantic Ocean traversed by the spreading Mid-Atlantic Ridge. The bedrock is composed of volcanics, mainly basaltic lava flows and hyaloclastites (móberg) of upper Tertiary and Pleistocene age. The bedrock is largely covered with glacial till and glaciofluvial- and lacustrine sediments formed at the end of the last glaciation (Jóhannesson and Saemundsson 1998, Saemundsson 1980). On average volcanic eruptions occur every five years. Explosive eruptions are common, producing tephra (volcanic ash), as many volcanoes/volcanic systems are partially or totally covered by glaciers or submerged by water (Thorarinsson and Saemundsson 1980). Icelandic soils have mainly formed in aeolian and tephra deposits overlying the glacial till and lava bedrock.

Material and Methods

Soils formed in tephra material classify mainly as Andosols. Andosols constitute 70-80 % of the soils of Iceland (Figure 1). Among their main characteristics are high CEC, considerable organic carbon content (1-10 %) and high clay content (20-40 %) (Arnalds 1999, Arnalds *et al.* 1995, Wada *et al.* 1992). All these factors influence the ability of soil to fix Cs-137.

Soil samples from 17 sampling sites were collected (Table 1). Soils were sampled with a 17 mm sampling probe. Twenty cores were collected at each site at even intervals along a 20 m long line. Compilation of sampling methods in use in the Nordic countries has been made by Isaksson (2000). The concentration of Cs-137 was measured using HPGe spectrometry. Results are reported as Bq/kg and Bq/m².

Table 1. Soil sampling in Iceland 2000

Sampling site/ abbreviation	Location- GPS	Soil type	Vegetation	Bedrock type ¹
Rjúpnað RJU	N64 05.093 W21 50.450	Andosols	Grass	Lavas/sedim. < 0.8 m.y.
Keflavík KEF	N64 01.591 W22 35.129	Andosols	Willow, moss, sedge	Lavas/sedim. < 0.8 m.y.
Grindavík GRI	N63 49.236 W22 31.224	Andosols (vitric)	Grass, hayfield	Lavas, postglacial
Stykkishólmur STH	N65 02.211 W22 46.105	Histosols (fibric)	Heath, moss	Lavas, Upper Tertiary
Barkarstaðir BAR	N65 12.387 W20 46.763	Histosols	Sedge, moss	Lavas, Upper Tertiary
Nautabú NAU	N65 27.533 W19 22.030	Andosols	Grass, hayfield	Lavas, Upper Tertiary
Mýri MYR	N65 22.602 W17 22.802	Andosols	Grass, infield	Lavas/sedim. 0.8-3.3 m.y.
Mánarbakki MAN	N66 12.001 W17 05.916	Andosols	Heath, moss, sedge	Lavas/sedim. 0.8-3.3 m.y.
Grímsstaðir GRF	N65 39.876 W16 05.789	Andosols	Grass, hayfield	Lavas/sedim. < 0.8 m.y.
Grímsárvirkjun GRV	N65 08.552 W14 31.132	Andosols	Heath, moss, sedge	Lavas, Upper Tertiary
Teigarhorn TEI	N64 39.191 W14 18.547	Andosols	Moss, sedge	Lavas, Upper Tertiary
Hólar HOL	N64 18.003 W15 11.685	Andosols	Grass	Lavas, Upper Tertiary
Fagurhólmýri FAG	N63 53.081 W16 37.703	Andosols/ Arenosols	Grass/sedge	Holocene sediments
Kirkjubæjarklaustur KIR	N63 46.833 W18 04.939	Andosols	Sedge, moss	Lavas/sedim. 3.3-0.8 m.y.
Vík in Mýrdal VIK	N63 24.635 W19 01.742	Andosols	Moss, sedge	Hyaloclastics < 0.8 m.y.
Hella HEL	N63 51.291 W20 23.940	Andosols	Heath, sedge, moss	Lavas/sedim. < 0.8 m.y.
Stóra-Ármót ARM	N63 59.632 W20 56.729	Andosols	Heath	Holocene lava

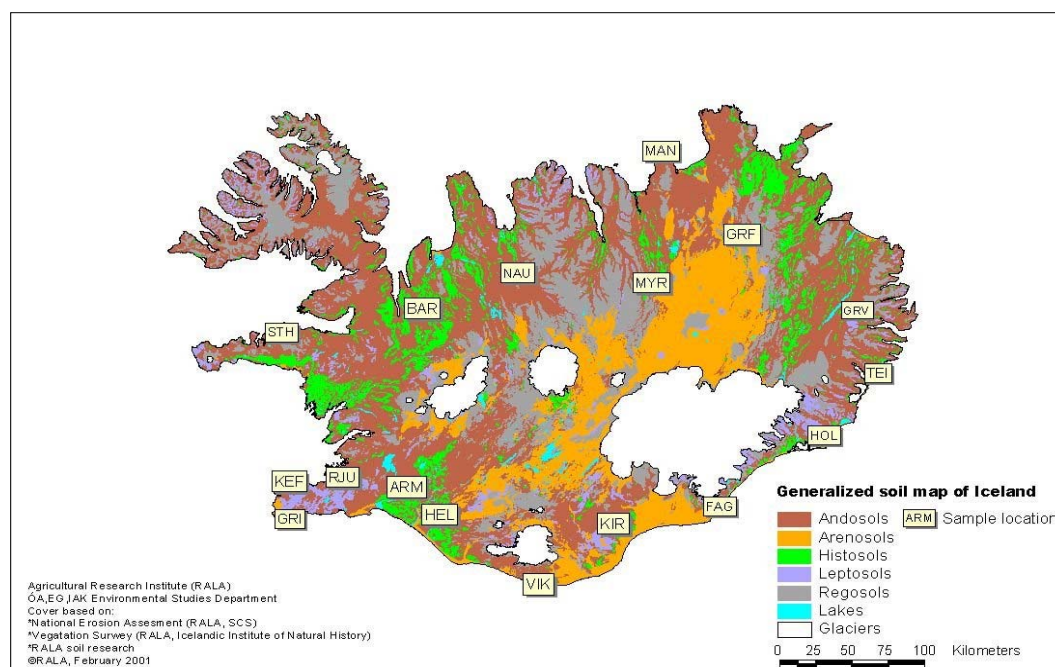


Figure 1. Generalized soil map of Iceland. Sampling sites from 2000 are shown.

¹ Ref: Jóhannesson and Saemundsson 1998

Table 2. Soil sampling in Iceland 2001.

Sampling site/ abbreviation	Location- GPS	Soil type	Vegetation (field obs.)	Bedrock type
Arnardalur ARD	N65 09.233 W15 57.446	Vitrisols	Willow, heath, moss	Lavas/sedim. < 0.8 m.y.
Arnarfjörður ARN	N65 45.833 W23 19.817	Andosols	Grass, sedge	Lavas, Upper Tertiary
Blöndulón BLO	N65 11.123 W19 44.024	Andosols	Heath, sedge, willow	Lavas/sedim. < 0.8 m.y.
Brú BRU	N65 06.384 W15 32.408	Andosol (vitric)	Grass, sedge	Lavas/sedim. 3.3-0.8 m.y.
Fagurhólmýri FAGa	N63 56.061 W16 26.846	Vitrisols	Sedge, moss	Holocene sediments
Fagurhólmýri FAGb	N63 52.931 W16 41.594	Vitrisols	Sedge, moss	Holocene sediments
Grafarlönd GRA	N65 20.257 W16 03.329	Vitrisols	Willow, heath, sedge	Lavas/sedim. < 0.8 m.y.
Grimsey GEY	N66 31.762 W17 59.357	Andosols	Grass, moss	Lavas/sedim. 3.3-0.8 m.y.
Gufuskálar GUF	N64 50.194 W23 56.950	Andosols	Moss, sedge	Lavas, postglacial
Gullfoss GUL	N64 18.213 W20 10.456	Andosols	Grass, moss	Lavas/sedim. 3.3-0.8 m.y.
Heimaey HEI	N63 24.045 W20 17.519	Andosol	Grass, moss	Lavas/sedim. < 0.8 m.y.
Hestur HES	N64 35.040 W21 38.405	Andosol	Sedge, moss	Lavas, Upper Tertiary
Hraun á Skaga HRA	N66 06.382 W20 06.567	Histosols	Sedge, moss	Lavas/sedim. 3.3-0.8 m.y.
Hvannalindir HVA	N64 53.273 W16 18.425	Vitrisols	Willow, moss	Lavas, postglacial
Hveravellir HVE	N64 51.943 W19 35.471	Andosols	Sedge, moss, heath	Lavas/sedim. < 0.8 m.y.
Hveravellir HVE1	N64 52.545 W19 31.749	Vitrisols	Desert	Lavas/sedim. < 0.8 m.y.
Kerlingarfjöll KER	N64 42.828 W19 25.907	Vitrisols	Moss, willow, sedge, heath	Lavas/sedim. < 0.8 m.y.
Kverkfjöll KVE	N64 50.353 W16 24.381	Vitrisols	desert	Lavas/sedim. < 0.8 m.y.
Kvísker KVI	N63 58.104 W16 26.567	Andosols	Grass, moss	Holocene sediments
Látrabjarg BJA	N65 30.188 W24 31.893	Andosols	Moss, sedge	Lavas, Upper Tertiary
Mýrar BOR	N64 41.739 W21 54.235	Histic Andosol	Sedge, moss, willow, heath	Lavas, Upper Tertiary
Mývatn MYV	N65 39.514 W16 59.559	Andosols	Heath, moss	Lavas/sedim. < 0.8 m.y.
Norðurárdalur BOR1	N64 42.708 W21 36.723	Andosols	grass	Lavas, Upper Tertiary
Nýidalur NYI	N64 43.668 W18 02.814	Vitrisols	Moss, willow, sedge	Hyaloclastics < 0.8 m.y.
Raufarhöfn RAU	N66 24.455 W15 53.261	Andosols	Sedge, moss	Lavas, postglacial
Reykholar REY	N65 26.965 W22 11.616	Histosols	Sedge	Lavas, Upper Tertiary
Sauðárdalur SAU	N64 55.231 W15 52.766	Andosols	Heath, willow, lichen	Lavas/sedim. < 0.8 m.y.
Sauðárdalur SAU	N64 55.331 W15 53.800	Vitrisols	desert	Lavas/sedim. < 0.8 m.y.
Vatnadalur VAT	N66 07.214 W23 34.195	Andosols (histic)	Moss, sedge	Lavas, Upper Tertiary

Results and Discussion

The Cs-137 retention for three depth intervals, i.e. 0-5 cm, 5-10 cm and >10 cm, is compared with the texture of the soils (Figure 2). The soils group into four different classes, loamy sand, sandy loam, loam and organic material (peat). Significant differences are observed in Cs retention between the soil texture classes indicating that binding of Cs is highly dependant on soil properties (Sigurgeirsson *et al.* 2001). A deeper penetration of Cs-137 is observed for sandy soils than finer loams.

The main results obtained so far are the following:

- Total activity per unit area of Cs-137 was found to be 700-4800 Bq/m².
- In all cases > 85 % of Cs-137 in soils is fixed in the uppermost 15 cm of the soil cover.
- In loam and sandy loam 60-95 % of Cs-137 is fixed in the top layer of the soil. In sandy loam and loamy sand (Vitric Andosols) considerable amount of Cs is fixed in deeper layers, i.e. below 5 cm depth, indicating that mobility of Cs-137 is dependent on grain size distribution and clay content of the soil. Former studies indicate that Cs is rather mobile in Icelandic soils (Oughton *et al.* 1997).
- In peat (Histosols) 80-85 % of Cs-137 is bound in the top layer of the soil (0-5 cm) and 10-15 % within the 5-10 cm depth range.
- Comparison of samples obtained from undisturbed hayfield (MYRa in Figure 2) and uncultivated land at the same site (i.e. MYR) showed lower concentration of Cs-137 in the top layer of the hayfield but on the other hand higher total activity per unit area. Sample from GRI, collected in a hayfield, shows similar behaviour of Cs-137 as in MYRa.
- Good correlation was obtained between precipitation and total activity of radiocaesium in soils (see Pálsson *et al.* 2001).

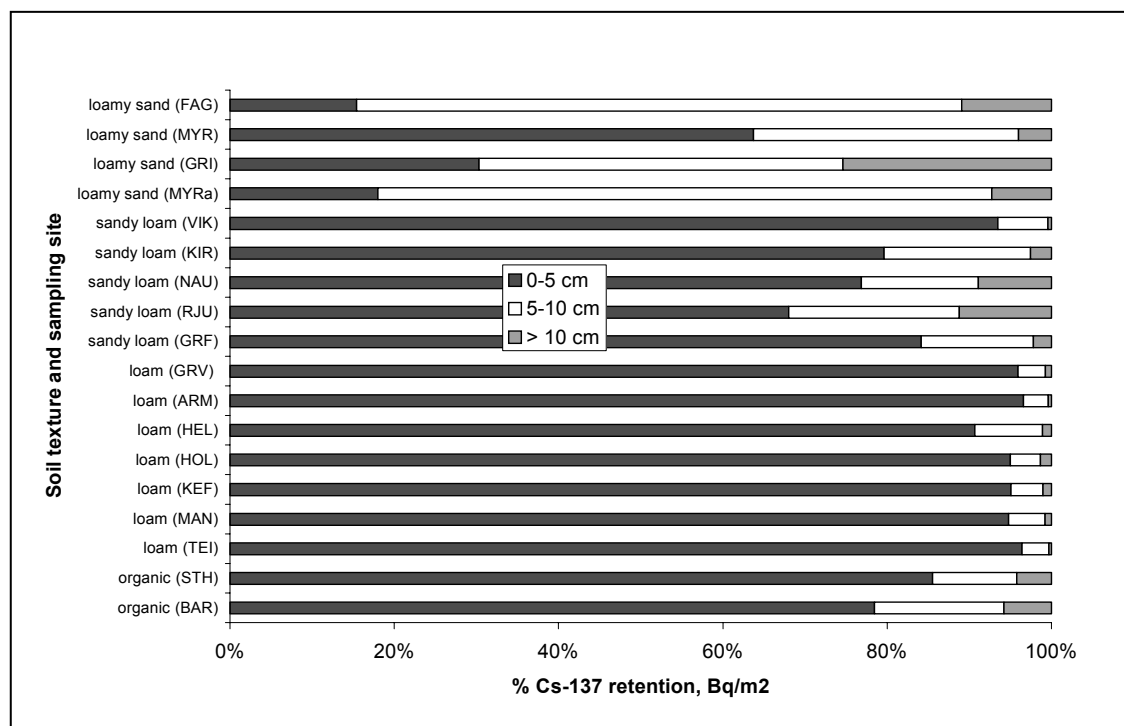


Figure 2. Radiocaesium retention of soils in relation to soil textures (data from 2000 included).

Conclusions

Good correlation was obtained between Cs-137 concentrations and precipitation. The behaviour of Cs in soils are highly dependant on soil properties, esp. clay content, indicated by good correlation between soil textures and Cs retention at various depth intervals. Several unexpected behaviour patterns were observed, e.g. at undisturbed hayfields and at sites close to the shores. Deeper penetration of radiocaesium was observed at these sites compared to other sampling sites. The causes are assumed to be some undefined soil properties and environmental factors. More detailed studies are needed to resolve these and other raised questions.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research and the Icelandic Research Council. The project is carried out in co-operation between the Icelandic Radiation Protection Institute and the Agricultural Research Institute.

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Transfer of Cs-137 from soil to plants and lamb meat in Iceland

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Introduction

The Cs-137 in the Icelandic terrestrial ecosystem is almost entirely from the nuclear weapons tests carried out in the atmosphere in the late fifties and early sixties. Relatively little reduction has been seen in Cs-137 concentrations in agricultural products during the last decade, and the reduction has been especially slow in lamb meat. Lambs roam freely and graze mostly in uncultivated highland areas during the summer time, from the time they are born in May until they are slaughtered in September or October. Gathering sheep from these areas is laborious and erosion of highland areas due to overgrazing is becoming more recognised as a problem. The importance of lowland pastures is thus likely to increase, not at least during the autumn grazing period.

Studies of transfer from soil to plants and lamb meat have been conducted within NKS projects previously, in the RAD-3 project 1990-1993 (Hove *et al.*, 1994) and in the EKO-2.1 part of the EKO-2 project 1994-1997 (Bergan, 2001). The studies in the other Nordic countries were mainly focusing on the long term trends (this was e.g. reflected in the title of the EKO-2 project: “Ecological Half-Lives of Radioactive Elements in Semi-Natural Systems”). During that project period it had become quite clear that in Iceland the change in Cs-137 concentration with time was very slow and that it was in most cases masked by other sources of variability. In the paper summarising the results from the Icelandic studies, it was concluded that understanding of these other sources of variability should therefore be the first priority in lamb studies in Iceland (Pálsson *et al.*, 2002). The lamb studies were allowed by the NKS to continue during the BOK-2 project period, but only as relatively low key studies. In Iceland it was therefore decided to build on the previous work and to focus mainly on studies at Stóra Ármót, with the aim of gaining a better understanding of the sources of variability in the lamb data sets.

Since 1995 work within the EKO-2.1 and later BOK-2.1 projects was carried out at the experimental farm Stóra Ármót in the Selfoss area, South Iceland. Previously, from 1990 to 1994, the project was carried out at the experimental farm Hestur in the Borgarfjörður area, in West Iceland. These two sites differ in many respects (Table 1). A more comprehensive description of the two sites and conditions there can be found in the paper summarising results from the Icelandic part of the EKO-2.1 lamb project (Pálsson *et al.*, 2002).

Icelandic sheep are mostly free ranging during the summer and thus have access to abundant types of uncultivated land. They therefore tend to vary their diet considerably during each grazing season as they select species with the highest nutritional content and quality at any given point of time. It is thus obvious that grazing research, conducted in a single small experimental pasture cannot be considered to be representative for conditions in a highly diverse environment. Therefore by using several distinct sampling sites a broader spectrum of data for interpretation and more interesting results will undoubtedly be gained. Additionally to the experimental farms Stóra Ármót and Hestur, the RAD-3 experiment in 1990-1993 was also being conducted at Auðkúluheiði, a common highland grazing district in north central Iceland. The experiment at that site had to be stopped due to the construction of a hydroelectric power station. The area is now a reservoir for the station and under water.

Both the Hestur and Stóra Ármót sites are typical for lowland grazing areas, Hestur as wetland and Stóra Ármót as dryland.

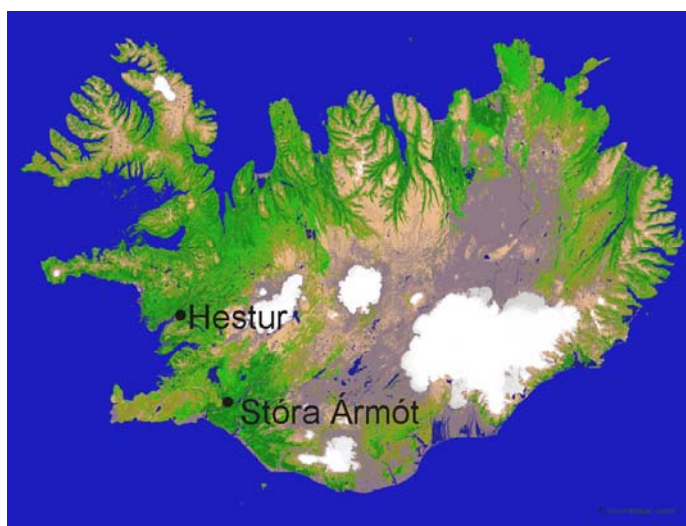
Material and Methods

Sampling and analyses

For the last decade, the NKS/RAD-3 procedure of sampling vegetation and soil had been used. Soil and composite vegetation samples were taken in 3 areas at Stóra Ármót. In each area, 3 cores of soil were taken with a coring tool of 5 cm diameter and to a depth of 15 cm. The cores were cut into 3 slices of 5 cm each and the sections from the 3 cores corresponding to the same depth were mixed together. This resulted in 3 samples from each area, corresponding to different depths. One sample of composite plant species from each area (0,75 m²) was cut close to the soil surface before soil sampling. All samples were dried in a slow-airflow, low temperature (50°C) drying cabinet. In the autumn of 2000 a new method of soil sampling, mostly adopted from the EML procedures, was started. A corer with an inside diameter of 17 mm and capable of taking 30 cm long cores was taken in use. A composite sample of about 20 cores is used (see Isaksson 2000, Sigurgeirsson *et al.* 2002a). For a while the old method will be used for comparison for some samples.

Lamb meat samples were taken at the time of slaughter, usually in the end of September or in the beginning of October. Samples were kept at –18°C until they could be analysed for radiocaesium.

Figure 1. Vegetation map of Iceland. The locations of sampling sites are shown.



The study area at Stóra Ármót is a 12 ha uncultivated lowland pasture. The surface is level, but considerably hummocky. With respect to vegetation, the whole area is homogenous in appearance. Dominating species are *Rhacomitrium hypnoides* (moss), low growing shrubs like *Salix lanata* (woolly willow) and *Vaccinium uliginosum* (bog bilberry), grasses like *Festuca vivipara* (northern fescue) and *Poa pratensis* (common meadow-grass). Other abundant species are *Calluna vulgaris* (ling, heather), *Carex bigelowii* (stiff sedge) and *Carex nigra* (common sedge). The pasture has been divided into three very similar looking sub-areas, where soil and vegetation has been sampled.

Figure 1 shows the location of the experimental farms and vegetation map of Iceland. The sampling sites are located in the two main agricultural production areas in Iceland. Some site statistics can be seen in Table 1. More information can also be found in the paper on the Icelandic EKO-2 study in the previous period (Pálsson *et al.*, 2002).

Table 1. Site statistics for the experimental farms Stóra Ármót and Hestur.

	Stóra Ármót	Hestur
location (lat.&long.)	63°60'N 20°55'N	64°25'N 21°38'W
size (km ²)	0.12	0.12
altitude (m.a.s.)	20	20
dist. to ocean (km)	19.5	5.5
type of climate	coastal	coastal
soil type	freely drained mineral soil	peat, gravelly
vegetation type	lowland heath type	lowland mire

The bedrock at Stóra Ármót is composed of an 8000 years old lava flow originating just 4 km north of the sampling area. Some outcrops are visible in the pasture. The thickness of the soil cover varies a lot, with a maximum thickness of approximately 2 meters.

Radiocaesium measurements were carried out at the Icelandic Radiation Protection Institute using gamma spectrometry (HPGe detectors).

Results

During the current period samples of soils, composite vegetation and lamb meat have been collected each year. The results obtained are summarized in Tables 2-6. These results should be compared with the previously obtained results (Pálsson *et al.* 2002) for comparison and interpretation.

Soil at Stóra Ármót

Table 2. Cs-137 concentration in 1998-2001 soil samples.

Sample	1998* Bq/kg dw	1998 Bq/m²	1998** Bq/kg dw	1998 Bq/m²	1999 Bq/kg dw	1999 Bq/m²	2000 Bq/kg dw	2000 Bq/m²	2001 Bq/kg dw	2001 Bq/m²
Area 1										
0-5 cm	230	3470	220	3279	220	2780	166	3294	123	2696
5-10 cm	6	185	14	351	14	145	4	96	12	347
10-15 cm	1	20	2	38	2	27	< 1	16	0,5	16
sum		3675		3668		2952		3407		3060
Area 2										
0-5 cm	119	2521	198	3820	198	3780				
5-10 cm	2	63	6	162	6	363				
10-15 cm	1	28	1	32	1	43				
sum		2613		4015		4186				
Area 3										
0-5 cm	136	2642	144	2452	144	2790				
5-10 cm	32	771	32	767	32	406				
10-15 cm	3	75	4	121	4	22				
sum		3488		3339		3218				

* samples taken in June

** samples taken in September

Vegetation

Composite vegetation samples

Table 3. Cs-137 concentration in 1998-2001

Year	Sampling area	Bq/kg dw
1998	Area 1	4,5
	Area 2	3,3
	Area 3	7,3
1999	Area 1	5,8
	Area 2	5,5
	Area 3	4,8
2000	Area 1	5,9
2001	Area 1	6,5

Individual plant species

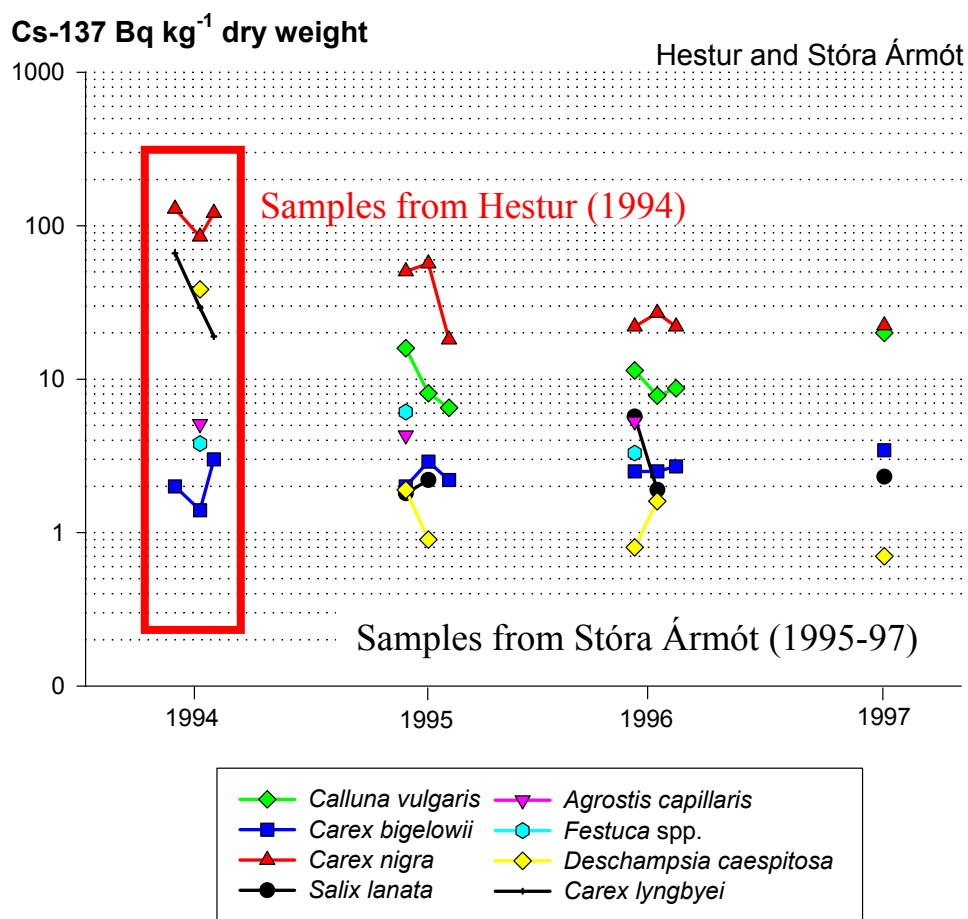


Figure 2. Cs-137 concentration in individual plant species from Hestur (1994) and Stóra Ármót (1995-97). Samples were taken late July, beginning of September and end of September / beginning of October every year. The 1997 samples are taken in September. (Data from the paper on the Icelandic results in the EKO-2.1 lamb project (Pálsson et al.2002)).

Meat

Table 4. Cs-137 concentration in 1999 lamb meat samples.

day born	weight 16 June (kg)	weight 6 Sept. (kg)	carcass weight (kg)	Bq/kg (fresh weight)
04.06.99	8	27	9,5	7,8
29.05.99	6	28	10,2	5,6
28.05.99	10	38	14,1	5,8
12.06.99	4	26	11,2	8,4
28.05.99	12	38	14,6	5,3
25.05.99	10	33	12,2	6,4

Table 5. Cs-137 concentration in 2000 lamb meat samples.

day born (exact dates not known)	weight 20 June (kg)	weight 19 Sept. (kg)	carcass weight (kg)	Bq/kg (fresh weight)
20.05.2000	9	32	11,2	4,0
20.05.2000	8	27	11,6	4,7
10.06.2000	7	29	11,8	7,8
10.06.2000	9	27	11,6	6,4
10.06.2000	7	23	10,3	4,9
10.06.2000	11	36	16,5	7,7

Table 6. Cs-137 concentration in 2001 lamb meat samples.

day born (exact dates not known)	weight 19 June (kg)	weight 19 Sept. (kg)	carcass weight (kg)	Bq/kg (fresh weight)
02.06.2001	6	28	10,8	6,2
02.06.2001	6	29	10,9	5,8
01.06.2001	6	38	11,5	5,2
02.06.2001	6	29	12,6	8,0
10.06.2001	5	35	14,3	5,9
15.06.2001	4	34	13,9	6,4
02.06.2001	5	32	11,6	6,1

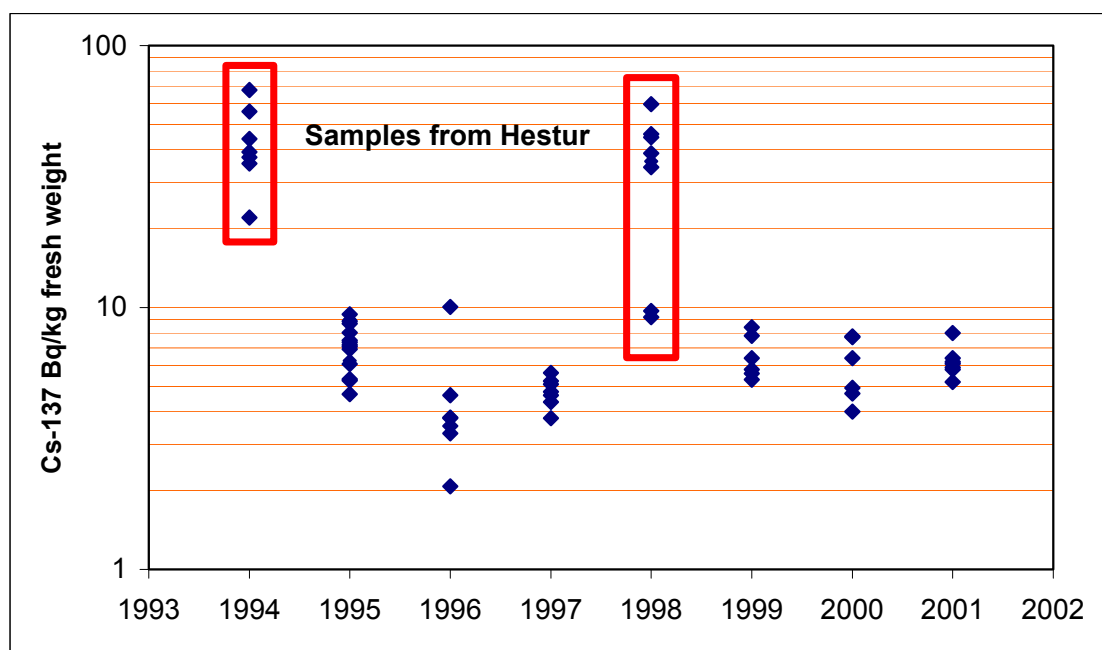


Figure 3. Cs-137 in lamb meat samples from Stóra Ármót 1995-1997 and 1999-2001. Samples from the farm Hestur are shown for comparison (from 1994 and 1998, enclosed in rectangles).

Discussion

The current results and results obtained in the previous periods (EKO-2 and RAD-3) show that:

- There is no significant reduction of Cs-137 concentration observed with time at Stóra Ármót in the sample types measured, the reduction is masked by other sources of variability.
- There were no significant differences observed in Cs-137 levels in the 3 areas at Stóra Ármót (therefore the decision was taken to concentrate on area 1 during the last two years).
- The concentration of Cs-137 in *lamb meat* at Hestur is generally an order of magnitude (10 x) higher than at Stóra Ármót.
- The same is true for *ewes milk* and *faeces*, concentration at Hestur is generally an order of magnitude higher than at Stóra Ármót. Thus the Cs-137 intake of lambs seems to be an order of magnitude higher at Hestur.
- The **composite plant samples** from Hestur also have a Cs-137 concentration an order of magnitude higher than those from Stóra Ármót.

The reason for this difference could e.g. be a combination of:

- a) Higher levels of Cs-137 deposition in the Hestur region
- b) Higher transfer rates of caesium from soil to plants in the Hestur region.
- c) Difference in relative abundance of plant species, with higher activity species being more abundant in the Hestur region.
- d) Difference in plant species selected for grazing by the sheep (the sheep can be highly selective in their grazing, so the intake does not have to reflect the general concentration in plants in the area).

The levels of Cs-137 in soil can be quite variable at each site, but there is no great difference the two sites indicating a major difference in Cs-137 deposition. Thus explanation (a) above does not seem to be likely in this case.

The Icelandic soils are unusual in many respects and have poor binding properties for caesium (Sigurgeirsson *et al.*, 2002b). Difference in soil properties could lead to higher transfer to plants at Hestur. Figure 2 shows results of analysis of individual plant species. Generally the levels are not higher at Hestur (data from 1994) than at Stóra Ármót (data from 1995 – 1997) when plants of the same species are compared, with the exception of *deschampsia caespitosa* and possibly *carex nigra*. Thus difference in soil properties and transfer to plants (explanation b) does not seem to be a major contributing factor.

Difference in selection of plants for grazing by the sheep can be a contributing factor, but not a major one, since the order of magnitude difference was also seen in the composite plant samples. These were collected by sampling all plant material above a certain height over the surface in a frame of fixed size. The order of magnitude difference can thus be found in this material. Explanation (d) can thus only explain part of the difference.

It seems that difference in relative abundance of plant species (explanation c) can explain the observed differences in the data sets from Hestur and Stóra Ármót. It was already shown in table 1 that Stóra Ármót has a freely drained mineral soil with lowland heath type vegetation, whereas Hestur is a relatively wet area with peat, gravelly type of soil and lowland mire vegetation. The greater abundance of the higher activity plants, commonly found in this type of environment, is thus a very plausible cause for the observed differences in Cs-137 concentration. In the cases of Hestur and Stóra Ármót, the higher activity vegetation species can be expected to be relatively more abundant at Hestur.

Conclusions

Icelandic sheep graze mainly on uncultivated land with many plant species. The availability of species is governed by various environmental factors, whether the area is dry or wet, soil types, average annual precipitation, height above sea level etc. The lamb studies within the NKS projects (RAD-3, EKO-2, BOK-2) have shown that fallout Cs-137 still remains readily available to plants, with little signs of reduction during the past decade. The special properties of the young volcanic soils of Iceland can explain this (Sigurgeirsson et al. 2002b). Further studies of their properties are recommended in order to gain better understanding of the processes involved.

The root of the observed variability in Cs-137 concentration seems simply to be the ecological diversity of the uncultivated semi-natural ecosystem where the sheep are grazing. Since the concentration of Cs-137 varies greatly between different plant species, difference in their relative abundance can cause considerable differences in agricultural products as have been observed here.

Much remains still to be done to understand better the behaviour of caesium in the Icelandic terrestrial ecosystem. The lamb study has been an important step in understanding roots of variability in this sub-arctic environment and it serves as a reminder to include the ecological aspects in a study of this type, not only focusing on processes.

Acknowledgements

The project was financially supported by Nordic Nuclear Safety Research (NKS). The project is carried out in cooperation between IRPI and the Agricultural Research Institute.

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Radiocaesium in Soil, Grass and Lamb at Ribe

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Introduction

Studies on the transfer of Chernobyl radiocaesium through the soil-grass-lamb foodchain have been carried out in NKS projects since 1990 (Hove et al, 1994; Bergan 1998). The purposes of these studies have covered the following items:

- to collect data from different Nordic environments on the time-dependent uptake of radiocaesium from soil to grass and further transfer to lamb,
- to make comparisons between locations and years of the uptake and transfer of radiocaesium and investigate reasons for variability (e.g. soil type, lamb's intake of soil and fungi),
- to establish time-series of data in order to document long-term trends,
- to use the data for radioecological modelling, and
- to estimate radiation doses to humans from consumption of lamb.

The present work covers follow-up during 1998-2000 on the collection of data from Ribe, Denmark. Furthermore, a special investigation on ^{129}I was carried out in 2000.

Material and Methods

Sampling

The study area is a coastal site of about 9 ha permanent grassland with sandy soil. Sampling was carried out annually of soil, grass and lamb. Soil cores were collected to 10 cm depth and sectioned into two 5-cm depth zones. Grass was cut a few cm above ground from an area of 4 m². Samples of lamb's neck were obtained from three individual animals. The study in 2000 on ^{129}I covered thyroids from 20 lambs and one grass sample.

Analyses

Caesium-137 was determined by gamma-ray spectrometry analysis based on Ge detectors. Due to the low concentrations of ^{137}Cs in grass and lamb these samples were ashed at 450°C prior to analysis. Iodine-129 was determined by neutron activation analysis.

Modelling

A radioecological model for the transfer of radiocaesium through the soil-grass-lamb system was developed during previous NKS projects (Nielsen, 1994) and used for the present data comparison.

Results and Discussion

The ^{137}Cs concentrations found in soil, grass and lamb during 1998-2000 follow the declining trend seen from previous years. The values have been compared with the corresponding levels predicted from model calculations. The results are shown in Fig. 1 showing a comparison between observed and predicted concentrations of ^{137}Cs for the entire time period 1990-2000. Soil concentrations (kBq m^{-2}) are shown with diamond marks, grass concentrations ($\text{Bq kg}^{-1} \text{ dw}$) with triangles, and lamb concentrations ($\text{Bq kg}^{-1} \text{ fw}$) with circles. The observed values from the present project period are shown with open marks and those from previous years with closed marks. This comparison shows that the observed results from 1998-2000 are well described by the values predicted by the model.

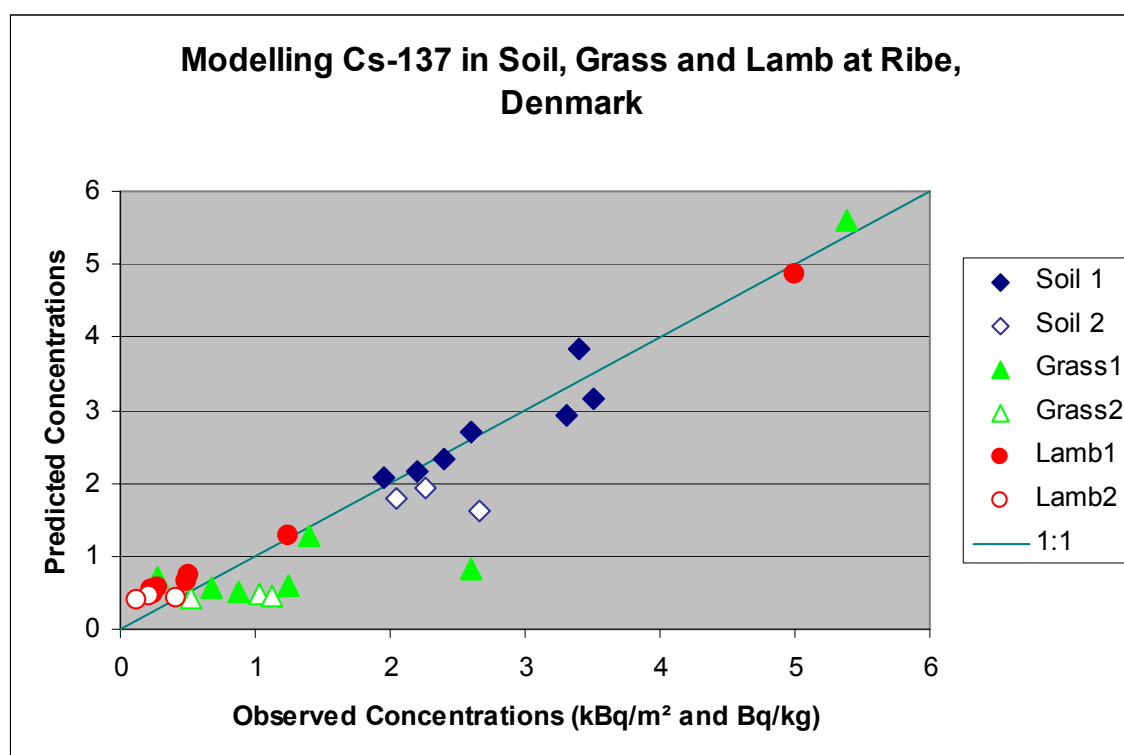


Fig 1. Comparison of observed and predicted concentrations of ^{137}Cs in soil, grass and lamb.

The ^{129}I results from the Ribe site show an average concentration of 1.3 Bq kg^{-1} in lamb's thyroids and 1.3 mBq kg^{-1} in grass. Studies in Nordic and North European countries indicate that environmental ^{129}I is due to atmospheric emissions and discharges to sea from the reprocessing plants Sellafield in the UK and La Hague in France.

Conclusions

The results observed during 1998-2000 of ^{137}Cs in soil, grass and lamb in Denmark at the Ribe site are in good agreement with values predicted by model calculations.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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¹³⁷Cs in the food-chain of lamb in the Faroe Islands. Measurements in the period 1990-2000.

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Introduction

The paper presents results from measuring ¹³⁷Cs in soil, grass and lamb meat in nine uncultivated pastures in the Faroe Islands for the years 1990-2000. Effective ecological half-lives and transfer factors are presented. Chemical characteristics of the soil have earlier been considered in connection with the observations (Joensen, 1999). Results for 1990-93 have been presented in a Nordic context by Hove *et al.* (1994). The study is part of the BOK 2.1 programme (Important Nordic food chains) of NKS.

The Faroe Islands is a mountainous rocky country consisting of 18 islands at 62°N and 7°W. The total land surface area is about 1400 km². Because of the Gulf Stream, the climate of the Faroes is milder throughout the year than the position of 62°N would normally permit. Based on measurements at synoptic weather stations, the yearly average air temperature is around 6-7 °C, with average winter and summer air temperatures around 3-4 °C and 9-10 °C, respectively (Lysgaard, 1969; Cappelen & Laursen, 1998). There are only minor temperature differences between different parts of the archipelago, but significant geographical variation is observed in precipitation rates because of combined effects from wind and topography. There are no woods in the Faroe Islands, but plenty of grass. About 94% of the land is reserved for grazing of around 70000 sheep and some cattle.

Material and Methods

Sampling

Soil, grass and lamb meat have been collected from nine uncultivated pastures across the country in the period 1990-2000 (Fig. 1). Funningur was, however, only included the first four years, and Hvalba and Sumba were not included in 2000. The first soil sampling in Hvalba took place in 1991, i.e. one year later than other pastures. Soil and grass were sampled in July-August from four randomly chosen square 0.25m² microplots in each pasture. The grass was cut from each microplot before taking three soil cores with 5.7cm diameter and length 10cm. Lamb meat (neck muscle) was collected in October from typically 5 lambs in each pasture except from Funningur, where no meat sample has been taken. The lamb carcass weight was around 12-13kg. Lamb faeces were collected in the years 1995-99, from fresh manure on the pasture ground, and measured as one sample per pasture each year.

Analyses

Grass samples from the microplots were dried at 105°C and ground before measured separately. Soil samples were dried at room temperature before measurement. The meat samples were kept frozen, thawed and measured separately. All measurements were carried out with a lead shielded Ge detector, and the software Omnigam from EG&G Ortec was used for the spectrum analyses.

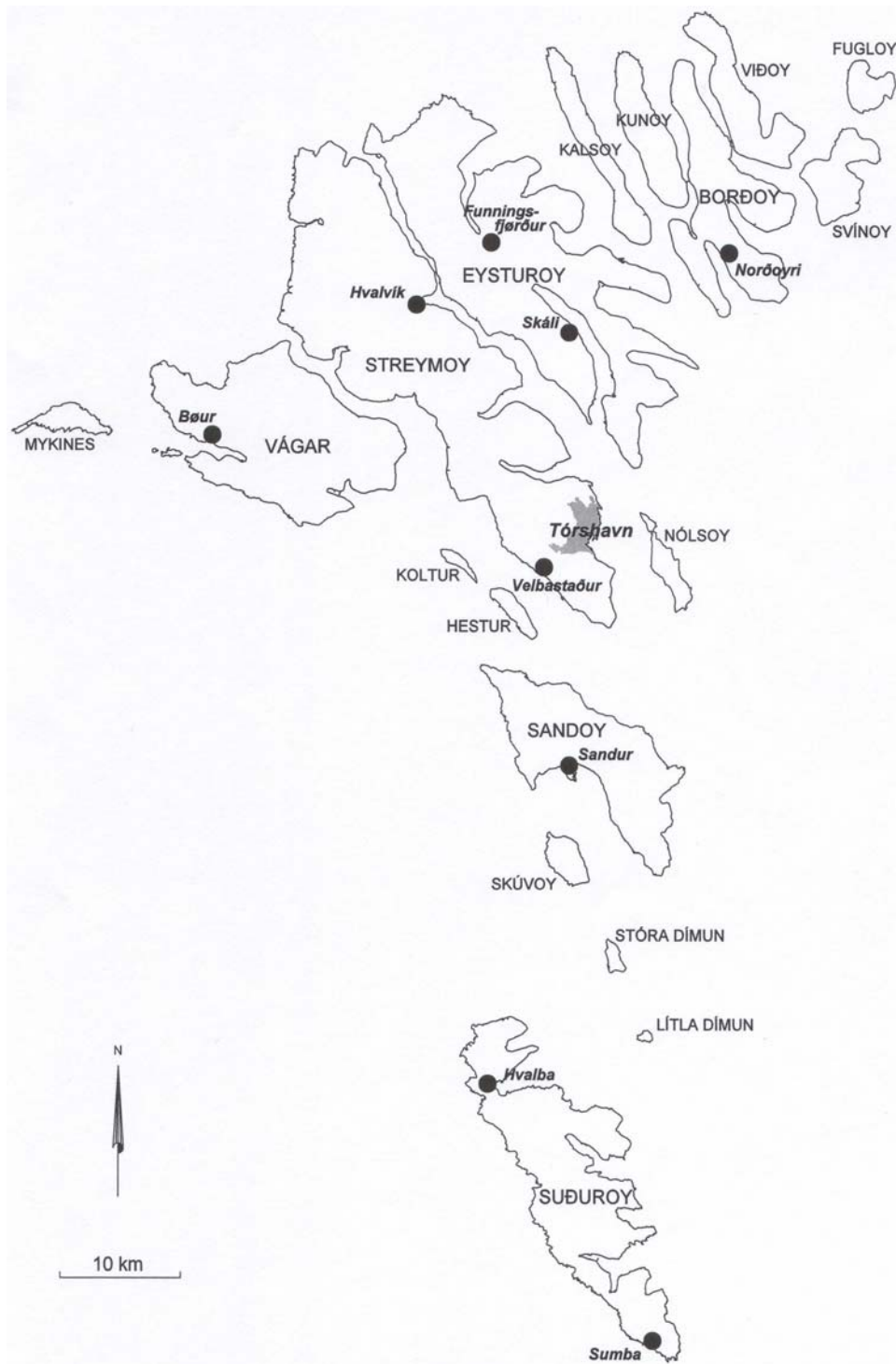


Figure 1. Map of the Faroe Islands showing the locations of the pastures selected for the study.

Results and Discussion

The ^{137}Cs deposition in the uppermost 10cm soil layer is presented in Fig. 2, showing large temporal and spatial variation within and between pastures. An exponential decay model was only acceptable for Hvalvík and Bøur, where the effective ecological half-lives, $T_{1/2}$, (VAMP, 1992), were estimated to 9.9 years ($R^2 = 0.63$) and 11.6 years ($R^2 = 0.40$), respectively. R^2 was below 0.30 for other pastures. If the deposition value for Bøur in 1990 is considered an outlier, we get $T_{1/2} = 11.6$ years for Bøur ($R^2 = 0.40$). 50-80% of the deposition in the top 10 cm soil layer is found to be in the uppermost 5 cm. The ^{137}Cs deposition, averaged across the pastures, decreased from 5.8 kBq/m² in 1990 to 5.1 kBq/m² in 1999 (all pastures were not included in 2000).

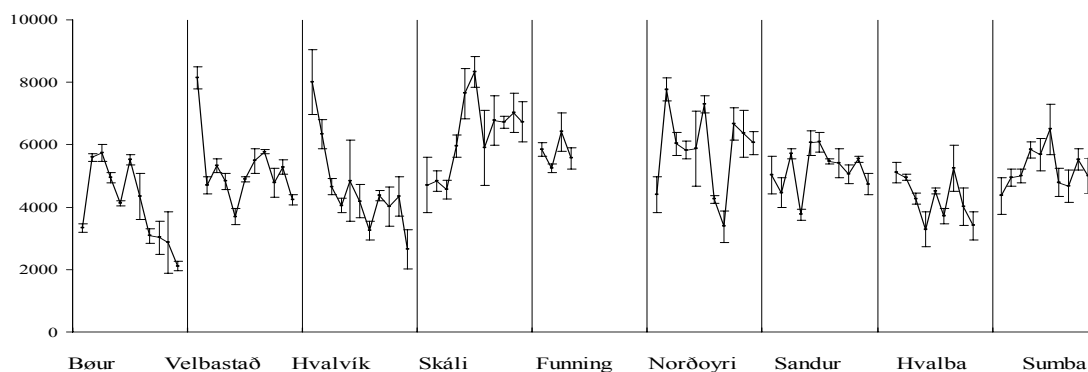


Figure 2. ^{137}Cs (Bq/m²) in 0-10cm soil layer 1990-2000. Yearly averages \pm 1 std. error. Hvalba and Sumba end in 1999.

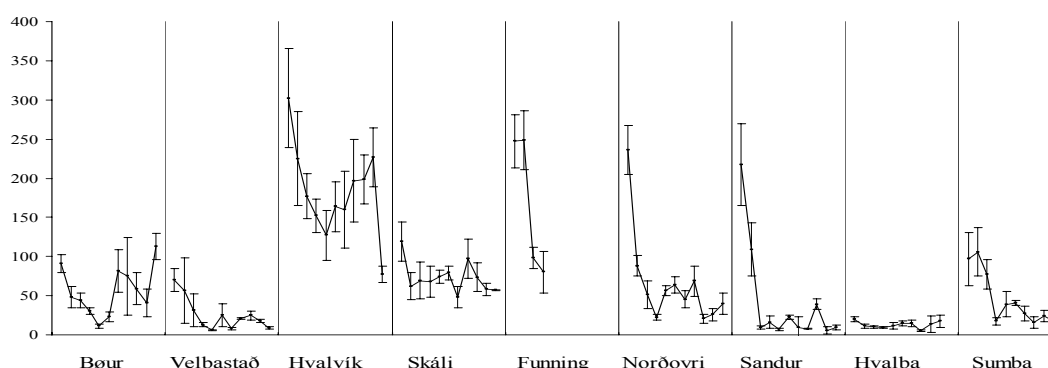


Figure 3. ^{137}Cs (Bq/kg dw) in mixed grass 1990-2000. Yearly averages \pm 1 std. error. Hvalba and Sumba end in 1999.

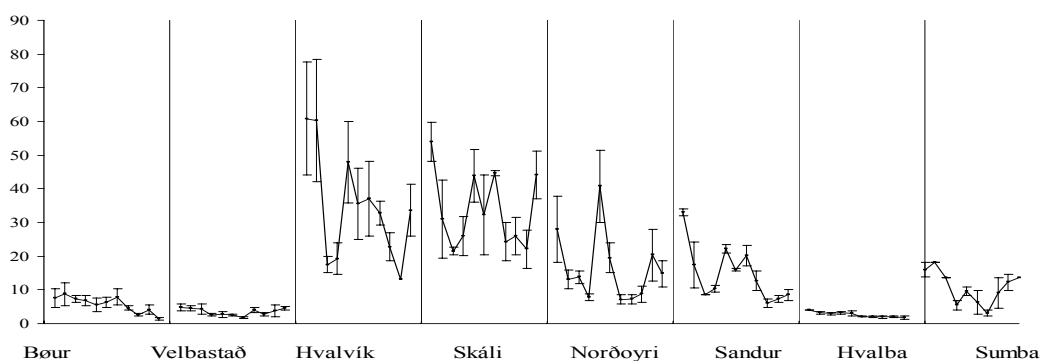


Figure 4. ^{137}Cs (Bq/kg ww) in lamb meat 1990-2000. Yearly averages \pm 1 std. error. Hvalba and Sumba end in 1999.

The pH in the soil was between 4.4 and 5.3, and loss on ignition was 50-70% (Joensen, 1999), both reflecting conditions for high uptake of radiocaesium.

The ^{137}Cs concentration in mixed grass (i.e. undetermined botanical composition) is observed to decrease in most pastures, with highest concentration in Hvalvík and lowest concentration in Hvalba (Fig. 3). Estimated effective ecological half-lives are presented in Table 1. The average ^{137}Cs concentration across the pastures was 155 Bq/kg(dw) in 1990 and 51 Bq/kg(dw) in 1999 (all pastures were not included in 2000).

The ^{137}Cs concentration in lamb meat 1990-99 is presented in Fig. 4. Large standard errors express large variations between animals. An effective ecological half-life could be estimated in four pastures (Table 1). The average concentration across the pastures was 26.0 Bq/kg(ww) in 1990 and 10.4 Bq/kg(ww) in 1999 (all pastures were not included in 2000).

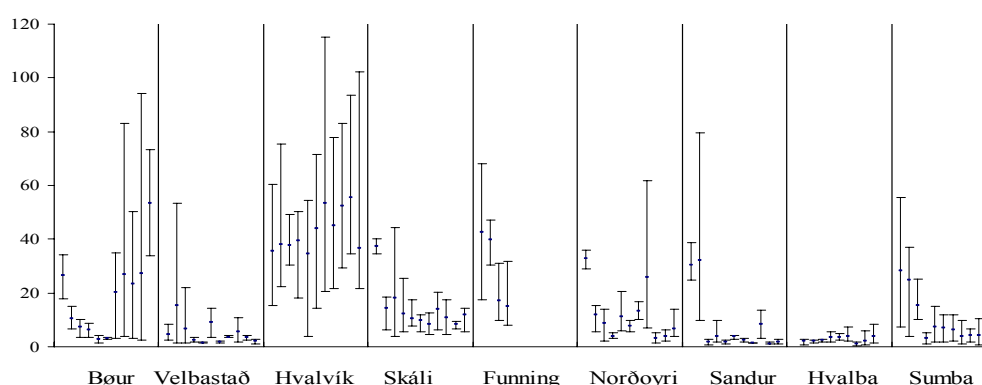


Figure 5. Soil-to-grass transfer factor of ^{137}Cs 1990-2000 ($10^{-3} \text{ m}^2/\text{kg dw}$). Yearly averages and ranges 1990-2000.

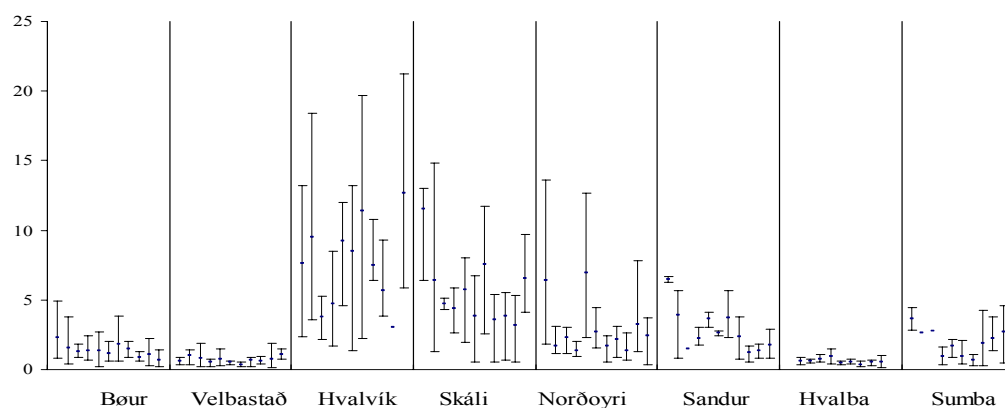


Figure 6. Soil-to-meat aggregated transfer factor of ^{137}Cs ($10^{-3} \text{ m}^2/\text{kg ww}$). Yearly averages and ranges 1990-2000.

Table 1. Effective ecological half-life in years, based on 11 year time series (1990-2000). Numbers in brackets represent R^2 from a linear regression between time and natural logarithm of ^{137}Cs concentration in the samples. No estimates if $R^2 < 0.3$.

	Bøur	Velbastað	Hvalvík	Skáli	Norðoyri	Sandur	Hvalba	Sumba
Grass	- (0.027)	5.3 (0.306)	- (0.235)	- (0.167)	5.3 (0.93)	3.1 (0.379)	- (0.005)	3.6 (0.667)
Meat	5.1 (0.668)	- (0.033)	- (0.199)	- (0.031)	- (0.060)	6.9 (0.392)	8.0 (0.873)	- (0.069)

The soil-to-grass transfer factor has been calculated for each 0.25 m² microplot used for grass and soil sampling, as the ¹³⁷Cs concentration, Bq/kg(dw), in grass divided by the ¹³⁷Cs deposition, kBq/m², in the top 10 cm soil layer (Fig. 5). The soil-to-meat aggregated transfer factor has been calculated similarly as the ratio of the ¹³⁷Cs concentration in lamb meat, Bq/kg(ww), to the ¹³⁷Cs deposition (Fig. 6). The averages across the pastures showed the following results: the soil-to-grass transfer factor decreased from 30·10⁻³ m²/kg(dw) in 1990 to 13·10⁻³ m²/kg(dw) in 1999, and the soil-to-meat aggregated transfer factor decreased from 5.5·10⁻³ m²/kg(ww) in 1990 to 2.0·10⁻³ m²/kg(ww) in 1999 (all pastures were not included in 2000).

Results from multiple linear regression between transfer factors and chemical parameters in the top 10 cm of the soil are presented in Table 2 (adapted from Joensen, 1999). Of the three chemical predictors, loss on ignition is found to be most significant. The regression coefficient (not reported) was always negative for pH and potassium, and positive for loss on ignition.

Table 2. Results from multiple linear regression between transfer factors and chemical parameters in the top 10 cm soil layer. P-values from t-test of the coefficients for the chemical parameters are given in brackets in the same order as the parameters in the header.

	Potassium and pH	Potassium and Loss on Ignition	Potassium, pH and Loss on Ignition
Soil-to-Grass Transfer	R ² = 0.15, p=0.007 (0.020; 0.043)	R ² = 0.29, p<0.0005 (0.002; 0.0005)	R ² = 0.30, p<0.0005 (0.003; 0.262; 0.001)
Soil-to-Meat Aggregated Transfer	R ² = 0.12, p=0.019 (0.264; 0.011)	R ² = 0.13, p=0.015 (0.114; 0.009)	R ² = 0.19, p=0.007 (0.156; 0.054; 0.040)

¹³⁷Cs has been measured in lamb faeces in the period 1995-99. The concentration in the samples was between 18 and 126 Bq/kg(dw), and the yearly average across the pastures was between 53 and 72 Bq/kg(dw). The average across the pastures for the faeces/meat concentration ratio and of the faeces/grass concentration ratio was 5.0 [Bq/kg(dw) per Bq/kg(ww)] and 2.1 [Bq/kg(dw) per Bq/kg(dw)], respectively.

Conclusions

Large temporal and spatial variations are found for the measured ¹³⁷Cs activities and the calculated transfer factors, both within and across pastures.

The effective ecological half-life, T_{1/2}, could not be estimated for all the sample types in every pasture. T_{1/2} for ¹³⁷Cs activity in grass and lamb meat could be estimated to 3.1-5.3 years and 5.1-8.0, respectively. Deposition of ¹³⁷Cs has not decreased significantly over the study period, except for two pastures with estimated effective ecological half-lives of 9.9 years and 11.6 years. These are all rough estimates, as large variability is associated with the measurements.

Soil characteristics are, as expected, important for the transfer of radiocaesium in the food chain of lamb. Among the three predictors potassium, pH and loss on ignition, loss on ignition is found to be most significant for transfer.

Acknowledgements

Thanks are extended to the Nordic Nuclear Safety Research (NKS) for financial support, and to my colleagues at the Faculty of Science and Technology at the University of the Faroe Islands, Trygvi Vestergaard, Johanna Zachariassen and Marjun Mortensen for taking samples, laboratory work as well as valuable discussions. Thanks are also extended to Johanna Zachariassen for her assistance in presenting data.

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Transfer of ^{137}Cs from soil to plants and sheep at Tjøtta, Norway, 1988-1998

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INTRODUCTION

Norwegian agricultural production was heavily affected by the Chernobyl accident. Large effort has been put into countermeasures and restrictions to reduce the radiocaesium levels in animals and animal products originating from areas with high deposition of ^{137}Cs

Knowledge concerning the long term behaviour of radiocaesium in the pasture-sheep ecosystem is essential to control the activity levels and to choose the most cost effective countermeasures in case of future radioactive fallout. This study was conducted at Tjøtta Rural Development Centre located on the coastline in Nordland county (66.6°N). The study site is a fenced area of 0.04 km² of a natural pasture with highly organic peaty soil and a vegetation dominated by *Poa spp*, *Festuca spp*, *Trifolium spp* and *Deschampsia cesp*. Each year 5-10 ewes and 10-20 lambs were let out in the beginning of June to graze in the area having a ^{137}Cs deposition level of about 35 kBq/m². This study started in 1988 and continued until 1998.

MATERIALS AND METHODS

The animals were live monitored every week by taking them «indoor» and separately measure them by placing the detector vertically against *on sacrum* on the standing animal. Some years the live monitoring were additionally controlled by laboratory measurements of meat samples. Total radiocaesium (^{137}Cs + ^{134}Cs) were measured in Bq/kg. The Chernobyl radiocaesium is dominating the old fallout from the nuclear weapons tests in the fifties and sixties. Data presented as ^{137}Cs were obtained by subtracting ^{134}Cs in accordance with the reported Chernobyl ratio, measured in Norway, $^{137}\text{Cs} : ^{134}\text{Cs} = 2.0$ by 1. July 1986.

RESULTS

The activity levels in lambs (fig. 1) are based on live monitoring of lambs in the period 8-12 weeks of grazing. The effective ecological half-life of ^{137}Cs in lambs 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 ^{137}Cs . The effective ecological half-life for ^{137}Cs in lambs was calculated to 10 years during the period 1988-1998 with an R^2 of 0.71. Similar calculations were done for the ewes showing an effective ecological half-life of 7 years. The correlation showed an R^2 of 0.76. As shown in figure 1, a marked decrease in activity levels in lambs was observed in 1995. This decrease was also observed for the activity levels in ewes.

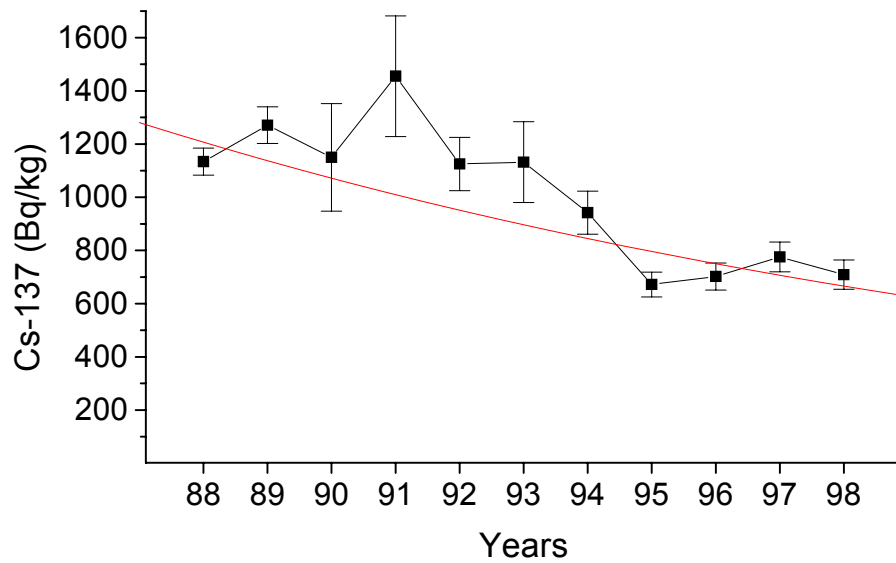


Fig. 1 Cs-137 (Bq/kg) in lambs. The figure shows the average activity levels in lamb, from 1988 to 1998, in the period 8-12 weeks after they were let out on pasture. Standard deviation of the single measurements is also shown.

Since 1988, six exclusion cages (No. 1-6) were placed on the pasture each year before the animals started grazing. The marked decrease in 1995 was also confirmed by ^{137}Cs levels in the single species from the cages showing lower levels than the previous year for all species (Table 1). The activity levels in 1996 and 1997 showed a tendency of increased levels compared to 1995. *Festuca spp* and *Rumex spp* showed the highest ability to accumulate ^{137}Cs with levels of 4200 Bq/kg and 6200 Bq/kg respectively. These levels were obtained in 1990 and 1991. In most of the species it is very difficult to observe any decrease in activity levels during the time period 1990-1997. Only for two of the eight species, *Agrostis spp* and *Festuca spp*, a decreasing activity level could be found.

PLANT	1990	1991	1992	1993	1994	1995	1996	1997
	Cs-137 (Bq/kg DW)							
<i>Trifolium spp</i>		2380	1244	1035	911		1129	1278
<i>Poa spp</i>	623	1210	133	878	950	210	543	997
<i>Agrostis spp</i>	2045	1720	256	2557	2215	260	845	999
<i>Deschampsia cespitosa</i>	1165	1700	185	159	514	180	817	809
<i>Festuca spp</i>	4201	2340	619	1648	636	255	701	662
<i>Rumex acetosa</i>	1620	6210		948	6074			4697
<i>Ranunculus spp</i>	736	1480	819	1235	1176	463	1005	935
<i>Carex spp</i>				857	3210	558		417

Table 1. Cs-137 (Bq/kg) in single species from the exclusion cages collected in late August in the period 1990-1997.

In 1995 six additional cages (No. 7-12) were put out to estimate the biomass-production and for collection of mushrooms. These additional cages were, except for 1998, cut four times

each season in intervals of one month. They were turned each month and vegetation above 3 cm not eaten by the sheep was cut down to ensure the correct biomass production to be sampled by next cutting. All vegetation samples were cut at a height of 3 cm in a predefined area. The total biomass production ranged from 260-460 (g/m²) in the years 1995-1998. During the four-year period it was shown that the biomass production was highest in the first two months. In 1995 it was a very high biomass production the first month and very low production later in the season compared to years 1996-1998.

Year	June	July	August	Sept.	Total
1995	303	72*	43	9	427
1996	92	63	60	47 ⁺	262
1997	110 ± 27	167 ± 34	80 ± 14	39 ± 12	396 ± 76
1998	154 ± 42	142 ± 39		169 ⁺ ± 56	464 ± 133

Table 2. Biomass production (g/m² d.w.) as an average from six cages. The vegetation is sampled in the middle of each month (* only from three cages. ⁺ sampled 2 September). For the years 1997 and 1998 the standard error of the mean is shown.

The data on precipitation and temperature were taken from the weather station located at Tjøtta, 4-500 m from the experimental field (table 3). It is shown that for 1995 it was very dry in May and the precipitation was very high (nearly double) compared to the normal in August.

MONTH	Normal	1994	1994	1995	1995	1996	1996	1997	1997
	Precip. (mm)	Precip. (mm)	Temp (°C)	Precip. (mm)	Temp (°C)	Precip. (mm)	Temp (°C)	Precip. (mm)	Temp (°C)
May	58,2	23,9	6,6	13,1	8,4	56,9	6,6	82,2	6,7
June	78,3	92,5	9,1	82,5	10,1	122,7	10,1	15,2	13,0
July	87,6	41,8	13,7	114,3	11,7	80,7	12,1	46,1	15,0
August	89,9	15,6	14,1	165,3	11,1	110,7	16,0	78,7	15,2
September	148,3	87,4	9,7	100,5	10,5	50,9	9,7	279,4	11,1

Table 2. Precipitation (mm) and temperature (°C) at Tjøtta is shown for the period 1994-1997 as an average of each month.

CONCLUSION

The effective ecological half-life for transfer to sheep at Tjøtta is 10 years for lambs and 7 years for ewes in the period 1988-1998. However the reduction in activity levels do not follow an exponential curve during the time of study and hence, the effective ecological half-life is dependent on the time period for which it is calculated. Hardly any decrease was observed during 1988-1993 while a significant decrease in activity levels in sheep was detected in 1994 and especially in 1995. Not surprisingly, this decrease was also observed in vegetation samples. In particular vegetation species of *Festuca* spp and *Rumex acetosa* accumulate more ¹³⁷Cs than the others, with maximum average levels of 4200 and 6200 Bq/kg respectively. In the time period from 1990-1994 *Trifolium* spp, *Deschampsia cespitosa* and *Festuca* spp showed a decrease in activity levels which was not observed in the other species studied in this period.

During the time period of experiment the occurrence of mushrooms was low, and it was studied in closer details in the last three years. In collected specimens the activity levels of ^{137}Cs has ranged from 1000 to 18.000 Bq/kg d.w. Consumption of mushrooms is not expected to explain the time variation of ^{137}Cs levels in sheep at Tjøtta.

ACKNOWLEDGEMENT

This project has received funding from NKS (Nordic nuclear safety research) and the Norwegian Research Council.

^{137}Cs concentrations in mushrooms collected in 1989-2000 at different forest stands in Kivalo experimental area

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Introduction

Although the Chernobyl ^{137}Cs fallout in 1986 was low in Finnish Lapland, averaging only 1000 Bq/m², measurable concentrations are still found in nature. The sharp, additional radiocaesium labelling with a $^{137}\text{Cs}/^{134}\text{Cs}$ isotope ratio of 1.6:1, due to the accident has made it possible to determine ecological half-lives and to estimate how much of the ^{137}Cs in the Arctic environment still originates from the global fallout period. This paper presents the results of a study on ^{137}Cs concentrations in the fruiting bodies of mycorrhizal mushrooms collected during 1989 – 2000 in Finnish Lapland.

Materials and methods

Fruit-bodies of mycorrhizal macrofungi species *Lactarius*, *Russula*, *Suillus*, *Leccinum*, *Cortinarius* and *Rozites* were collected during 1989 - 2000 in the birch, spruce, mixed forest and pine stands at the Kivalo research area of the Finnish Forest Research Institute (Metla), located 70 km SE from Rovaniemi. The mushroom samples were cleaned from soil and plant litter, sliced and dried at 105°C temperature. Analogous dry mushroom samples from 1983 were obtained from Metla's archive and used as reference material for situation before the Chernobyl nuclear power plant accident in 1986.

In autumn 1993 and 1999 four quantitative samples were taken (25 cm x 25 cm plots, 5 m from each others) at the same stands from the surface vegetation, humus and underlying mineral soil layer down to the depth of 5-6 cm. The surface vegetation and soil layers were prepared separately and dried at 105°C temperature.

All the dry mushroom, vegetation and soil layer samples were homogenised and measured by gamma spectrometry with a high purity germanium semiconductor and a multichannel pulse-high analyser. The results were analysed using STUK's own computer program. The measurement geometry was either 550 ml Marinelli containers or 35 ml or 100 ml plastic jars.

Results and discussion

According to lichen samples collected in 1987 and 1989 in a young pine stand near the Kivalo area, the total amount of ^{137}Cs in the vegetation and 1-2 cm humus layer was 1300 – 1100 Bq/m², of which 400 – 500 Bq/m² was from Chernobyl fallout.

The ^{137}Cs concentrations in the surface vegetation and soil layers at four individual sampling plots collected in 1993 and in 1999 for this research could vary largely. In 1993 the 0-5 or 0-6 cm surface samples contained an average of 770- 980 Bq/m² and in 1999 750 Bq/m², except in pine stand, 1400 Bq/m², Fig. 1.

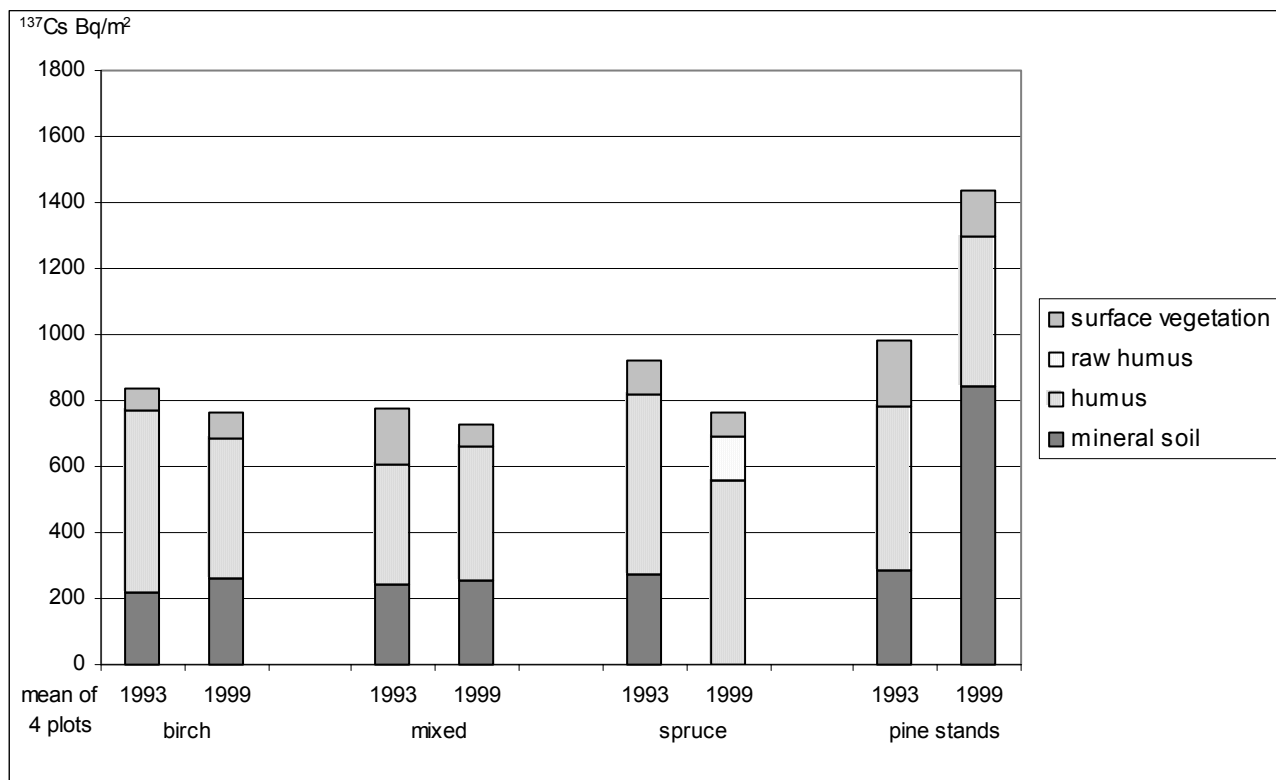


Fig 1. Average ^{137}Cs concentrations, Bq/m², in surface vegetation and soil layers at the forest stands in Kivalo in 1993 and 1999.

Time series of the ^{137}Cs concentrations, Bq/kg dry weight, in *Lactarius*, *Suillus*, *Leccinum* and *Russula* mushroom species are presented in Fig. 2 and in *Cortinarius* and *Rozites* species in Fig. 3.

Radiocaesium was found to have a long retention time in the soil layers and mushrooms. In 1993 measurable amounts of Chernobyl ^{134}Cs was still detected in most of the surface vegetation and humus layers. It was estimated that about 50 % of the total ^{137}Cs in the surface vegetation layer and 60 % in the humus layer, was derived from global fallout period. In the heavily grazed lichen carpet and surface vegetation of the pine stand, however only 20 % was old caesium.

In 1989 about 75 % and in 1990-1993 about 60 %, of the total ^{137}Cs in the mushrooms originated from the global fallout period. The calculations are based on Chernobyl fallout $^{137}\text{Cs}/^{134}\text{Cs}$ isotope ratio of 1.6:1. The high proportion of the global fallout caesium in the mushrooms was also evident when the mushroom concentrations in 1989 were compared to the reference samples from the same stands in 1983. The levels were almost the same in most of the species.

During the 12-year follow-up period the ^{137}Cs concentrations of *Lactarius*, *Russula*, *Suillus*, *Leccinum* and *Rozites* species decreased slowly, with a 7 to 10 or 18-year-long ecological half-life ($T_{1/2}$). For *Cortinarius armillatus*, a poisonous mushroom showing the highest caesium concentrations, the ecological half-life appears to be of the order of the physical half-life of ^{137}Cs , 30 years. Table 1 represents the estimated ecological half-lives of ^{137}Cs of the different mushroom species, and as an example the fresh weight concentrations measured in mushrooms collected in 1990 and 1991.

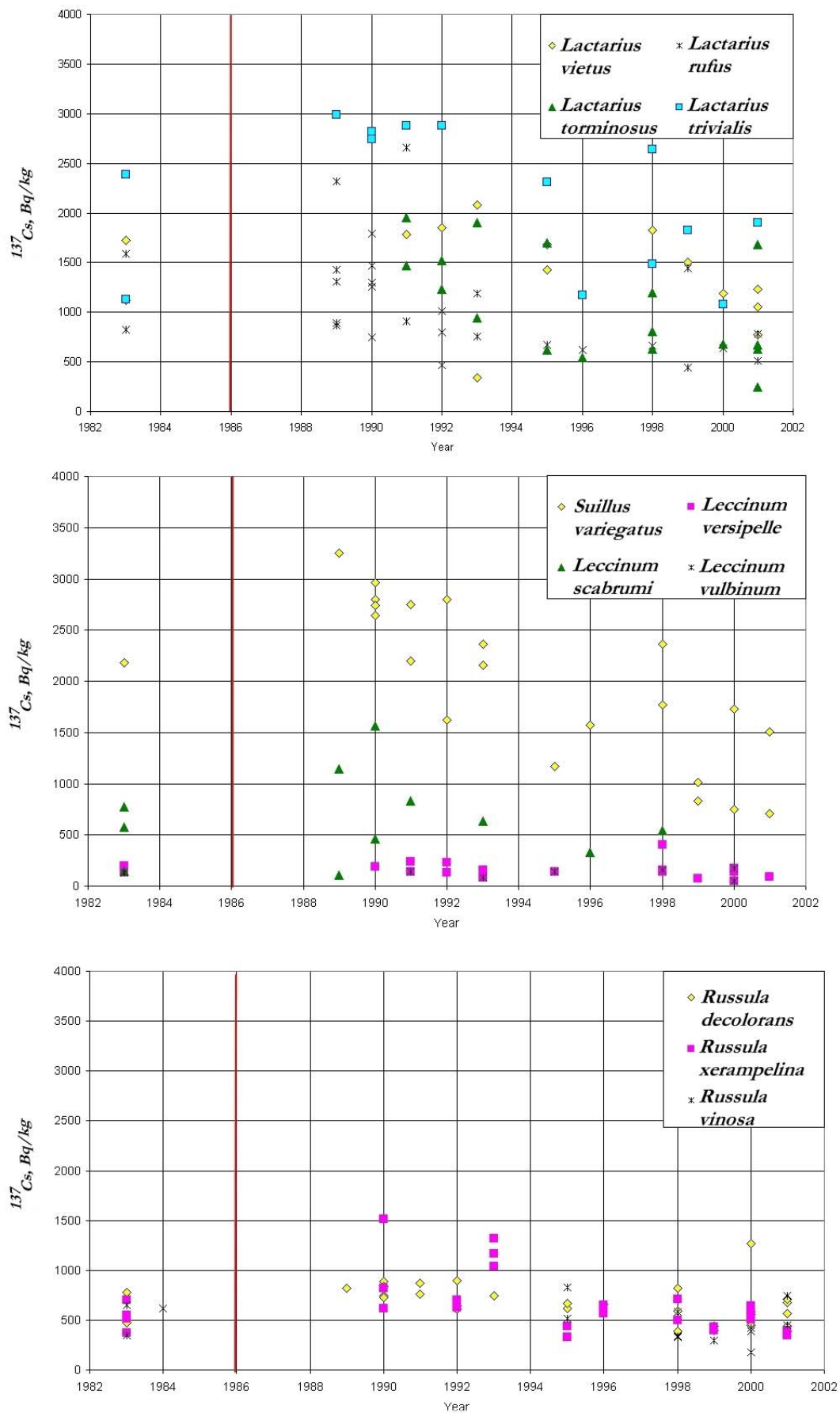


Fig 2. ^{137}Cs concentrations, Bq/kg dw, in 1983 and 1989-2001 collected *Lactarius*, *Suillus*, *Leccinum* and *Russula* samples at the Kivalo forest stands.

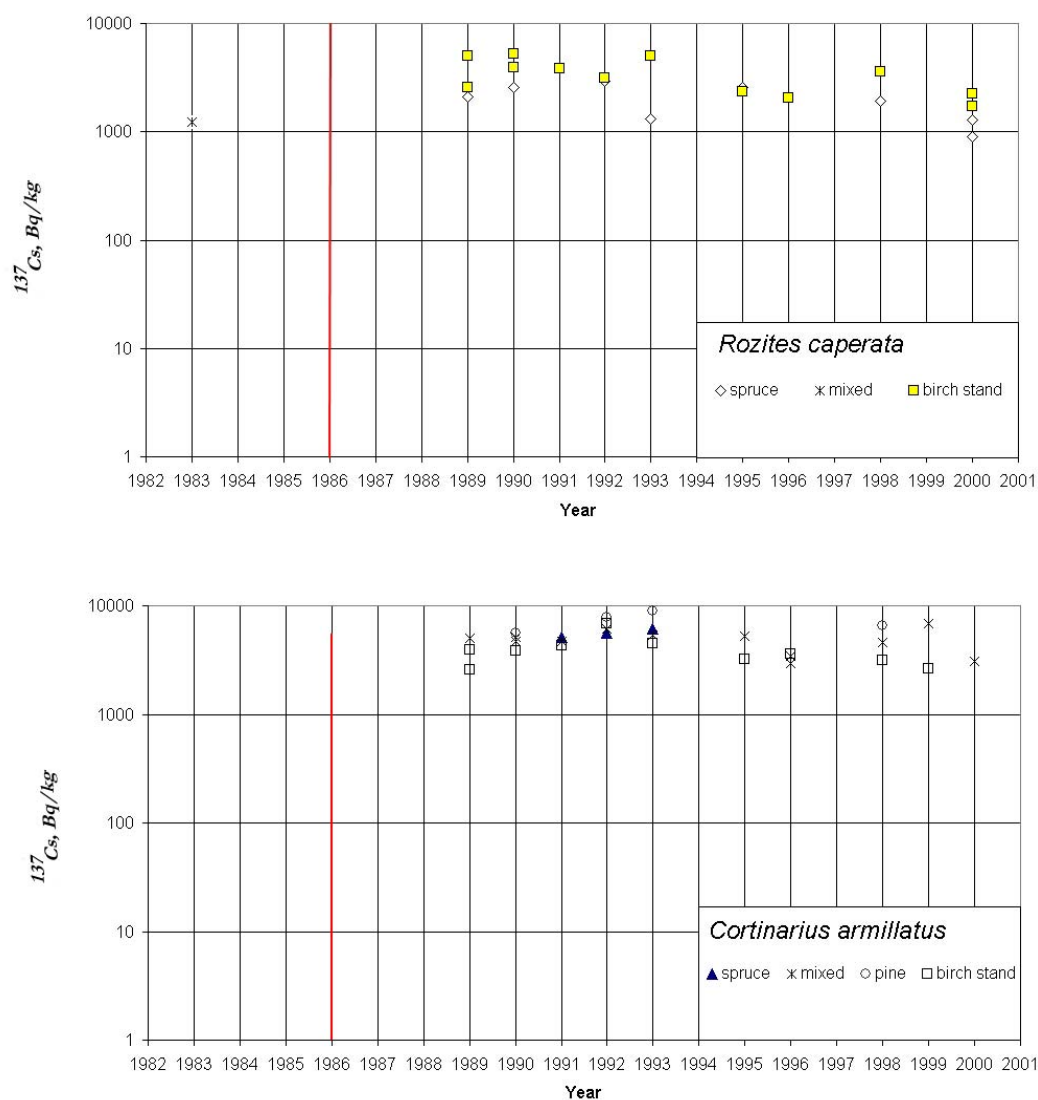


Fig 3. ^{137}Cs concentrations, Bq/kg dw, in 1983 and 1989-2001 collected *Cortinarius* and *Rozites* samples at the Kivalo forest stands.

The accumulation rate for different mushroom species varies, but there does not seem to be any clear correlation between the ^{137}Cs concentrations (Bq/kg fw) in the 1990-1991 mushrooms and the length of the ecological half-life. This is despite the fact that the highest concentrations have been measured in *Cortinarius armillatus*, the ecological half-life of which appears to be of the order of the physical $T_{1/2}$ of ^{137}Cs , 30 y. All the *Russula* species have had low concentrations, although the half-lives are rather long 11-18 y. *Lactarius trivialis* has higher levels than the other analysed *Lactarius* species but there is no correlation with the half-lives. The type of forest stand does not seem to have any effect on the caesium concentrations, except possibly for *Rozites*.

Table 1. ^{137}Cs concentrations, Bq/kg fresh weight, in 1990-1991 collected Kivalo mushrooms. The estimation of the ecological half-lives, $T_{1/2}$, of ^{137}Cs in mushrooms are based on results from 1989-2000.

Species (number of samples)	^{137}Cs 10-100 Bq/kg	^{137}Cs 100-200 Bq/kg	^{137}Cs 200-300 Bq/kg	^{137}Cs 300-400 Bq/kg	^{137}Cs 400-500 Bq/kg	$T_{1/2}$
<i>Leccinum vulpinum</i>						
<i>Leccinum versipelle</i> (3)	17					
<i>Leccinum scabrum</i> (3)	92					
<i>Suillus variegatus</i> (6)			220			8 years
RUSSULA						
<i>Russula vinosa</i> (1)		110				
<i>Russula xerampelina</i> (3)		130				11 years
<i>Russula paludosa</i> (1)		110				
<i>Russula decolorans</i> (7)		110				18 years
MILK CUP						
<i>Lactarius rufus</i> (7)		150				10 years
<i>Lactarius torminosus</i> (2)		150				7 years
<i>Lactarius vietus</i> (1)		170				16 years
<i>Lactarius trivialis</i> (3)			230			11 years
CORTINARIUS						
<i>Rozites caperata</i> (4)					440	10 years
<i>Cortinarius armillatus</i> (7)					480	30 years

Conclusions

The human consumption of mushrooms is not very common in northern Finland and is especially low by the Saami population. *Lactarius* species need parboiling, which decrease the radiocaesium concentration by 90 %. "Boiling and spoiling" treatment is too common also for other mushroom species when prepared as human food. After a nuclear accident people are able to avoid mushroom consumption.

Reindeer will continue to collect as many mushrooms as available, because the mushrooms are their most important autumn fodder. This can be noticed in the condition of the animals after a good and long mushroom growing season compared to the high death rate of reindeer during hard winter conditions after a poor and short mushroom period in autumns due to dry summer and early night frosts. Because radiocaesium is effectively accumulating in mushrooms and the rate of decrease is slow, mushrooms will contribute significantly, for a long period of time to the intake of reindeer following a severe nuclear accident. This phenomenon has clearly been visible, after good mushroom seasons, in reindeer and other animals that consume large amounts of mushrooms in those parts of Norway and Sweden most affected by the Chernobyl emissions.

The high accumulation of ^{137}Cs in mushrooms and their importance to the radiocaesium intake of humans, reindeer, deer, lam and other mushroom consuming species was not fully understood before the Chernobyl accident. During the atmospheric nuclear test period hardly any attention was paid to the radioactivity concentrations in mushrooms.

SR-90 IN COW'S AND GOAT'S MILK IN NORWAY

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Introduction

Since the nuclear weapons tests and the Chernobyl accident, substantial research has been conducted on the transfer and uptake of ^{137}Cs in milk from radioactive fallout. A considerable amount of data is also available for ^{90}Sr after the nuclear weapons tests, but data on ^{90}Sr after the Chernobyl accident are rather sparse. Usually, the fallout of ^{137}Cs will be larger than for ^{90}Sr for accidents involving nuclear bombs or reactors. Yet, there are nuclear accidents where ^{90}Sr may be the main contaminant of concern, for instance the Kyshtym accident in former USSR in 1957.

Since strontium is a calcium analogue, strontium will mainly be taken up in bone and milk. The main exposure pathway to humans will thus be consumption of contaminated milk. This paper summarises data on ^{90}Sr contamination in milk from 1964 up to present analysed by IFE and NRPA.

IFE has analysed milk samples from cows in the Kjeller area around their research reactor since 1964. The discharges of ^{90}Sr from the reactor are negligible compared to the nuclear weapons fallout (NWF) and the Chernobyl fallout, thus the reported levels originate from the NWF and the Chernobyl accident.

NRPA has analysed ^{90}Sr in milk samples from cows and goats since 1997. Cow's milk has been collected from 10 dairies nation wide and three livestock from areas moderately to heavily contaminated after the Chernobyl accident. Goat's milk has been collected from two dairies and two livestock likewise.

Materials and Methods

IFE samples

Since 1964 samples of milk have been collected weekly from different farms in the Kjeller area. The sampling has been carried out as a part of the environmental surveillance program for the nuclear activity of the Institute for Energy Technology at Kjeller. Until 1980, the samples were combined to monthly samples and analysed for ^{90}Sr . After 1980, the samples were combined to quarterly samples before analyses.

Before 1990, ^{90}Sr was separated from the matrix with fuming nitric acid, and after a period of ingrowth, ^{90}Y was counted on a low background (0.8 cpm) beta counter.

After 1990 the combined samples were dried at 125 °C and ashed at 500 °C. ^{90}Sr was analysed by separating and counting its daughter product, ^{90}Y . Y was separated from the matrix by successive extractions with tri-butyl phosphate, precipitated as the hydroxide and converted to the oxalate for counting. The ^{90}Y and ^{90}Sr was in radioactive equilibrium at the

time of separation, so no period of ingrowth was necessary. Titration with EDTA was used in chemical yield determinations. All samples collected after 1991 were counted on low-level anticoincidence beta detectors, GM-25-5 (background 0.15 cpm).

NRPA samples

One sample of approximately 1.5 litres was taken from each location in September each year at the end of the grazing period. The samples were freeze-dried and ashed in muffle furnaces. ^{90}Sr was analysed by separating and counting its daughter product, ^{90}Y . Fuming nitric acid was used for the chemical separations. After ingrowth of equilibrium, the samples were measured by low background anticoincidence beta counter, GM-25-5. ^{85}Sr and titration with EDTA was used in chemical yield determinations.

Results and Discussion

Activity concentration in milk from the Kjeller area

Figure 1 shows the annual average activity concentration of ^{90}Sr for all the farms where samples have been collected from 1964 to 2000. Through the 1960's, -70's and the first half of the -80's, the concentration of ^{90}Sr in milk was generally decreasing. In a nation-wide survey of ^{137}Cs and ^{90}Sr in milk (T. Hvinden and A. Lillegraven, 1970) the ^{90}Sr -concentration in milk in this area was reported to be 407 mBq/litre (11 pCi/litre), which is the same order of magnitude as our results.

The increase in ^{90}Sr -concentration in 1976 is caused by relatively high results for the samples from the third quarter of the year. The reason for this is not understood, and the results are not in agreement with other measurements performed that year. An increase in the fallout was first detected for the last three months of 1976. The fallout in the first nine months of 1976 was "normal" for that period.

In 1985, a 2-fold increase compared to 1984 was observed. No explanation has been given for this. The continuing increasing concentrations from 1986 to 1988 are caused by the fallout from the Chernobyl accident. It seems to be a delay between the time of the Chernobyl accident and the maximum ^{90}Sr -concentration in the milk. This might be due to a higher biological availability of ^{90}Sr for the cows after a period of time.

A new concentration peak was reached in 1992. No reasonable explanation has been given for this. After 1992 the concentration has decreased and was about 50 mBq/litre in 2000.

It should be noted, however, that the data presented in this paragraph are surveillance data and that the uncertainty in the measurements therefore is higher than for data obtained through research projects. This might be an explanation for some of the otherwise unexplained data.

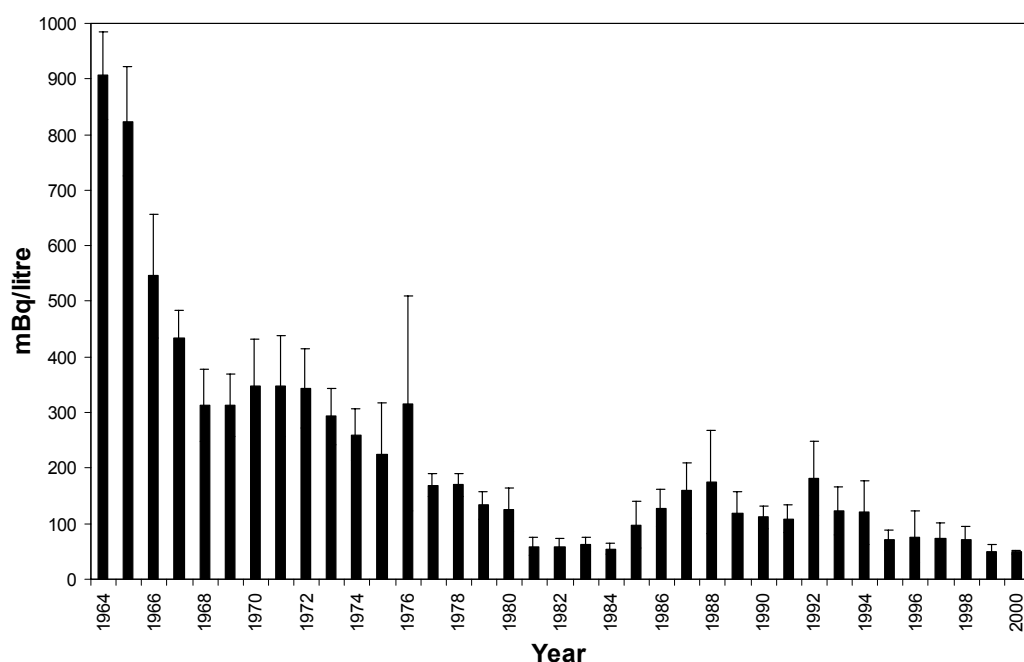


Figure 1. Annual average activity concentration of ⁹⁰Sr in cow's milk from selected farms in the Kjeller area.

Activity concentration in cow's and goat's milk from dairies and livestock

Figure 2 shows the location of the dairies where samples have been collected. The results are presented in table 1. Generally the data show low ⁹⁰Sr activity concentrations in cow's milk, but there are some elevated concentrations in samples from Fosheim and Østfold in 1997 (Liland et al., 2001). The concentration in milk from Fosheim probably reflects that this dairy is situated in an area of Norway receiving significant Chernobyl fallout. Furthermore, traditional grazing of dairy cattle on unimproved mountain pastures during summer is common in this area. No reason is known for the relatively high activity concentration in milk from Østfold, except that it may be a result of the larger uncertainty in the analysis. This is further confirmed in 1999 when the values at Østfold had dropped substantially. All locations except two show a decrease in ⁹⁰Sr activity from 1997 to 1999. For Sandnessjøen the increase is within the uncertainty, while for Tunga there was an increase of approximately 85 %. There is no apparent reason for this increase.

The ⁹⁰Sr contamination in goat's milk from the two dairies is generally higher than observed values for the cow's milk samples. This is consistent with a known higher transfer of ⁹⁰Sr to smaller ruminants than to larger ruminants (Russell, 1966).

Figure 3 shows locations where samples were taken from selected livestock. Animals in these livestock are part of a monitoring program for cow's and goat's milk during the grazing season in Norway. The results for ⁹⁰Sr are given in table 2.

In Øystre Slidre part of the livestock are given concentrates with AFCF¹ to reduce ¹³⁷Cs uptake while, for research purposes, others are not. The samples from these livestock are

¹ AFCF – Ammonium ferric hexacyano ferrate – also known as Prussian Blue is used as a caesium binder to reduce the uptake of radiocaesium by ruminants.

thus divided in two. The use of AFCF is not reported to influence on the ^{90}Sr uptake, however, it may seem that higher values for ^{90}Sr are recorded when AFCF is distributed to the animals in this project.

The results from the livestock confirm the higher transfer to goat's milk compared to cow's milk. For goat's milk in Øystre Slidre the ^{90}Sr activity concentration is generally in the range 0.8 - 1.2 Bq/l, while it is 0.6 – 1.0 for cow's milk except two higher values (1.2 and 1.35 Bq/l). In Vevelstad and Vega the ^{90}Sr activity concentration in cow's milk is in the range 0.1 - 0.2 Bq/l. The goat's milk from Namsskogan has ^{90}Sr activity concentration in the range 0.35 to 0.40 Bq/l.

When comparing the results from livestock 1998 to 2001, there is no general decreasing trend for ^{90}Sr in milk. Small annual variations are apparent for most livestock even when the analytical error is taken into account.

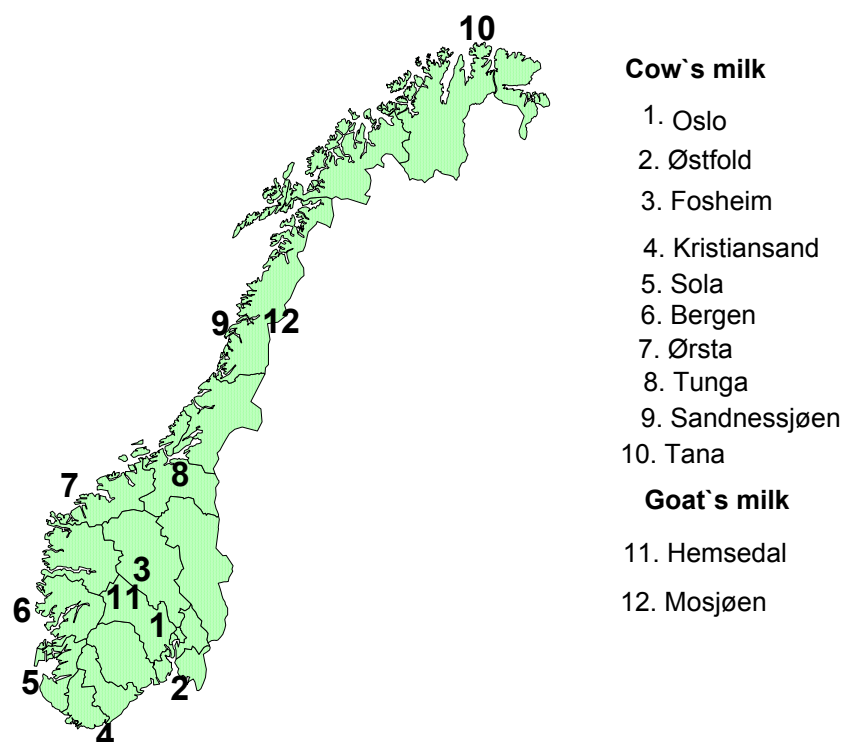


Figure 2. Sampling from Norwegian dairies

Table 1. ^{90}Sr activity concentration in cow's and goat's milk from Norwegian dairies.

Dairy	^{90}Sr (Bq/l) 1997 $\pm \sigma_{\text{total}}^*$	^{90}Sr (Bq/l) 1999 $\pm \sigma_{\text{total}}^*$
1. Oslo	0.055 ± 0.011	0.053 ± 0.011
2. Østfold	0.120 ± 0.030	0.040 ± 0.010
3. Fosheim	0.130 ± 0.020	0.089 ± 0.015
4. Kristiansand	0.070 ± 0.014	0.020 ± 0.005
5. Sola	0.038 ± 0.007	0.029 ± 0.006
6. Bergen	0.085 ± 0.015	0.075 ± 0.014
7. Ørsta	0.070 ± 0.013	0.050 ± 0.011
8. Tunga	0.070 ± 0.019	0.130 ± 0.020
9. Sandnessjøen	0.044 ± 0.010	0.050 ± 0.011
10. Tana	0.053 ± 0.008	0.047 ± 0.011
11. Hemsedal	0.201 ± 0.028	0.125 ± 0.019
12. Mosjøen	0.252 ± 0.034	0.115 ± 0.018

* σ_{total} - total analytical uncertainty

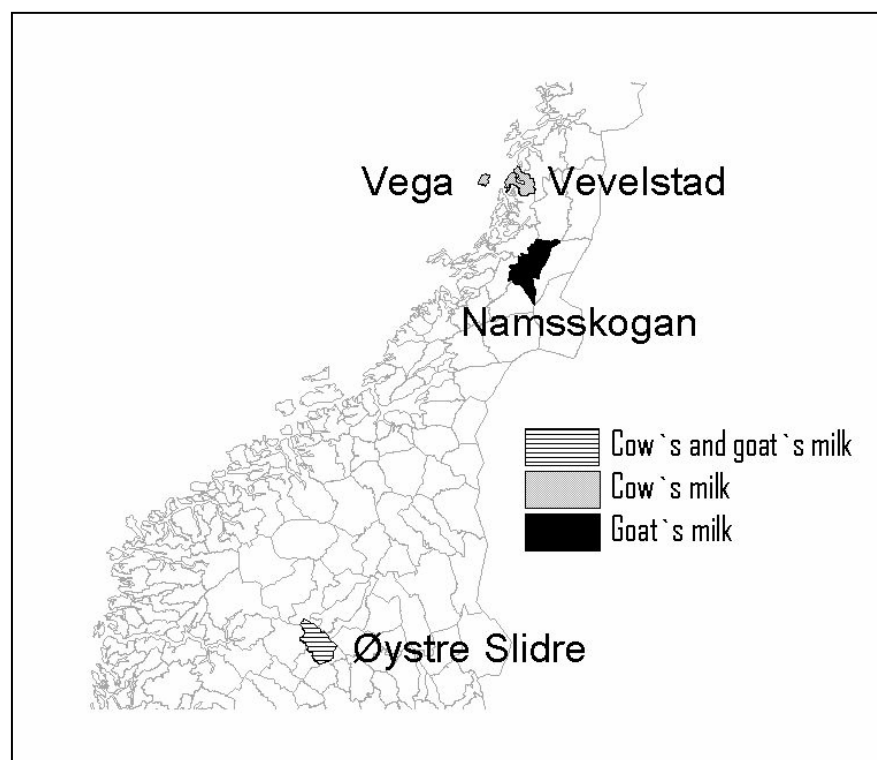


Figure 3. Municipalities where milk is sampled from selected livestock for ^{90}Sr analyses.

Table 2. ^{90}Sr activity concentration in cow's and goat's milk from selected livestock in Norway.

Milk from	Livestock or cow's number	Location	^{90}Sr (Bq/l) $\pm \sigma_{\text{total}}$ *			
			Sept.1998	Sept.1999	Sept. 2000	Sept.2001
Goat (AFCF)	544-0334	Øystre Slidre	1.08 \pm 0.07	1.01 \pm 0.08	1.21 \pm 0.07	0.91 \pm 0.05
Goat (no AFCF)	544-0334	Øystre Slidre	0.78 \pm 0.06	1.08 \pm 0.08	1.09 \pm 0.06	0.86 \pm 0.05
Cow (AFCF)	544-2013	Øystre Slidre	1.03 \pm 0.08		0.89 \pm 0.05	1.20 \pm 0.07
Cow (no AFCF)	544-2013	Øystre Slidre	0.59 \pm 0.05			
Cow - Kronos	Cow no.275	Øystre Slidre		0.61 \pm 0.05		
Cow - Lykkebot	Cow no. 293	Øystre Slidre		0.67 \pm 0.05		
Cow - Bota	Cow no. 313	Øystre Slidre			0.57 \pm 0.03	
Cow - Malik	Cow no. 318	Øystre Slidre			0.71 \pm 0.04	1.35 \pm 0.08
Cow - Kronild	Cow no. 321	Øystre Slidre		0.61 \pm 0.05	0.63 \pm 0.04	0.84 \pm 0.05
Cow - Krone	Cow no. 335	Øystre Slidre				0.73 \pm 0.04
Goat	1740-4985	Namsskogan		0.40 \pm 0.03	0.40 \pm 0.03	0.37 \pm 0.02
Goat	1738-2052	Namsskogan			0.34 \pm 0.03	0.37 \pm 0.02
Cow	16-0047	Vevelstad	0.20 \pm 0.02	0.13 \pm 0.01	0.070 \pm 0.009	0.114 \pm 0.007
Cow	16-0027	Vevelstad	0.12 \pm 0.01	0.10 \pm 0.01	0.101 \pm 0.007	0.107 \pm 0.006
Cow	15-0158	Vega	0.22 \pm 0.02	0.13 \pm 0.01	0.178 \pm 0.015	0.162 \pm 0.010

* σ_{total} - total analytical uncertainty

Conclusions

Samples from the Kjeller area show a general decreasing trend over the years according to deposition. Yet, there are some unexplained peaks. The samples from the dairies show a general decrease from 1997 to 1999 except for one location. By contrast, the samples from the livestock show no general decreasing trend over the years 1998-2001.

The project confirms a general higher uptake of ^{90}Sr in goats compared to cows. In addition, a possible higher uptake of ^{90}Sr in milk is observed when AFCF is distributed to the livestock. More data is needed, though, to confirm this.

The ^{90}Sr activity concentrations in the Kjeller area in the late 1990's are comparable to the dairy measurements from the nearby areas Oslo and Østfold. These values are among the lowest reported corresponding to a low impact from the Chernobyl accident in these areas. The reported values are higher in Vevelstad and Vega that experienced a medium impact, while the highest values are registered in Øystre Slidre, the most heavily affected municipality in Norway after the Chernobyl accident.

If further funding is available in the future, it will be interesting to investigate the ^{90}Sr deposition in Norway following the NWF and the Chernobyl accident, and estimate transfer factors to milk based on the monitoring data presented here.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research and The Norwegian Food Control Authority.

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A Study of Plutonium in different fresh water systems

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Abstract

Plutonium has been studied in different lake systems, spanning from clear water oligotrophic lakes in the high arctic to humic rich dystrophic lakes in the temperate regions. Correlation's between radio caesium, ²¹⁰Pb and the plutonium inventory in the lake sediments has been studied. In the high arctic lakes we find higher inventories of plutonium, radiocaesium and ²¹⁰Pb than expected from fallout data and the annual flux of ²¹⁰Pb at this latitudes. We hypothesize that this is due to that the lakes acts like traps for wind driven material which is caught by the water surface during the time of year when the lakes are ice-free. The influence of the Thule accident in the high Arctic lakes was also studied with respect to resuspension of debris to the lakes from the catchment area. Judging from the plutonium isotopic composition and ²³⁹⁺²⁴⁰Pu/¹³⁷Cs ratio, no detectable plutonium from the accident were found in the lakes situated in the Thule region.

The dystrophic lake studied shows very high levels of fallout plutonium (130-220 µBq l⁻¹, annual variation) in the water. This plutonium is largely associated to humic colloids (77% was in the fraction between 1 µm and 100 kD), obviously the fate of these colloids plays an important role for the behaviour of plutonium in this fresh water systems.

Introduction

Lakes show a much larger variation than the marine environment. A large number of parameters influence the behaviour of radionuclides, such as morphometry (lake area, catchment area, depth, shape, volume, water residence-time), physical properties (light conditions, temperature, turbidity, precipitation, sediment structure, suspended matter, stratification, ice formation), chemical properties (pH, conductivity, K⁺, Ca²⁺, Mg⁺², organic content, nutrients, sediments) and biological factors (littoral/profoundal, biological activity, transport to biota).

For the high arctic lakes an area around Thule, Greenland was selected. Thule is particularly interesting since in January 1968, a B-52 aircraft caught fire and crashed on the ice at Bylot Sound, 11 km west of the Thule air base. The aircraft carried 4 unarmed nuclear warheads, which performed a conventional explosion at impact and caused contamination of the sea ice.

Most of the plutonium was recovered but about 1800 GBq sank to the sea floor when the ice broke up in June (USAF, 1970, Eriksson *et al.*, 1999). It was also stated that about 30-180 GBq drifted west-south west with the cloud of smoke and debris (Hansson, 1980). The clean up operation started several weeks after the accident meanwhile the contaminated snow and ice were exposed to severe weather conditions, which caused resuspension of plutonium. Air data from Environment Measurement Laboratory (EML,

1999) showed increased concentrations of plutonium at the Thule air base up to one year after the accident. A 20-100-fold increase of plutonium in lichens and soil were found in 1974 at certain areas. (Hansson, 1980). It is therefore not unlikely that the accident could be recorded in sediments from lakes in that area. To study the magnitude of resuspended plutonium and the spatial distribution, four lakes with different distance to impact site were chosen.

For a temperate region we selected dysoligotrophic and oligotrophic lakes in southern Sweden. These lakes have seasonal effects with ice formation with seasonal or permanent anoxic sediments influencing the plutonium mobility. The choice of lakes is based on the idea that plutonium (together with other actinides) form humic complexes of importance for the processes in the lakes. The remobilization of actinides from the sediments might follow the iron manganese redox cycle (Alberts *et al.*, 1987) or forming humic complexes (Roos *et al.*, 1993).

Materials and methods.

High arctic lakes are generally difficult to get access to for sampling. For the Arctic lakes at Thule (Pitufikk) Greenland there is an infra structure and from time to time expeditions are undertaken following the B-52 accident in 1968. In this way equipment could be shipped from Denmark to Thule (Pitufikk) in connection with such an expedition in August 1997.

The Thule area is situated at 76.6 °N and has a low precipitation (130 mm a⁻¹). The sea and the lakes are frozen about 10 month per year. Four lakes were selected and these lakes are typical shallow clear water lakes with depths between 2 –12 m. Lake Pia (V1) (maximal depth 4 m) and Lake Lena (V2) (maximal depth 12 m) are quite similar in size while lake Pia has a larger catchment area. Lake Lena actually constitutes the water reservoir for the Thule base and has a high impact from human activities. Lake Catarina (V3) is a very small shallow lake (maximal depth 2 m) which probably gets bottom frozen during winter. For the 4th lake, V4 (Lake Sofie) only water was collected.

From the lakes, water samples of 200 l were pumped into a plastic drum. Plutonium was preconcentrated to about 10 l by hydroxide precipitation using ²⁴²Pu as yield determinant. Since conclusions about plutonium can be taken by using the Pu/Cs ratio subsequently radiocaesium was preconcentrated from an equal volume either by precipitation with copperferrocyanate (Cu₂Fe(CN)₆) or sorbtion on a cotton wound cartridge filter impregnated with cupperferrocyanate (Roos *et al.*, 1994). As radiochemical yield determinant ¹³⁴Cs was used.

A plastic boat was transported to the lakes on the roof of a Toyota Landcruiser. From the boat sediments were collected with a kayak-sediment sampler connected to a tube with an inner diameter of 64 mm. Later during the expedition winter came unusually early, the lakes got frozen and the ice was used as a sampling platform. Sediment sampling was then carried out through drilled holes in the ice. The cores were sliced immediately in 1-cm sections and stored in plastic bags for further analysis at home in the laboratory.

At the lakes in the temperate region an identical procedure was carried out, boats as well as the ice were used as sampling platforms. This region is situated at 56.5 °N in a forested area and has an annual precipitation of about 650 mm. The lakes are frozen about 3 months per year.

At the laboratory Cs precipitations were dried and filters were incinerated and radiocaesium was measured by HPGe gamma spectrometry. It was also possible to measure ^{210}Pb in the sediments at the same time, which was used for the dating of the sediments. The actinide precipitations were dissolved in HCl and coprecipitated as iron hydroxides. The radioanalytical procedure for plutonium then followed the procedure of IAEA (1989). Plutonium was measured by alpha spectrometry using solid state Si-detectors.

Results and discussion

Arctic lakes

Measurements of plutonium concentrations in air at Thule showed slightly elevated concentrations during 1968 (EML, 1999). The concentration pattern did not follow the usual annual cycling with maxima during March-April when the mixing of troposphere and stratosphere takes place. The areas where Hansson (1980) found increased concentrations were not in the direction of the lakes.

For the assessment of an impact on the lakes from the accident radionuclide and isotopic ratios can be used. Hansson also estimated the areal deposition at Thule in 1974 at non-contaminated sites to 13 Bq m⁻² for $^{239+240}\text{Pu}$ and 777 Bq m⁻² for ^{137}Cs . These values are representative for this latitude with such a low precipitation. This gives a decay-corrected value to 1997 of 0.028 for the $^{239+240}\text{Pu}/^{137}\text{Cs}$ activity ratio. The $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio in Thule debris was 0.015 while it would be 0.028 in 1997 in nuclear test fallout including SNAP-9A (the reentry of a satellite, with a ^{238}Pu power source, over the Mozambique channel in April, 1964). Another tool would be to use the $^{240}\text{Pu}/^{239}\text{Pu}$ ratio where the atomic ratio in stratospheric injection is 0.179 and the activity ratio 1.35 (Perkins and Thomas, 1980) while these ratios are 0.055 and 0.23 respectively in Thule debris. This would however require mass spectrometric measurements. The vertical distributions for plutonium, radiocaesium and ^{210}Pb for 3 of the lakes, V1, V2, V3 (Lake Pia, Lena and Catarina) studied at Thule are displayed in Fig. 1-3 and the deposition data and sedimentation rates in Table. 1.

The sedimentation rates are between 0.4 and 1.6 mm per year using ^{210}Pb data and the theory of Constant Rate of Supply (CRS) model (Appleby and Oldfield, 1978). One cm then corresponds to between 6 and 25 years. It is therefore not possible to resolve the fallout peak in 1963 and an eventual contribution from the Thule accident in 1968.

The integrated area content of ^{137}Cs should be around 400 Bq m⁻² in 1997 which is in fair agreement only with lake Lena (V2). The other lakes show about 2-3 times higher area content than expected from integrated fallout data, which is also, the case for plutonium. This is not unexpected since the lakes act as traps for snowdrift and litter. Furthermore the sedimentary material leaves the slopes of the lakes to the deeper regions where the samples were collected. The activity ratios $^{239+240}\text{Pu}/^{137}\text{Cs}$ for lake V1, V2 and V3 are 0.027, 0.024 and 0.020 respectively which are slightly lower than in integrated fallout from nuclear tests. This indicates that there has been a slightly higher run off of

caesium than plutonium from the catchment area and that there is no detectable impact from the accident.

The activity ratios $^{238}\text{Pu}/^{239+240}\text{Pu}$ varied between 0.028-0.056 for the sediment cores. This is higher than anticipated from fallout. The levels of ^{238}Pu are very low with analytical difficulties but the data indicate again that there is no impact from the accident.

The annual flux of ^{210}Pb to the sediments can be calculated to 149 ± 9 , 98 ± 7 and $45 \pm 3 \text{ Bq m}^{-2} \text{ a}^{-1}$. This flux is dependent on the exhalation of ^{222}Rn , which in turn depends on the type and amount of surrounding landmass and if the ground is frozen. Polar regions should have a low flux such as less than $20 \text{ Bq m}^{-2} \text{ a}^{-1}$ (El Daoushy, 1988) for the Arctic and $9 \text{ Bq m}^{-2} \text{ a}^{-1}$ for the Antarctic (Roos *et al.*, 1994) compared to about $200 \text{ Bq m}^{-2} \text{ a}^{-1}$ for continental lands far from the coast.

The flux of ^{210}Pb in the area is very high compared to expected values. There is also no correlation between the fluxes of ^{210}Pb and inventories of plutonium and caesium. This is probably due that the sources are different and that lead has different transport mechanisms compared to plutonium and caesium to the lakes from the catchment area. ^{210}Pb can therefore not be used as a normalising factor.

The peaks from $^{239+240}\text{Pu}$ and ^{137}Cs coincide in time with each other. They also agree very well with the time horizons in the sediments using the ^{210}Pb dating method if 1963 is the expected time for maximum from nuclear test fallout.

The concentrations of $^{239+240}\text{Pu}$ in filtered water from the lakes are between 3-6 $\mu\text{Bq l}^{-1}$. This might be a representative value for such lakes. It is however not clear if this activity originates from run off, remobilization processes or precipitation.

Temperate lakes

The dysoligotrophic lake Svartsjön with high humic content is very shallow (1-2 m) and the sediments are mechanically disturbed by wave actions. The water concentrations show very interesting data as can be seen in Table 1. The levels 130 – 220 $\mu\text{Bq l}^{-1}$ are extremely high compared to most other natural waters contaminated from nuclear test fallout. Such levels can otherwise only be found close to nuclear fuel reprocessing plants. The areal deposition of $^{239+240}\text{Pu}$ in the area is about 40 Bq m^{-2} and the contribution from the Chernobyl accident was only 0.02 Bq m^{-2} (Holm, 1992). The plutonium is in a colloidal form and the major fraction passes a $1 \mu\text{m}$ or a GF/A filter. Ultrafiltration showed that 77% was between $1 \mu\text{m}$ and 100 kD and 7 % associated with particles larger than $1 \mu\text{m}$. The remainder, 16% is then associated with colloids smaller than 100 kD. Clearly the large humic colloids then to a large extent governs the behaviour of Pu in this lake. The origin of these colloids may either be directly from the nearby peatbog or indirectly via decomposition of the released peat in the lake sediments.

During the winter the lake is frozen. The ice was analysed showing that the ice contained no measurable concentrations of plutonium or radiocaesium. The freezing process gives a perfect cleaning of radionuclides, which are frozen out with the humic material not fitting into the ice crystal lattice.

Alberts *et al.* (1987) found an increase of actinide concentrations in the deepest waters of ponds during periods of intense anoxia as well as increased concentrations of plutonium in anoxic, meromictic lake. The theory that this was a consequence of the iron and manganese redox cycle, which occurred in the ponds annually, was not proven but other mechanisms might be involved. Roos *et al.* (1993) showed the remobilization of

plutonium and other actinides from sediments in a permanently anoxic fjord and that this was due to association with humic complexes.

For this particular lake (Svartsjön) the major source of plutonium is an adjacent large peat bog. The decomposition of the *Sphagnum* moss releases plutonium together with colloidal form, previously trapped during the nuclear test period. This plutonium becomes associated with humic colloids as mobile species. The low turnover of the water in the lake result in high concentrations of plutonium in the water column. A substantial fraction of the water is frozen during winter and this process increases concentrations in the water phase.

Lake Mien is an oligotrophic lake in the area, i.e. the impact from nuclear test fallout and Chernobyl is the same. This lake is much deeper (max 47 m) and is supposed to be the result of a meteor impact 150 million years ago.

A typical sediment profile for an oligotrophic lake (Mien) at 30 m depth for $^{239+240}\text{Pu}$, ^{137}Cs and ^{210}Pb in the lake is shown in Fig. 4. The integrated area contents of $^{239+240}\text{Pu}$ and ^{137}Cs in the profile are 21 and 975 Bq m⁻² respectively. This is much lower (a factor 2-3) than anticipated from nuclear test fallout and the Chernobyl contribution. The sedimentation rate is estimated to 1.1 mm per year and the annual flux of ^{210}Pb to the sediments is 124 Bq m⁻² a⁻¹. The flux of ^{210}Pb to the sediments is about the same as for the arctic lakes but should generally be higher for this area. This relatively low flux can partly be explained by that the lake has a small catchment area. It is more difficult to explain the “losses” of plutonium and radiocaesium. There must be an accumulation area in the lake. The water concentration of $^{239+240}\text{Pu}$ was $15.1 \pm 1.5 \mu\text{Bq l}^{-1}$ and the inventory in the water is no more than 2.5% of the total inventory.

We find an activity ratio of $^{239+240}\text{Pu}/^{137}\text{Cs}$ of about 0.021 and nuclear test fallout it is expected to be 0.027 and the lower value shows fallout of radiocaesium from the Chernobyl accident. The activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ in the entire sediment core was 0.023 ± 0.005 which is in agreement with expected from fallout and SNAP-9A. The distribution of ^{137}Cs is different than that of plutonium at around 1 cm depth corresponding to the Chernobyl accident in 1986. The fractional contribution of ^{137}Cs from the Chernobyl accident in the total sediment core can be estimated to 30%.

Conclusions

For the high Arctic lakes at Thule, sedimentation rates are such that the peak from nuclear test fallout, 1962-1963, can not be resolved from eventual contribution from Thule/Pitufikk accident in 1968. There is no detectable contamination of plutonium in the lakes following the accident using deposition data and Pu/Cs ratios. Plutonium and radiocaesium are effectively transferred to the sediments in these lakes, which are ice covered 10 months of the year. The integrated depositions of radiocaesium, plutonium and the annual flux of ^{210}Pb are much higher than anticipated from fallout data and typical fluxes for the area of ^{210}Pb . This is due to that the lakes act as traps for wind driven snow and other material. This is then collected at the deeper areas of the lakes where sedimentary material can be found.

Dysoligotrophic lakes in temperate regions show seasonal variations and high concentrations of plutonium in the water, 10-100 fold higher than other lakes. The high concentrations depend on that plutonium is associated with humic colloids released from the catchment area. This phenomenon plays a vital role for the mobility of plutonium and

other actinides in fresh water system from catchment areas into the lakes and further to rivers and to the sea. Annual variations depend on the seasonal run off, freezing and melting processes and possibly also on seasonal anoxia following the Fe/Mn redox cycle.

A deeper oligothropic lake with a small catchment area shows low area content in the sediments while sedimentation rates are similar to arctic lakes. The vertical distribution of radionuclides in such a lake as well as a more whole covering sedimentation study should be undertaken.

Acknowledgements

This study was made possible by grants from the Swedish Radiation Protection Institute (Temperate lake part) and the Nordic Nuclear Safety Research (NKS). We also wish to thank various people at the Thule Airbase (especially Jan Trysø) for their logistic support, which was essential for undertaking the sampling.

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Table 1. Sediment inventories, water concentrations of ^{137}Cs and $^{239+240}\text{Pu}$, sedimentation rates and annual flux of ^{210}Pb in arctic and temperate lakes.

Lake	Inventories		Water conc.	Sed. rate	Annual flux
	^{137}Cs	$^{239+240}\text{Pu}$	$^{239+240}\text{Pu}$		^{210}Pb
	Bq m^{-2}		$\mu\text{Bq l}^{-1}$	mm a^{-1}	$\text{Bq m}^{-2} \text{ a}^{-1}$
V1, Pia	1190	32.4	6.9	1.6	149
V2, Lena	363	8.7	3.3	0.4	98
V3, Catarina	1090	21.9	5.9	0.4	45
V4, Sofie	-----	-----	2.4	---	---
Mien	1045	20.5	15.1	1.1	117
Svartsjön	-----	35.2	130-220	4	48

Sediment profile from lake V1

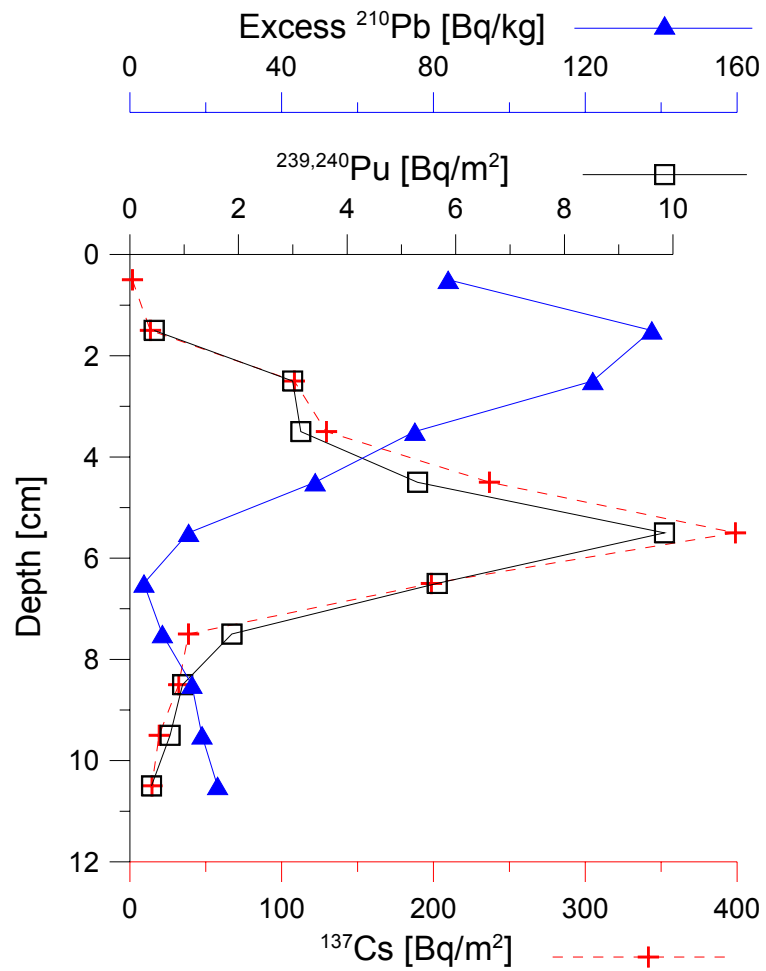


Fig. 1. Distribution of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{210}Pb in a sediment core from Lake V1 (Lake Pia), Thule, Greenland.

Sediment profile from lake V2

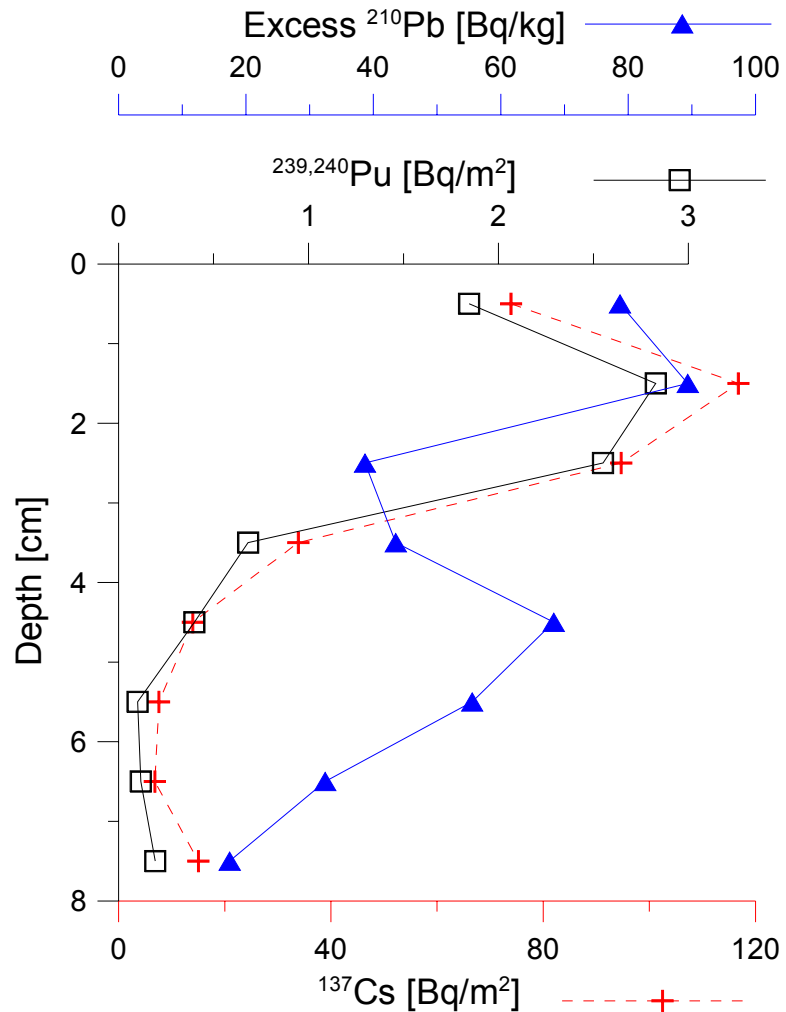


Fig. 2. Distribution of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{210}Pb in a sediment core from Lake V2 (Lake Lena), Thule, Greenland.

Sediment profile from lake V3

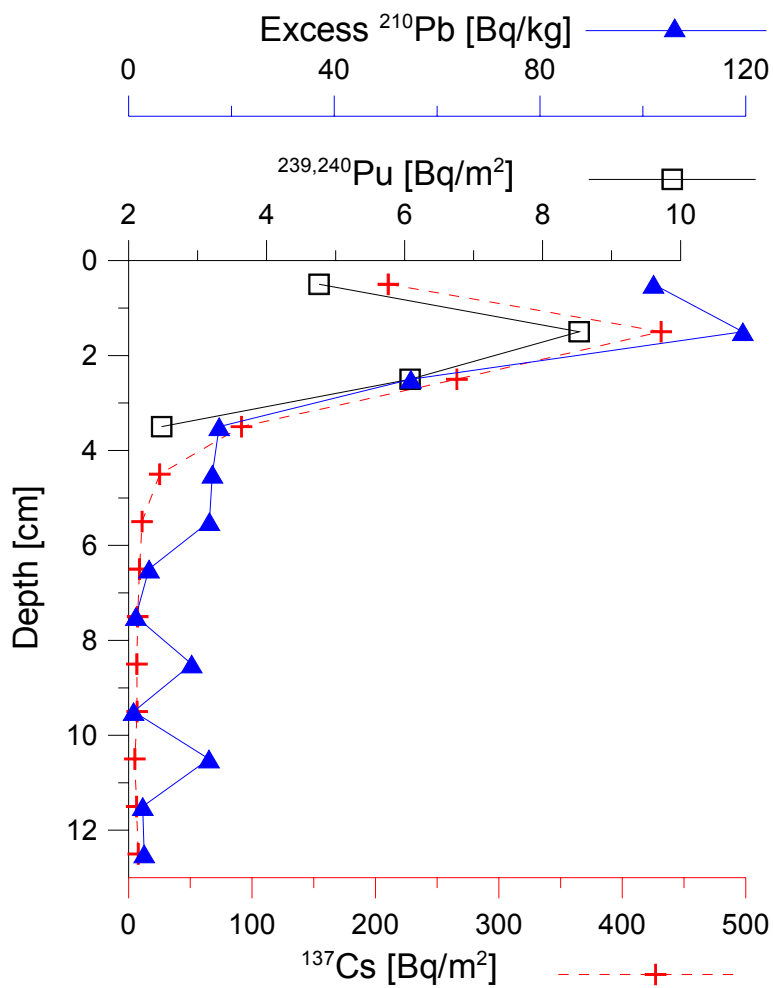


Fig. 3. Distribution of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{210}Pb in a sediment core from Lake V3 (Lake Catarina), Thule, Greenland

Sediment profile from lake Mien

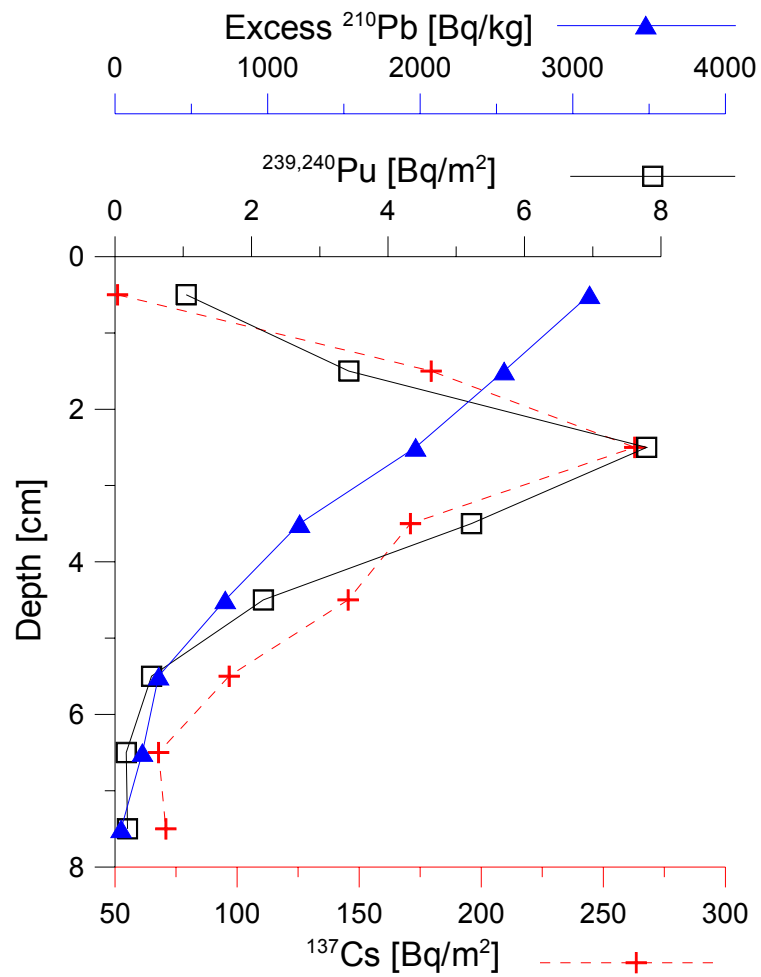


Fig. 4. Distribution of ^{137}Cs , $^{239+240}\text{Pu}$ and ^{210}Pb in a sediment core from Lake Mien, S. Sweden.

Field Sampling, Preparation Procedure and Plutonium Analyses of Large Freshwater Samples

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Introduction

This study is part of an investigation of the mobility of plutonium in freshwater systems containing humic substances. A well-defined bog-stream system located in the catchment area of a subalpine lake, Øvre Heimdalsvatn, Norway, has been studied. The area has been thoroughly investigated with respect to other radionuclides (i.e. ^{137}Cs and ^{90}Sr) in several studies after the Chernobyl accident (e.g. Brittain et al (1992), Brittain et al (1997), Hongve et al (1995)).

During the summer of 1999, six water samples were collected from the tributary stream Lektorbekken and the lake itself (Figure 1). However, plutonium concentrations were below the detection limit in all the samples. Therefore renewed sampling at the same sites, but using new and improved field techniques, was carried out in August 2000.



Figure 1. Map of sampling locations. Subalpine birch forest is indicated in green (dark grey).

Material and Methods

Fieldwork

A new sampling system was used to enable larger volumes to be sampled, and at the same time reducing the need for transporting large volumes back to the laboratory. Five large volume water samples were collected from Lektorbekken and Øvre Heimdalsvatn (Table 1).

Table 1. Samples collected from Øvre Heimdalsvatn and Lektorbekken, August 2000

Sample	Sample site	Volume
ØH – out	Øvre Heimdalsvatn, outlet	195 litres
ØH – in	Øvre Heimdalsvatn, inlet	178 litres
LeC	Lektorbekken, site C	174 litres
LeB – bog	Lektorbekken, site B, bog water	170 litres
LeB – stream	Lektorbekken, site B, stream water	160 litres

The water was lead through a filter (0.45 μm) and a flow meter into a large plastic container with the use of a water pump. The total volume of each sample was between 160 and 200 litres. One sample was collected of the bog water, which contained more humic substances than water from the lake or the stream. The bog water was therefore subjected to pre-filtration to prevent clogging of the 0.45 μm filter. The sampling set-up is shown in Figure 2.



Figure 2. Sampling set-up at Lektorbekken, site B

A ^{242}Pu -spike, concentrated HCl (to adjust acidity to pH 2), FeSO_4 and $\text{Na}_2\text{S}_2\text{O}_5$ were then added to the sample. The pH was controlled after a couple of hours and NaOH was added to adjust alkalinity to pH 10. The solution was thoroughly mixed after each step. The sample was left overnight. The next day the water was decanted and the precipitate transferred to a 10 l bucket. After further decanting, the total volume of the sample was reduced to one litre, which was a suitable sample size for easy transportation.

Laboratory analyses

The samples were brought to IFE's laboratory. Concentrated HCl was added to the samples to dissolve the precipitate. The samples were then analysed for $^{239,240}\text{Pu}$ using a controlled valence procedure, and counted by alpha spectrometry (Chen et al (1991), modified by IFE).

Results and Discussion

Table 2. Results of analyses

Sample	$^{239,240}\text{Pu}$ [$\mu\text{Bq/l}$]
ØH – out	-
ØH – in	3.3 \pm 0.7
LeC	2.1 \pm 0.5
LeB – bog	77 \pm 7
LeB – stream	2.2 \pm 0.5

Analyses of water samples collected in Heimdalen in 1991 showed that the concentration of $^{239,240}\text{Pu}$ was in the range 5-8 $\mu\text{Bq/l}$ (Brittain, unpublished data). The present concentrations are about 40 % of the levels found in 1991.

In 1996, water samples from different streams and small lakes were collected in Himdalen near Kjeller, 250 km south-east of Heimdalen and close to the then planned repository for low and intermediate radioactive waste in Norway. The purpose of the study was to establish background values for the concentration of different radionuclides in the area before operation of the repository. The results showed $^{239,240}\text{Pu}$ concentrations in stream water in the range 200-500 $\mu\text{Bq/l}$ (Varskog and Bergan, 1998), a factor 100 higher than found in Heimdalen. The concentration in the small lakes was a little lower, 90-200 $\mu\text{Bq/l}$. Both Himdalen and Heimdalen have received Pu from fallout, mostly from the atmospheric nuclear weapon tests in the 1950s and 60s.

Water samples have also been collected annually as a part of the environmental surveillance program for the nuclear activity of the Institute for Energy Technology at Kjeller. In 1999, the concentration of $^{239,240}\text{Pu}$ in water from the Nitelva River close to the institute was between 25 and 330 $\mu\text{Bq/l}$ (IFE-report IFE/I-2000/015). These results are also higher than what was found in Heimdalen.

Himdalen is mainly a boggy area; consequently the water contains large amounts of humic substances which, due to the presence of carboxyl, hydroxyl and phenolic groups, form strong complexes with plutonium. This increases the mobility of plutonium and might be one explanation for the higher concentrations of plutonium in Himdalen compared to what has been found in Heimdalen. This is supported by the considerably higher concentration in the bog water in Heimdalen compared to the streams.

A Swedish study (Eriksson 2001) within the NKS BOK-2 programme documented Pu-concentrations that were similar to our results. The humic rich lake Svartsjön showed $^{239,240}\text{Pu}$ -concentrations in the range 130-220 $\mu\text{Bq/l}$, whereas four oligotrophic arctic lakes that were studied showed $^{239,240}\text{Pu}$ -concentrations in the same range (2.4 - 6.9 $\mu\text{Bq/l}$) as found in Heimdalen.

The Heimdalen area is covered with snow for eight months of the year. Consequently, a substantial part of the fallout plutonium has been deposited directly on snow. During the spring flood, much of this plutonium has possibly been washed out with the melt water. This was observed for radiocaesium in Heimdalen in connection with Chernobyl fallout (Forseth

et al., 1993). In the lowlands, the ground is covered with snow for a period of only 4-6 months; consequently the plutonium will have had more time to adsorb to soil particles. Lowland soils will therefore, to a higher degree than in the mountains, be a continuing source of remobilised plutonium to lakes and rivers.

Conclusions

The results so far are in agreement with previous analyses from the Heimdalen area. However, up to 100 times higher concentrations were found in the lowlands in the eastern part of Norway. The reason for this is not fully understood, but may be caused by differences in the concentrations of humic substances, the fact that the mountain areas are covered with snow for a longer period of time every year and the flushing effect of the spring snowmelt.

Acknowledgements

The study was financially supported by the Institute for Energy Technology, the University of Oslo and Nordic Nuclear Safety Research.

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Plutonium in coniferous forests

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Introduction

Our aim was to study the uptake of plutonium by trees, undervegetation and some wild foods. The ratio of ^{238}Pu / $^{239,240}\text{Pu}$ in soil samples was determined for comparisons of the fallout origin.

Material and Methods

Soil, trees, dwarfs, berries and mushroom samples were collected at a pine-dominated site in Central Finland (Fig. 1, site 1). Litter and humus, and 0-2 cm mineral soil layers were analysed for ^{238}Pu and $^{239,240}\text{Pu}$ in five profile samples (Taipale, Tuomainen 1985). The layers 2-5 cm and 5-10 cm were studied in two of these core samples (Fig. 2). A 20-year old Norway spruce (*Picea abies*) was sectioned by age of needles and branches. The stem was divided in three parts. Wood and bark of the two uppermost stem parts were studied. Needles from the two uppermost quarters of five Scots pine canopies (*Pinus sylvestris*) were analysed. Mixtures of meat from both adults and calves of moose (*Alces alces*) from site 2 and from locations which received dry deposition in spring 1986 were analysed (Fig. 1). Dwarfs and berries of *Vaccinium vitis-idea* and berries of *Vaccinium myrtillus*, as well as mushrooms *Cantharellus cibarius* and *Cantharellus tubaeformis* were examined.

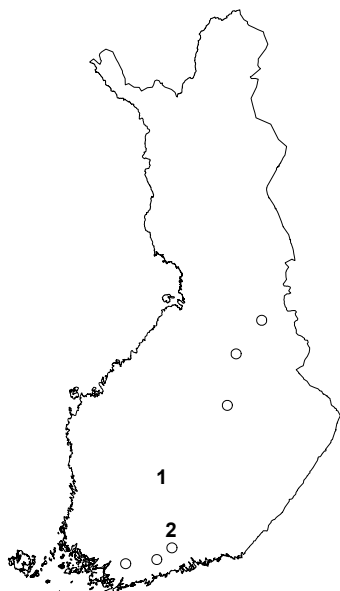


Figure 1. Sampling sites 1 and 2 representing wet deposition in 1986 and dry deposition locations for moose sampling.

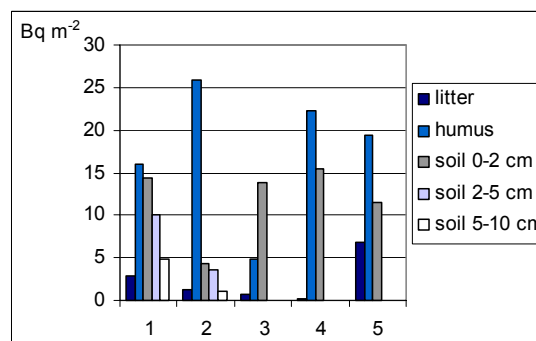


Figure 2. $^{239,240}\text{Pu}$ (Bq m^{-2}) in five soil profiles from site 1.

Results and Discussion

Forest floor. In litter, humus and mineral soil layer 0-2 cm the total $^{239,240}\text{Pu}$ activities per surface area were almost equal (Fig. 2). In deeper mineral soil layers additional plutonium was found, and a total of about 40 Bq m^{-2} was estimated for the site no 1. The maximum activity ratio $^{238}\text{Pu} / ^{239,240}\text{Pu}$ in litter, 0.20, was closest to the values found for plutonium in the Chernobyl fallout, 0.47 suggested by Holm et al (1992), and 0.55 by Paatero and Jaakkola (1998), see Fig. 3. In humus almost the same ratio was occasionally measured, whereas in the mineral soil the ratio was equal to 0.03, estimated for nuclear weapons fallout by Holm et al. (1992).

Trees. $^{239,240}\text{Pu}$ was found in the bark of the middle part of the stem, $18 \text{ mBq kg}^{-1} \text{ dw}$ and in the sample of dead branches and needles, $27 \text{ mBq kg}^{-1} \text{ dw}$. Detection limit for other tree fractions varied between 3 and $20 \text{ mBq kg}^{-1} \text{ dw}$.

Needles. $^{239,240}\text{Pu}$ was not detected, the range of the detection limits was 2 - $5 \text{ mBq kg}^{-1} \text{ dw}$. Neither did $^{239,240}\text{Pu}$ activities in stem wood or bark exceed the detection limit $2 \text{ mBq kg}^{-1} \text{ dw}$.

Wild foods and dwarfs. In dwarfs the detection limits of the activity concentration of $^{239,240}\text{Pu}$ ranged from 10 to $20 \text{ mBq kg}^{-1} \text{ dw}$. Plutonium was not found in wild food samples. The detection limit of $^{239,240}\text{Pu}$ ranged between 0.7- $3 \text{ mBq kg}^{-1} \text{ fw}$. The transfer of Pu from soil to wild foods was of the order of $10^{-5} (\text{m}^2 \text{ kg}^{-1} \text{ fw})$ or less, comparable with the findings by Suomela et al. (1999). The ingestion dose estimate for adults through average consumption (Markkula, Rantavaara, 1996) of wild foods was less than 4 nSv a^{-1} . Per unit deposition this upper limit corresponds to $0.1 \text{ nSv a}^{-1} \text{ per Bq m}^{-2}$.

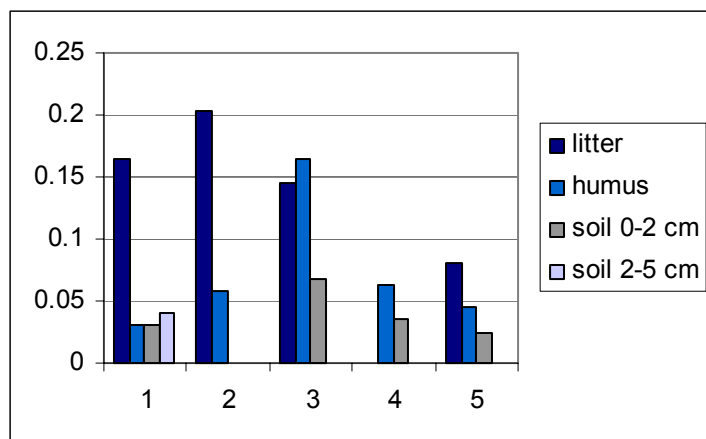


Figure 3. Activity ratio $^{238}\text{Pu} / ^{239,240}\text{Pu}$ in soil samples from the site 1.

Conclusions

In twelve years the Chernobyl derived plutonium had not reached the mineral soil. This refers to a very slow downward migration in podsollic soil. The study confirmed also the low Pu uptake by vegetation and an insignificant contribution to human doses through wild foods.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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BOK- 2.1.2 Assessment of internal doses

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1. Introduction

The assessment of internal doses is based on direct measurements of people, on indirect measurements of excreta or on measurements of activity in food (dietary studies) and, for eg. Iodine, air concentration. These measurements are mostly used to estimate the intake. The internal dose is then calculated using suitable metabolic and dosimetric models. Quality assurance in whole-body measurement and the associated calculation of internal doses is a complex matter. The measuring process is complicated and the uncertainty in the measurement geometry between the radiation source and the detector is unavoidable when measuring on humans.

In the European Union, the "Basic Safety Standards" [1] and the directives are followed. In what are known as the "Outside Workers" directive, it is stated that the internal dose measurement shall be included in the Union's supervision system for radiation. There are several bilateral agreements on Nordic assistance in emergency situations. It is therefore essential that all Nordic countries have intercalibrated their measurement systems. In order to be able to carry out calibration and intercomparison exercises, a so-called phantom simulating the human body is required.

2. BOK-2.1.2.1 Exercises on internal dose calculations

An intercomparison on internal dose calculation within the European countries was conducted, showing a need for harmonisation of dose calculation procedures.[2, 3] As only a couple of laboratories from each European country were allowed to participate a course in internal dose calculation was arranged with the intention to train experts in internal dose calculation. The course was held at STUK, Finland 13 –15 October 1999 where 29 persons from all Nordic countries attended the course. The content of the course is seen in the agenda below

Agenda for the Workshop		
Wednesday, October 13		
Before lunch	Arrival and information of NKS projects .	
After lunch	Lectures on Internal dosimetry, Introduction, Basic definitions, Biokinetic and dosimetric models and software MIRDOSÉ	
Thursday, October 14		
Before lunch	Lectures on Respiratory tract model and demonstration of software LUDEP, Exercise with LUDEP and MIRDOSÉ	
After lunch	Lectures on Bioassay and LUDEP, Exercise with LUDEP and MIRDOSÉ, Calibration exercises	
Friday, October 15		
Before lunch	Lecture on the use of simple compartment models for dose calculations. Exercise with LUDEP and MIRDOSÉ, Calibration exercises	
After lunch	Exercise with LUDEP and MIRDOSÉ, Calibration exercises	
The lectures were held by		
Lennart Johansson Institututinen för Radiofysik, Umeå	Alan Birchall NRPB, UK	Sigurður Emil Pálsson Geislavarnir Ríkisins IS

The course was successful and a second similar course was held at STUK, Finland 7 – 8 March 2002 where 21 persons from Nordic countries and Estonia, Latvia and Lithuania attended the course. This second course contained also information on an "Integrated Modules for Bioassay Analysis (IMBA)"

3. BOK-2.1.2.2 Calibration and intercomparison of equipment for direct measurements of radioactivity in the body and iodine in thyroid.

Quality assurance includes quality control with; inter alia, procedure and protocol descriptions, detector calibrations, instrument control, documentation of measurement results and evaluation internally or by outside persons. This evaluation also involves regularly repeated comparative measurements.

Earlier such exercises within the NKS framework are:

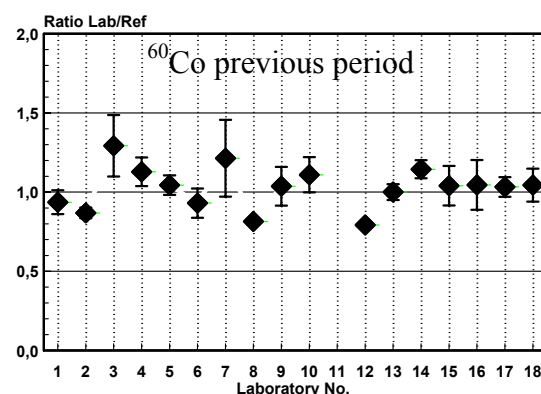
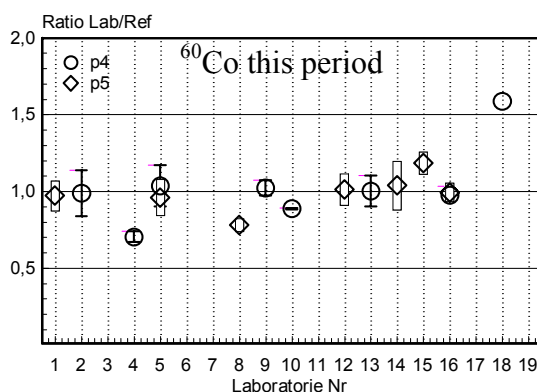
- NKA-1984-85. A phantom from Finland with an activity content unknown to the participants was circulated between whole-body measurement laboratories in the Nordic countries. The conclusion: Intercalibration was required.
- NKS-1990-93. An "ice-pack phantom" from Finland containing known quantities of ^{137}Cs , ^{134}Cs and ^{60}Co was circulated between the whole-body measurement laboratories in the Nordic countries. 20 laboratories participated and the results were published together with the corresponding results from the NKA- 1984- 85 project with the title "Intercalibration of whole-body counting systems".

- NKS 1996-97. A whole-body phantom consisting of inactive plastic modules, which can be fitted with rods containing radioactive substances, was used for the intercomparison exercise. The phantom is of the same type as that used in the concluded EU project "Intercomparison of *in-vivo* monitoring systems" [4]. Seventeen Nordic laboratories participated and they reported their results. [5].

The whole-body phantom used for this intercomparison exercise is the same as used during the intercomparison exercise 1996-97. The phantom was circulated among the participants together with 4 different sets of radionuclides. The activity content was unknown by the participants. Also a neck phantom with a source of ^{133}Ba was circulated at the same time to provide a possibility to calibrate for measurements of ^{131}I in thyroid. The ^{133}Ba source was used instead of a ^{131}I source, as we wanted all participants to use same source. 19 laboratories have participated and 15 have submitted results for the intercomparison shown in the following graphs.

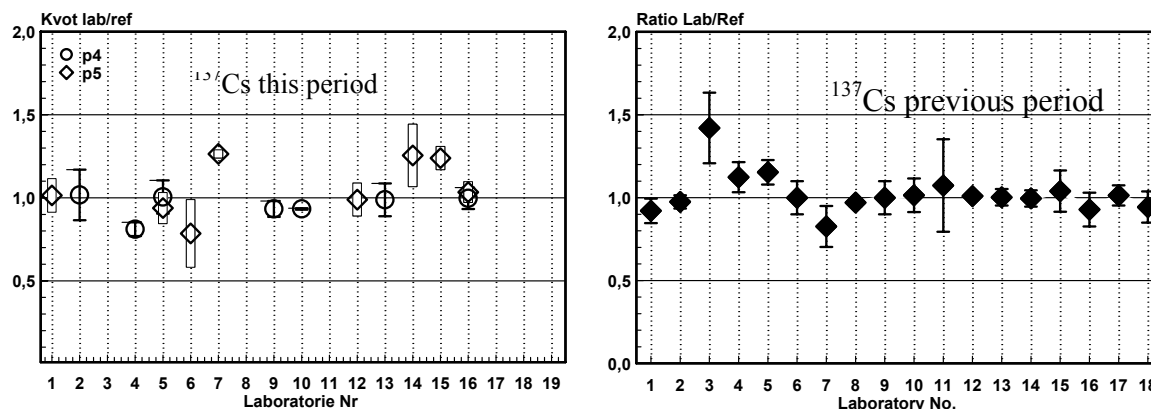


The phantom used is built of tissue equivalent blocks for easy size and position configurations. In each block can be loaded with two tiny rods with activity. Picture to right show "chair-geometry" with simultaneous thyroid measurement



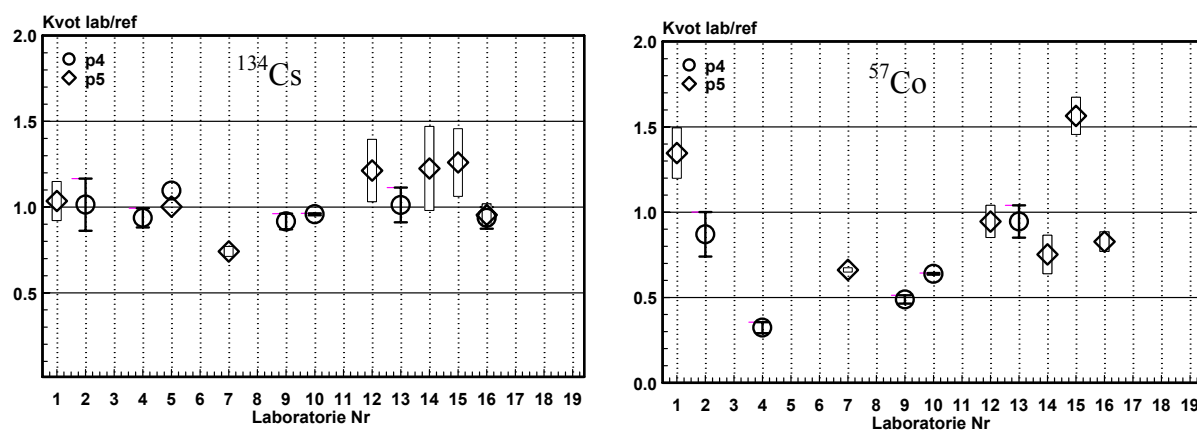
Two different sizes of the phantom (P4 and P5) corresponding to 61.5 kg and 77.8 kg bodyweight could be assembled and used for the intercomparison measurement. The error-bars are the overall 1 SD uncertainty given by the laboratory. The reported ratio ^{60}Co -

activity/reference activity is shown above. Current result to the left and previous (1996-97) results to the right.



The reported ratio ^{137}Cs -activity/reference activity is shown above. Current result to the left and previous (1996-97) results to the right.

As can be seen in the graphs most of the reported values are within a factor of two, but they also show that regularly intercomparison exercises are necessary as the previous (1996-97) results demonstrated less divergence.



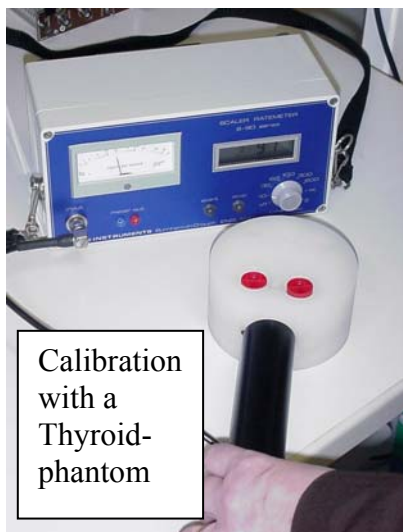
The results for the less common nuclides ^{134}Cs and ^{57}Co show a larger divergence

4. BOK-2.1.2.3 Rapid monitoring of people and preparation of handbook for emergency situations

Experiences from accidents where many people might be internally contaminated show a need for rapid monitoring for screening purposes but also for later estimates of dose.

Calibration of hand-held instruments for measurement ^{131}I in thyroid and ^{137}Cs body-burden was arranged in conjunction with the workshop on internal dose calculation, STUK, Oct. 1999.

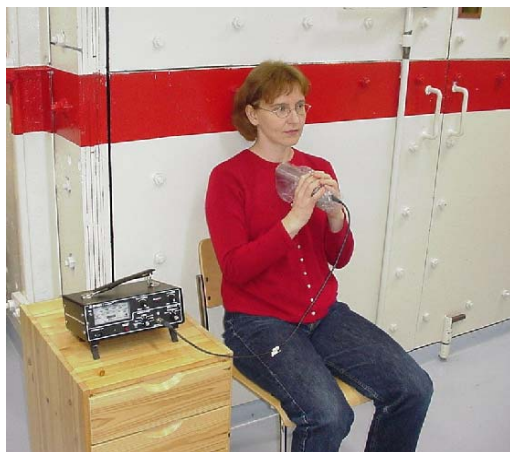
18 participants participated and the results from the exercise clearly showed that with a proper calibration many hand-held common gamma instruments are usable for in-vivo measurements in an emergency situation.



The participants were:

Mats Hjelm
Lennart Johansson
Björn Sandström
Monica Eklöf
Felix Kuffner
Heidi Anderson
Sigurdur Emil Palsson
Michael Tillander
Sauli Pusa
Eero Illukka

Annette Löfvefors Daun
Göran Ågren
Rolf Falk
Kjell Lindberg
Peter Hofvander
Aud Raaum
Alan Birchall
Tua Rahola
Marketta Puhakainen
Matti Suomela



For instruments with NaI-detectors a few kBq of ^{131}I in thyroid can be measured, corresponding to a dose less than 1 mSv. For ^{137}Cs , body-burden of 20-50 kBq can be measured, corresponding to a dose of about 1 mSv. Instruments with GM-tubes are about 5 times less sensitive.

Details for different instruments and measuring procedures can be found in "Handbook for rapid measurements of internal contamination of people in emergency situations [6].

The Contents of the Handbook is:

- Introduction
- Measurements with handheld instruments
- Measurement of ^{131}I in thyroid with handheld instrument
- Measurement of ^{137}Cs in the body with handheld instruments
- Measurements of ^{131}I and ^{137}Cs with gamma-camera and iodine-monitor
- Measurements with gamma-camera
- Measurements with iodine-monitor
- References

The handbook will be written in Swedish with strong emphasis on Nordic emergency preparedness methods. Possible translations to other Nordic languages will be considered later.

5. BOK-2.1.2.4 Comparison of methods of estimating internal doses to Nordic populations- whole- body measurements and dietary investigations

The work in BOK - 2.1.2.4 aims at a better understanding of the connection between the important Nordic foodchains and the internal doses originating in these food-chains. The reasons for differences in internal dose estimates depending on the methods used - whole-body counting or dietary surveys- will be investigated. The discrepancies were demonstrated in the earlier project (1990 - 1993) on "Internal doses to the Nordic populations". The reason for discrepancies still is an open question also internationally. In this project existing material as well as results from measurements within the frame of this project in Finland, Norway and Sweden will be used. People consuming much produce from nature will be whole-body counted and their diets surveyed. This investigation will be done on individual base as opposed to the earlier one in which statistical data on diets were used. The statistical data do not give a good enough intake estimation. The internal dose calculated using statistically obtained dietary data seem to give an overestimation of dose, which is a conservative approach.

Results of fieldwork in Norway were reported earlier by Lavrans Skuterud [9] A short summary is given below.

In Finland, Jenni Murto did a master's degree work under supervision of Tua Rahola [10]. This work giving the results of the dietary investigations in Padasjoki was written in Finnish and is not included here.

Recent assessments of ^{137}Cs intake by populations in Norway

Two separate assessments of ^{137}Cs ingestions doses to population in Norway has been carried out as part of the BOK-2.1.2 project, one among average rural populations in Southern Norway, the other among reindeer keepers in Northern Norway.

Average rural population

The area selected in Gudbrandsdalen and Valdres (the municipalities Sel, Øystre Slidre, Vestre Slidre and Vang in the county of Oppland) is situated in central Southern Norway, an area that was heavily affected by Chernobyl radiocaesium fallout. Totally 416 persons were interviewed about their dietary habits. The questionnaire was based on frequency and amount of consumption ("quantitative food frequency questionnaire"), and had separate sections

focussing on individual and household intake, respectively. Of the 416 persons, 266 randomly chosen persons were whole body monitored in September and October. Information on radiocaesium activity concentrations in totally 753 food samples was obtained from the local food control authorities, dairies and the national “food basket” programme, in addition to samples received from the interviewees.

The results of the dietary habits interviews and the activity concentrations in food samples were combined to give estimates of the daily radio-caesium intake. These estimates were compared to the results of the estimates obtained from the whole body monitoring results, using the ICRP-56 retention function for caesium in the human body (and assuming that 100 % of the ^{137}Cs taken in with food is absorbed in the GI-tract). From the whole body counting results female and male intake rates were estimated to 3.4 and 4.6 Bq/day respectively, about 1/4th the estimates based on diet and contamination of food products, 19 and 14 Bq/day.

Possible reasons for the discrepancy in intake estimates were discussed (i.e., underestimates based on the whole body counting, overestimation of food consumption, and possible problems with food sampling). Despite a number of ‘expert judgements’ and assumptions about how representative the food samples were, and about losses of activity in foodstuffs during preparation, the lowest estimated intake based on the dietary survey and the food samples still gives twice as high intake as that indicated by the whole body counting results. The later analysis for Kautokeino suggests that the discrepancy is reduced some 20 % if the caesium retention model proposed by Melo et al. [11] is applied instead of ICRP’s model.

Reindeer keepers

Reindeer keepers in Kautokeino (in Finnmark county) have been whole body monitored annually in the period 1965-1990, thereafter with a 3-year cycle. In most years, the reindeer keepers have provided samples of reindeer meat for radiocaesium analysis. In 1989-1990 and 1999 dietary surveys were performed in conjunction with the whole-body monitoring. Earlier diet information is available from a separate study in 1963. The aim of this assessment was to review and evaluate the available information from Kautokeino, and to derive some conclusions regarding the reindeer meat consumption by today’s reindeer keepers.

The dietary survey results and analysis of reindeer meat samples was used to estimate dietary radiocaesium intake, and from this average whole body activity concentrations were estimated using the ICRP-56 model. These estimated whole body activity concentrations were compared with the results of the whole body measurements. The ratio between estimated and observed concentrations ranged from about 1 in 1999, to nearly 7 in 1990. The seemingly nice agreement for 1999 is assumed to be an underestimate, since the intake estimate did not take into account any loss of radiocaesium during food processing and preparation.

Similar estimates using the revised caesium retention model proposed by Melo et al. (1997) give less discrepancy between the estimated and observed whole body activity concentrations (values corresponding to the above mentioned ratios for 1999 and 1990 were 0.6 and nearly 4).

As in the case for the rural population, some assessments of losses and how representative the reindeer meat samples are have been carried out. Accepting that the meat consumed on average has lost 50 % of its radio-caesium activity due to preparation (boiling, and discarding broth), and that only applying results of winter slaughtered reindeer in the intake estimates gives a 30 % overestimate, it may be concluded that a ratio around 2.6 between estimated and observed ^{137}Cs whole body activity concentrations is acceptable. Using a ratio of 2.6 suggests that the reindeer meat consumption in 1999 was about 240 and 310 g d⁻¹ for females and males

respectively. This is in agreement with an average consumption of 192 g d^{-1} (the result for 1999 based on household procurement) if the population consume twice as much reindeer meat during November-April as during May-October, and assuming that male meat consumption is 30 % higher than female.

The results of the dietary studies among reindeer keepers in Kautokeino suggest that a questionnaire based on household procurement works best, but that some additional information is needed on differences between female and male consumption habits.

Internal doses from ^{137}Cs calculated from whole-body counting results and from dietary studies

Whole-body counting and dietary studies

In Finland the group chosen for measurement and individual diet investigation was one of the groups that have been followed since the Chernobyl accident. The persons in the group live at Padasjoki in the middle of Finland where the deposition of Cs-137 was highest. They were selected according to consumption habits. All of them using local produce from nature such as fish from small lakes and wild berries from the forest. The results show body burdens much higher than the mean Cs-137 burden in Finland. Since the intake of caesium is mainly from produce from nature it is not necessary to register all foodstuffs of the diet at each measurement occasion. For this purpose a special questionnaire was prepared including only fish, berries, mushrooms and game. This questionnaire was sent to each participant in advance to remind them of what they had been eating during the two weeks prior to the test. They delivered the questionnaire when coming for whole-body measurement and were further interviewed at the same occasion.

The questionnaires used in Finland, Norway and Sweden after the Chernobyl accident were also collected and compared. All of them included the foodstuffs of importance for this project. So also earlier obtained results could also be used.

At Padasjoki whole-body counting measurements were performed twice in 1998, 1999 and once in 2000. Each year in June a dietary expert interviewed the participants. The results of the interview and those obtained by the questionnaire sent out in advance agree well. The results of whole-body counting measurements of the Padasjoki group from 1987 to 2000 are shown in Fig. 1. The mean body burden for men and women of the Padasjoki group are more than ten times higher than the mean body burden of the Finnish population.

Internal doses

The internal radiation dose from ^{137}Cs was estimated at 0.58 mSv for men and 0.28 mSv for women during the period summer 1998 to summer 2001 using the whole-body counting method(method 1). This gives an mean annual dose of 0,19 for men and 0,09 mSv for women. The corresponding results using the intake method (method 2) were 0,77 and 0,49 mSv or 0,26 and 0,16 mSv annually. Figure 2 shows the correlation between doses calculated with the two different methods. The correlation factor was found to be 0.92.

The doses estimated using the intake method were higher by 1.3.for men and 1.7 for women than those estimated with the whole-body counting method. In an earlier investigation in

Finland the factor was higher than 2. (Suomela et al. 1991). Also in studies made in Norway the intake method gave higher values the factors varying from 2 to 6 (Strand et al. 1992, Bøe et al. 1991).

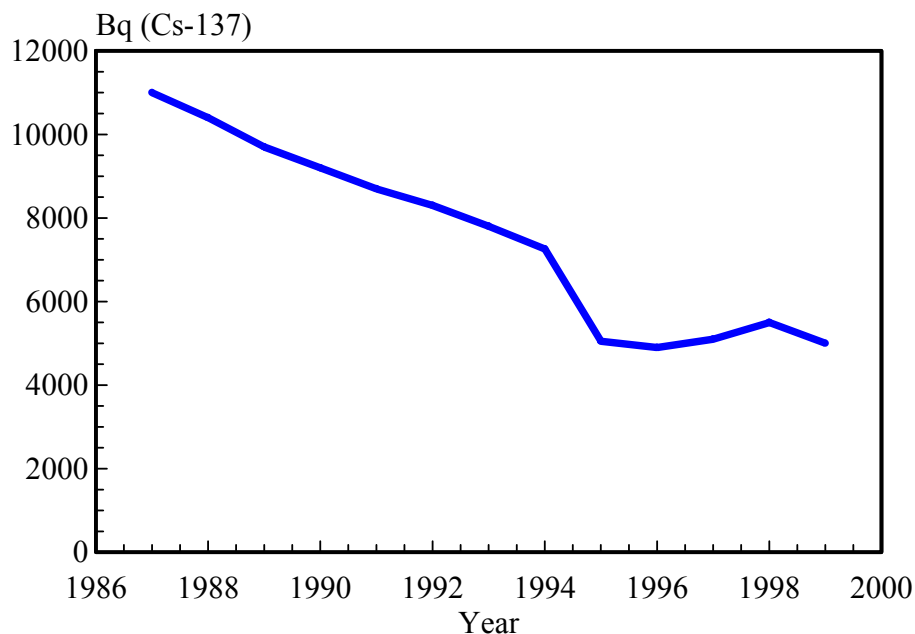


Fig 1. The mean ¹³⁷Cs body burden of the Padasjoki group

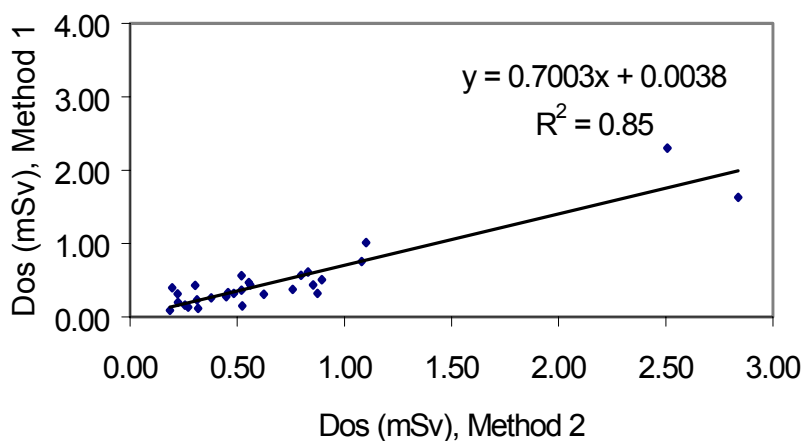


Fig 2. Correlation of doses estimated using the whole-body counting method(1) and the intake method(2). The doses are total doses for the period 1998-2001.

6. BOK-2.1.2.5 Activity losses during food preparation

One explanation to the discrepancies between dose estimation from whole body measurements compared with those made from dietary surveys can be a result of the uncertainties in the amount of activity in the diet after food processing. Correction factors for losses in food preparation have been presented earlier [7, 8]. For meat the correction factors given as retention factors (defined as the total amount of activity in the processed food divided by the total amount of activity in the original food) normally are around 0.5. For fish and berries around 0.7 and for mushrooms around 0.3. As an attempt to estimate the retention factors after food preparation during normal food preparation of local produce from nature a study of the losses of activity under those conditions have been started. The results up to now show that for cooking of meat the losses are rather large with a retention factor of about 0.4. For frying of meat the losses are less, with retention factors around 0.8 -1.0 dependent on the size of the pieces of meat. So far the number of prepared meals are rather low, but the study will continue decreasing the uncertainties.

7. Conclusions

There is a definite need for training in dose calculation. Our first course was successful and was followed by a second, both courses were fully booked. An example of new tools for software products for bioassay analysis and internal dose assessment is the “Integrated Modules for Bioassay Analysis (IMBA) were demonstrated at the second course. This suite of quality assured code modules have been adopted in the UK as the standard for regulatory assessment purposes.

The intercomparison measurements are an important part of the Quality Assurance work. In what is known as the "Outside workers ' directive it is stated that the internal dose measurements shall be included in the European Union's supervision system for radiation protection.

The emergency preparedness regarding internal contamination was much improved by the training with and calibration of handheld instruments from participants' laboratories. More improvement will be gained with the handbook giving practical instructions on what to do in case of emergency.

The comparison of results of different dose calculation methods show that on an individual basis both methods can be used. Using statistical data only for intake calculations will overestimate the internal dose. This has to be kept in mind when using such estimations in emergency situations. The results obtained in this project will also be published in the open literature.

8. Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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NKS-BOK-2.1.2

Mätning av radioaktiva ämnen i människa i beredskapssituationer.

En metodhandbok vid jod och cesium-kontaminering

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1 Inledning och syfte

Uppskattningar av interna stråldoser baseras på direkta mätningar av människor eller också indirekt på mätningar av utsöndringsprodukter eller födoämnen. För jod kan uppskattningarna även bygga på mätningar av luftkoncentration. Mätningarna utförs oftast i syfte att bestämma (uppskatta) intaget av radioaktiva ämnen och den interna stråldosen beräknas sedan utifrån metaboliska och dosimetriska modeller.

Erfarenheterna från olyckor där många människor kontaminerats internt av radioaktiva ämnen har visat på ett behov av att snabbt kunna kontrollmäta många människor. Både för kontroll av kontaminationssituationen men också för att i efterhand kunna uppskatta stråldoserna från kontaminationen.

Med en adekvat kalibrering, kan många handinstrument användas för mätningar av intern kontamination vid en olyckshändelse som resulterar i att radioaktiva ämnen sprids till omgivningen. Instrument med NaI-detektorer kan registrera ett fåtal kBq ^{131}I i sköldkörteln, vilket motsvarar en effektiv dos på under 1 mSv. För ^{137}Cs , kan helkroppsinnehåll på 20-50 kBq registreras (ger <1 mSv). Instrument med GM-rör kräver ca. 5 gånger högre aktivitet.

Beroende på situationen kommer mätningarna att tjäna olika syften. Vid situationer då internkontaminationen kan tänkas vara hög blir det mätningar för att klarlägga om det behövs medicinsk behandling eller om det krävs andra mätningar för noggrannare dosimetri.

Mätningarna är också viktiga för kommande dosuppföljning. Vid de tillfällen när exponeringen kan vara utsträckt i tiden är det lämpligt att genomföra upprepade mätningar.

För jod kan mätningarna upprepas med en veckas mellanrum och för cesium 4 ggr per år. På grund av den relativt korta halveringstiden för jod-131 är det viktigt att mätningarna startar så snart som möjligt.

Handboken innehåller instruktioner och beskrivning av procedurer för dessa situationer, men behöver kompletteras med detaljerade instrumentspecifika instruktioner för de aktuella instrument som avses att användas. I första hand har situationer med jod eller cesiumkontaminering varit i åtanke när handboken skrivits. Handboken kan ses som en del i en beredskapsplan och anpassas till lokala förhållanden.

2 Mätningar med handinstrument

2.1 Thyroideamätning (sköldkörtel) av ^{131}I med handinstrument

2.1.1 Förberedande skede

- Välj lämplig lokal för mätningarna.
 - Lokalen bör helst vara belägen i ett område som inte drabbats av nedfall. Om detta inte är möjligt väljs en lokal i en byggnad med hög skärningsfaktor, t.ex. källare i stenhus.
 - Lokalen bör vara så stor att väntrum och mätrum kan iordningsställas. Tvättmöjligheter (dusch) är önskvärt.
- Arrangera lokalen för mätning och omhändertagande av personer som ska mätas.
 - Ställ i ordning väntrum med skogrens och utrymme för ytterkläder.
 - Avgränsa ett utrymme för mätning. Hotet är att utrymmet blir kontaminerat. Personer som kommer för mätning lämnar skor och ytterkläder redan innan de kommer till det avgränsade området.
 - Arrangera för inlämning av kontaminerade kläder och skor.
- Granska tvättmöjligheter (om sådana finns).
 - Kontrollera i så fall att det finns tillräckligt med handdukar, mätdräkter, tvål, schampo etc. Ordna service för tvätt.
- Förbered mätinstrumenten.
 - Kontrollera apparaturen. Skydda detektorn med en plastpåse. Placera, om möjligt, ut "limmattor" på strategiska ställen.Se till att mätinstruktioner för instrumenten finns tillgängliga.

2.1.2 Aktionsskede

Minns att det är viktigt att behandla dem som kommer till mätning lugnt och vänligt. De är sannolikt mera bekymrade än du. Visa inte din egen oro, prata inte för mycket, koncentrera dig på vad du gör och förklara gärna varför.

- Kontrollera personernas eventuella ytkontamination redan före skogrens. Var lugn och vänlig. Minns kontaminationsrisken, undvik kontamination på golv, sittplatser, instrument. Använd ditt sunda förnuft
- Personerna klär av sig ytterkläderna och tar av sig skorna vid ytterdörren. Tvättar händerna om det är möjligt. Överväg i annat fall att förse personerna med engångshandskar. Skydda stolarna med plast eller annat material som går lätt att byta.

2.1.3 Mätning:

- Koppla på apparaten.
- Välj mättid (100 s eller motsvarande för det instrument som används).
- Skriv in personens namn och andra uppgifter i mätblanketten (Bilaga 1)
- Skydda detektorn med en plastpåse som byts efter varje mätning.
- Håll detektorn vid halsgropen på den person som ska mätas, se bild. (För cesium se separat instruktion nedan)
- Starta mätningen.
- Notera resultatet och fyll i mätblanketten.

- Håll (tryck lätt) detektorn mot personens lår och gör en bakgrundsmätning under samma tid som personmätningen, se bild.
- Notera resultatet och fyll i mätblanketten.
- Kontrollera att blanketten är korrekt ifylld.

Kom ihåg lagar och förordningar angående registrering av personuppgifter. Inga uppgifter ges åt utomstående utan medgivande av personen det gäller. Minns din tystnadsplikt. Inga utomstående i mätutrymmena. Mediarepresentanter får närvara endast med särskilt tillstånd.



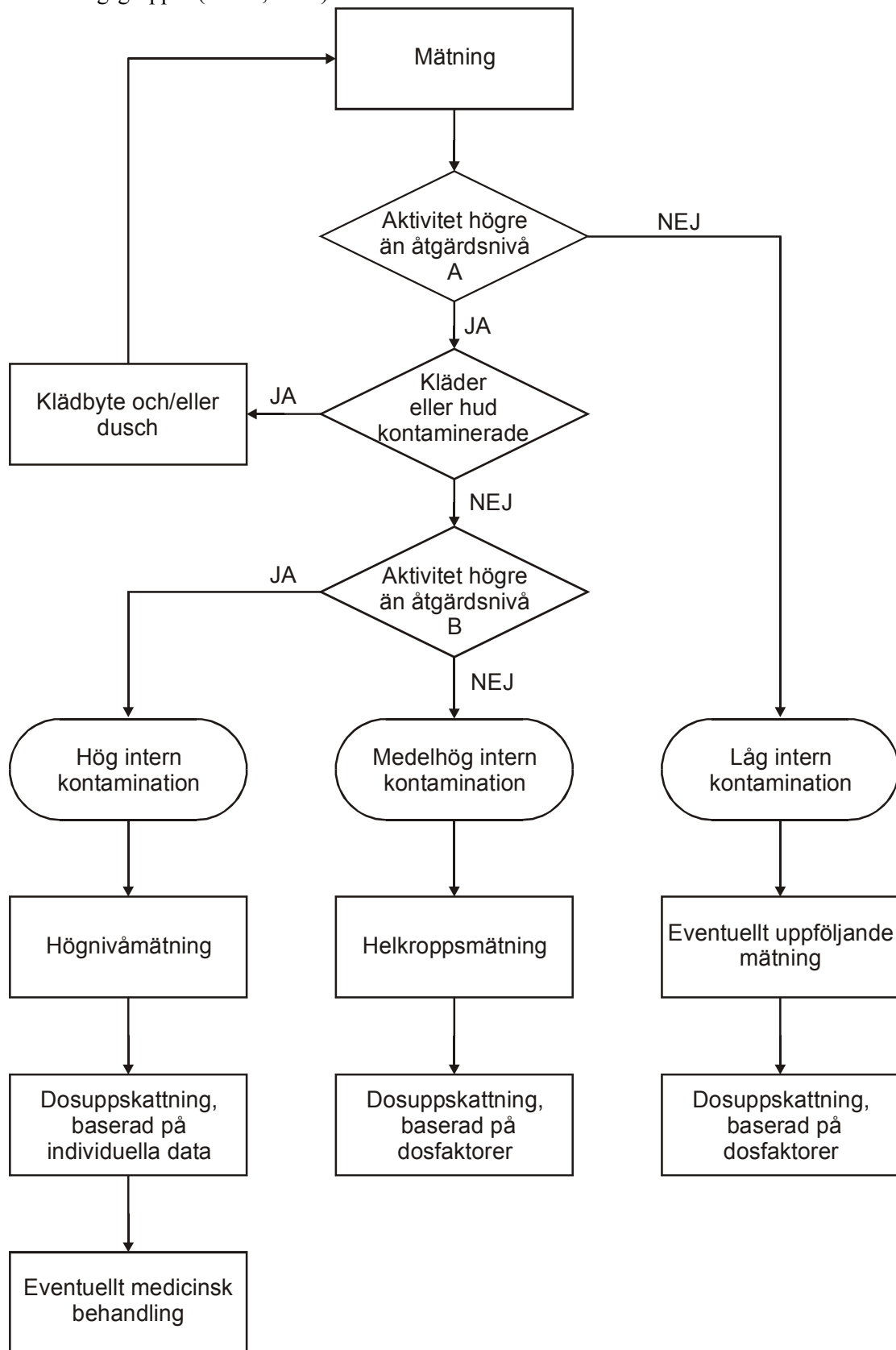
Mätning av jod-131 i sköldkörteln



Bakgrundsmätning till mätning av jod-131 i sköldkörteln

2.1.4 Beslut om åtgärder

Figur 1 nedan anger IAEA:s rekommendation för åtgärder i samband med kontrollmätningar av stora befolkningsgrupper (IAEA, 1994).



Figur 1 Rekommenderade åtgärder vid mätning.

Enligt Figur 1 bestäms två åtgärdsnivåer, A och B, sådana att dessa aktiviteter av ^{131}I i sköldkörteln ger en viss stråldos. Åtgärdsnivåerna och motsvarande aktiviteter för jod eller eventuellt andra radionuklider meddelas vid den uppkomna situationen av landets strålskyddsmyndighet.

De personer som har ett sköldkörtelinnehåll av ^{131}I med aktivitet mindre än A löper inte en oacceptabelt stor risk för skador på grund av strålningen, men kan behöva följas upp med känsligare utrustning på ett senare stadium för exempelvis epidemiologiska studier.

Personer med en sköldkörtelaktivitet mellan A och B bör undersökas med känsligare utrustning (helkroppsräknare) för en noggrannare bestämning av aktiviteten. För dessa personer beräknas stråldosen utgående ifrån tabellerade data (doskoefficienter).

För de som har en intern kontamination som överstiger aktiviteten B krävs i regel särskild mätutrustning för noggrann dosbestämning. Dessa personer kan också komma att behöva medicinsk vård för att minska risken för framtida men.

2.1.5 Sammanställning av några mätinstrument

Tabell 1 och Tabell 2 nedan visar resultatet av en kalibrering av några förekommande instrument för stråldosmätning. Kalibreringsfaktorer anges för vuxna respektive barn, då mätinstrumentet är i kontakt med halsen. Beroende på hur respektive instrument visar mätresultatet anges kalibreringsfaktorn i enheterna $\mu\text{Sv}/\text{h}/\text{Bq}$ respektive cps/Bq .

Tabell 1. Kalibrering av handinstrument för mätning av ^{131}I i tyreoidea (vuxna) med instrumentet i kontakt med halsen. Aktiviteten beräknas som (mätvärde-bakgrundsvärde)/kalibreringsfaktor.

Fabrikat	Typ	Detektortyp	Detektorstorlek	Kalibreringsfaktor ($\mu\text{Sv}/\text{h}/\text{Bq}$ alt. cps/Bq)
BICRON	ANALYST	NaI(Tl)	50 × 50 mm	0,041
SAPHYMO-STEL	SPP2	NaI(Tl)	25 × 50 mm	0,012
Exploranium	Gr-110s	NaI(Tl)	38 × 38 × 50 mm	0,021
Mini Instruments	6-90 Scaler-Ratemeter	NaI(Tl)	25 mm	0,0045
RNI	10	GM-rör		0,000016
SAPHYMO-Phy	ADB/AD-6	Plastscint.	76 × 76 mm	0,013
SAPHYMO-Phy	ADB/AD-3R	GM-rör		0,000013
Automess	AD-b/AD-6	Plastscint.	76 × 76 mm	0,013
Automess	6150 AD3 R	GM-rör	18 × 8 mm Ø	0,000013
Okänt fabrikat	SRP-88	NaI(Tl)	25 × 40 mm	0,0012
Morgan	Minimonitor 900	NaI(Tl)	25 × 19 mm Ø	0,0042
Morgan	Minimonitor 900	NaI(Tl)	25 × 32 mm Ø	0,0093

Tabell 2. Kalibrering av handinstrument för mätning av ^{131}I i tyreoidea (barn) med instrumentet i kontakt med halsen.

Fabrikat	Typ	Detektortyp	Detektorstorlek	Kalibreringsfaktor ($\mu\text{Sv}/\text{h}/\text{Bq}$ alt. cps/Bq)
SAPHYMO-STEL	SPP2	NaI(Tl)	25 × 50 mm	0,020
Exploranium	Gr-110s	NaI(Tl)	38 × 38 × 50 mm	0,036
RNI	10	GM-rör		0,000039
SAPHYMO-Phy	ADB/AD-6	Plastscint.	76 × 76 mm	0,013
Automess	AD-b/AD-6	Plastscint.	76 × 76 mm	0,022
Okänt fabrikat	SRP-88	NaI(Tl)	25 × 40 mm	0,0019
Morgan	Minimonitor 900	NaI(Tl)	25 × 19 mm Ø	0,0059
Morgan	Minimonitor 900	NaI(Tl)	25 × 32 mm Ø	0,015

2.2 Helkroppsmätning av ^{137}Cs med enkel apparatur

Mätning av ^{137}Cs i hela kroppen kan också göras med enkel utrustning, t.ex. enkanals spektrometrar, i situationer där det är viktigt att kunna mäta en stor grupp människor snabbt utan stora krav på noggrannhet. Vid dessa mätningar placeras detektorn i knät på personen som ska mätas och personen kan antingen sitta upprätt eller böja sig över detektorn (s.k. Palmer- eller Lappgeometri), se bild. Om personen böjer sig över detektorn blir känsligheten högre, men å andra sidan ger den upprätta positionen möjlighet att mäta även mindre flexibla personer, t.ex. barn, gravida kvinnor, äldre och sjuka. Även s.k. ryggeometri kan användas, se bild.

Den minsta detekterbara aktiviteten i denna typ av mätningar har uppskattats till 1 – 2 kBq för ^{137}Cs ($\pm 50\%$) [Zvonova *et al.*, 1995]. Osäkerheterna i mätningen beror på dels personens läge vid mätningen och dels på bakgrunden som skärmas till viss del av personen. Eftersom mätningarna ofta utförs i områden med hög bakgrund kan den senare faktorn vara problematisk då den är svår att uppskatta. Jämförelser med reguljära helkroppsmätare har dock visat en tillfredställande överensstämmelse och metoden kan betraktas som tillförlitlig om man är medveten om de korrekationer (bakgrund m.m.) som måste göras.

2.2.1 Förberedande skede

- Välj lämplig lokal för mätningarna.
 - Lokalen bör helst vara belägen i ett område som inte drabbats av nedfall. Om detta inte är möjligt väljs en lokal i en byggnad med hög skärmningsfaktor, t.ex. källare i stenhus.
 - Lokalen bör vara så stor att väntrum och mätrum kan iordningsställas. Tvättmöjligheter (dusch) är önskvärt.
- Arrangera lokalen för mätning och omhändertagande av personer som ska mätas.
 - Ställ i ordning väntrum med skogräns och utrymme för ytterkläder.
 - Avgränsa ett utrymme för mätning. Hotet är att utrymmet blir kontaminerat. Personer som kommer för mätning lämnar skor och ytterkläder redan innan de kommer till det avgränsade området.
 - Arrangera för inlämning av kontaminerade kläder och skor.
- Granska tvättmöjligheter (om sådana finns).
 - Kontrollera i så fall att det finns tillräckligt med handdukar, mätdräkter, tvål, schampo etc. Ordna service för tvätt.
- Förbered mätinstrumenten.
 - Kontrollera apparaturen. Skydda detektorn med en plastpåse. Placera, om möjligt, ut "limmattor" på strategiska ställen.Se till att mätinstruktioner för instrumenten finns tillgängliga.

2.2.2 Aktionsskede

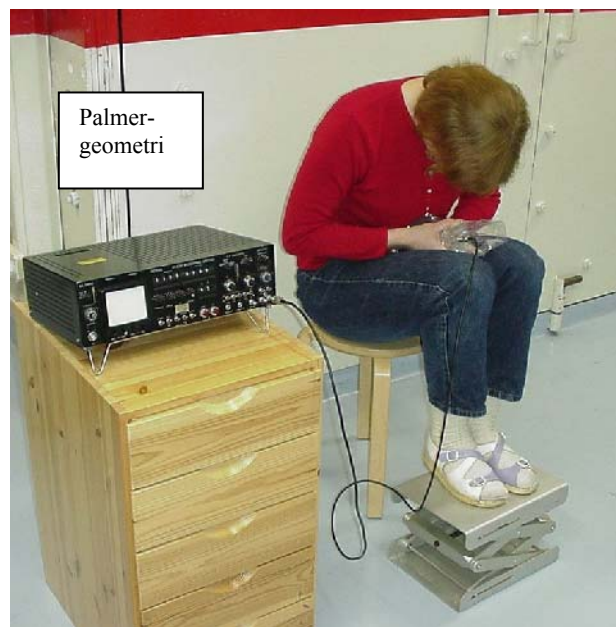
Minns att det är viktigt att behandla dem som kommer till mätning lugnt och vänligt. De är sannolikt mera bekymrade än du. Visa inte din egen oro, prata inte för mycket, koncentrera dig på vad du gör och förklara gärna varför.

- Kontrollera personernas eventuella ytkontamination redan före skogränsen. Var lugn och vänlig. Minns kontaminationsrisken, undvik kontamination på golv, sittplatser, instrument. Använd ditt sunda förnuft
- Personerna klär av sig ytterkläderna och tar av sig skorna vid ytterdörren. Tvättar händerna om det är möjligt. Överväg i annat fall att förse personerna med engångshandskar. Skydda stolarna med plast eller annat material som går lätt att byta.

2.2.3 Mätning:

- Koppla på apparaten.
- Välj mättid (100 s eller motsvarande för det instrument som används)
- Skriv in personens namn och andra uppgifter i mätblankett (Bilaga 2)
- Skydda detektorn med en plastpåse som byts efter varje mätning
- Starta mätningen.
- Notera resultatet och fyll i mätblanketten.
- Gör en bakgrundsmätning på en person som inte är kontaminerad under samma tid som personmätningen.
- Notera resultatet och fyll i mätblanketten.
- Kontrollera att blanketten är korrekt ifylld.

Kom ihåg lagar och förordningar angående registrering av personuppgifter. Inga uppgifter ges åt utomstående utan medgivande av personen det gäller. Minns din tystnadsplikt. Inga utomstående får närvara i mätutrymmena. Mediarepresentanter får närvara endast med särskilt tillstånd.



2.2.4 Sammanställning av några mätinstrument

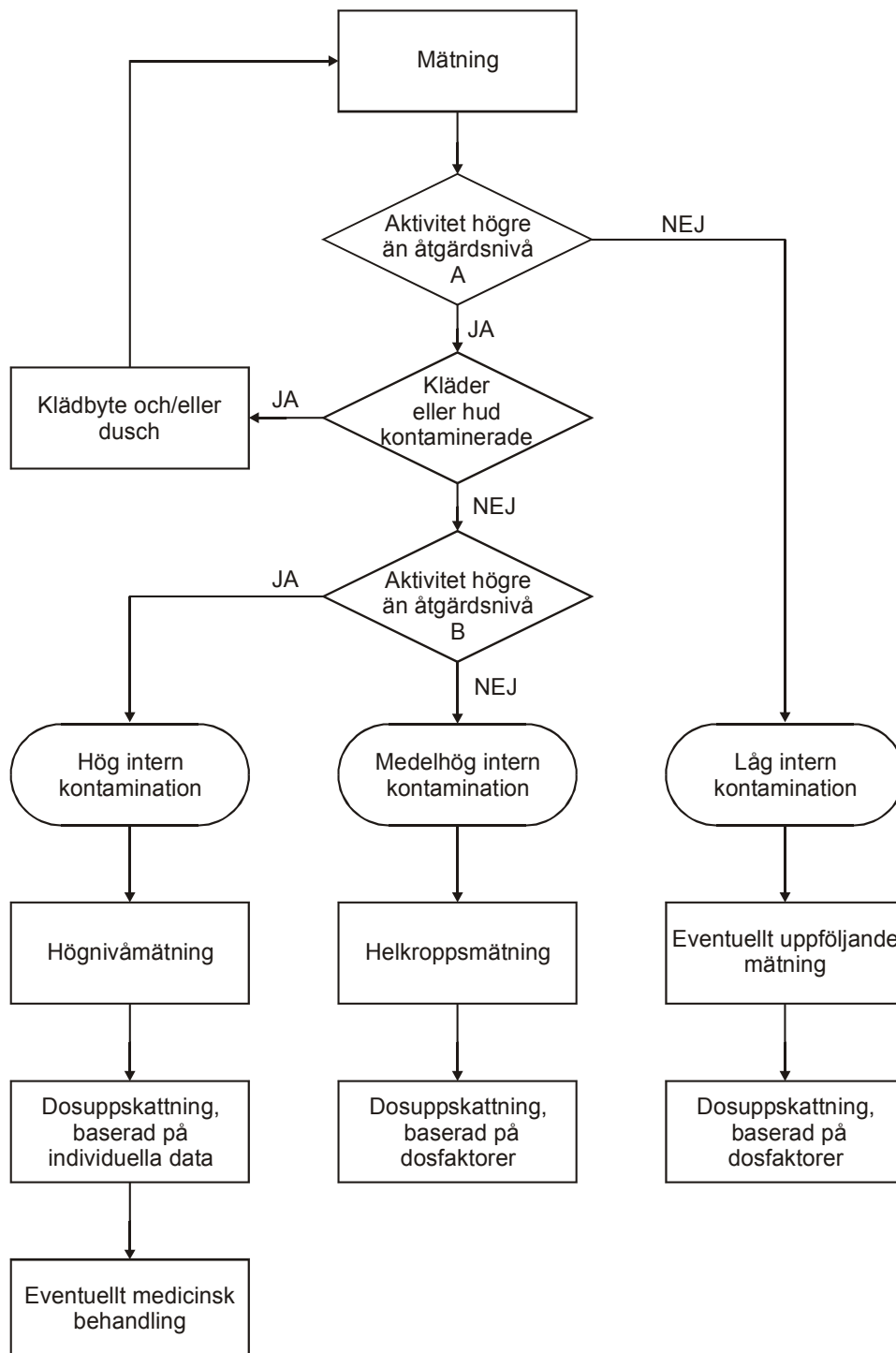
Tabell 3 nedan visar resultatet av en kalibrering av några förekommande instrument för stråldosmätning. Kalibreringsfaktorer anges för vuxna för två olika mätgeometrier, ”Palmer-geometri” och ”Rygg-geometri”. Beroende på hur respektive instrument visar mätresultatet anges kalibreringsfaktorn i enheterna $\mu\text{Sv/h/Bq}$ respektive cps/Bq .

Tabell 3. Kalibrering av handinstrument för mätning av ^{137}Cs i människa (vuxna) för två olika mätgeometrier. Aktiviteten beräknas som (mätvärde-bakgrundsvärde/kalibreringsfaktor).

Fabrikat	Typ	Detektortyp	Detektorstorlek	Mätgeometri	Kalibreringsfaktor ($\mu\text{Sv/h/Bq}$ alt. cps/Bq)
BICRON	ANALYST	NaI(Tl)	50 × 50 mm	Rygg	0,0025
BICRON	ANALYST	NaI(Tl)	50 × 50 mm	Palmer	0,0039
SAPHYMO-STEL	SPP2	NaI(Tl)	25 × 50 mm	Palmer	0,00079
RADOS	SRV-2000	GM-rör		Palmer	0,0000006
Exploranium	Gr-110s	NaI(Tl)	38 × 38 × 50 mm”	Palmer	0,0010
Exploranium	Gr-110s	NaI(Tl)	38 × 38 × 50 mm	Rygg	0,0011
RNI	10	GM-rör		Palmer	0,0000013
RNI	10	GM-rör		Rygg	0,00000063
Morgan	Minimonitor 900	NaI(Tl)	25 × 19 mm Ø	Rygg	0,000095
Morgan	Minimonitor 900	NaI(Tl)	2,5 × 32 mm Ø	Rygg	0,00038
Morgan	Minimonitor 900	NaI(Tl)	25 × 19 mm Ø	Palmer	0,000095
Morgan	Minimonitor 900	NaI(Tl)	25 × 32 mm Ø	Palmer	0,00038
Okänt fabrikat	SRP-88	NaI(Tl)	25 × 40 mm	Rygg	0,000057
Okänt fabrikat	SRP-88	NaI(Tl)	25 × 40 mm	Palmer	0,00010

2.2.5 Beslut om åtgärder

Figur 3 nedan anger IAEA:s rekommendation för åtgärder i samband med kontrollmätningar av stora befolkningsgrupper (IAEA, 1994). Enligt figur 3 bestäms två åtgärdsnivåer, A och B, sådana att dessa aktiviteter av ^{137}Cs ger en viss stråldos. Åtgärdsnivåerna och motsvarande aktiviteter för cesium eller eventuellt andra radionuklider meddelas vid den uppkomna situationen av landets strålskyddsmyndighet.



Figur 3 Rekommenderade åtgärder vid mätning.

3 ^{131}I och ^{137}Cs mätning med gammakamera och jod-monitor

3.1 Mätning med gammakamera

Gammakameror som finns på många sjukhus kan utgöra en viktig resurs i samband med kontaminationsmätningar av ett stort antal människor i en beredskapssituation. På sjukhusen finns också personal som har erfarenhet av dessa mätningar.

Wallström et al. (1995) har vid undersökningar av två olika kameror funnit att minsta detekterbara aktivitet (MDA) beror på (1) avståndet från mätobjektet, (2) det energiintervall som används vid mätningen och (3) om kameran används med eller utan kollimator. MDA var lägst då kameran placerades nära kroppen och användes utan kollimator. Nackdelarna med denna mätkonfiguration är att kameran kan behöva balanseras (med avseende på mekanisk stabilitet) för att kompensera för kollimatoren och att känslighetens variation med mätobjektets storlek blir större vid små avstånd.

Bakgrundens variation, vid mätningar med gammakamera i en beredskapssituation, utgör den främsta källan till mätresultatens osäkerhet. Denna beror i första hand på variationer i omgivningens strålnivå och skärmning av personer som befinner sig i närheten av kameran. Speciellt skärmningseffekten kan vara omfattande och även svårhanterlig eftersom den varierar med storleken på de skärmande personerna.

Som ett exempel på MDA och känslighet vid mätning med gammakamera visas i Tabell 4 några uppmätta data från fantommätningar (Wallström et al., 1995).

Tabell 4. Minsta detekterbara aktivitet och känslighet vid mätning av ^{137}Cs och ^{131}I med gammakamera i olika konfigurationer (kollimatorval och detektorhöjd). Aktiviteten beräknas som (mätvärde-bakgrundsvärde)/kalibreringsfaktor.

Energi-intervall (keV)	Kollimator	Fantom-storlek (kg)	MDA ^{137}Cs i helkropp (kBq)		MDA ^{131}I i thyroidea (kBq)		Känslighet ^{137}Cs i helkropp, centralt (cps/kBq)		Känslighet ^{131}I i thyroidea, över halsen (cps/kBq)	
			5 cm	35 cm	10 cm	40 cm	5 cm	35 cm	10 cm	40 cm
50-450	Ingen	14	0,25	0,90			78	24		
		61	0,40	1,0	0,10	0,45	46	21	170	47
		93	0,45	1,1			39	18		
	LEGP	14	0,80	2,3			9,1	3,3		
		61	2,1	3,5	0,80	2,5	3,8	2,2	9,8	3,4
		93	2,3	4,2			3,0	1,7		
	HEGP	14	3,6	8,5						
		61	12		13	17				
		93	11	17						
550-750	Ingen	14	0,30	1,3			12	3,2		
		61	0,70	2,0	1,5	9,0	5,4	2,2	2,6	0,47
		93	1,6	3,2			2,2	1,2		
	LEGP	14	1,1	2,9			2,9	1,1		
		61	2,5	4,9	4,5	20	1,2	0,64	0,67	0,16
		93	3,0	5,8			0,93	0,52		
	HEGP	14	7,0	17						
		61	13	31	26	47				
		93	11	27						

3.2 Mätning med jod-monitor.

Jod-monitorer som också finns på många sjukhus kan utgöra en viktig resurs i samband med kontaminationsmätningar av ett stort antal människor i en beredskapssituation. På sjukhusen finns också personal som har erfarenhet av dessa mätningar.

Jodmonitorerna används i diagnostiskt syfte i sjukvården och är speciellt avsedda för mätning av ^{131}I i sköldkörteln. Ett exempel på sådan utrustning är Nukleus – jodmonitor (256-kanals analysator) försedd med 50 mm NaI(Tl)-detektor och bakgrundsskydd av bly där känsligheten är 0.001 cps/Bq.

3.3 Mätprocedur och rapportering

Mätningarna utförs i enlighet med de procedurer som normalt används vid sjukhuset/lasarettet. Rapportering sker förslagsvis enligt blanketten i bilaga 3.

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Personblankett: Mätning av ^{131}I med handinstrument

Bilaga 1

Personuppgifter

Namn		Personnummer
Hemadress		Telefonnummer hem/Mobil
		Telefonnummer arbetsplats
Arbetsplatsadress		Yrke
Vikt (kg)	Längd (cm)	

Övriga uppgifter/anmärkningar

--

Mätuppgifter

Mätplats		
<table border="1"> <tr> <td>Monitortyp</td> <td>Serienummer</td> </tr> </table>	Monitortyp	Serienummer
Monitortyp	Serienummer	
Ytterligare uppgifter		

Resultat

Datum	Tidpunkt	Avstånd (cm)	Avläst resultat thyroidea ($\mu\text{Sv/h}$) alt. (cps/Bq)	Avläst resultat lårbakgrund ($\mu\text{Sv/h}$) alt. (cps/Bq)	Nettoresultat ($\mu\text{Sv/h}$) alt. (cps/Bq)	Kalibreringsfaktor ($\mu\text{Sv/h/kBq}$) alt. (cps/Bq)	Aktivitet (kBq)

Mätningen utförd av
Sign.

Rekommenderade åtgärder
Datum (yy-mm-dd), Sign.
Namnförtydligande

Personblankett: Mätning av ^{137}Cs med enkel apparatur

Bilaga 2

Personuppgifter

Namn		Personnummer
Hemadress		Telefonnummer hem/Mobil
		Telefonnummer arbetsplats
Arbetsplatsadress		Yrke
Vikt (kg)	Längd (cm)	

Övriga uppgifter/anmärkningar

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Mätuppgifter

Mätplats		
<table border="1"> <tr> <td>Monitortyp</td> <td>Serienummer</td> </tr> </table>	Monitortyp	Serienummer
Monitortyp	Serienummer	
Ytterligare uppgifter		

Resultat

Datum	Tidpunkt	Mätgeometri Rygg/Palmer	Avläst resultat ($\mu\text{Sv/h}$) alt. (cps/Bq)	Avläst resultat bakgrund ($\mu\text{Sv/h}$) alt. (cps/Bq)	Nettoresultat ($\mu\text{Sv/h}$) alt. (cps/Bq)	Kalibreringsfaktor ($\mu\text{Sv/h/kBq}$) alt. (cps/Bq)	Aktivitet (kBq)

Mätningen utförd av
Sign.

Rekommenderade åtgärder
Datum (yy-mm-dd), Sign.
Namnförtydligande

Bilaga 3

Personblankett: Mätning av ^{137}Cs och ^{131}I med gammakamera och jodmonitor

Personuppgifter

Namn		Personnummer
Hemadress		Telefonnummer hem/Mobil
		Telefonnummer arbetsplats
Arbetsplatsadress		Yrke
Vikt (kg)	Längd (cm)	

Uppgifter om exponeringen

Exponeringsdatum (yy-mm-dd)
Exponeringsförhållanden

Mätuppgifter

Mätplats	Mätdatum (yy-mm-dd)	Mätning nummer
Mätutrustning och geometri (avstånd, gammakamera med/utan kollimator, etc.)		
Mättid (s)	Ytterligare uppgifter	

Resultat

Radionuklid	Räknehastighet, netto (cps)	Bakgrund (cps)	Känslighet (Bq/cps)	Aktivitet (kBq)

Mätningen utförd av	Rekommenderade åtgärder
Sign.	Datum (yy-mm-dd), Sign.
	Namnförtydligande

Timing of fluxes in the Nordic Seas - overview

Svend Aage Malmberg

Hafrannsóknastofnunin (Marine Research Institute, Reykjavík, Iceland)

The large-scale near-surface circulation in the Nordic Seas is dominated by the warm northward flowing Atlantic inflow, mainly on the eastern side, and the cold East Greenland Current flowing southwards on the western side (Figure 1). Atlantic water enters the Norwegian Sea mainly through the Faroe-Shetland Channel along the Scottish slope feeding the eastern branch of the Norwegian Atlantic Current, and between the Faroes and Iceland feeding the Faroe Current which flows further into the Norwegian Sea as a western branch of the Norwegian Atlantic Current. The eastern branch of the Norwegian Atlantic Current divides into an eastern flow into the Barents Sea and a northwards flow towards Spitzbergen. Some Atlantic water from the south is also carried into the Iceland Sea through the Denmark Strait by the North Icelandic branch of the Irminger Current. In the north off Spitsbergen the Atlantic water of the eastern branch of the Norwegian Current named West Spitsbergen Current, divides into a branch flowing into the Arctic Ocean and another one recirculating counterclockwise into the southward flowing East Greenland Current. Together with the main southward flowing branch of the East Greenland Current it also branches off counterclockwise to the east firstly as Jan Mayen Polar Current, which brings the waters into a cyclonic circulation in the Greenland Sea, and secondly and further south as the East Icelandic Current flowing eastwards into the Iceland and Norwegian Seas. Thus in general, the circulation in the upper layers of the Nordic Seas is fed by the warm Atlantic water from the south and the cold Polar water from the north, a circulation steered by the topographic features into cyclonic circulation's, both overall in the Nordic Seas and regionally in the separated Norwegian, Greenland and Iceland Seas or Basins (Dickson *et al.* 1988, Poulain *et al.* 1996, Belkin *et al.* 1998, Blindheim *et al.* 1999, Hansen & Østurhus 2000).

Contaminants in the waters from Western Europe reach the eastern areas of the Nordic Seas from the south and the western areas from the north by the cyclonic current systems. The timing of this pathway from the North Sea area and around the Nordic Seas into North Icelandic waters was established according to time-series on salinity as well as radionuclides in the 1970's and 1980's 4-7 years but 7-10 years all the way from Sellafield (Dahlgaard 1994). Ongoing research on the fate of the recent Tc-99 radionuclide released in the latter half of the 1990's may reveal its appearance in North Icelandic waters early in the first decade of the new millennium.

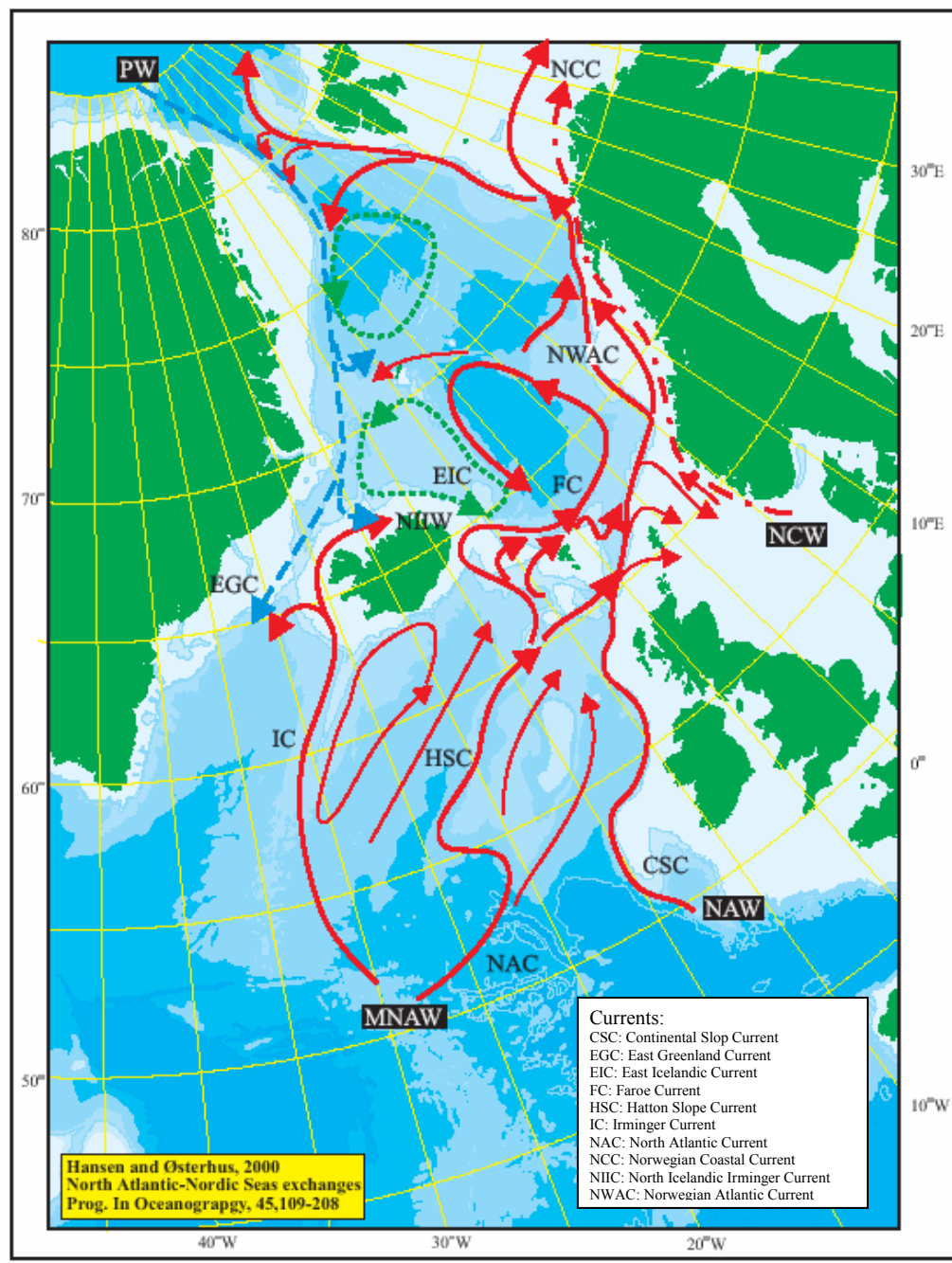


Figure 1. Ocean currents in the Nordic seas.

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Long term study of ^{99}Tc in brown seaweed from the Swedish west coast

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Abstract

Technetium was analyzed in brown seaweed samples from Särö, located at the Swedish west coast. Since 1967 this location has been used for regular sampling of seaweed (Fucus serratus and Fucus vesiculosus). The activity concentration of ^{99}Tc in seaweed is influenced by the liquid discharge from the nuclear fuel reprocessing plant in Sellafield, UK. Transit time and seasonal variations were estimated by comparing the concentration of technetium in seaweed with the annual discharge of that radionuclide from Sellafield. In this work samples collected from 1994 to 2000 were analyzed and the results were combined with data from 1967 to 1993. The concentration of ^{99}Tc in the 1994-2000 seaweed samples varied between 10 and 230 Bq/kg dry weight. The current high values of ^{99}Tc in seaweed samples originate from the increased liquid discharge of technetium starting when the Enhanced Actinide Removal Plant (EARP) at Sellafield came into operation in 1994.

Introduction

Technetium-99 is a long-lived ($T_{1/2} = 213\,000\text{ y}$) pure β -emitter ($E_{\beta\text{max}} = 293\text{ keV}$). It is formed as a daughter product of ^{99}Mo , which is produced by fission of ^{235}U or ^{239}Pu or neutron activation of ^{98}Mo . Tc is very conservative under aerobic seawater condition with a K_d factor of 1×10^2 for coastal sediment (IAEA, 1985) and is most likely present as pertechnetate, TcO_4^- (Shulte and Scoppa, 1987). It has therefore been used in several tracer studies in the marine environment to monitoring different oceanographic parameters.

Seaweed is an excellent bioindicator to monitor the variations of Tc in seawater because of its high uptake of Tc as pertechnetate. Because of the high concentration factor ($\sim 10^5$) technetium can easily be determined in seawater with low activity concentration.

The technetium concentration at the Swedish west coast is dominated by the controlled liquid discharge from Sellafield nuclear fuel reprocessing plant with a minor contribution from La Hague reprocessing plant and the Chernobyl accident in 1986. The contribution from nuclear bomb test fall-out in these waters is about 1 Bq/kg in seaweed (Holm et al., 1984a).

The Sellafield nuclear reprocessing plant has been releasing liquid discharge, contaminated with radionuclides, into the sea from the beginning of the operation in 1952. When the EARP (Enhanced Actinide Removal Plant) began operation in 1994 the estimated liquid discharge of ^{99}Tc increased from 2-6 TBq/year in 1981-1993 up to 190 TBq/year in 1995. This is due to that the highly soluble pertechnetate ion is not effectively removed in the process.

The aim of this study was to investigate the time variations of ^{99}Tc activity concentration in seawater by using seaweed as a bioindicator.

Material and Methods

Sampling

Sampling was performed at Särödal (56.76 °N, 12.63 °E) located at the Swedish west coast (figure 1). The collection of brown seaweed (*Fucus serratus* and *Fucus vesiculosus*) at this location started in 1967 (Mattsson, 1984, Mattsson and Erlandsson, 1991) and is still going on. In this study samples collected from 1994 to 2000 were analyzed for technetium and the results were compared with data for technetium concentration until 1993 (Roos et al., 1993).

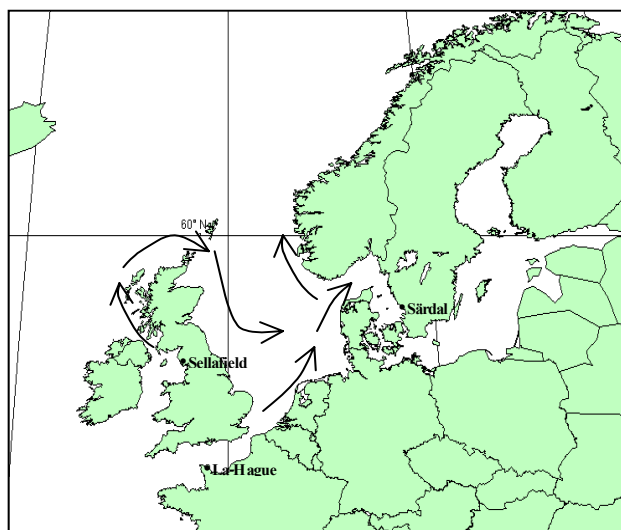


Figure 1. Map of northern Europe showing the Särödal sampling site, Sellafield (UK) and La Hague (France) and major surface currents in the North Sea.

Analyses

The samples were weighted (3-5 g d.w.) and ashed at 550 °C after addition of oxalic acid until all the carbon are removed. According to Momoshima et al., 1991, the loss of ^{99}Tc from dry ashing is in the range of 5 to 10 %. About 20 kBq of $^{99}\text{Tc}^{\text{m}}$ in the form of pertechnetate, TcO_4^- , were added as a yield determinant. The analytical separation procedure used was a modified version developed by Ballestra et al., 1987. The ash was dissolved in hydrogen chloride acid and Tc was oxidized to +VII oxidation state with potassium persulphate and hydrogen peroxide. The samples were transformed to 3 M sulphuric acid and Tc was extracted with tributylphosphate and hydrogen fluoride. The technetium was then back-extracted with 2 M sodium hydroxide solution and xylene, electroplated on to stainless steel discs, and counted by an anticoincidence shielded GM-counter. The radiochemical recovery was determined by counting the activity of $^{99}\text{Tc}^{\text{m}}$ ($E_\gamma = 140 \text{ keV}$) with a HPGe-detector and was normally between 20 and 70 %.

Results and Discussion

As can be seen in figure 2 the ^{99}Tc concentration in *Fucus serratus* varied with the Sellafield discharge. The activity concentration increased from about 10 Bq/kg in 1995 up to 230 Bq/kg in the year 2000. This indicates a transit time from Sellafield to the Swedish west coast of 4-5 years, which is in good agreement with earlier estimations by Dahlgaard, 1995.

The seasonal variation of technetium concentration in the 1994-2000 samples are not so pronounced as in previous investigations from the same locations (Holm et al., 1984b). The explanation for this is still under investigation. One explanation can be that the seawater movement has changed over the years and altered the salinity and the routes of transport in the coastal area.

Comparing the activity uptake of Tc in *Fucus serratus* with *Fucus vesiculosus* sampled at the same time show a ratio of 0.52 ± 0.06 . It agrees well with earlier measurements of 0.56 ± 0.15 (Holm et al., 1984a).

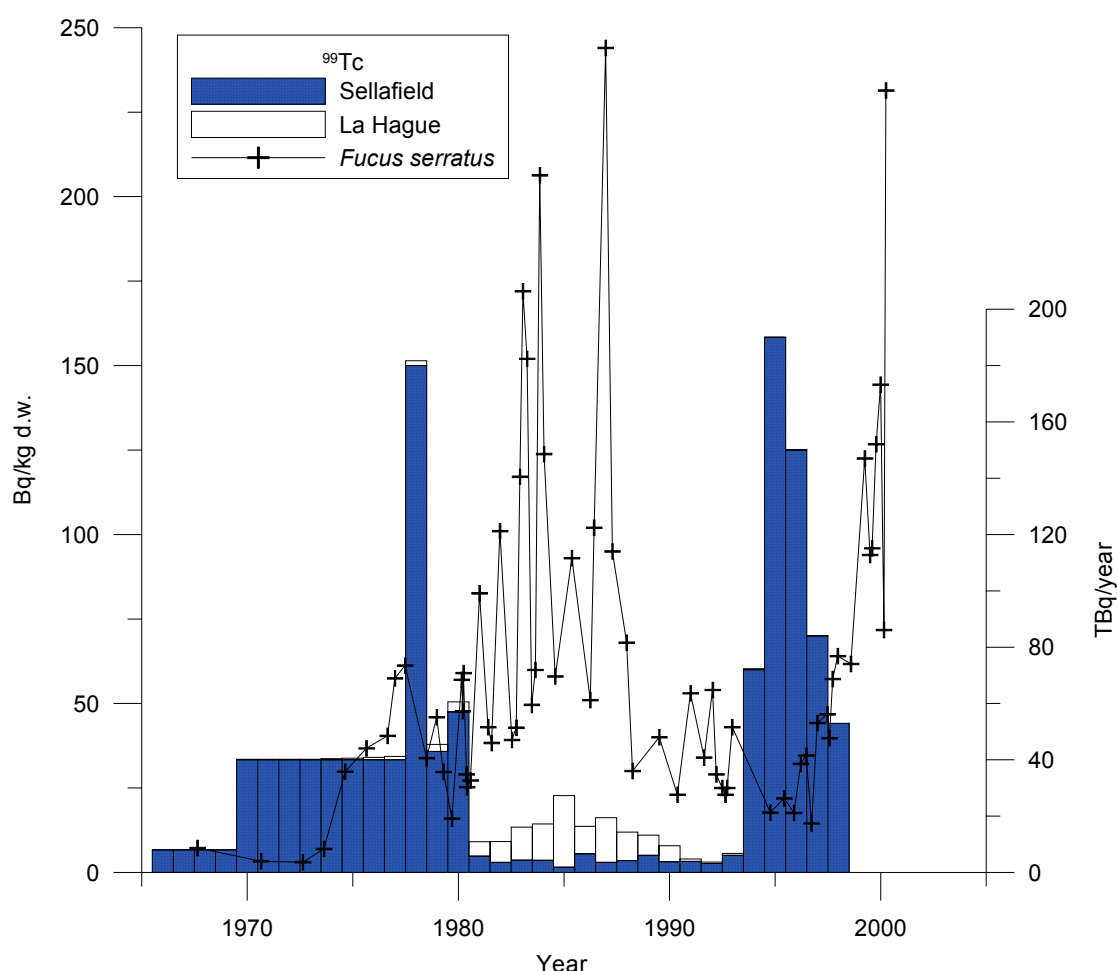


Figure 2. Activity concentration of ^{99}Tc (primary axis) in *Fucus serratus* from Särðal (56.76 °N, 12.63 °E) and annual ^{99}Tc discharge from Sellafield and La Hague (secondary axis).

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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Radioactive Tracers in Nordic Waters, ^{99}Tc , ^{137}Cs and ^{129}I

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Introduction

The discharges of ^{99}Tc from Sellafield increased dramatically late 1994 when the new Enhanced Actinide Removal Plant, EARP, started operation. Earlier studies based on ^{137}Cs discharges from Sellafield indicated a 4 year transport time from the discharge to the inner Danish waters, the Kattegat and the Belt Sea.

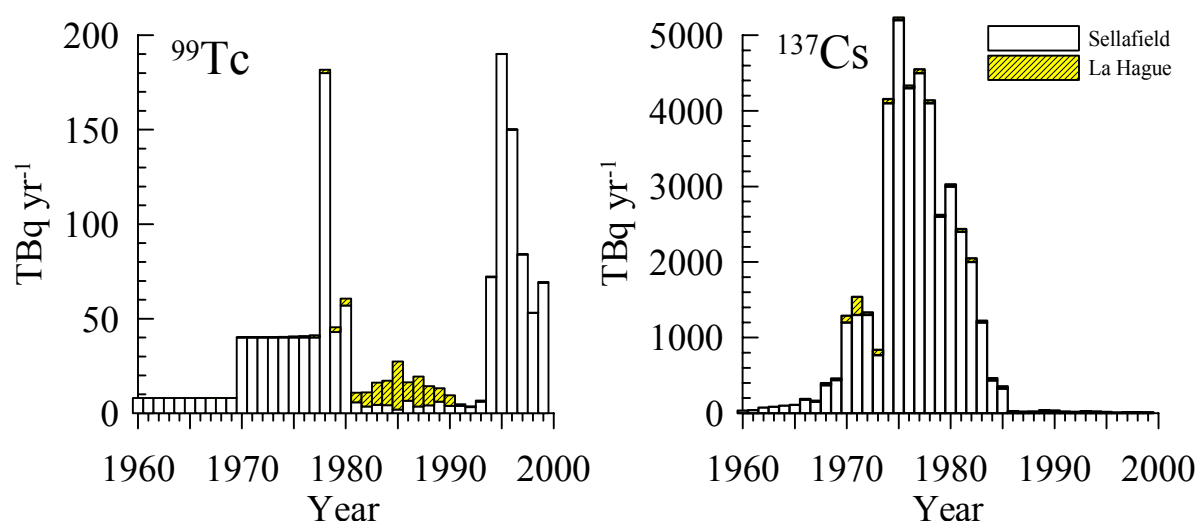


Figure 1. Annual discharges of ^{99}Tc and ^{137}Cs to the marine environment from Sellafield and La Hague.

Objectives

The work will focus on a re-evaluation of the transfer of technetium from Sellafield to the Danish Straits

Material and Methods

Sampling

Seawater samples from near bottom and surface has been collected at least twice annually since the 1960's at a number of locations in the Danish Straits. Since 1992, the samples have been taken by the Royal Danish Navy.

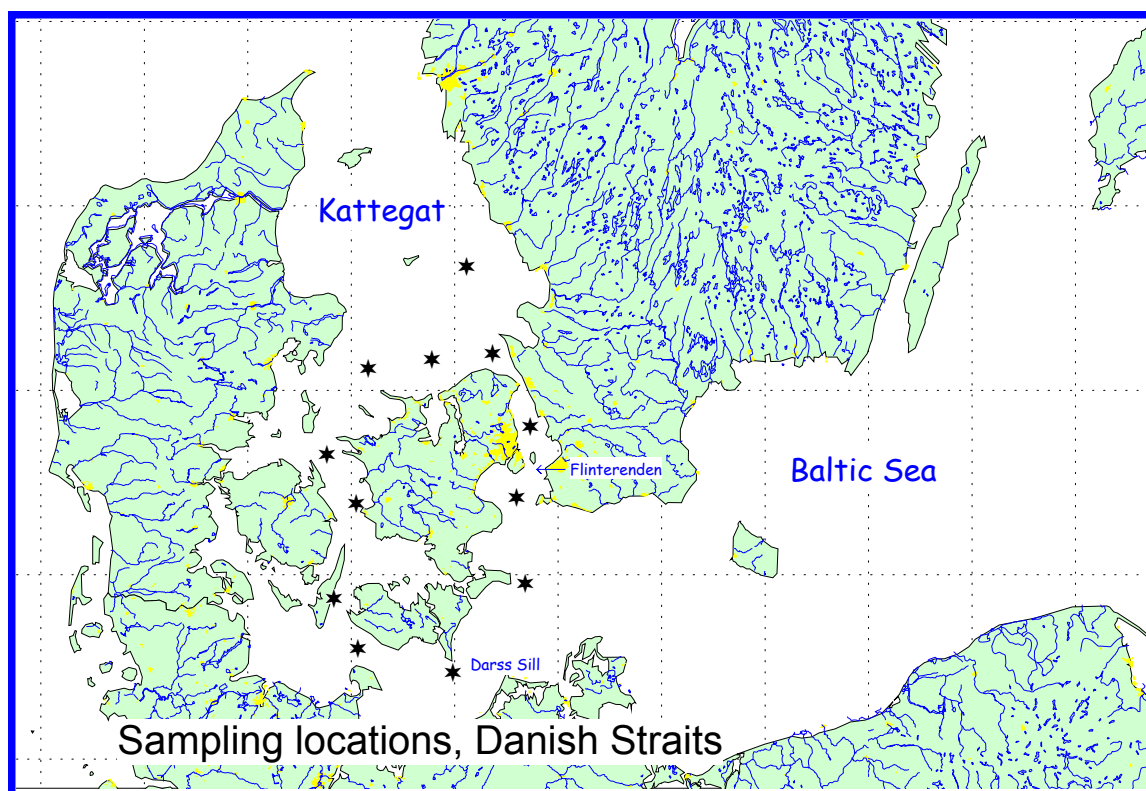


Figure 2.: Sampling locations in Danish straits normally taken around May and November. ^{137}Cs data have been analysed since 1972; ^{99}Tc since 1998. The two low sills controlling the water exchange with the Baltic: Flinterenden (8 m) and Darss Sill (18 m), are shown.

Analyses

Technetium-99 is analysed according to Chen *et al.*, (1994): the technetium is concentrated by anion exchange directly from seawater after adding $^{99\text{m}}\text{Tc}$ as a yield determinant. After appropriate cleaning and electroplating the yield is determined by gamma counting, and the ^{99}Tc is determined on a proportional β -counter after the decay of the yield determinant. ^{137}Cs is measured by gamma counting on low background semiconductor (Ge) counters after concentration on ammonium-molybdo-dodeca-phosphate (AMP). ^{129}I has been determined by radiochemical neutron activation analysis (Hou *et al.*, 1999).

Results and Discussion

The Baltic outflow of ^{137}Cs through the Danish Straits

The Baltic water exchange is slow. The connection to the North Atlantic is restricted through the Danish straits with two shallow sills determining the border between the Baltic and the transition zone. A driving force in the Baltic water exchange is the fresh water surplus from river run-off estimated as $473 \text{ km}^3 \text{ yr}^{-1}$ (Sehested Hansen *et al.*, 1990). This amount of water constitutes the net outflow leaving the Baltic through the surface layer in the Danish Straits. Using the “Knudsen theorem” (Knudsen, 1900; Jacobsen, 1980), Sehested *et al.* calculated the mean Baltic outflow as $1107 \text{ km}^3 \text{ yr}^{-1}$ with a salinity of 8.6 ‰, and the inflow to $634 \text{ km}^3 \text{ yr}^{-1}$ with a salinity of 15 ‰. The inflowing North sea water is gradually entrained in the outflowing surface water leaving the Baltic. Therefore the surface water

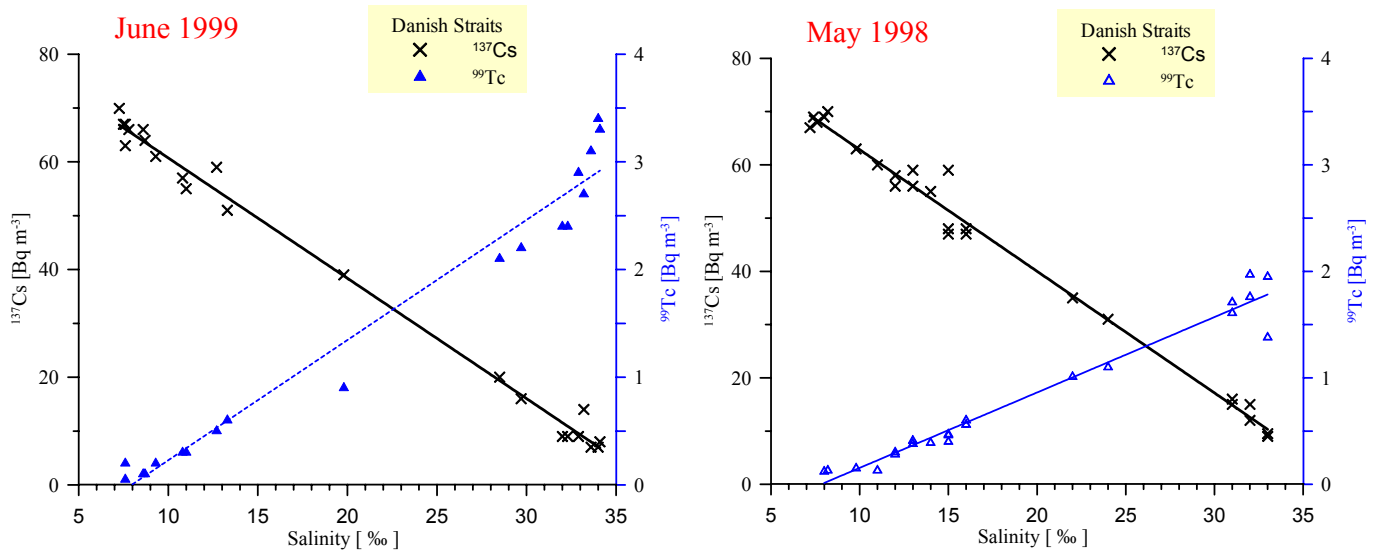


Fig. 3: Concentrations of ^{137}Cs and ^{99}Tc in the Danish Straits as a function of salinity June 1999 and May 1998.

salinity is gradually increasing from the Baltic to the North Sea. The entrainment means that part of the $634 \text{ km}^3 \text{ yr}^{-1}$ are "recycled" Baltic outflow. Correcting to 34 ‰, i.e. full salinity Atlantic water, gives a value of $280 \text{ km}^3 \text{ yr}^{-1}$ of new inflow, i.e. the total water exchange rate – the freshwater surplus plus the new inflow – is $753 \text{ km}^3 \text{ yr}^{-1}$. A mean residence time for the 21580 km^3 Baltic water volume may then be calculated as 29 years equivalent to a "half-life" for the water volume of 20 years.

A semi-annual time series of ^{137}Cs data from the Danish straits have been collected since 1972. Linear regressions of ^{137}Cs concentrations versus salinity (Fig. 3) are used to estimate the time-trend in the different water masses characterised above. It is seen that ^{137}Cs is dominated by the low-saline Baltic outflow whereas ^{99}Tc is linked to the inflowing salt North Sea water. The dominant ^{137}Cs source in the area is the Chernobyl contamination in the Baltic, whereas the ^{99}Tc originates from Sellafield discharges to the Irish Sea. A net outflow from the Baltic can be calculated as 39 TBq in year 2000. This value shows an exponential decrease 1991 – 2000 with an observed halflife of 13.4 years. When the physical half-life of ^{137}Cs (30.17 yr) is subtracted, the "stable" caesium net outflow may be described by an exponential decrease with a halflife of 24 years. This is equivalent to the Baltic water half-life indicated above as 20 years.

In the calculation above, there is no room for a net sedimentation of ^{137}Cs . Caesium is known to show a significant sedimentation – especially in the brackish, particle rich Baltic. Thus before the Chernobyl accident, sedimentation of ^{137}Cs was a significant parameter for inventory calculations (Salo & Tuomainen, 1986). If the indication of zero net sedimentation is valid, the explanation probably is that the sedimentation is at present counterbalanced by remobilisation of earlier sedimented ^{137}Cs and of the ^{137}Cs content in the river inflow. As the current runoff of ^{137}Cs to the Baltic is modest (Saxen & Ilus, 2001), remobilisation is suggested as the major responsible parameter. Remobilisation of previously sedimented ^{137}Cs has earlier been demonstrated from the Irish Sea (Hunt & Kershaw, 1990; Cook *et al.*, 1997).

Iodine-129

The concentration of ^{129}I in seawater from Bornholm and Møen in the Baltic Sea had reached 6.0×10^{-13} g/L and 16×10^{-13} g/L, respectively in 2000, and from the Kattegat, 270×10^{-13} g/L was found. The $^{129}\text{I}/^{127}\text{I}$ ratio in seaweed samples from Bornholm increased continuously from 2.8×10^{-8} in 1992 to 4.9×10^{-8} in 1999. This is more than two orders of magnitude higher than the global fallout level. By comparison with the level of ^{129}I in lake water and precipitation in this region, it is estimated that more than 95% of ^{129}I in the Baltic Sea originates from reprocessing emissions, especially from the French nuclear fuel reprocessing plant at La Hague. Further results are given in Hou *et al.*, (2000).

Conclusions

The Danish straits – the transit area between the North Atlantic and the Baltic Sea – has been monitored for ^{99}Tc and ^{137}Cs twice per year in the project period. Furthermore, several samples of seawater and seaweed have been analysed for ^{129}I by neutron activation analysis. The Sellafield discharge rate of ^{99}Tc showed a distinct peak in 1995. The peak was present in the in-flowing bottom water in the Danish straits in June 1999, i.e. 4 years after the discharge. Since then, concentrations have decreased in accordance with the discharge rate. This confirms an earlier estimate of the transit time based on the large ^{137}Cs discharges before the Chernobyl accident. The ^{137}Cs concentration in the Danish straits is now dominated by the outflow of low-saline water from the Baltic and is thus inversely related to the salinity, whereas ^{99}Tc concentrations are positively correlated with salinity as the concentration is dominated by the Sellafield discharges.

^{129}I discharges to the sea have increased sharply during the 1990's especially from La Hague in France. Results from time-series seaweed (*Fucus vesiculosus*) samples show, that the ^{129}I signal is penetrating not only to the Kattegat (Klint) but even to Bornholm in the Baltic

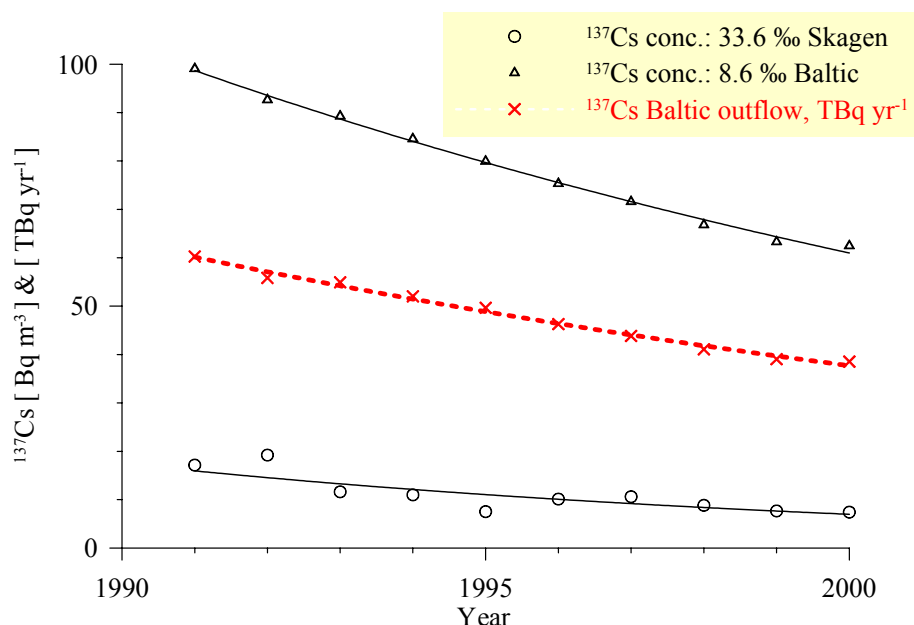


Figure 4: Calculated net ^{137}Cs outflow from the Baltic 1991 – 2000 and concentrations in two water masses, 33.6 ‰ and 8.6 ‰. The trend lines are exponential fit to the data.

proper.

It is concluded that data on environmental concentrations of ^{129}I as well as of ^{99}Tc in combination form a strong tracer tool as it makes a comparison of the transport from the two European reprocessing plants to Nordic waters possible.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research project BOK-2

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Outflow of radiocaesium from the Baltic Sea detected in brown algae along the southern Norwegian coast

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Introduction

The sources of artificial radionuclides in the Norwegian coastal waters during the last three decades have mainly been fallout from the atmospheric nuclear bomb tests, releases from European reprocessing plants (Sellafield and Cap de la Hague) and fallout from the Chernobyl accident. For the study of radionuclides in seawater along the Norwegian coast, brown algae have been used as bioindicators. The algae rapidly reflect changes in seawater concentrations and also accumulate most radionuclides. Analysis of algae is therefore often more convenient and has lower detection limits than analysis of the seawater itself.

The outflow water from the Baltic Sea flows northwards along the Swedish west coast into the southern Oslo Fjord and then turns southwest and joins the North Sea currents west of Cape Lindesnes. The ^{137}Cs transported out of the Baltic Sea may therefore significantly influence the concentration of this nuclide in the algae along the Norwegian coast.

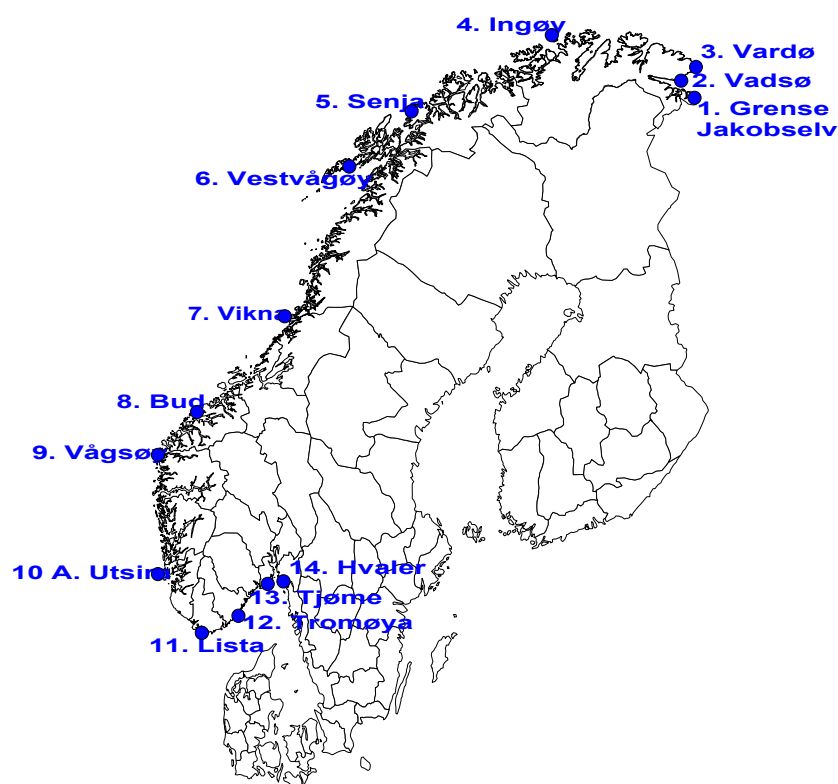


Figure 1. Map of Norway showing the sampling locations.

Sampling and measurements

The brown alga *Fucus vesiculosus* has been collected annually at several sampling locations since 1980 (Figure 1), mainly in August and September (Christensen (1986), Christensen and Selnaes (1995)). At the location Utsira in the southwest, monthly samples have been collected regularly since January 1986. The *Fucus* samples were dried at 105 °C, homogenised and analysed for gamma emitters using high purity germanium gamma spectrometry and computerised spectrum analysis.

In the early 1980's, the concentration of radiocaesium was highest at the locations Utsira and Lista in the south, decreasing both eastwards along the Skagerrak coast and northwards along the west coast to the Barents Sea (Figure 2). In the 1990's the highest concentrations have generally been found in the Oslo Fjord and Skagerrak and decreases northwards. The higher concentrations in 1983 and 1994 at the location Hvaler compared to the nearby Tjøme may probably be explained by differences in salinity. While the salinity at Tjøme is about 30 ‰, it is about 15 ‰ at Hvaler due to influence from the Glomma River, Norway's largest river (Figures 2 and 3).

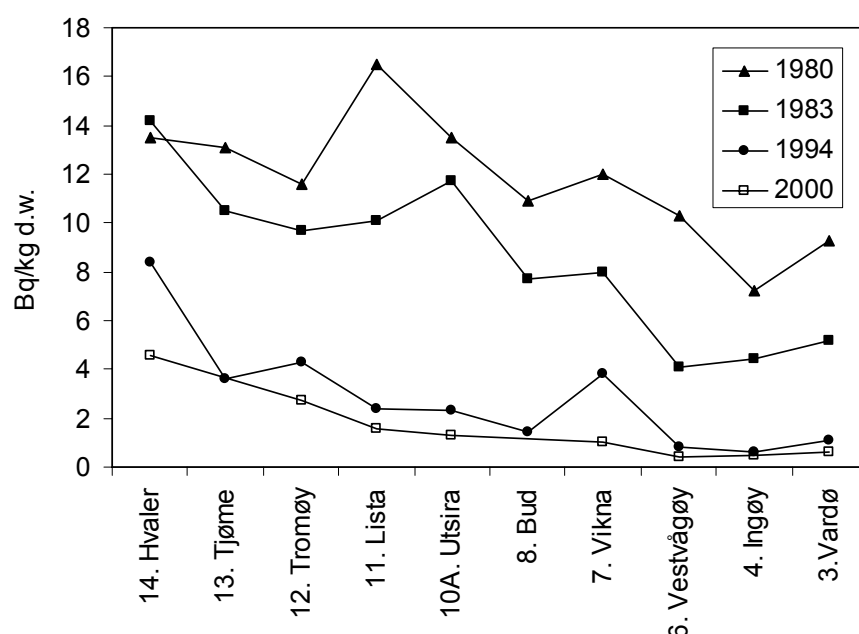


Figure 2. Concentrations of ^{137}Cs in *Fucus vesiculosus* along the Norwegian coast in 1980, 1983, 1994 and 2000.

Between Utsira and the northernmost location Ingøy, a distance of about 2000 km, the concentration decreased by a factor 2-3. The concentration of radiocaesium in the *Fucus* clearly reflected the decrease of the discharges of the reprocessing plant at Sellafield. The influence of radiocaesium from the similar plant at Cap de la Hague has been negligible. Comparison of the $^{134}\text{Cs}/^{137}\text{Cs}$ ratios in the algae and in the Sellafield discharges, indicated a transport time from Sellafield to Utsira of about 3-4 years and another 2-3 years to the Barents Sea and the Kara Sea (Christensen and Selnaes (1995), Dahlgaard et al. (1997)).

The Chernobyl accident in 1986 caused markedly increased concentrations of radiocaesium during the first 2-3 months after the accident (Figure 4). During the next 3-4 years, the activity of ^{137}Cs in the *Fucus* at the Utsira location decreased with a radioecological half-life

of about one year. In the 1990's, the ^{137}Cs concentration has shown only a small decrease, mainly due to the outflow of radiocaesium originating from the Baltic Sea.

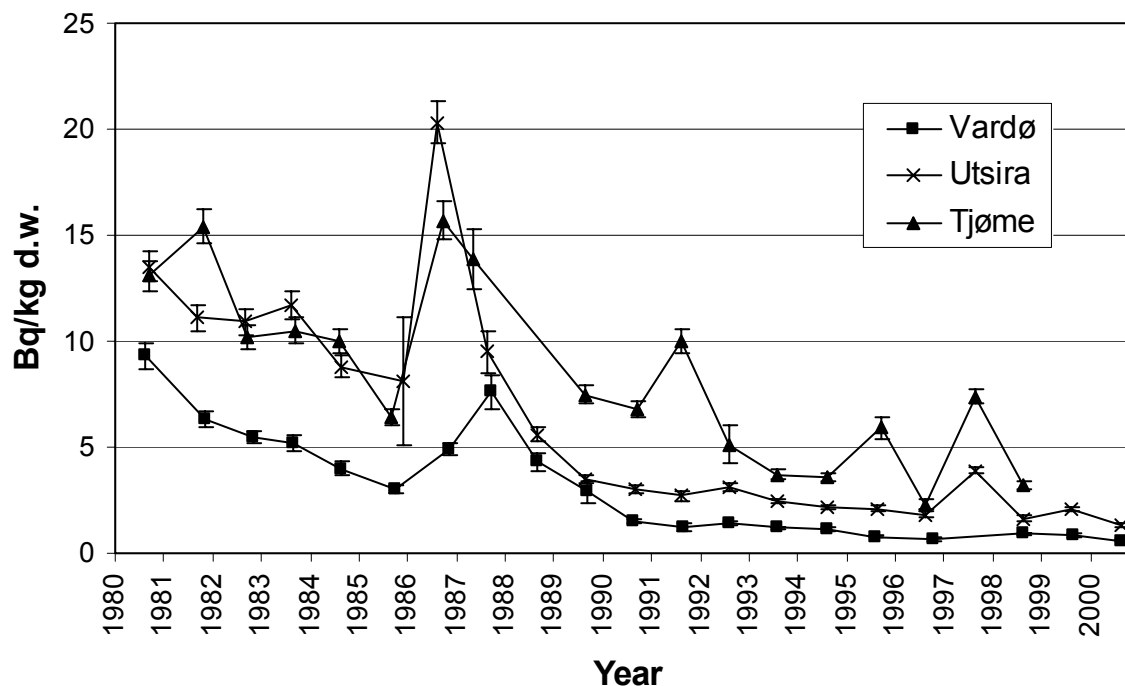


Figure 3. Concentrations of ^{137}Cs in *Fucus vesiculosus* at the sampling locations Vardø, Utsira and Tjøme. The samples were collected in August or September. Each date indicated along the time axis marks January of that year.

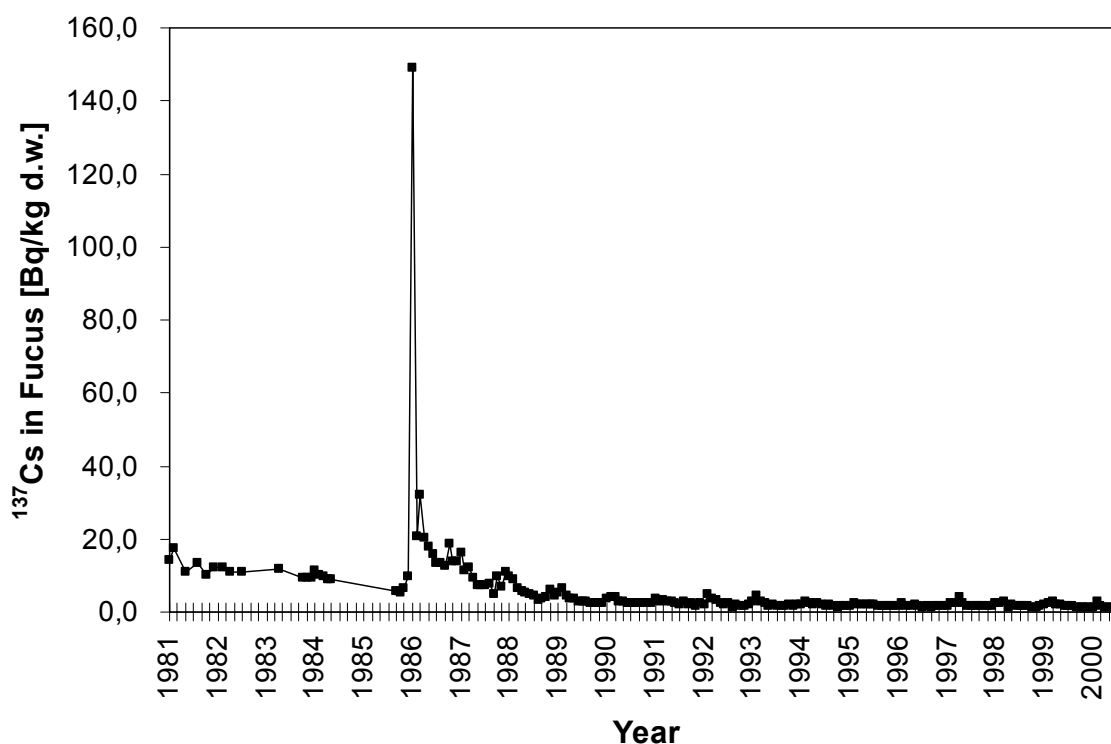


Figure 4. Concentrations of ^{137}Cs in *Fucus vesiculosus* at Utsira 1981-2000.

Fucus transfer factors

The term “transit time” has been used for the supposed maximum effect of a single discharge (Dahlgaard et al. (1997)). Having estimated a representative transit time (t years), a *Fucus* transfer factor (FTF) can be calculated as the quotient between observed concentrations in the *Fucus* ($\text{Bq kg}^{-1} \text{ d.w.}$) at the sampling site and an average discharge rate (PBq yr^{-1}) t years earlier. The unit for the FTF then becomes $\text{Bq kg}^{-1} \text{ d.w.} / \text{PBq yr}^{-1}$.

The transit time from Sellafield to the southern coast of Norway has been estimated to 3-4 years (Dahlgaard et al. (1997)). Knowledge of the annual discharge of ^{137}Cs from Sellafield year i (BNFL (1995)) allow us to calculate FTFs from Sellafield to the four locations given in Table 1 based on concentrations in *Fucus* in year $i+4$. The calculations were done for the years 1980-85 when other sources than Sellafield were negligible.

For transport of radiocaesium from the Baltic Sea, the transit time from the Kattegat (box 71 of the Risø box model (Nielsen (1998))) has in this work been estimated to be one year to the three locations Lista, Tromøya and Tjøme, and 2 years to Utsira. The annual net water outflow from the Belt Sea to the Kattegat has been estimated to 1170 km^3 (Nielsen (1998)). By using published data for the ^{137}Cs concentrations (Bq m^{-3}) in the Kattegat waters (box 71) for year i (Hermann (1998)) and measured concentrations of ^{137}Cs in *Fucus* ($\text{Bq kg}^{-1} \text{ d.w.}$) at the chosen locations for year $i+1$ or $i+2$, FTFs have been calculated. These calculations were done for the years 1990-95 when other sources than the Chernobyl fallout were negligible.

These *Fucus* transfer factors were used to calculate the contribution from the different sources compared to the total amount of radiocaesium present in the seaweed. The results are shown in Figure 5.

Table 1. *Fucus* transfer factors (FTF) in $\text{Bq kg}^{-1} \text{ d.w.} / \text{PBq yr}^{-1}$. The FTF values for Sellafield and the Baltic Sea are mean values from the first half of the 1980's and 1990's, respectively.

Sampling location	FTF Sellafield	FTF Baltic Sea
10A Utsira	3.1	35
11 Lista	3.8	38
12 Tromøya	2.8	46
13 Tjøme	3.2	59

Discussion

The amount of radiocaesium originating from Sellafield in *Fucus vesiculosus* has steadily decreased since 1980. Our calculations (Figure 5) show that in the early 1980's, approximately all the radiocaesium in the seaweed at these sampling locations originated from Sellafield. After 1990, the contribution from Sellafield is nearly negligible. Since 1986, fallout from the Chernobyl accident makes the most important contribution to radiocaesium in *Fucus*.

Based on measured ^{137}Cs concentrations of $50\text{-}60 \text{ Bq m}^{-3}$ in Kattegat sea water (Hermann (1998)) and a net annual outflow of 1170 km^3 (Nielsen (1998)), calculations show that about 60 TBq of ^{137}Cs is now transported out of the Baltic Sea annually.

The calculated *Fucus* transfer factors for the Baltic outflow do in fact also include the outflow of radiocaesium via Norwegian rivers to the Oslo Fjord, as these two sources both will influence on the concentration of ^{137}Cs in the *Fucus*. The relevant Norwegian rivers have their major catchment areas within the three counties Hedmark, Oppland and Buskerud. These counties received a total of 870 TBq ^{137}Cs in 1986 because of the Chernobyl accident (Backe et al. (1986)). Corrected for decay, this corresponds to 710 TBq in 1995. Based on investigations done in 1995, one has calculated a 1995-inventory in the upper 5 cm soil layer of 500 TBq ^{137}Cs in the same counties (Steinnes and Varskog (1998)). Thus, a total 210 TBq has "disappeared" from this soil layer during the first 9 years after the accident, giving an average annual decrease of about 23 TBq.

This decrease represents the maximum annual contribution from Norwegian rivers to the concentration of radiocaesium in the Oslo Fjord, corresponding to about one third of the annual input to these waters from the Baltic Sea. The actual contribution is probably much lower.

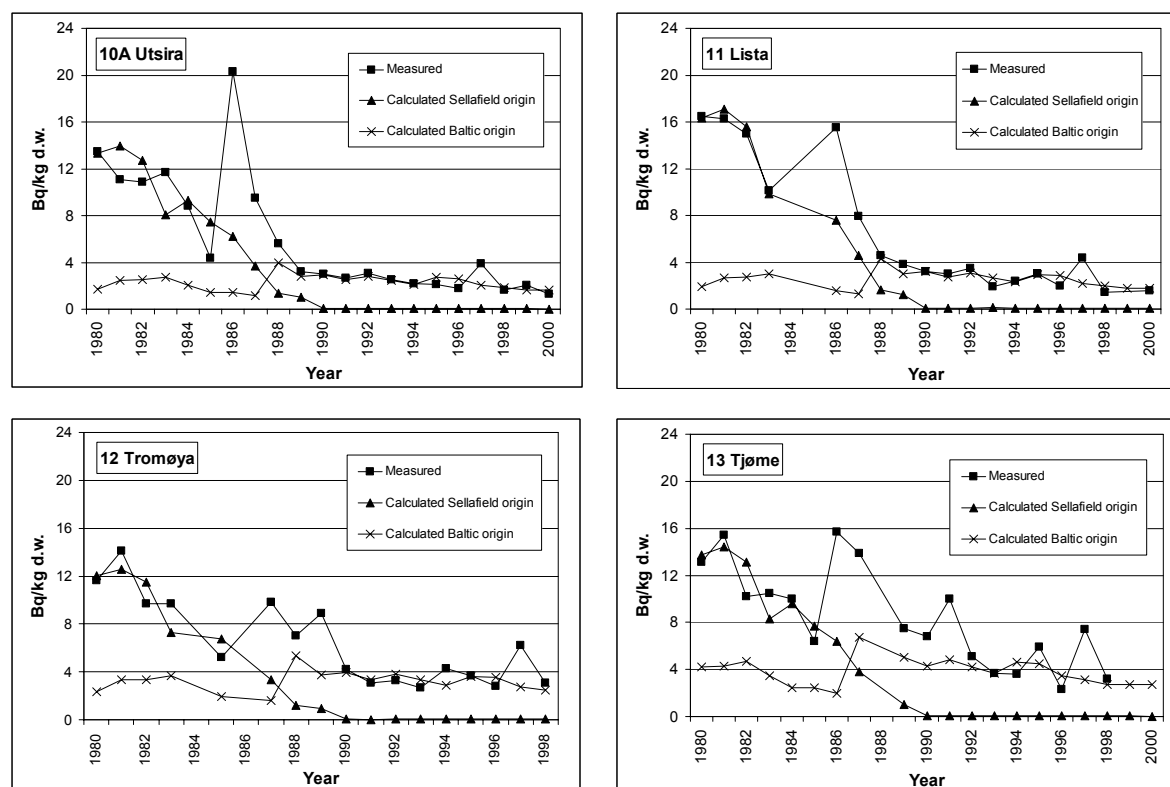


Figure 5. Calculated contributions from Sellafield and the Baltic Sea to the concentrations of ^{137}Cs in *Fucus vesiculosus* at the locations Utsira, Lista, Tromøya and Tjøme.

In 1997, a small increase in the ^{137}Cs concentrations was observed at all four locations, but in 1998, they were down to the 1996 levels. The reason for this is not known, but the summer of 1997 was unusually warm, and the seawater temperature was several degrees higher than normal from June to September.

Conclusions

Our calculations, based on measurements of ^{137}Cs in *Fucus vesiculosus*, show that at present the major source of radiocaesium in the seaweed at the coast of southern Norway is the Chernobyl fallout. The major part of this activity is due to the outflow from the Baltic Sea. A maximum of 25 % of the radiocaesium in the seaweed may originate from Norwegian rivers.

Acknowledgements

The study was financially supported by Nordic Nuclear Safety Research.

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⁹⁹Tc in Norwegian Marine Environments

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Introduction

Increasing discharges of ⁹⁹Tc from Sellafield reprocessing plant in the early 1990s have led to higher levels of ⁹⁹Tc in sea water and biota samples along the Norwegian coast. Samples of sea water were collected during expeditions to the Norwegian Sea and the Barents Sea in 1999 and activity levels of ⁹⁹Tc are reported. At the sampling stations Utsira (IFE) and Hillesøy (NRPA), monthly samples have been collected regularly since respectively January 1986 and July 1997. The objectives of the investigations are to study time variations of the ⁹⁹Tc concentrations in the algae and the sea water and to assess transfer factors and transit times, from Sellafield. Previous studies have indicated that the uptake of ⁹⁹Tc by seaweed shows a seasonal variation, with higher concentrations being observed in winter as compared to summer months. In this respect stable analogue analysis have been carried out at LAK to offer insights into the possible causes of such variations. The elements having the closest chemistry to Tc are Re, Mo, W and Ru, all of which can be determined simultaneously using ICP-MS. Variation of ⁹⁹Tc levels in *Laminaria Hyperborea* stipes and leaves with time have also been measured by using ICP-MS and AMS. An additional factor of interest in this study is the use of *Laminaria Hyperborea* in the alginate industry. Levels of ⁹⁹Tc in two alginate samples from the west coast of Norway are reported. Levels of ⁹⁹Tc in a few samples of mussels and crustacea are also reported.

Material and Methods

Sampling

The Norwegian Radiation Protection Authority, NRPA, in collaboration with The Institute of Marine Research collected sea water samples in the Norwegian Sea and in the Barents Sea in 1999. NRPA has since July 1997 been sampling sea water and seaweed samples on a monthly basis at Hillesøy in Northern Norway (see Figure 1.).

Simultaneous sampling of sea water and *Laminaria hyperborea* was carried out at three major harvesting *Laminaria* areas at Kvitsøy, Buskøy and Smøla in August/September 1998 and furthermore two alginate samples produced from *Laminaria* at Buskøy and Kvitsøy were collected and ⁹⁹Tc analysis were carried out by beta counting. All samples were obtained

from a Norwegian company involved in the commercial extraction of alginate from seaweed, FMC Biopolymer a.s. (former Pronova Biopolymer a.s.).

In addition, samples of *Laminaria Hyperborea* have been collected at monthly intervals from Kvitøy since December 1996. For a period of two years, AUN has studied the variation of the ^{99}Tc levels in stipes and leaves with time by using ICP-MS and AMS. Although samples are available up to 2001, to date only samples up to October 1998 have been analysed. IFE has annually collected the brown alga *Fucus vesiculosus* at several sampling locations since 1980 (Figure 2.), mainly in August and September (Christensen, 1986; Christensen and Selnaes, 1995). Blue mussels have been sampled every three months at Hillesøy and once per year in Grundvik. In addition, a few samples of crabs and shrimps have been collected from the Barents Sea and at the western coast of Norway in 1998 and 1999.



Figure 1 Map showing NRPA sampling sites along the Norwegian coast.

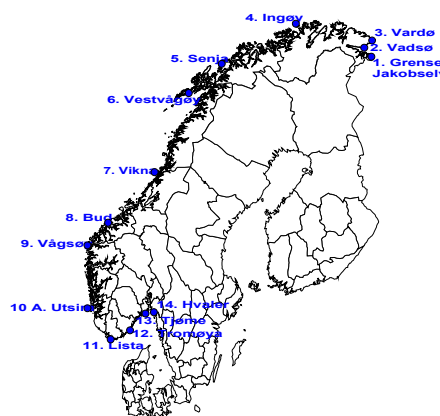


Figure 2 Map showing IFE sampling sites

Analysis

At IFE, samples of dried and homogenised *Fucus vesiculosus* have been analysed for ^{99}Tc . The pretreatment has been performed at IFE, Kjeller and the analysis at the University of Lund, Sweden, with extraction of technetium by TBP from sulphuric acid-hydrogen fluoride media. Backextraction is done from sodium hydroxide solution from which media technetium is electrodeposited onto stainless steel discs. As radiochemical yield determinant $^{99\text{m}}\text{Tc}$ is used. After decay of the yield determinant technetium is measured by an anticoincidence shielded GM counter (Holm et al., 1984).

At NRPA, normally samples of 50 litres of sea water or 10- 20 gram dried biota are analysed using a modified analytical method based on the method of Chen et al. 1990. $^{99\text{m}}\text{Tc}$ is used as a yield monitor and ^{99}Tc are electrodeposited on stainless steel discs and beta counted on low background GM-25-5 beta-counter after separation and purification (Kolstad et al., 1999).

AUN in collaboration with Rachel Carling and Philip Day from Manchester University, have analysed samples for ^{99}Tc by ICP-MS and AMS. Subsamples of the monthly collections of *Laminaria* at Kvitøy were dried at 60°C , ground and portions of approximately 1g were prepared for analysis by AMS and ICP-MS (see accompanying report). The ICP-MS instrument at the Agricultural University of Norway, LAK, is an Elan 6000 Perkin Elmer fitted with 4 different sample introduction techniques. Work carried out in 1998 and 1999 has

focused primarily on the electrothermal vapourisation, ETV, system. It requires low sample volumes (30-60µl) and determination limits of 20µBq ^{99}Tc has been achieved for this system.

Results and Discussion

Barents Sea- and Norwegian Sea Cruises in 1999

The results are shown in Figure 3. and Table 1. The results reflect the general circulation of water masses in the Norwegian Sea and the Barents Sea, resulting in elevated levels of ^{99}Tc along the coast.

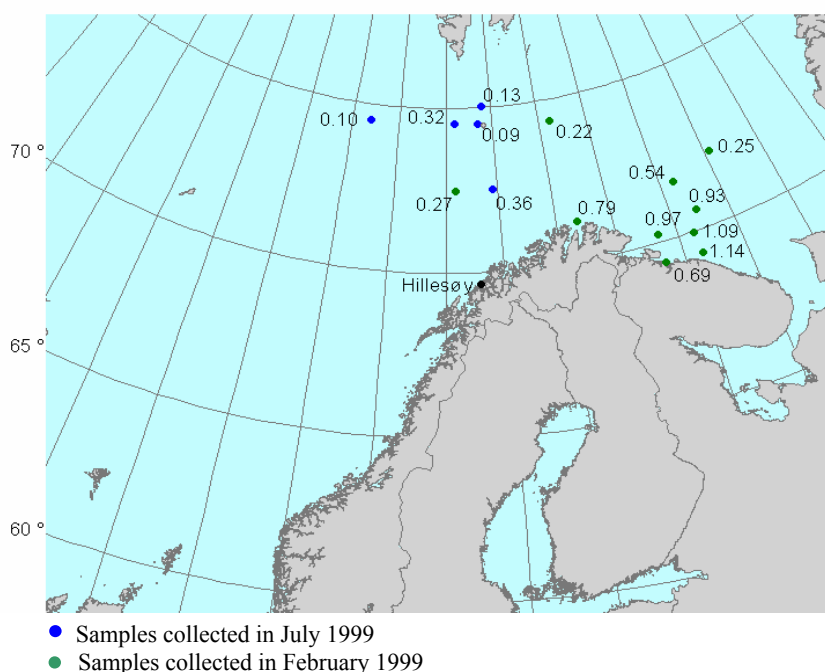


Fig.3. Levels of ^{99}Tc (Bq m⁻³) in sea water for the Barents Sea and the Norwegian Sea respectively in February and July 1999.

At two stations the vertical distribution of ^{99}Tc demonstrated that concentrations were higher in surface water than in near-bottom-water (Table 1.).

Table 1. Levels of ^{99}Tc (Bq m⁻³) in surface- and bottom water from station 3 and 6.

Station	Sampling place	Salinity (‰)	^{99}Tc -activity (Bq m ⁻³)
3. surface	69°56.26' N, 36°21.37'Ø	34.63	0.97 ±0.08
3. bottom	- "-	34.62	0.54 ±0.04
6. surface	71°35.79' N, 36°16.26'Ø	34.80	0.52 ±0.04
6. bottom	- "-	34.87	< 0.100

Utsira

The results for ^{99}Tc in the *Fucus* samples from Utsira from January 1995 to October 2000 are shown in Figure 4. The discharges of ^{99}Tc from the EARP plant at Sellafield started late 1994. It is evident from the results that the discharge *front* has reached Utsira waters already in the winter 1996/97. This is faster than the *average* transit time of 3-4 years calculated for ^{137}Cs (Dahlgard et al., 1997). The discharges reached a peak in 1995/96 as shown

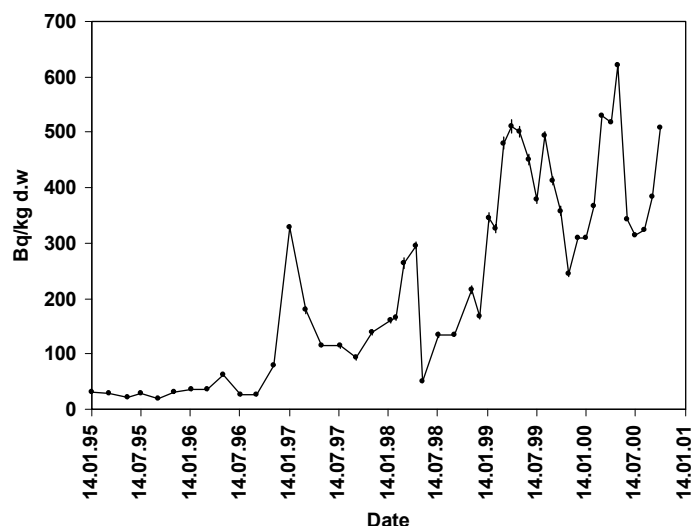


Figure 4. ^{99}Tc in *Fucus vesiculosus* at Utsira January 1995- October 2000.

in Figure 5. (BNFL, 1995, Dahlgard, 2001). If the transit time for ^{99}Tc is shorter than for ^{137}Cs , which one might expect because Tc behaves more conservatively than Cs, then the average *Fucus* concentration of ^{99}Tc should be lower in 2000 than in 1999. The results showed however that the concentration increased from 1998 to 1999 and then remained at the same level in 2000, as shown in Figure 6. The average concentrations in 1998, 1999 and 2000 were 176 (73), 401 (87) and 422 (112) Bq kg⁻¹ d.w. respectively (standard deviations are shown in brackets). If the transit times are roughly equal (3-4 years), one should expect a decrease in the concentration in 2001.

The results for 1997 and 1998 show an annual variation in the ^{99}Tc concentration with a peak in the winter. The lower concentrations in the summer season may be explained with dilution effect because of growth. In comparison, ^{137}Cs has a concentration peak in the summer season, and therefore one might expect that some different biophysical/-chemical mechanisms are governing this behaviour. For the years 1999 and 2000 the results are however different. The trend seen in 1997-1998 is not found here. As shown in Figure 6. the years 1995-2000 show no clear seasonal variation trend. The observed variations may instead be related to varying seawater concentrations due to large variations in the monthly Sellafield discharges. This will be the object of further studies. The results of the stable analogues study described below seems however to support this.

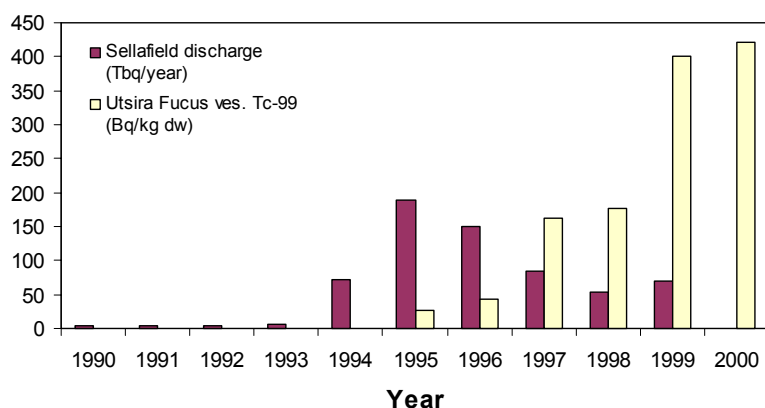


Figure 5. Total annual ^{99}Tc discharges from Sellafield 1990-1999 and annual average ^{99}Tc concentrations in *Fucus vesiculosus* at Utsira 1995-2000

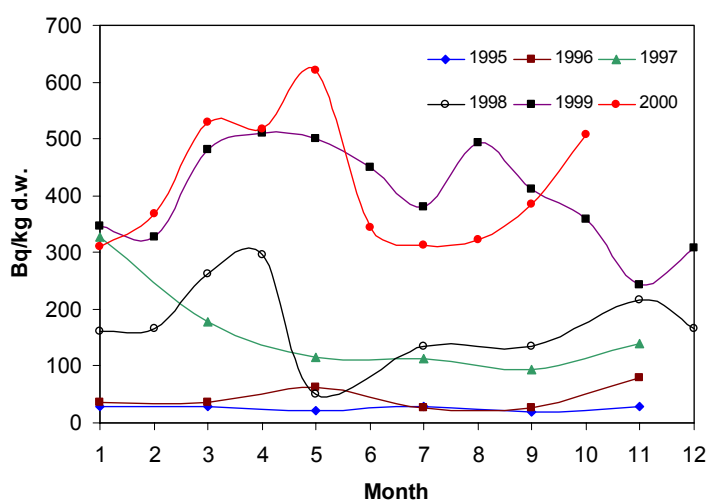


Figure 6. Annual variation of ^{99}Tc -concentration in *Fucus vesiculosus* at Utsira, 1995-2000

Hillesøy, Troms

The variation in the ^{99}Tc -level for *Fucus vesiculosus* is shown in Figure 7. An increasing trend of the concentrations of ^{99}Tc in *Fucus vesiculosus* at Hillesøy has been observed, from about 60 Bq kg^{-1} (d.w.) in July 1997 to a maximum of 393 Bq kg^{-1} in May 2000. Substantial variations are observed, and there seem to be a seasonal variation with higher concentrations of ^{99}Tc in winter and spring seasons.

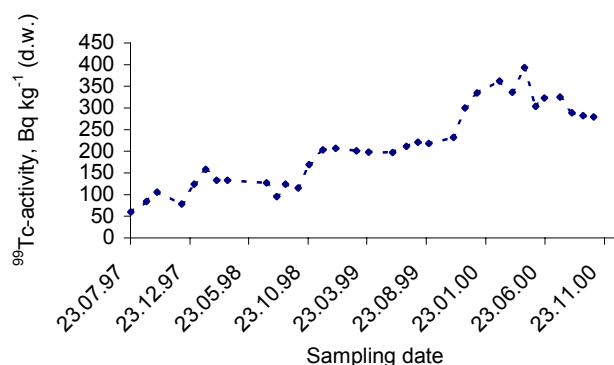


Figure 7. Concentrations of ⁹⁹Tc in *Fucus vesiculosus* at Hillesøy.

Use of Stable Analogues (Re, Mo, W) to study temporal variations in ⁹⁹Tc uptake in Seaweed

The hypothesis is that physiologically based factors influencing the variability of ⁹⁹Tc such as spring growth would be reflected in analogue seasonality, whereas variations due to ⁹⁹Tc concentrations in seawater and ocean currents would not. Figure 8. shows ICP-MS measurements on a series of seaweed (2 years) collected from Hillesøy. Analyses were carried out on 0.5 g microwave digest of the seaweed. Interestingly the results show that Re (the closest analogue to Tc) shows a slight seasonal variation, yet with peaks in the summer rather than winter months as observed for ⁹⁹Tc. Explanations could include a different uptake or accumulation mechanism for Re and Tc in seaweed, competition between Re and Tc or that the ⁹⁹Tc variability is more a reflection of seawater concentrations than physiological factors. Further samples are under analysis to confirm the observations.

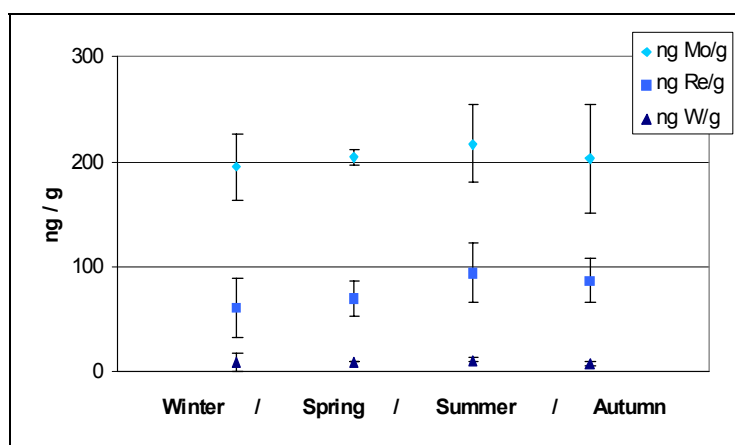


Figure 8. ICP-MS determination of Re, W, Mn and Mo in a series of seaweed from Hillesøy.

Transit times and transfer factor

The sea water concentrations increased from a level of 0.45 Bq m^{-3} in July 1997 to a maximum of 2.0 Bq m^{-3} in January 2001 (Figure 9.). In order to determine transit times and transfer factors cross correlation analysis has been utilised (Brown et al., July 1999, Brown et al., September 1999). The technique basically involves the comparison of ^{99}Tc discharge data with the temporal trend of ^{99}Tc activity concentrations in water, for a range of transit times, until a close match is found. Monthly discharge data have been provided by BNFL. These data have been subsequently detrended or smoothed to partly account for the dispersion of the Sellafield ^{99}Tc signal. The closest match was produced when the smoothing time approached 9 months. Data up to and including December 1998 have been used in this particular analysis. Regional levels of ^{99}Tc in sea water in the early 1990s,

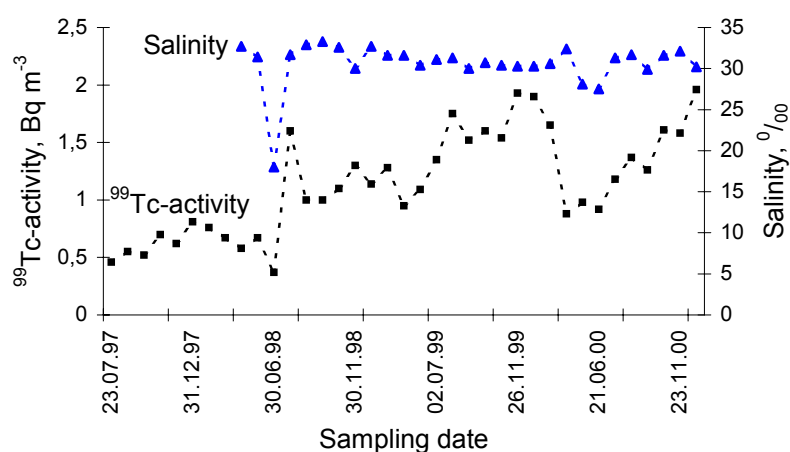


Figure 9. Graph showing - (i) activity concentration of ^{99}Tc (Bq m^{-3}), (ii) salinity in sea water at Hillesøy in northern Norway.

derived from global fallout and earlier releases from western European nuclear reprocessing operations, are believed to have fallen between $0.2\text{-}0.4 \text{ Bq m}^{-3}$. This was the range assumed in the present study. Recent information has shown that regional ^{99}Tc concentrations in 1994 for the Northern Norwegian Coastal Current and Western Barents were actually in the range $0.03\text{-}0.14$ (Kershaw et al., 1999).

A best-fit transit time of 42 months (3 years, 6 months), with an estimated uncertainty of ± 9 months commensurate with the smoothing function, combined with a best-fit transfer factor of 0.07 Bq m^{-3} per TBq month^{-1} (or 6 Bq m^{-3} per PBq a^{-1} assuming steady state) has been derived. The transit time derived from this study falls within the range reported by Dahlgaard (1995), who reported a transit time of 3-4 years from Sellafield to the North-west Norwegian current for conservative radionuclides. The ^{99}Tc transfer factor calculated in the present study falls below a corresponding Transfer factor range for a generic conservative radionuclide reported in the same article, i.e. 6 Bq m^{-3} per PBq a^{-1} in the present study compared with $10\text{-}50 \text{ Bq m}^{-3}$ per PBq a^{-1} reported by Dahlgaard (1995). The degree to which the Sellafield discharges are reflected in sea water in north Norway are presented in Figure 10.

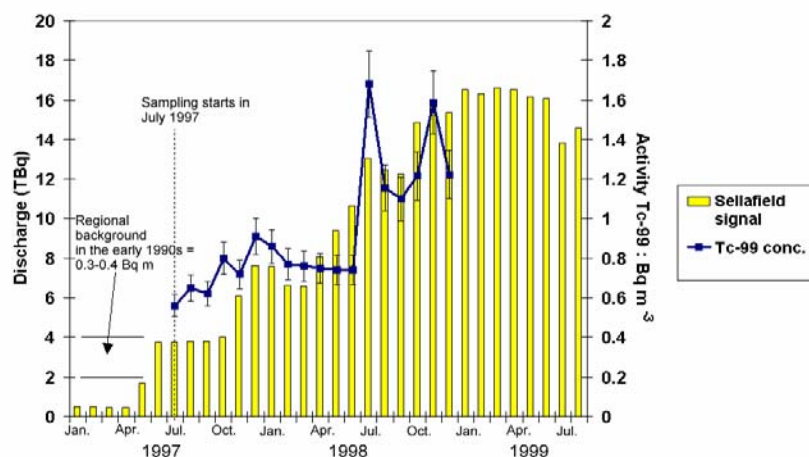


Figure 10. A comparison of «smoothed» ^{99}Tc discharges from Sellafield with ^{99}Tc activity concentrations (Bq m^{-3}) in sea water at Hillesøy.

Kvitsøy, Buskøy and Smøla

The purpose of this study was firstly to study the transport of ^{99}Tc from Sellafield discharges using time series data of ^{99}Tc in *Laminaria*, secondly to study the accumulation and temporal trends of ^{99}Tc in the stems and leaves of this seaweed species, and the possible transfer of ^{99}Tc by this route to the human food chain. Alginates are used in the food industry as low price thickeners for products such as ketchup, mayonnaise, ice creams, soups, puddings and mashed potato. Two samples of alginate from Kvitsøy and Buskøy showed low levels of ^{99}Tc below 1 Bq/kg (d.w.). The observed ^{99}Tc concentrations in *Laminaria Hyperborea* from the Kvitsøy, Buskøy and Smøla are in the range $20\text{--}72 \text{ Bq/kg}$ (d.w.), corresponding to concentration factors of about $7,000\text{--}9,000 \text{ Bq kg}^{-1}(\text{d.w.})/\text{Bq l}^{-1}$ for the leaves and $11,000\text{--}17,000 \text{ Bq kg}^{-1}(\text{d.w.})/\text{Bq l}^{-1}$ for the stems.

There are several conclusions, which can be drawn from Figure 11. Firstly, the concentration factor of *Laminaria Hyperborea* for technetium is comparable to that of other brown seaweeds. The concentration of ^{99}Tc in *F. vesiculosus* from Stavanger in Norway was determined to be 172 Bq kg^{-1} dry weight in May 1998. For *Laminaria Hyperborea* the average concentration (stipes and leaves) in May 1998 was 54.6 Bq kg^{-1} dry weight, which is approximately one third that of *F. vesiculosus* and would correspond to a concentration factor in the order of 2.5×10^4 .

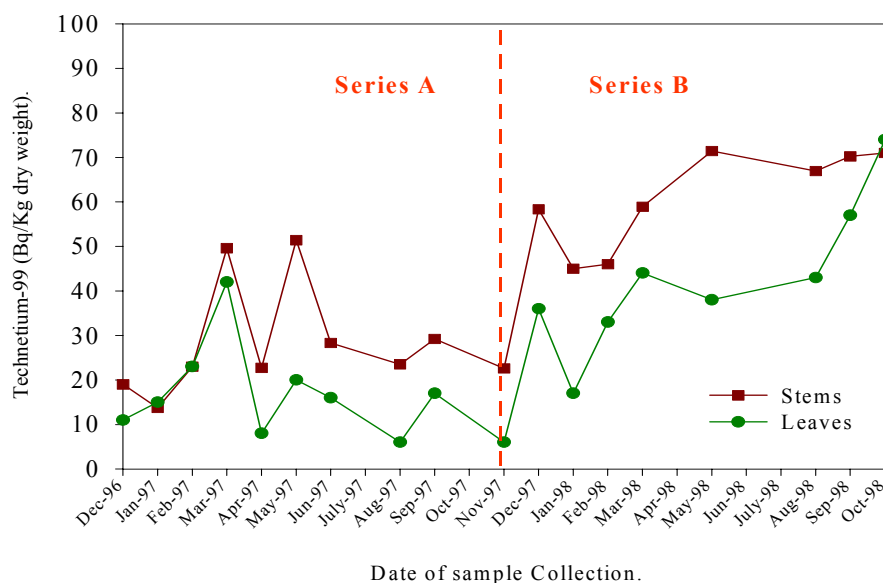


Figure 11. The Variation of ^{99}Tc in *Laminaria Hyperborea* Stipes and Leaves with time.

The distribution of technetium in the seaweed is clearly not homogenous with the stipes containing more technetium than the leaves. There are two possible explanations for this. First, the concentration of technetium in seaweed may exhibit a seasonal fluctuation due to a “dilution” effect from new growth in the spring. Unfortunately, in this case, any seasonal fluctuation is probably masked by the overall upward trend in concentrations due to the Sellafield discharges. Furthermore, any such trend would have been difficult to interpret due to the fact that the time series is incomplete. However, the “dilution” theory may be a possible explanation for the consistent difference in concentration between a given pair of leaves and stipes. With the exception of the results for May 1998, which may not be real, the pattern of variability is replicated in both the leaves and stipes with the stipes exhibiting a consistently higher concentration of ^{99}Tc . The second explanation, and probably the more likely one, is that the reproductive cells contained in the stipes accumulate technetium to a greater extent than the leaf tissues. This would agree with the observation by Bonotto that technetium is more rapidly accumulated by the parts of the plant which bear reproductive cells.

There appears to be an overall upward trend in the amount of ^{99}Tc present in *Laminaria Hyperborea*. Prior to November 1997 (data set, Series A) the average value of ^{99}Tc in both the leaves and stipes is significantly ($p < 0.01$) lower than that post December 1997 (data set, Series B). The average value of ^{99}Tc in the leaves increased by a factor of 2.5 between Series A and B, whereas the stipes increased by a factor of just over 2. Between December 1996 and October 1998 the concentration of ^{99}Tc increased almost seven fold in the leaves and almost four fold in the stipes.

However, the transit time is defined as the time period between a specific discharge event and the occurrence downstream of the maximum contribution of that event. The time series ended in October 1998 and at that point the levels of technetium did not appear to have reached their peak. The discharge of ^{99}Tc was continuously high from January 1995 to September 1996, so to ensure a good estimate of the transit time the time series should be extended up until the levels of technetium begin to fall. This would provide a far clearer indication of the transit time. However, in this case the maximum event has not yet occurred

and only the sharp edge of the increase is present, but based on this evidence alone, the data indicate that the discharges which occurred at Sellafield in the first quarter of 1995 had reached Stavanger by December 1997, just three years later.

Reports in the literature indicate that the transit time to the Norwegian Coastal Current is in the order of 3 to 4 years (Brown et al, 1998; Dahlgaard et al 1997). The Utsira data will probably confirm this.

The transit time for ^{99}Tc , from Sellafield to the Norwegian coastal current, is estimated to be about 3 years from the results of the Laminaria study. This value is reasonably similar to that obtained by other people. The transit times estimated from technetium data are generally faster than those estimated using caesium because caesium behaves less conservatively than technetium. However, transit times depend upon a variety of factors some of which do not remain constant. Climatic changes can affect coastal currents, which in turn can influence the preferred route and rate of transport. Mixing with additional sources of contaminated water can also occur.

^{99}Tc in mussels and crustacea.

A few samples of lobsters, shrimps, crabs and blue mussels have been analysed for ^{99}Tc . The results are given in table 4. The highest concentrations are seen in lobster, in agreement with earlier observations from the Irish Sea.

Table 4. Levels of ^{99}Tc in Mussels and Crustacea (soft parts)

Sample	Sampling area	Date	^{99}Tc Bq kg ⁻¹ , dry weight	^{99}Tc Bq kg ⁻¹ , wet weight
Shrimps (<i>Pandalus borealis</i>)	Rogaland	30.01.98	0.49 ± 0.03	0.11 ± 0.01
Shrimps	Harøyfjord	13.02.98	0.48 ± 0.03	0.11 ± 0.01
Shrimps	Barents Sea	Feb.99	*bdl	bdl
Shrimps	Barents Sea	Feb.99	bdl	bdl
Blue mussels	Grundvik	26.11.98	3.2 ± 0.2	0.55 ± 0.03
Blue mussels (<i>Mytilus edulis</i>)	Hillesøy	1.02.98	1.90 ± 0.11	0.44 ± 0.03
Crab (<i>Brachyura</i>)	Nordfjord	23.02.98	8.4 ± 0.5	1.10 ± 0.07
Crab	Nordfjord	23.02.98	8.7 ± 0.5	0.95 ± 0.05
Crab	Barents Sea	Feb. 99	bdl.	bdl
Lobster (m., <i>Homarus gammarus</i>)	Ytre Oslofj.	15.11.98	76.3 ± 3.8	14.4 ± 0.9
Lobster (f.m.)	Ytre Oslofj.	15.11.98	140 ± 7	26.2 ± 1.6

*bdl – below detection limit

Conclusions

In February and July 1999, the ^{99}Tc concentrations varied between 0.09 Bq m⁻³ and 1.14 Bq m⁻³ in the Barents Sea and in the Norwegian Sea. The highest levels of ^{99}Tc were observed in the eastern part of the Seas. The results reflect the general circulation of water masses in the Norwegian Sea and the Barents Sea.

It is evident from the seaweed data from Utsira that the discharge *front* from Sellafield reached Utsira waters already in the winter 1996/97. This is in agreement with seawater data from late 1996 and 1997 (Brown et al, 1998). Sea water data from Hillesøy show a best-fit transit time of 42 months (3 years, 6 months), with an estimated uncertainty of ± 9 months. The transit time for ^{99}Tc , from Sellafield to the Norwegian coastal current (Kvitsøy), is estimated to be about 3 years from the results of the *Laminaria Hyperborea* study. The Utsira data indicates a longer transit time, probably 3-4 years.

The distribution of technetium in *Laminaria Hyperborea* is clearly not homogenous with the stipes containing more technetium than the leaves. The observed ^{99}Tc concentrations in *Laminaria Hyperborea* from the Kvitsøy, Buskøy and Smøla are in the range 20-72 Bq kg^{-1} (d.w.), corresponding to concentration factors of about 7,000- 9,000 Bq kg^{-1} (d.w.)/ Bq l^{-1} for the leaves and 11,000-17,000 Bq kg^{-1} (d.w.)/ Bq l^{-1} for the stems.

In 1997 and 1998, the seaweed results from Utsira show an annual variation in the ^{99}Tc concentration with a peak in the winter, but for the years 1999 and 2000 the results are however different. The observed variations may instead be related to varying seawater concentrations due to large variations in the monthly Sellafield discharges. The results of the stable analogues study for seaweed show that Re (the closest analogue to Tc) shows a slight seasonal variation, yet with peaks in the summer rather than winter months as observed for ^{99}Tc . At Hillesøy substantial variations are observed, and there seem to be a seasonal variation with higher concentrations of ^{99}Tc in winter and spring seasons.

Acknowledgements

In addition to the support from the NKS BOK 2.2 project, the Ministry of Environment in Norway has supported this work financially.

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Cs-137 and Tc-99 in the ocean and at the shores around Iceland related to timing of fluxes in the Nordic Seas

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Introduction

Watermasses

Iceland is located on the Greenland-Scotland Ridge; the boundaries of the North Atlantic and the Nordic Seas. Northwest of Iceland, boundaries are found between relatively warm and saline Atlantic water (AW) of the Irminger Current (a branch of the North Atlantic Current System) and the cold, low-saline Polar water (PW) of the East Greenland Current (Fig. 1). Between Iceland and the Faroes Atlantic water meets Arctic waters including the Arctic surface water (ASW) of the cold East Icelandic Current. The cold currents from the north originate in the Arctic (Polar water of the East Greenland Current) but are influenced by the eastern Arctic intermediate water (AIW) and the Arctic surface water (ASW) of the northflowing warm branches of the Norwegian Current. The PW thus brings properties mainly from the Arctic and the AIW and ASW from the eastern part of the Nordic Seas (Norwegian Sea) and West European waters. The warm Irminger Current on the other hand brings properties into Icelandic waters from the open ocean to the South and Southwest of Iceland and further into North Icelandic waters (Stefánsson and Ólafsson 1991, Stefánsson 1994, Valdimarsson and Malmberg 1999, Malmberg 2002). There the Atlantic water (AW) mixes with the waters from the north (PW, AIW, ASW) and forms in winter a new water mass, North Icelandic Winter water (NIW) (Stefánsson 1962).

All the above water masses are bound to the upper layers of the Nordic Seas, but they are overlaying cold deep water masses (DW) of different origin. The upper layer of the DW flows from the Nordic Seas across the submarine ridges into the North Atlantic (so-called "overflow"). This flow through the Denmark Strait, more or less influenced by intermediate water masses, called Denmark Strait overflow water (DSOW), contributes to the deep water of the western North Atlantic. In Table 1 the main water masses in the seas around Iceland and their temperature/salinity (t/s) properties are listed.

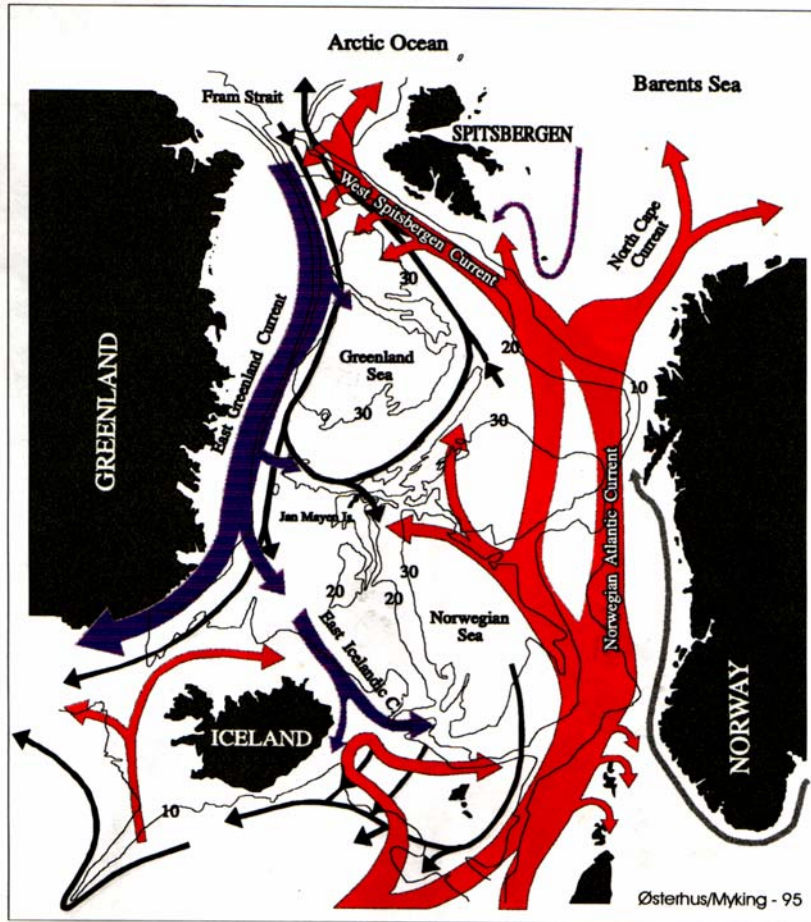


Figure 1. Ocean currents in Icelandic waters and in the Arctic.

All the above water masses are bound to the upper layers of the Nordic Seas, but they are overlaying cold deep water masses (DW) of different origin. The upper layer of the DW flows from the Nordic Seas across the submarine ridges into the North Atlantic (so-called "overflow"). This flow through the Denmark Strait, more or less influenced by intermediate water masses, called Denmark Strait overflow water (DSOW), contributes to the deep water of the western North Atlantic. In Table 1 the main water masses in the seas around Iceland and their temperature/salinity (t/s) properties are listed.

Table 1. Properties of the different Icelandic water masses sampled.

Water masses in Icelandic waters	t	s
Atlantic water (AW)	3-6 °C	> 34.9
Polar water (PW)	< 0°C	< 34.4
Arctic intermediate water (AIW)	0-2°C	34.8-35.0
Arctic surface water (ASW)	1-3°C	34.6-34.8
Deep Water (DW)	< 0°C	34.89-34.93
Denmark Strait overflow water (DSOW)	< 1°C	34.8-34.9
North Icelandic winter water /North Icelandic shelf area (NIW)	2-3°C	34.8-34.9

Radionuclides

Cs-137 (radiocaesium) has a relatively long half-life and carries chemical characteristics identical to K (potassium). It therefore enters the biosphere easily. Gamma rays are emitted in the course of the decay of Cs-137, which makes it easy to measure. The main source of Cs-137 in the Icelandic marine environment is fallout from atmospheric nuclear weapons testing in the 1950's and 1960's. The concentration of Cs-137 in seawater is higher north and east of Iceland than south and west. This is mainly due to liquid discharges into the Irish Sea from the Sellafield plant in the U.K. which is the main contributor of the West European reprocessing plants. In recent years, the Sellafield plant has substantially reduced liquid discharges, particularly of Cs-137 (Fig. 2).

Tc-99 is a soft beta-emitter with a half-life of 213 000 years. It is formed in significant quantities by the fission processes that occur in nuclear reactors. In 1994 liquid discharges of Tc-99 from the nuclear facilities in Sellafield were greatly increased (Fig. 1) in 1994. This increase may reveal its appearance as higher Tc-99 values in North Icelandic waters in near-by times.

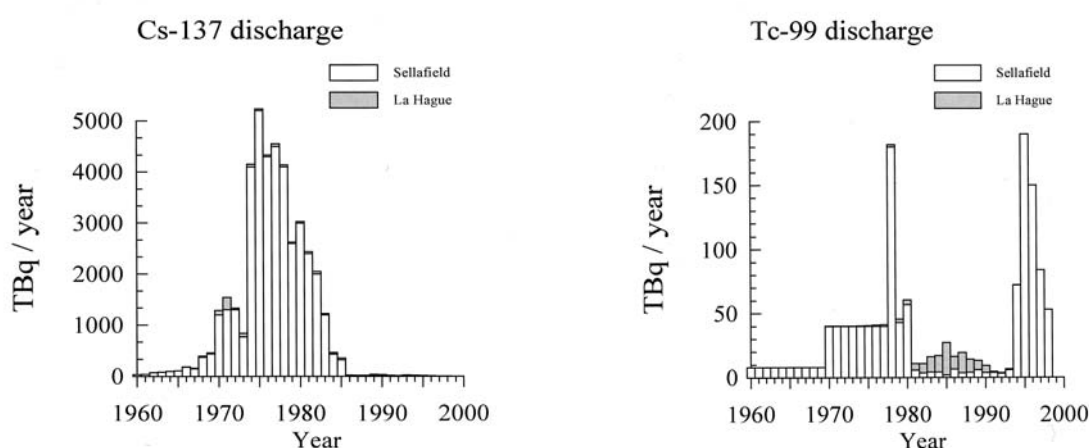


Figure 2. Discharges of Cs-137 and Tc-99 from the Sellafield nuclear reprocessing plant (obtained from Dahlgard, H.D., personal communication).

Material and Methods

Sampling

The research vessel *Bjarni Sæmundsson*, owned by the Marine Research Institute in Reykjavík (MRI) was used for collecting seawater samples. Nearly all the samples were taken from standard sections (Fig. 3). Most of the samples collected were taken from a depth of 4-5 meters with a pump from the ship. Some samples from sub-surface and deep water were taken as well. Due to low concentration of Cs-137, large samples of 100 - 200 litres were used for radionuclide studies.

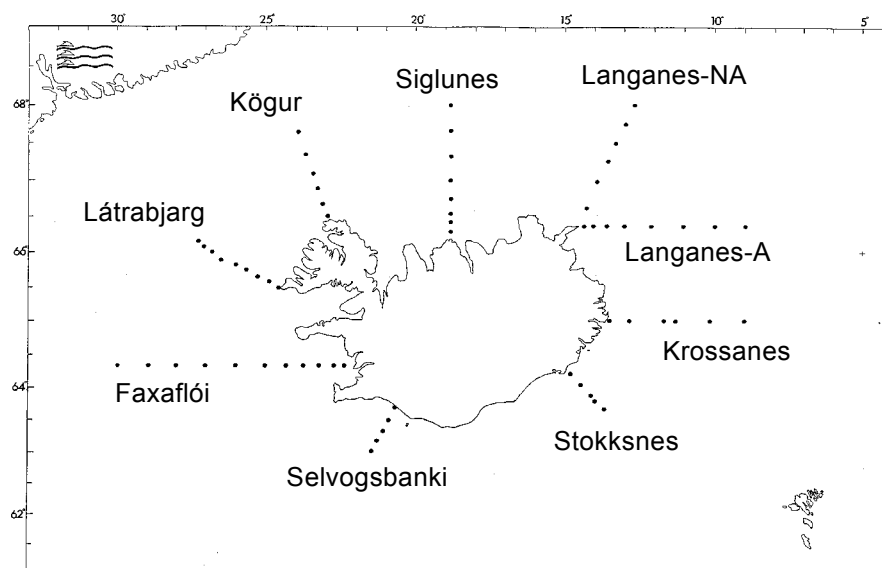


Figure 3. Standard sections for collecting seawater samples by MRI. Stations within each section have a serial number starting with one closest to shore.

Seaweed for Cs-137 analyses has been sampled at six different locations (Table 2, Fig. 4) in Iceland since 1989. Sampling was carried out four times per year (in March, June, September and December). The sampling frequency was decreased to once per year (in September) 1993-1998, but the same locations were used. In the beginning of 1999 the sampling frequency was increased again to four times per year. The reason for the increased sampling frequency in the beginning of 1999 was the decision to start measuring Tc-99 in seaweed. The most recent samples are being processed, measured and analysed.

Table 2. Sampling locations for seaweed in Iceland.

Location	Longitude N	Latitude W	Species
Ólafsvík	64°54'	23°56'	<i>Ascophyllum nodosum</i>
Ísafjörður	66°06'	23°02'	<i>Fucus vesiculosus</i>
Grímsey	66°33'	18°02'	<i>Fucus vesiculosus</i>
Fáskrúðsfjörður	64°54'	13°55'	<i>Ascophyllum nodosum</i>
Stokksnes	64°14'	14°59'	<i>Fucus vesiculosus</i>
Vestmannaeyjar	63°25'	20°17'	<i>Fucus vesiculosus</i>

Samples of different species of fish; cod, herring and dab, have been taken since 1989. Those samples were collected at different fishing grounds around the island.

Radionuclide analyses of seawater, seaweed and fish from 1997-2000 have been compiled and published (Sigurgeirsson *et al.* 2001).

Analysis

The concentration of Cs-137 in each sample was measured by a gamma spectrometry, using a HPGe- gamma spectrometric system. Samples of seawater were prepared by using standard

radiochemical techniques (AMP-method). Samples of seaweed were dried at 105°C and grounded before counting. Each fish sample was made up from sub-samples of flesh from 25 individuals. The flesh was minced and one kg from each sample measured.

Results and Discussion

Cs-137 in Seawater

All samples were divided into water masses based on the criteria outlined above. A few samples did not fit with any of the water mass definitions in Table 1 (e.g. a mixture of two or more water masses). Those samples were not included in these results.

Table 3. Cs-137 in different water masses around Iceland in 1990-2001

Water mass	Section	1990-2001		Values from 1990-1997*	
		Cs-137 Geometric mean (Bq/m ³)	Number of measurements	Cs-137 Geometric mean (Bq/m ³)	Number of measurements
AW		2,7	44	2,7	44
AIW		3,3	9	3,3	3
ASW		4,1	15	4,3	20
DW		2,2	1	2,2	1
DSOW		3,3	8	4,1	3
NIW		3,4	5	3,3	8
PW	a	7,1	26	7,3	16
	b	5,5	12	5,6	4
	c	4,1	4	4,2	2
	d	3,5	7	3,2	4

* Ólafsdóttir, E. D. *et al.* 1999

Compared to earlier data on Cs-137 in seawater (Ólafsdóttir *et al.* 1999) no significant changes were encountered in the 1998-2000 data.

Analysis of variance showed a significantly greater variation in Cs-137 values between water masses than could be expected at random ($P = 5,2 \cdot 10^{-18}$). Within each water mass, no significant variation between sections could be observed except in the case of Polar Water (PW) ($P = 1,2 \cdot 10^{-6}$). Parameters such as seasons, salinity and temperature do not explain the variation within each water mass to a significant degree.

No obvious correlation with time was observed for any of the water mass types except for DSOW (Fig. 4).

The geometric mean for DSOW is 4,1 Bq/m³ for the years 1991-1997 compared to 3.3 Bq/m³ for 1991-2001. This delayed response is in agreement with the description of fluxes and currents given in this paper.

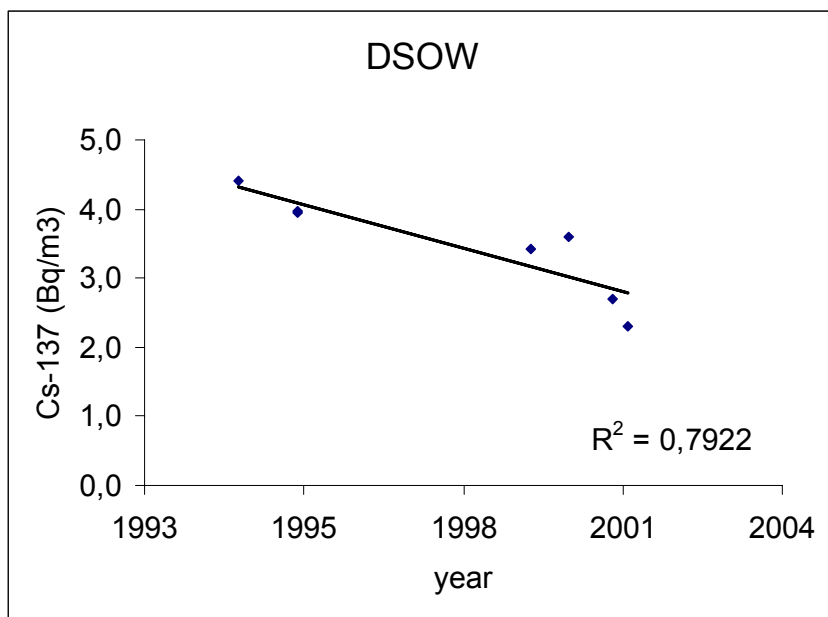


Figure 4. Cs-137 in DSOW North West of Iceland in the years 1994-2001.

The lowering in Cs-137 values is expected to be detected first in AIW. Dahlgaard (1994) gives the value 5,5 and 5,2 Bq/m³ for AIW in 1988 and 1990. The measured concentration 1995-2001 seem to be stable (3,3 Bq/m³). This indicates that the decrease in Sellafield discharges 1975-1985 (Fig. 1) had fully appeared in AIW in 1995. This is in accordance with transit time from Irish Sea to Icelandic waters.

Cs-137 in Seaweed

Results for seaweed are shown in Figure 5. Analyses of variance of Cs-137 in seaweed samples, shows a statistically significant difference between values from different sampling stations ($P = 4,1 \cdot 10^{-6}$).

Figure 5. Cs-137 in seaweed at the Icelandic shore 1989-2001.



Samples of *Fucus vesiculosus* and *Ascophyllum nodosum* taken at same time in Ólafsvík show same values of Cs-137 (0,13 Bq/kg d.w.). No correlation was obtained for variance in Cs-137 values at each sampling station for salinity or season. A significant decrease in Cs-137 values is obtained in the years 1989-1992 (Fig. 6). This is in agreement with the reduction in release in 1975-1985 (Fig. 2) corresponding to a delayed response of about 8 years.

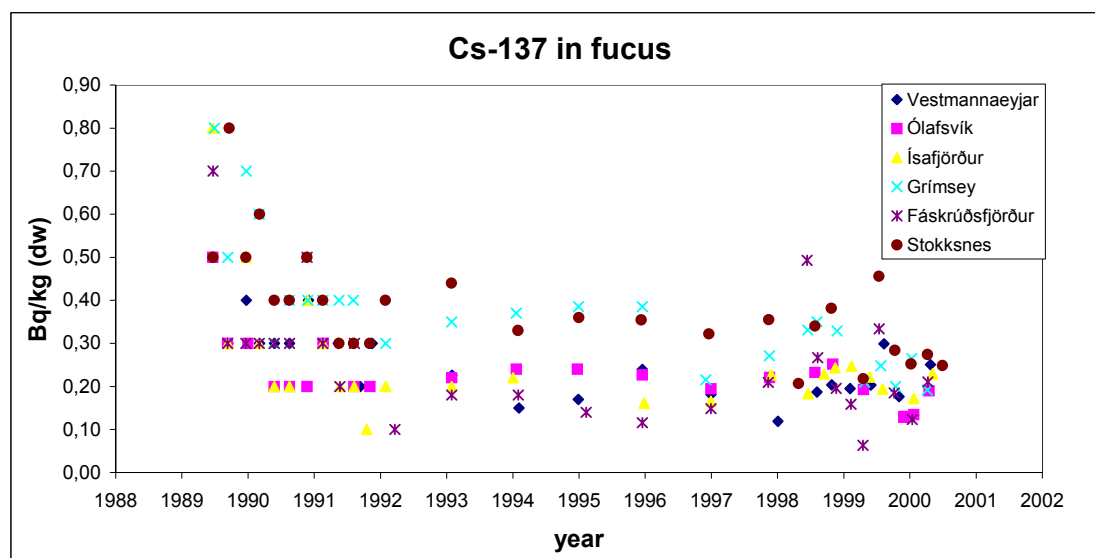


Figure 6. Cs-137 in seaweed at the shore around Iceland in 1989-2001.

Cs-137 in Fish

A few samples of fish have been measured since 1989. The values are very low (Table 4). These results show nearly twice as high values for cod than for dab.

Table 4. Cs-137 in fish from Icelandic fishing grounds

Type	Cs-137 Bq /kg	Number of sub-samples
Cod	0,23	14
Dab	0,13	10
Herring	0,13	1

Tc-99 in Seaweed

Samples of seaweed collected in 1998 - 2000 have been analysed for Tc-99 at Geislavarnir. These show similar values for Tc-99 compared to values of samples from 1990-1992 measured at Risø for Vestmannaeyjar. Similar results show higher values at Grímsey in 1998-2000 compared to 1990-1992. This could be accordance with increasing discharges of Tc-99 from Sellafield, increasing rapidly in 1994 (Fig. 2). Though measurements from Dahlgaard

1999 in the East Greenland Current do not confirm any increase in Tc-99 values at that same time.

Table 5. Tc-99 in seaweed at the shore around Iceland in 1990-2000

	<i>1998-2000^a</i>		<i>1998-2000^b</i>	
	<i>Cs-137 Bq/kg d.w.</i>	<i>Number of measurements</i>	<i>Cs-137 Bq/kg d.w.</i>	<i>Number of measurements</i>
Grímsey	3,8	6	2,5	6
Vestmannaeyjar	1,6	6	1,3	5
Fáskrúðsfjörður	8,2	6		
Ísafjörður	1,6	7		

^asamples measured at Geislavarnir, Iceland

^bsamples measured at Risö, Denmark

Acknowledgements

This study was financially supported by Nordic Nuclear Safety Research (NKS).

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^{137}Cs and ^{99}Tc in seawater and seaweed around the Faroe Islands

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Introduction

Radionuclides like ^{137}Cs , ^{90}Sr and ^{99}Tc are transported over large distances in the marine environment by ocean currents. For the Faroe Islands, particular concern regards the concentration of these isotopes in the Irish Sea. The surface currents from the Irish Sea go north of Scotland into the North Sea towards Scandinavian coasts, continue north along the Norwegian west coast with branches to the Barents Sea, the Fram Strait and towards the East Greenland coast, following the East Greenland Current southwards. Radionuclides from European releases will most likely enter Faroese waters by ocean currents heading the islands from north.

Seawater samples have been collected from different water masses around the Faroe Islands in 1990 and 1995. ^{137}Cs and ^{90}Sr were measured together with hydrographic parameters (Dahlgaard et al., 1999), showing increased ^{137}Cs levels at intermediate depths.

In the context of BOK 2.2, we focus on the isotopes ^{99}Tc and ^{137}Cs . ^{137}Cs , having a physical halflife of 30 years, is measured by gamma spectroscopy. ^{99}Tc is a pure beta emitter with a physical halflife of 210000 years. A laboratory for analysing ^{99}Tc in seawater and biota has been established at the University of the Faroe Islands, and results from trial runs are presented in this report. Gamma spectroscopy has, on the other hand, been carried out by the laboratory for more than 20 years.

Material and Methods

Sampling

The results from a pilot project in 1990 and 1995 clearly suggested profile sampling at selected positions, particularly north of the Faroes. However, this has not been practically possible since 1995, mainly because of tight time schedule for the research vessel and the demand for calm weather in order to handle the big water sampler (270 litre Hydro Bios Large Volume Water Sampler).

For the BOK 2.2 programme, it has only been possible to get samples of seawater from the sea surface and samples of seaweed. The seawater has been sampled at the location Kirkjubøur on the west side of the country. The seawater samples are expected to be representative for the Faroese shelf area, as Kirkjubøur is open to the North Atlantic. Because of low radionuclide concentrations in Faroese seawater, 200 litres were collected for the analyses. Seaweed was sampled at the same location as the seawater.

Analyses

Radiocaesium

For the ^{137}Cs analyses in seawater, CsCl is added to the samples. pH is adjusted to 1.5-2.0, whereupon caesium is precipitated on AMP (Ammonium Molybdo Phosphate) for 24 hours.

The precipitate is vacuum filtered and washed with 0.025 M NH_4NO_3 and with 0.01 M HNO_3 , whereupon it is dried at 105°C for 24 hours. The dried material is analysed by gamma spectroscopy.

Seaweed samples are dried at 100°C and ground before they are analysed by gamma spectroscopy.

Technetium

The seawater samples were spiked with $^{99\text{m}}\text{Tc}$, fetched from the local Hospital from a $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generator. In the laboratory the sample was decontaminated for interfering β -emitters and concentrated in Tc by anion exchange, liquid-liquid extraction and precipitation at controlled valency (Chen et al., 1990). The chemical yield of $^{99\text{m}}\text{Tc}$ was determined by γ -counting within 24 hours after adding the $^{99\text{m}}\text{Tc}$ spike, using a Canberra Series 10 Plus system with a NaI detector. A new $^{99\text{m}}\text{Tc}$ spike was added to the stripped sample, and the chemical yield of $^{99\text{m}}\text{Tc}$ was determined at the end of the procedure. Metallic Tc was deposited on a steel disc and counted in a GM-25-5 β -multicounter.

Seaweed was dried at 100°C and ground before carbonised by concentrated H_2SO_4 and digested by concentrated HNO_3 after which normal procedures for seawater were followed.

Results and Discussion

Radiocaesium

Results from ^{137}Cs measurements of seawater samples from north of the Faroe Islands in 1990 and 1995 can be found in Table 1. The results show significant variation with depth in August 1990. The relative high value at 400m depth in 1990 has been related to the water mass characterised as the Arctic Intermediate Water (Dahlgard et al., 1999). The Overflow Water from the north passing out of the Faroe Bank Channel below 500m depth showed also increased caesium levels in 1990 (Dahlgard et al., 1999). The results suggested Sellafield as a possible source. A similar profound signal was not observed in May 1995. A possible reason for this difference could be that the samples in 1990 and 1995 do not derive from exactly the same oceanographic watermasses. The results do, however, indicate a decrease in the ^{137}Cs activity from 1990 to 1995.

Table 1. Results from measurements of ^{137}Cs in seawater north of the Faroe Islands in 1990 and 1995. The volume per seawater sample was 200 litre.

	August 1990					May 1995					
	Position: 63°20'N 6°05'W. Bottom depth: 2400m					Position: 63°30'N 6°04'W. Bottom depth: 1831m					
Obs. depth; m	50	200	400	700	1000	50	100	150	200	300	470
^{137}Cs ; Bq/m ³	3.5	3.7	4.9	3.5	2.3	2.9	2.6	2.6	2.9	2.9	2.5
St.dev; Bq/m ³	0.4	0.4	0.4	0.4	0.3	0.3	0.3	0.2	0.3	0.2	0.2

^{137}Cs has been measured again in seawater samples from November 2000 and January 2001. The results are presented in Table 2. The results agree fairly well with the results presented in Table 1. The samples were measured by Risø for practical reasons at the laboratory.

The ^{137}Cs activity has been measured in *seaweed*, sampled 8 May 2000 and 19 October 2000 at the location Kirkjubøur. The results were 0.49Bq/kg(dw) and 0.40Bq/kg(dw), respectively, but it should be considered that the counting uncertainty was more than 50% in both cases.

Table 2. Results from measurements of ^{137}Cs in surface seawater collected at Hoyvík, located in the central part of the Faroe Islands. Sample size in November: 10litre. Sample size in January: 200litre.

Sampling date	28 November 2000	4 January 2001
^{137}Cs ; Bq/m ³	1.71±0.67	2.06±0.14

Technetium

Measurements have been carried out on two samples of surface seawater, each 200 litre, and six samples of seaweed, using 10 g dry weight of seaweed for each sample. The samples were collected in May and June 2000 at the location Kirkjubøur. The results are presented in Table 3.

Table 3. Preliminary results from measurement of ^{99}Tc in Faroese samples. Chronology of analysis from left to right for each type of sample.

	Seaweed						Seawater	
	8 May 2000						8 May 2000	8 June 2000
^{99}Tc ; Bq/kg(dw)	1.65	1.86	8.26	5.82	4.58	5.95		
^{99}Tc ; mBq/m ³							7.0*	590

* ^{99}Tc deposited on the steel disc was below detection limit.

Aarkrog et al. (1994) reports ^{99}Tc levels around 20-30 mBq/m³ in surface seawater from the Faroe Islands, indicating that the values in Table 3 for seawater are out of scale. For comparison, the ratio between the activity in seawater and the activity in seaweed at Klint in Denmark is found to be around 60 m³/kg (Aarkrog et al., 1992). Further analyses are needed in order to obtain better estimates of the Tc-levels in the Faroese marine environment. The transit time from the Irish Sea to the East Greenland Current has been estimated to 7-10 years (e.g. Dahlgaard et al., 1991), resulting in even longer transit time to the Faroe Islands. The aim of this study is to get a reference level of ^{99}Tc in the Faroese marine environment, to be compared with if the new release from Sellafield should reach Faroese waters.

It is important to emphasise that the measurements presented in Table 3 should only be considered as trial runs. As experience was gained during the runs, so did Tc-yields. Recovery of the added $^{99\text{m}}\text{Tc}$ spike improved from 35% to 55% during the trial runs in the case of seaweed. Chen et al. (1990) report yields of 70-80% for seawater and 80-95% for seaweed.

Conclusions

The observed ^{137}Cs levels in seawater and seaweed are low, and a decreasing trend has been observed for the activity in seawater. ^{137}Cs activity in seawater have shown significant variation with depth north of the Faroe Islands. It is, however, very difficult to carry out profile sampling in the area, and it may become necessary more or less to stick to surface samples of seawater.

The introductory technetium part of the study has mainly aimed at building up competence in analysing seawater and seaweed for ^{99}Tc . The obtained data are not satisfactory, but the work so far documents that the laboratory is able to manage the selected procedure, and that the laboratory is ready for further Tc-analyses.

Acknowledgements

Thanks are extended to the Nordic Nuclear Safety Research (NKS) for financial support. Thanks are also extended to colleagues in the BOK-2.2 group for valuable discussions, and to Risø for assistance in measuring samples.

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A sediment trap experiment to study seasonal variations in vertical fluxes in the Baltic sea.

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Introduction

The continental shelf is an important regime for vertical export of particles and associated constituencies and a commonly used method for estimating vertical fluxes in surface waters is sediment traps. However, artefacts such as hydrodynamic biases, preservation problems and the presence of 'swimmers' have questioned the quality of the flux data. These problems have in later years been accentuated, partly as an effect of obtaining more accurate data on the drawdown of atmospheric CO₂ into the oceans in order to estimate potential climate changes. In this context the flux of particulate organic carbon (POC) formed by biological activity in the upper water masses has been of particular interest. The drawdown of atmospheric CO₂ is related to the primary production but the formed POC may mineralize back into CO₂ unless the POC is exported to deeper water as sedimenting matter. The rate of surface water export of POC and subsequent (temporal) burial of organic carbon in the sediments is in other words an important measure of the rate at which excess levels of atmospheric CO₂ may return to 'normal' levels again.

An alternative way of determining fluxes of reactive substances is to use a marker or tracer which attaches to the particulate matter and which is easily measured. Fortunately such a marker exist in the form of ²³⁴Th. This isotope is constantly produced in the water as a result of the decay of ²³⁸U. Uranium behaves almost completely conservative in salt water (concentration of ²³⁸U is directly proportional to salinity in the world oceans within the salinity interval 3-40‰) while thorium quickly becomes attached to surfaces in the water, in particular particulate organic matter. The activity ratio ²³⁴Th/²³⁸U is thus a measure of the steady-state removal rate of particulate matter (onto which ²³⁴Th is adsorbed). The production of ²³⁴Th during steady-state is simply balanced by the decay and removal and the residence time of ²³⁴Th may thus be calculated. By analysing ratios ²³⁴Th/X in particulate matter where 'X' is the constituent of choice (eg ²³⁹⁺²⁴⁰Pu) removal rates of 'X' may also be determined. These rates, or fluxes, may be compared with the flux measured by analysing the trap material. In this way the sediment traps may become 'calibrated'. In particular, analysis of ²³⁴Th in the trap material is of great use if the water column disequilibrium ²³⁴Th-²³⁸U has been determined over the collection period (1-3 months is suitable when using ²³⁴Th having a half-life of 24d). The degree of over- or undertrapping may then become determined.

Although coastal seas (such as the Baltic Sea) only constitute a few percent of the total water surfaces of the world oceans they play an important role due to the much higher biological productivity as compared to the open oceans. In general, the scavenging of particle reactive constituents in coastal waters are orders of magnitude higher than in the open ocean. In the open ocean settling material may also mineralise before reaching the sediments and scavenged particulate substances may return to solution and return to the surface waters as an effect of water mass mixing. In coastal shallow seas settling particulate matter don't mineralise before reaching the bottom and the scavenged substances will thus become buried in the sediments.

An additional problem in determining residence times for reactive substances in the water column is the dynamics in both biological productivity and water transport leading to large changes in suspended matter concentrations and particle quality over the year. Normally we only obtain 'snapshots' of the system and extrapolate data to cover all seasons.

To conclude, most sediment trap studies of vertical fluxes of reactive constituents for the purpose of estimating residence times in the water column suffer from at least two problems; Sediment trap artefacts and seasonal variations. Also other effects should be considered, in particular resuspension events.

I was for the purpose of studying these two (three) effects on the vertical flux of particulate matter that this experiment was conducted as a result of a two year long sediment trap study in the Landsort basin (Baltic Sea).

Materials and methods

Sediment traps, consisting of straight gyro-mounted cylinders, 10 cm diameter and an aspect ratio (height/width) of 5, were deployed below the mixed surface layer at 40m depth in the Landsort deep, Baltic Sea, some 40 km offshore. Collection of the trap material was done biweekly to monthly between July 1998 to August 2000, thus over a period of more than two years. The trap material was allowed to settle in conical settling tubes and later centrifuged. After drying the material (in the range of 20-500 mg dry salt corrected matter) was transferred to test tubes and gamma counted for ^{234}Th and ^{137}Cs in a well type HPGe-detector.

Samples for water column ^{234}Th analysis were collected, in the middle of the mixed layer, by filtering large volumes (500-1500 l) of water through 0.2 μm polypropylene filters to separate particles and, through two identical cartridges impregnated with Mn-oxide to extract dissolved thorium.

The filters were ashed and thorium separated by standard radiochemical methods using ^{229}Th as a yield determinant, the purified sample was deposited on to stainless steel discs and counted for its beta activity using an anticoincidence-shielded multi GM-counter (Risö-type).

The discs were then alpha counted, using PIPS-detectors, in calibrated geometries to determine the other thorium isotopes (^{232}Th , ^{230}Th and ^{228}Th) as well as to determine the radiochemical recovery (necessary in order to determine the ^{234}Th activity).

Results and Discussion

The result for the ^{234}Th determinations in the sediment traps are shown in figure 1. Clearly the flux ($\text{Bq m}^{-2} \text{ d}^{-1}$) of ^{234}Th is depending on the season. During spring (march) a strong flux peak is seen. This correlates well with the increased biological productivity seen during this month. Although only a limited number of the water column data for ^{234}Th are available at present, fluxes calculated from the ^{234}Th - ^{238}U disequilibrium agrees surprisingly well with what is found in the traps. The traps thus seems to be able to collect what we consider to be 'actual' fluxes.

In figure 2 the ^{137}Cs data from the sediment traps are shown. As with the ^{234}Th data the seasonal variation is very strong and the spring-peak is present for both years. Since the ^{137}Cs found in the trap material most likely is related to the amount of resuspended matter we may here use the ^{137}Cs as an important tracer. We know from the weekly monitoring program performed at the BY-31 station in Landsort deep that the biological productivity much follows the pattern shown in figure 1 and 2. But is the biological productivity responsible for the fluxes of ^{234}Th measured? The proportion of ^{234}Th adsorbed on particulate matter certainly should correlate rather nicely with the amount of POC in the surface waters but we determine particulate matter (as well as POC) as the amount of matter caught via filtration. This matter does not necessarily sink, unless some 'anchors' becomes connected to it. The pure POC in itself has such a low density that it sinks very slowly. But it readily attaches to other suspended material, including resuspended 'heavy' mineral grains.

The two pictures below thus gives us some insight as to what governs the cyclicity observed in the fluxes. We believe that the ^{137}Cs figure shows how spring mixing of the water column brings up nutrients from shallow sediments (in this case most likely the nearby archipelago) together with fine clay and silty material which is transferred offshore by surface currents. The nutrients together with an increase in light triggers the biological productivity. The increase in POC during this time is manifold but were it not for the supplied 'anchors' in the form of mineral grains, as traced by the ^{137}Cs , there would not be such a large export flux of ^{234}Th . We believe that this effect is the reason to the commonly observed relation between mass flux and Th-flux. It is simply an effect of two completely different mechanisms (uptake of Th onto organic matter and drawdown of the organic matter through 'anchors') but which both has its origin in the supply of material derived from the sediments.

From this we may learn that measuring particulate matter concentrations alone may not be sufficient to correlate to vertical fluxes. We must also consider the type of particles present. We may also learn that tracers providing information on sediment born material may be of great help in interpreting data. This concerns both tracers for particulate and dissolved matter.

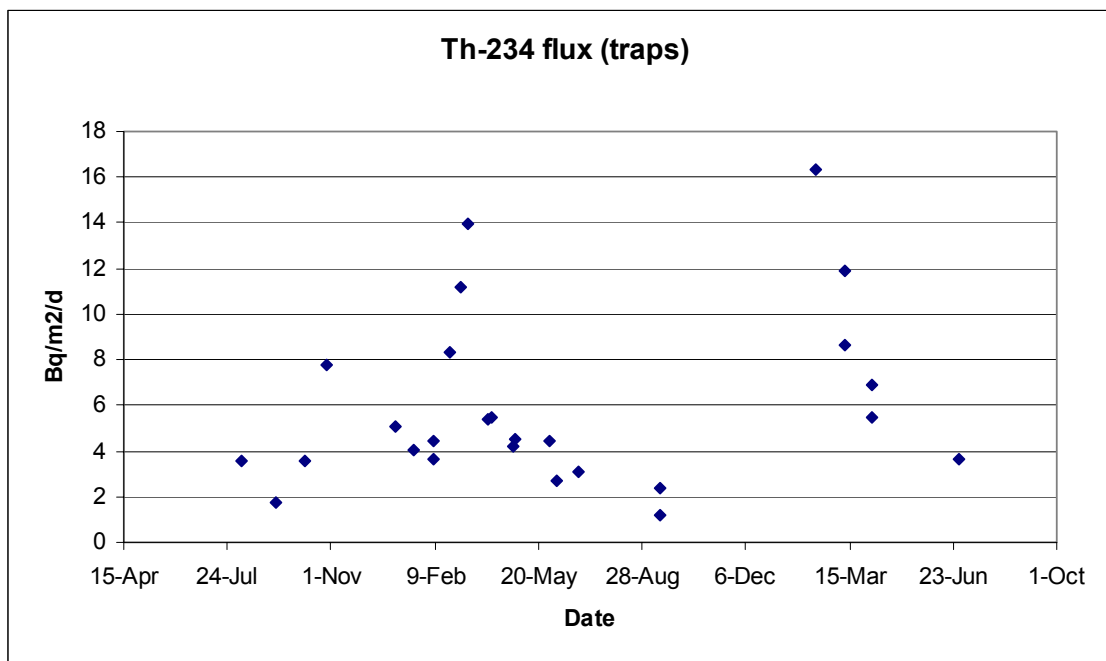


Figure 1.

Results for ^{234}Th -flux ($\text{Bq m}^{-2} \text{d}^{-1}$) as determined from measurements on sediment trap material.

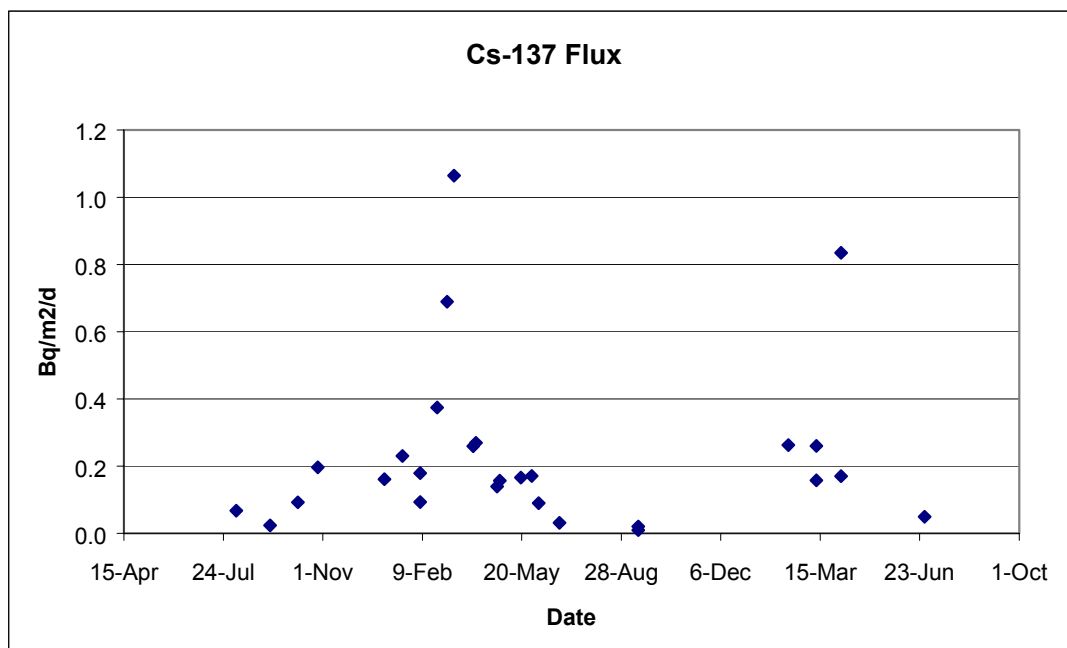


Figure 1.

Results for ^{137}Cs -flux ($\text{Bq m}^{-2} \text{d}^{-1}$) as determined from measurements on sediment trap material.

Acknowledgement

The radioisotope part of this work was financially supported by Nordic Nuclear Research.

The Finnish contribution to BOK-2.2 was divided into 4 sub-projects within 2 main topics of the Project, both of them focusing on the use radionuclides as tracers in the Baltic Sea.

Use of Tc-99 as tracer for Sellafield discharges in the Baltic Sea

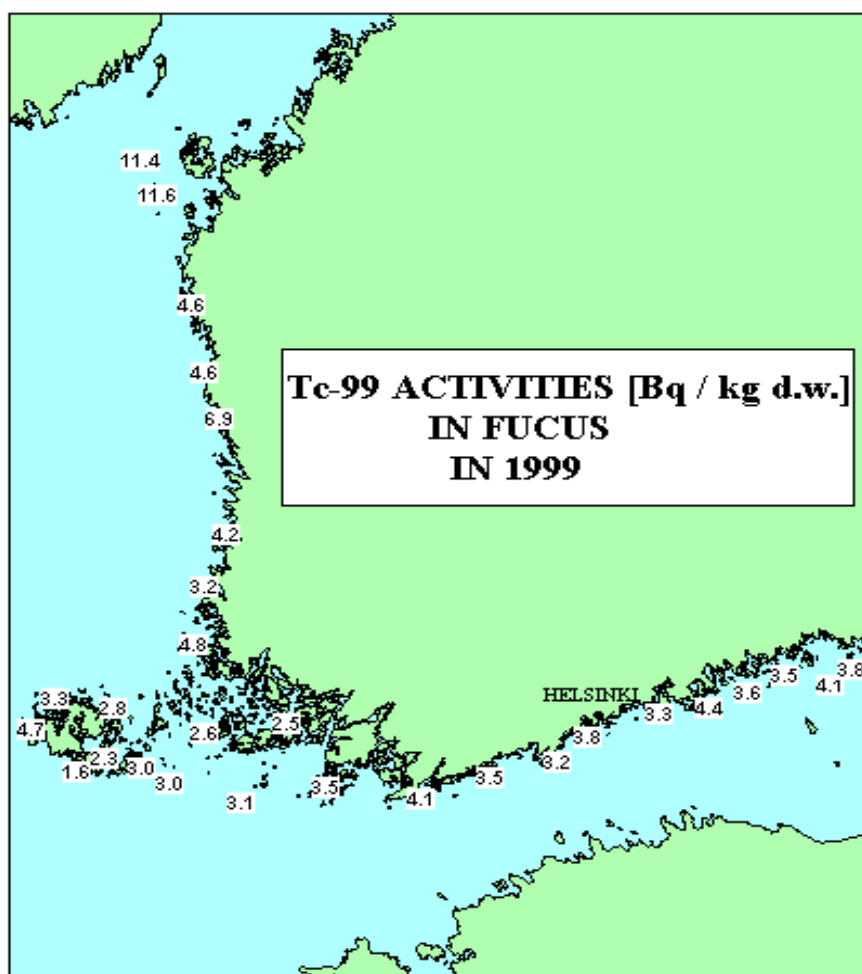
Technetium-99 in biota samples collected along the Finnish coast in 1999

Erkki Ilus, Vesa-Pekka Vartti, Tarja K. Ikäheimonen, Jukka Mattila and Seppo Klemola

STUK - Radiation and Nuclear Safety Authority, Finland

Abstract

The aim of the study was to develop a method for analysing ^{99}Tc at STUK, and to establish whether ^{99}Tc can be detected in Finnish coastal waters. Samples of *Fucus vesiculosus*, *Myriophyllum*, *Macoma baltica*, *Mytilus edulis*, *Saduria entomon* and seawater were collected at more than 30 stations along the western and southern coasts of Finland for ^{99}Tc analysis. Small amounts of ^{99}Tc were observed in all the *Fucus* samples collected; the activity concentrations ranged from 1.6 to 11.6 Bq kg⁻¹ dry wt. The highest concentrations were found in two samples taken from the northernmost stations in the Quark, probably for biological reasons. Due to the low salinity of the water, *Fucus vesiculosus* is very slow growing and small in this area, which is at the extreme limit of its permanent distribution range in the Baltic Sea. In seawater and in all the other biota samples, the concentration of ^{99}Tc was below the detection limit, which supports the use of *Fucus* as an indicator organism for ^{99}Tc in the marine environment. Global fallout from atmospheric nuclear weapons tests carried out in the 1950s and 1960s is certainly the most important source of ^{99}Tc detected in *Fucus* on the Finnish coast. In addition, the samples were analysed for gamma-emitting radionuclides.



The method developed and the results have been published in:

Ikäheimonen, T.K., Varti, V.P., Ilus, E., Mattila, J., 2002. Technetium-99 in *Fucus* and seawater samples in the Finnish coastal area of the Baltic Sea, 1999. *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 252, No. 2 (2002) 309-313, Budapest.

Ilus, E., Varti, V-P., Ikäheimonen, T.K., Mattila, J. and Klemola, S., 2002. Technetium-99 in biota samples collected along the Finnish coast in 1999. *Boreal Environment Research*, 7: 91-97, Helsinki.

Radionuclide processes in the Baltic Sea and its catchment and the adjacent areas

Discharge of ^{137}Cs and ^{90}Sr by Finnish rivers to the Baltic Sea in 1986-1996

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Abstract

The total amounts of ^{137}Cs and ^{90}Sr transported from Finland by rivers into the Gulf of Finland, Gulf of Bothnia and Archipelago Sea since 1986 were estimated. The estimates were based on long-term monitoring of ^{137}Cs and ^{90}Sr in river and other surface waters and on statistics of water discharges from Finnish rivers to the above sub-areas of the Baltic Sea.

The total amounts of ^{137}Cs and ^{90}Sr removed from Finland into the Baltic Sea during 1986 - 1996 were estimated to be 65 and 10 TBq, respectively. The results show that, although the deposition of ^{137}Cs was much higher than that of ^{90}Sr after the Chernobyl accident, the amount of ^{137}Cs removed from Finland is only six times as high as that of ^{90}Sr . This emphasizes the importance of ^{90}Sr while considering radiation doses from surface waters and of ^{137}Cs while estimating doses via pathways from catchment soil, lake sediments and biota after a fallout situation.

The results have been published in:

Saxén R., Ilus E., 2001. Discharge of ^{137}Cs and ^{90}Sr by Finnish rivers to the Baltic Sea in 1986-1996, *Journal of Environmental Radioactivity* 54 (2001), 275-291, Elsevier Science Ltd.

Old data on ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in Baltic Sea sediments prior to the Chernobyl accident (1966-1980)

Erkki Ilus and Jukka Mattila

STUK - Radiation and Nuclear Safety Authority, Finland

Background

The aim of the Finnish BOK-2.2 project was to give an overall view of radionuclide processes in the Baltic Sea, in its catchment and in adjacent areas. Plenty of data have been published considering quantities of certain long-lived radionuclides in the Baltic Sea sediments after the Chernobyl accident, while the data published from the period of the weapons tests fallout prior to the Chernobyl accident are relatively few.

STUK has long traditions in the Baltic Sea studies. Radioactive substances in its water, sediments and biota have been monitored since the late 1950s. However, considerable amounts of results from the 1960s and '70s concerning ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in sediments have not been published and collated with the existing data on seawater.

Objectives

The objectives of this study were

- to collect and prepare the existing old data for publication
- to compare the old data with that published after the Chernobyl accident
- to consider the earlier published inventory results of these nuclides in the Baltic Sea sediments prior to the Chernobyl accident in the light of the new knowledge
- to consider the role of sedimentation in losses of radionuclides from water column into the seabed

Results

The old sediment data consist of 114 sediment profiles taken in 1966-1980 from the Bothnian Bay, Bothnian Sea, Archipelago Sea, Gulf of Finland and the Northern Baltic Proper. A majority of the samples has been taken from the areas surrounding the sites of the Finnish nuclear power plants, Loviisa and Olkiluoto.

^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ were quite evenly distributed in the areas studied. In general, however, the total amounts of these nuclides (per m^2) were slightly higher in the coastal areas than at the open sea.

The total amounts of ^{90}Sr ranged from 10 to 260 Bq m^{-2} , those of ^{137}Cs from 50 to 13.000 Bq m^{-2} , and those of $^{239,240}\text{Pu}$ from 4 to 340 Bq m^{-2} . The amounts of ^{137}Cs were significantly lower than those reported after the Chernobyl accident (Fig. 1). The maximum value (13.000

Bq m⁻²) found from the Archipelago Sea was almost one order of magnitude lower than the highest values reported from the Bothnian Sea after the Chernobyl accident.

The material is in preparation for publication.

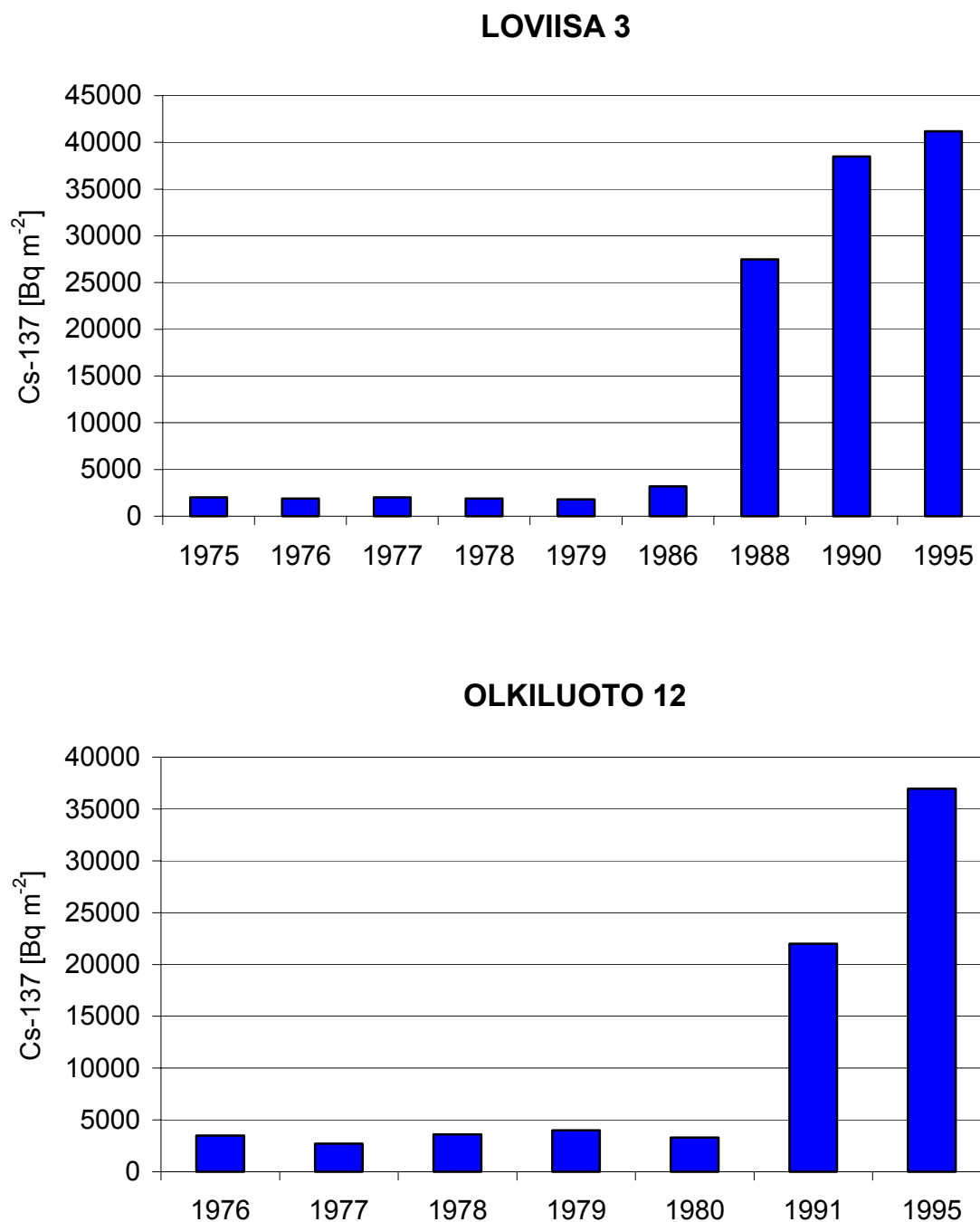


Figure 1. Total amounts of ¹³⁷Cs [Bq m⁻²] at two coastal station prior to the Chernobyl accident and after that (1986).

¹³⁷Cs in the sediments of the Gulf of Bothnia: additional data on total inventory in sediments

Jukka Mattila and Erkki Ilus

STUK - Radiation and Nuclear Safety Authority, Finland

Background

In 1993-1997 STUK and the Finnish Institute of Marine Research carried out an extensive study on the total inventory of ¹³⁷Cs in the Baltic Sea sediments. The study was linked with the Finnish EKO-1 Project: "Sedimentation rate in the Baltic Sea" supported by the NKS. The results were published in IAEA-TECDOC-1094 "Marine Pollution, Proceedings of a symposium held in Monaco, 5-9 October 1998". The study showed that the proportion represented by the Bothnian Sea was clearly dominant in the total Baltic Sea inventory of ¹³⁷Cs, but the areal representativeness of these results remained unverified.

Objectives

The objectives of this study were

- to improve the knowledge of ¹³⁷Cs amounts in different parts of the Gulf of Bothnia by taking some additional samples and by repeating sediment sampling at certain stations sampled before
- to improve the knowledge of the role of sedimentation in losses of radionuclides from water column into the seabed.

Results

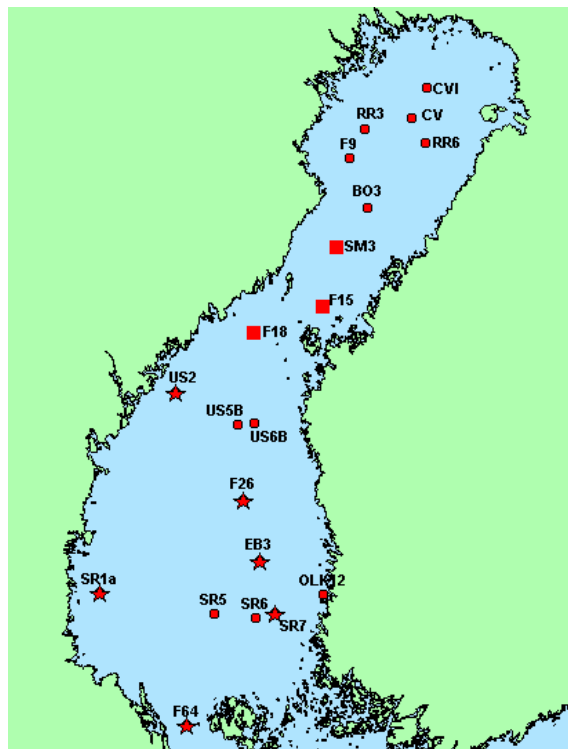


Figure 1. Sampling stations in the Gulf of Bothnia. New sampling stations are marked with stars, old stations with circles and stations where sampling has been repeated with squares.

Table 1. Total amounts of ^{137}Cs [Bq m^{-2}] at the sampling stations in the Gulf of Bothnia. Values are time corrected to the year 2001.

Station	Year	^{137}Cs [Bq m^{-2}]
CVI	1996	3900
CV	1996	5600
RR3	1996	8700
RR6	1996	5100
F9	1996	8800
BO3	1996	15800
SM3	1996	6500
SM3	1999	13100
F15	1996	88600
F15	1999	87700
F18	1996	104400
F18	1999	109400
US2	1999	117500
US6B	1996	7000
US5B	1996	90400
F26	1999	26700
EB3	1999	30800
OLK12	1995	31800
SR1a	1999	37700
SR5	1996	28400
SR6	1996	26300
SR7	1999	15700
F64	1999	12900

Table 2. Average and median values for total amounts of ^{137}Cs [Bq m^{-2}] based on samples collected in 1996 and 1999.

	1996	1996	1996 and 1999	1996 and 1999
Area	Aver.	Median	Aver.	Median
Gulf of Bothnia	30800	12300	33900	21100
Bothnian Sea	48100	30100	44300	29600

- the results confirm our previous results on the high amounts of ^{137}Cs in the sediments of the northern Bothnian Sea
- the results improve the knowledge of the areal distribution of ^{137}Cs in the sediments of the Gulf of Bothnia, and the role of sedimentation in losses of ^{137}Cs from the water column into the seabed.

The material is in preparation for publication.

Comparison of two ICP-MS systems for measuring Tc-99 in large volume water samples

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Paper published in the Analyst, 2002, **127**, 70-75

Abstract

Large volume fjord and seawater samples have been radiochemically prepared for ICP-MS analysis in order to test the robustness of the procedure and to carry out a comparison of two ICP-MS systems. A sector field instrument (MicroMass PT2) coupled with an ultrasonic nebuliser and a quadrupole ICP-MS (Perkin Elmer Elan 6000) coupled with an electrothermal vaporisation (ETV) unit were used. The results showed that the radiochemical procedure was robust, removing Ru and Mo to acceptable levels, and that the two instruments gave results that were in agreement. The correlation coefficient between the sets of 11 results was 1.0 ± 0.05 . The importance of establishing the matrix effect when using an ETV is discussed.

Introduction

Technetium-99 is a long-lived (2.1×10^5 y) beta-emitting radionuclide, present in the European seas primarily from reprocessing effluents discharged from Sellafield, UK, and Cap la Hague, France. There is widespread interest in measuring Tc in marine and estuarine waters, in order to utilise it as a marine tracer and understand its behaviour in the environment. Technetium-99 is currently measured in most environmental samples by beta-counting, following prior removal of all other beta-emitting nuclides from the sample, since beta-counting is not nuclide-specific. While beta-analysis has a reasonable detection limit (around 3 mBq (5 pg)¹) and has yielded a large amount of useful data, there is a drive to use alternative methods with lower detection limits and without the uncertainty associated with completely removing all other beta-emitting nuclides from every sample.

Different types of mass spectrometry have been used to measure Tc, and the results have been generally very good. TIMS and accelerator mass spectrometry have shown low instrumental detection limits, at approximately 10 - 20 fg^{2,3}, but most work has been carried out using inductively couple plasma mass spectrometry (ICP-MS),

because of the relative availability of these instruments and the lower cost of running samples. Detection limits for ICP-MS range considerably according to the age and design of the instrument, the sample introduction system used and, possibly, the calculation used. Generally, quotes lie in the range 3 pg l^{-1} ⁴ to 0.6 ng l^{-1} ^{5,6}, which correspond to absolute detection limits of 0.02 mBq (30 fg) – 3.8 mBq (6 pg) , assuming that the samples were prepared in 10 ml where appropriate. Significantly better detection limits have been reported by Becker and Dietze⁷ using a sector field instrument with a torch shield and a MicroMist microconcentric nebuliser with a minicyclonic spray chamber, of 2.5 fg . However, despite the number of papers reporting measurements of pure Tc solutions or high specific activity samples, there are very few which describe the use of the technique for the routine analysis of low-level, and especially, large volume samples.

The main interferences in the mass spectra of ^{99}Tc are ^{99}Ru and, to some extent, ^{98}MoH (see also table 1). These can be removed in the chemical separation procedure, for example by using TEVA.spec resin, but when working with large samples and using large amounts of reagents, absolute clean up can be difficult. Some ^{99}Ru can be tolerated as it can be accounted for by measuring at mass 101 for ^{101}Ru . Mass 101 experiences very little, if any interference, and so this is a reasonable correction. However, despite being able to account for Ru, its presence affects the minimum detectable activity of ^{99}Tc in a sample, especially when working with the most sensitive techniques⁸. Other potential problems involve the use of $^{99\text{m}}\text{Tc}$ as the chemical tracer for the separations procedure, since low levels of ^{99}Tc may build up in the tracer unless the ^{99}Mo generator is rigorously cleaned prior to yield monitor elution⁹. The problem increases for low-level, large volume analysis, since the complicated separation procedure often requires the addition of multiple aliquots. There is no stable isotope of Tc to use as an internal standard in mass spectrometry, but suitable elements include Rh⁸ and Nb³ which can be selected according to the chemical composition of the sample. For example, when analysing seawater Nb may not be favoured because of the Nb content of the samples. Beals⁵ proposed use of the long-lived ^{97}Tc isotope as both a yield monitor and internal standard, but difficulties arise from obtaining high purity ^{97}Tc at a reasonable expense, as well as the isobaric interference from ^{97}Mo . Additionally, in our laboratory we have observed an unidentified polyatomic interference at 97.

In this study we compare two different ICP-MS systems for measuring Tc, using large volume water samples, of 5 – 50 litre volume, taken from a fjord in southern Norway and the Kattegat. This is in order to assess the suitability of a chemical procedure and the capability of both instruments for analysing “difficult” samples. The ICP-MS systems used were a MicroMass PT2 ICP-sector field mass spectrometer, or high resolution ICP-MS, coupled with an ultrasonic nebuliser (USN) and an Elan 6000 quadrupole ICP-MS coupled with an electro thermal vaporisation unit (ETV). The different sample introduction systems used gives the Elan 6000 an advantage, since up to 80% of the sample reaches the plasma as opposed to approximately 20% via an ultrasonic nebuliser¹⁰. However, the ICP-SFMS magnet is too slow to scan in conjunction with an short transient signal from an ETV, meaning that both instruments are running at their, theoretically, most sensitive configurations with the introduction systems we had available.

Methods

Radiochemical Procedure Technetium-99m was used as the yield monitor during the chemical separations. A ^{99}Mo generator was obtained from the isotope laboratory at Risø National Laboratory, and washed thoroughly before the actual spike solution was eluted with 0.9% NaCl. The washing consisted of five 24 ml rinses of 0.9% NaCl on obtaining the generator and on every Monday morning, followed by 5 washes prior to every spike elution. An aliquot of the spike solution was gamma counted 3 times in standard geometry in a well detector, and then the samples were spiked with aliquots of the same solution, approximately $1 \text{ kBq sample}^{-1}$. A portion equivalent to 20 kBq of each $^{99\text{m}}\text{Tc}$ solution was prepared for analysis by ICP-MS to assess the ^{99}Tc content of the yield monitor itself. 10 ml of 0.1 M HNO_3 was added to the yield monitor solution and this was applied to a 0.5 ml TEVA.spec column. The column was washed with 10 ml 2M HNO_3 to remove the NaCl and the Tc eluted with 4 ml 8 M HNO_3 . The elution was gamma counted in standard geometry and the yield calculated.

The water samples analysed were of different compositions, ranging from oxic seawater through to anoxic waters, and so it was necessary to break down any organic complexes or precipitate phases, to release the entrained Tc into the solution phase. The first step was therefore to acidify the sample with 14.4 M HNO_3 to a pH less than 1.5 and to heat it strongly for 2 hours. Some samples had precipitated $\text{FeO}_x(\text{OH})_y$ onto the walls of the container during storage, and so 12 M HCl was used to dissolve any precipitate from the walls of the containers, and this was added to the rest of the sample prior to heating. During heating, the $^{99\text{m}}\text{Tc}$ yield monitor was added, ensuring that the spike was left to equilibrate for at least 30 minutes. After heating, saturated NaOH solution was added to precipitate $\text{Mg}(\text{OH})_2$ (pH~10), and remove the precipitable components of the sample. The supernatant was filtered through a paper filter, acidified to pH 6 and then pumped through a BioRad AG1-X4 column¹ (see table 2 for column volume and flow rate for the different sample volumes). The columns were then washed with 5 column volumes of 0.5 M HNO_3 and then the Tc eluted with 5 column volumes of 10 M HNO_3 .

In the method development stage we found that the BioRad resin contains a large amount of Ru, which bleeds from the resin when concentrated nitric acids are applied to it. Since Ru is the major interference in mass spectrometry, it was necessary to pretreat the resin with 14.4 M HNO_3 , leave it to stand for 2 hours and wash it thoroughly with nitric acid and deionised water to remove most (approximately 95%) of the Ru. Using 10 M rather than 14.4 M HNO_3 to elute the Tc also reduces the amount of Ru leached by a factor of approximately 15. During the course of sample preparation, an additional wash of a solution of NaOH, EDTA and NaClO was used for some samples. This is routinely used at Risø to remove radioactive Ru-isotopes prior to Tc elution and so seemed a quick way of achieving Ru decontamination prior to the final clean up, described below. However, the samples prepared in this way actually showed stable Ru levels enhanced by a factor of approximately 30, seriously affecting the counting statistics at mass 99. It is therefore concluded that the BioRad column should simply be used as a pre-concentrating step, rather than included in the Ru removal method. It is important not to use only radioactive Ru to test for Ru decontamination in method development for ICP-MS as it overlooks any contamination from the reagents and laboratory.

The 10 M HNO₃ elutions from the Bio-Rad column were heated down to 0.25-0.5 ml and then 25 ml deionised water (18.2 MΩ) was added. The solution was applied to a 2 ml TEVA.spec column, washed with 40 ml 2 M HNO₃, and eluted with 7 ml 8 M HNO₃, as described in Tagami and Uchida¹¹. When the initial volume of the samples was 25 or 50 l, this step was repeated. The samples were then counted in standard geometry to obtain the yield. The 25 and 50 l samples required double spiking, the first for the preconcentration stage and the second for the TEVA.spec columns.

Samples Two sets of samples were used in this study, firstly a set of 11 samples from two stratified fjords which were initially prepared for analysis on the Elan 6000 with ETV, as described by Skipperud *et al.*¹². The samples were dried very slowly at < 80 °C in AA cups, then 100 µl of 1% HNO₃ was added to each and they were left to stand for 1 hour. Finally, 100µl of 1M NH₃OH was added and the sample was agitated to ensure thorough mixing and dissolution of the sample. The total volume was therefore only 200 µl, maximising the amount of sample Tc reaching the plasma per 20 µl injection. Smaller volumes can be used, but this can lead to pipetting difficulties. The resultant solution is alkaline, which helps stabilise the volatility of Tc, minimising losses in the drying stages of the heating program prior to the vaporisation temperature of 2600 °C¹². Three 20 µl aliquots were analysed on the Elan 6000 with ETV. The remaining 140 µl of the samples were dried slowly at < 80 °C, redissolved in 10 ml of 1% HNO₃ with 50 ng l⁻¹ ¹⁰³Rh internal standard, and measured on the PT2.

An internal standard is not necessary when using the Elan 6000 with ETV as the signal is stable, and 3 replicates are analysed to account for any small variation. However, when using the ICP-SFMS with USN, the signal noise is greater than with the standard pneumatic sample introduction, and the signal can decrease over time as particulate matter builds up on the cones. Furthermore only one replicate is analysed, so the internal standard is very important. ¹⁰³Rh, as suggested by Fifield *et al.*⁸, was selected as the best internal standard for these samples, because it fulfils all the necessary criteria; it is close to Tc in both mass and first ionisation potential (7.46 vs 7.28 eV), it has a minimal memory effect, the background at 103 is very low and there are very few interferences at 103, and our samples contained very little Rh. Moreover, each TEVA.spec column was found to remove 98% of the Rh present, so by adding a significant amount of internal standard (50 ng l⁻¹) there will be very little error, either from additional Rh from within the sample or from measurement error.

The second set of samples consisted of 5 sub-samples (5 l) of Kattegat seawater. These were prepared radiochemically and analysed on the two instruments to assess the reproducibility of these measurements and to establish matrix effects on both systems. Although this seawater is not directly representative of the fjord sample compositions, it tested the sensitivity of the systems to a sample composition that is at least similar to the bulk of our samples, and that had been through the same chemical preparation. The unique samples of low Tc atom concentrations from the fjord could not be directly tested for matrix effects, as the maximum amount of sample had to be retained in order to measure them on both ICP-MS instruments.

Different approaches were used for testing the matrix effects on the two instruments. For the PT2, one Kattegat sample (prepared in 10 ml) was subdivided into five 2 ml aliquots and 4 of these were spiked with 100 µl of Tc standards of increasing

concentration, so that a calibration plot could be drawn with the sample concentration as the 'blank' (method of standard addition). The counting time was reduced in line with the smaller volumes (2.1 ml) of these sub-samples. The gradient of this calibration curve was compared with that of the standard calibration curve, to establish if the signal was enhanced or suppressed by the sample matrix. For the Elan 6000 with ETV, our samples were dissolved in a very small volume, 200 µl total volume per sample. Testing the matrix effect was therefore not as accurate as for the PT2, since we wanted to look at the matrix effect when the Kattegat samples were also dissolved in 200 µl, and so could not practically create a parallel calibration curve by sub-dividing and spiking a sample. Instead, the sample was dried down slowly after analysis and then redissolved in 140 µl of a spiked solution.

Running Protocols

Elan 6000 ICP-QMS The Elan 6000 with ETV was run using the parameters in tables 3 and 4, utilising the development work of Skipperud *et al.*¹². The samples were run semi-manually, with washes, blanks and standards run regularly.

PT2 ICP-SFMS The PT2 ICP-SFMS procedure had been developed prior to the comparison exercise, while the chemical method was also being developed. The ICP-SFMS method is similar to the method previously published for the measurement of trace levels of Pu in environmental samples¹³. The operating conditions are shown in table 5. The memory effect, or carry-over between samples, was eliminated using washes of 0.8 M HNO₃, as described by Richter *et al.*⁶. This brought the background down to the original level, and so we included a wash "sample" between every sample and standard. The wash time used was therefore the same as the time taken to analyse a sample (~8 minutes), and is longer than deemed necessary by Richter *et al.*⁶. However, this reliably reduced the memory effect and allowed unknown Tc concentrations to be analysed in randomly ordered samples. Standards were run every 3-5 samples.

Results and Discussion

Radiochemical methodology The results show that the chemical preparation successfully reduced Ru in the samples to a level where it did not interfere significantly with the Tc measurement. For example, on the Elan 600 with ETV the counts at 99 from Ru showed a mean of 313 ± 115 counts, whereas the Tc contributed $1\,600 - 20\,000$ counts. However, when working close to the detection limit, this amount of Ru would affect the minimum detectable activity in a sample. The analyses of the ^{99m}Tc yield monitor solutions, 20 times the activity used for each sample, showed that the amount of ^{99}Tc per spike aliquot was negligible. All of these were below detection limit, and so each spike aliquot must have contained less than approximately $10\ \mu\text{Bq}$ ($16\ \text{fg}$), which would be indistinguishable from the error on the background. The chemical yield of the first part of the procedure shows an average of 80.5%, with slightly higher yields obtained for the smaller samples. This is presumably because of lower losses during the $\text{Mg}(\text{OH})_2$ precipitation since the supernatant can be removed from the sample with more ease. The average yield from the TEVA.spec separation was 96%.

Stability The Elan 6000 showed great stability over the course of the first run, with the external standards showing a standard deviation of only 3.4%. The PT2 ICP-SFMS is less stable *per se*, but the internal standard compensates for this, reducing the standard deviation of the external standards from 12.2% to 3.5%. ^{103}Rh therefore seems to be a better internal standard in the PT2 than reported for AMS, which shows within-run variation of 12% between standards⁸. On the second run on the Elan 6000, however, there was a slow upward drift throughout the run, followed by a rapid decrease in signal when the tip sealing the ETV had corroded. This illustrates the day-to-day variability one may encounter in machine performance.

Interferences The potential interferences are shown in table 1. There are more potential interferences when using an ETV than a USN because of the formation of carbides from the graphite of the furnace, but the interferences from hydrides and oxides should be lessened. The probabilities of these polyatomic interferences forming in the plasma of the PT2 ICP-SFMS were found experimentally. Increasing concentrations of Mo and Co were measured, monitoring the change in signal both at 99 and 98 and 59 respectively. Once the signal at 98 or 59 started to trip, the signal was estimated from the signal for lower concentrations, and the concentration was raised until the polyatomic interference was clearly visible. The formation probabilities were calculated to be; $^{98}\text{Mo}^1\text{H}/^{98}\text{Mo} = 3 \times 10^{-5}$ and, $^{59}\text{Co}^{40}\text{Ar}/^{59}\text{Co} = 2 \times 10^{-6}$.

However, it is not always desirable to put very high concentrations of an element into the instrument, and so it is often sufficient to know that a polyatomic will not form at the concentrations present in your samples. Therefore, when using the Elan 6000, the concentrations of ^{59}Co , ^{98}Mo , ^{87}Rb and ^{87}Sr (see table 1) were increased until they caused the machine to trip at the respective mass. No extra counts were observed at mass 99. None of our samples caused the Elan 6000 to trip when measured at mass 59, 98 or 87, and so polyatomic signals would not interfere with the measurement of these samples. Comparing this with the formation probability found using the PT2 system of $^{98}\text{Mo}^1\text{H}/^{98}\text{Mo} = 3 \times 10^{-5}$, we should have only seen only 12 extra counts, which would have been indistinguishable from the background variation. The

formation probability of $^{98}\text{Mo}^1\text{H}$ can therefore be said to be less than or equal to this value.

Matrix effects The PT2 with USN showed no matrix effect for either the raw Tc data or that corrected by the Rh internal standard. The gradients of the standard addition and calibration curve slopes were within one standard deviation of one another, as shown in table 6.

The Elan 6000 with ETV, however, showed an average signal enhancement of 1.32 ± 0.02 for the Kattegat samples. Interestingly, the $^{99\text{m}}\text{Tc}$ yield monitor solutions measured using the ETV showed a much greater signal enhancement of 2.2 ± 0.15 when they were tested for a matrix effect. The matrix therefore has a very distinct effect on the transmission of Tc into the plasma and must be assessed very carefully when using an ETV.

Detection limits, background and sensitivity Typical sensitivities and detection limits and are shown in table 7. They are given in counts/mBq, mBq/sample and fg/sample to avoid the confusion of using ng l^{-1} , given that the sample volumes were 10 ml and 200 μl for the PT2 and the Elan 6000, respectively.

There is a significant amount of daily variation in the sensitivity of an instrument, depending on the optimisation of the machine and factors such as the time of the last service. Equally, the sensitivity can be improved by maximising the amount of time counting on a particular mass. For example, if Tc alone is measured, then the time spent counting the available atoms will be more than double when 99 and 101 are measured. There is a finite time for the magnet to settle when it changes mass, hence the more masses counted, the more time is "wasted", as well as there being less time available for measuring at mass 99. Including an internal standard therefore raises the detection limit for Tc, although it is necessary when using an USN. The detection limit is affected by the sensitivity and the background, and so the cleanliness of the machine is also very relevant, and does vary somewhat from day to day. Therefore, it is extremely difficult to give an accurate, let alone exact, detection limit of any instrument.

The practical detection limit, quoted as 3 times the standard deviation of the background, of measuring large batches of samples is even harder to assess, since the background may increase as a run progresses, or the signal will fall if the cones block slightly with build up of deposits. Moreover, for Tc, it depends on the amount of Ru in the sample. With this in mind, the data in table 7 are given for the background amount of Ru (the signal at mass 101 in the blanks and standards), at the beginning of a run, when the calibration curve is made. The background variation was determined in two ways, firstly as the square root of the initial background signal, and then as ± 50 counts, which was seen to be a typical variation in background over the course of a run. The second detection limits are therefore the more realistic for a normal set of analyses. The sensitivity is monitored by external standards run at regular intervals throughout the course of a run, and these did not fall significantly for either machine. The data reflect the same conditions as used when analysing the samples, *i.e.* the same number of sweeps, the same dwell times and the same masses measured. They are a function of both the sample uptake system and the instrument itself, and are given per

sample, on the basis that samples for the ETV were dissolved in 200 µl solution, despite the fact that only 60 µl was used.

Given the above discussion, the detection limits and sensitivity of the two overall systems are remarkably similar, with both capable of measuring samples with a Tc content an order of magnitude lower than is possible to measure by beta counting (~ 3 mBq (5 pg l^{-1})). Comparing these detection limits with those in the literature, they are certainly among the better values for ICP-MS, but are at least one order of magnitude higher than quoted for AMS³ and TIMS². The detection limits on our PT2 instrument could probably be improved by approximately a factor of 10 by utilising a shielded torch, and reduced even further by using MicroMist nebuliser, as demonstrated by Becker and Dietze⁷.

Comparison of the results from the two systems Initially, a scatter plot was drawn of the results obtained for the fjord samples on the two ICP-MS systems, with no correction for the matrix effect when using the ETV. The correlation coefficient (R^2) of the scatter plot was 0.978, with a 1σ standard deviation of 4.9%, showing that the results are consistent between the two systems. However, the slope of the line was 1.29, with the results from the Elan 6000 with ETV being 29% higher than obtained using the PT2 with USN. This agrees well with the matrix effect observed for the Kattegat water samples, which showed no matrix effect when using the USN but a 30% signal enhancement when using the ETV. Thus, when working with an ETV the matrix effect must be monitored closely and, for these samples, the data must be divided by a correction factor of 1.3 to obtain accurate results. However, once the matrix effect has been accounted for, the two systems show good agreement. Figure 1 is the scatter plot of the corrected results, of slope 1.0 ± 0.05 , and Figure 2 shows the residuals of the scatter plot, demonstrating that there is no systematic error.

The Kattegat seawater used to test the reproducibility of the measurements had a concentration of 1.2 mBq l^{-1} . The PT2 measurements showed a standard deviation of 6.1 % over 3 measurements and the Elan 6000, 5.8% over 2. After correcting the data from the Elan 6000 with ETV for the matrix effect (dividing by 1.3), the standard deviation for the five samples was lower than those from the individual instruments, at 5.2%. This shows that the two systems agree within the precision of the individual instruments and procedural reproducibility.

Overall, the Elan 6000 and PT2 ICP-SFMS are both capable of measuring Tc at low levels and with good precision. The PT2 ICP-SFMS was one of the first ever constructed, and lower detection limits have been quoted in the literature for similar machines, indicating that ICP-MS technology is improving all the time. Equally, the Elan 6000 is a relatively new instrument and shows much better results than usually quoted for quadrupole machines. The detection limit of the Elan 6000 with ETV could be improved by not counting on Mo, for example, or by dissolving the sample in a smaller volume. However, such changes could only improve the sensitivity by a factor of 2 or so, and would create different problems, such as how to assess for matrix effects for that sample without analysing parallel samples. While Ru is a limiting problem, the detection limits and sensitivity of these instruments are probably as good as could be required for measuring low-level, large volume samples by ICP-MS. However, the use of matrix modifiers may reduce the Ru interference when using ETV, as described by Song and Probst¹⁴. With AMS, it has been demonstrated that

99% of the Ru in a sample can be prevented from reaching the detector for a 50% loss in ^{99}Tc signal⁸. This is not possible using ICP-MS, but represents a way in which the low detection limits of AMS may be utilised without Ru continually masking the benefits.

Both instruments have detection limits around an order of magnitude lower than beta counting, and a series of low activity samples have been measured on both with excellent agreement between the two sets of measurements. The sample activities, while not being at the detection limit, would have been generally unmeasurable by beta-counting, demonstrating that ICP-MS is a viable, reliable and sensitive method for carrying out low-level Tc analysis of large volume samples.

Acknowledgements

We thank the NKS for supporting this collaboration, Lise Stower-Roston for technical support when using the Elan 6000 and Lis Vinther Kristensen for technical support when using the PT2.

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Table 1. Potential interferences at mass 99

Interference (m/z=99)
^{99}Ru
$^{98}\text{Mo}^1\text{H}$
$^{59}\text{Co}^{40}\text{Ar}$
$^{87}\text{Sr}^{12}\text{C}$
$^{87}\text{Rb}^{12}\text{C}$

Table 2. Column volumes of BioRad AG1-X4 and flow rates used for different initial sample volumes of sea- or fjord water.

Initial sample volume (l)	5	25	50
Volume of BioRad AG1-X4 (cm ³)	21	59	79
Flow Rate (ml/min)	6	30	55

Table 3: Data acquisition parameters and ICP conditions for the Elan 6000 ICP-MS

Dwell time (^{99}Tc ; ^{100}Mo , ^{101}Ru)	15 ms; 5 ms; 10 ms
Read delay	0.7 s
Sweeps per reading	1
Readings per replicate	100
Number of replicates	3
Points per peak	1
ICP RF power	1100 W
Plasma Ar flow rate	0.98 l min ⁻¹
Auxiliary Ar flow rate	0.3 l min ⁻¹
Nebuliser Ar flow rate	0.68 l min ⁻¹
Sample uptake volume	20 μl

Table 4: ETV heating program

Cell temperature (°C)	Ramp time (s)	Hold time (s)
80	10	10
120	5	10
2600	0	3
2650	1	4

Table 5: Data acquisition parameters and ICP conditions for the PT2 ICP-SFMS

Dwell time (at masses 99, 101, 103)	4 ms
Sweeps per reading	80
Readings per replicate	1
Number of replicates	1
Points per peak	15
ICP RF power	1350 W
Plasma Ar flow rate	12.5 l min ⁻¹
Auxiliary Ar flow rate	0.8 l min ⁻¹
Nebuliser Ar flow rate	0.7 l min ⁻¹
Solution uptake rate	1.15 ml min ⁻¹

Table 6: Testing the matrix effect on the PT2 Instrument

Rh correction?	Curve type	Gradient	1 sigma standard deviation
No	Calibration Curve	2358	27
	Standard Addition	2373	51
Yes	Calibration Curve	8.03×10^{-3}	9×10^{-5}
	Standard Addition	8.30×10^{-3}	2×10^{-4}

Table 7: The detection limits of the two instruments (N.B. Elan 6000 with ETV detection limits given for two separate occasions)

	PT2 /per sample	Elan (1) /per measurement	Elan (2) /per measurement	Elan (1) /per sample	Elan (2) /per sample
Typical background (c/measurement)	200	200	300		
Typical sensitivity (c/mBq)	700	8000	12000		
Typical detection Limit (3 x sqrt; mBq)	0.089	0.006	0.004	0.06	0.04
Typical detection limit (3 x 50 c; mBq)	0.29	0.018	0.013	0.18	0.13
Typical detection Limit (3 x sqrt; fg)	140	10	6	96	64
Typical detection limit (3 x 50 c; fg)	460	30	20	290	210

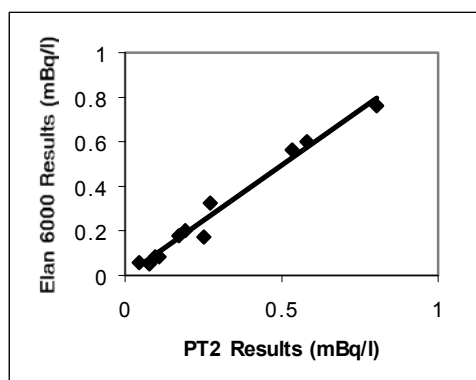


Figure 1: The scatter plot of the results for the fjord samples measured on both systems

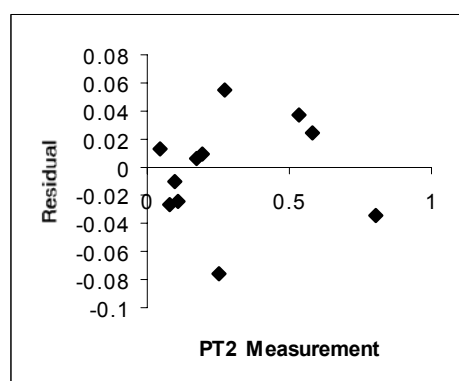


Figure 2: The residuals of Figure 1

Test of using ICP-MS for fast determination of ^{99}Tc in Icelandic Coastal waters

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Introduction

^{99}Tc has been routinely measured in seaweed at the Icelandic Radiation Protection Institute for the past two years. Samples from various locations around the island have been measured dating back to 1998 and a reasonable estimate of ^{99}Tc concentration in seaweed has been obtained. Because of the ocean current system around the island the expected Sellafield peak will appear much later in seaweed than in seawater collected at the East Greenland current, which is the main source of ^{99}Tc in Icelandic coastal waters. Seawater measurements using radio counting methods are in the early stages but as an alternative some experimental work has been done on using ICP-MS for ^{99}Tc measurements. The advantage of using ICP-MS rather than traditional radio counting for seawater is that smaller sample volumes can be used on ICP-MS because of lower limit of detection. For radio counting, seawater samples 200L or larger are needed for low concentration measurements but with ICP-MS the volume could possibly be reduced to 50L. That reduction in sample volume is of great concern both for sampling and sample preparation.

The aim of this experiment was to measure ^{99}Tc in Icelandic coastal water with ICP-MS using simple chemical separation process. Coastal water was chosen because it is easy to sample and has low concentration of ^{99}Tc . The idea was that if Coastal water could be measured then other seawater types would be measurable. The Coastal water would also give information about the correlation between ^{99}Tc in seaweed versus seawater. Emphasis was made to have the chemical separation process as simple as possible to avoid contamination of the sample with metals coming from various reagents.

Material and Methods

Sampling :

Sampling took place in May 2001 when 11 samples were collected from various locations around the island. Sample volume was ranging from 50 to 60 litres. All the samples were Coastal water samples which is Atlantic water diluted with rivers coming from the mainland. Atlantic water brings properties into Icelandic waters from the ocean waters South and Southwest of Iceland. In the north part of the island cold Polar water sometimes mixes with the Coastal water. The cold Polar water brings with it properties originating from eastern part of the Nordic Seas (Norwegian Sea) and West European waters [1] [2]. As a result higher concentration of ^{99}Tc has been measured in samples taken at the north part of the island compared with other parts of the island. The ocean current system around Iceland [3] is shown in figure 1 and the sample locations are shown in figure 2.

To be able to estimate the validity of our results we measured Seawater sample used in intercalibration exercise in BOK-1.2. That sample had been analysed for ^{99}Tc at various Nordic laboratories using radio counting measurements

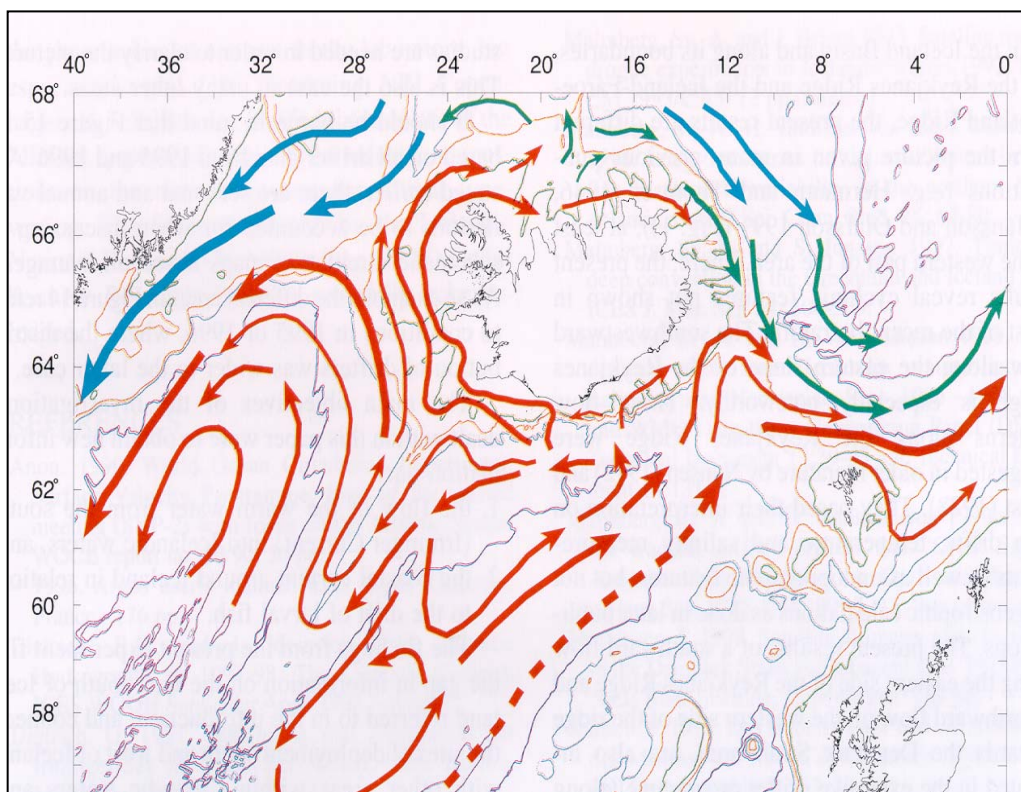


Figure 1. Ocean currents around Iceland. Red arrows: Atlantic water; Blue arrows: Polar water; Green arrows: Mixed water. Polar water has higher concentration of ^{99}Tc than Atlantic water. Even though Atlantic water surrounds the island Polar water and Atlantic water mix to some extent north of the island. As a result, higher concentration of ^{99}Tc have been measured in seaweed there than at other sample locations.

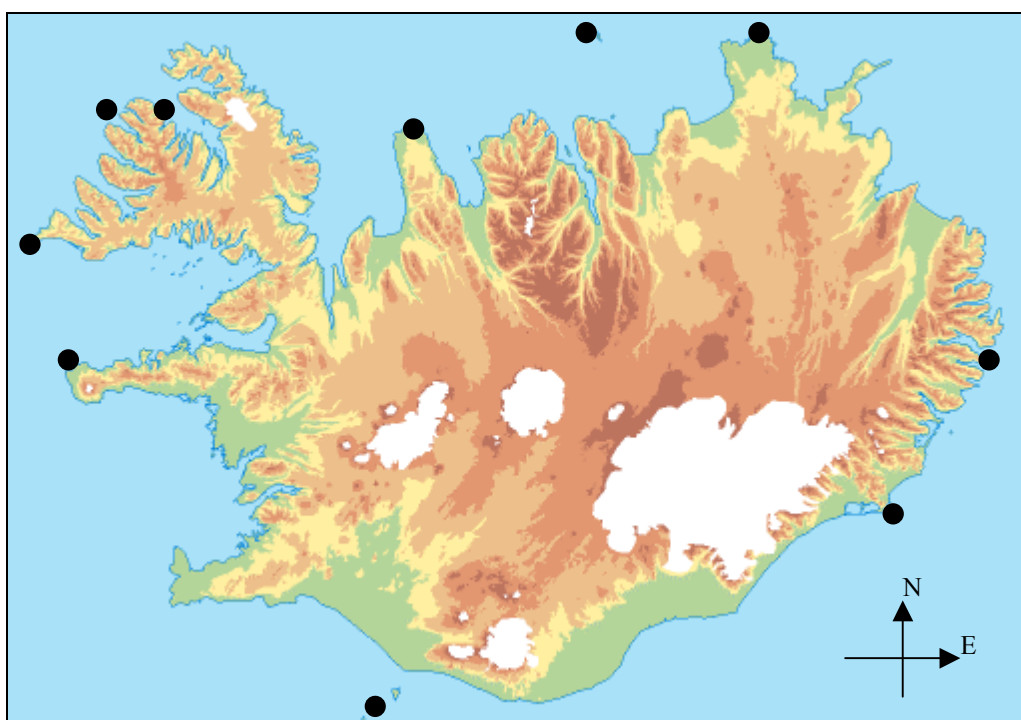


Figure 2. The eleven sample locations are shown with black dots.

Analyses

Sample preparation:

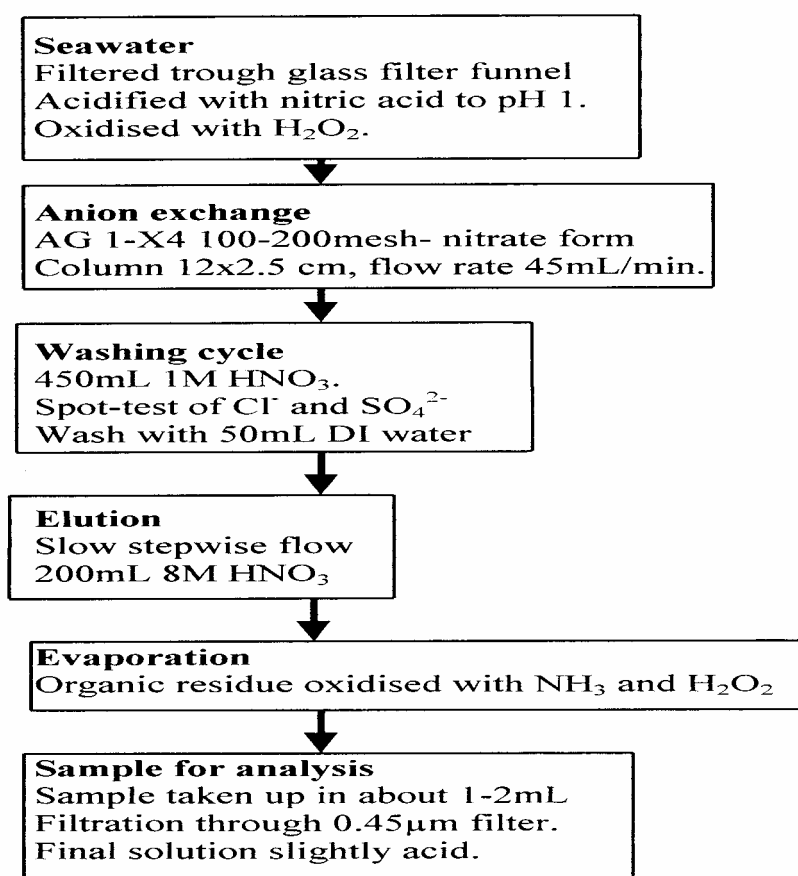
The seawater was filtered through a glass filter funnel (no 3) and acidified with Nitric acid to a pH below 1.5. Small amount of hydrogen peroxide (H_2O_2) was then added. This was done to dissolve various organic complexes and precipitates. The hydrogen peroxide makes the seawater solution oxidising and ensures that all ^{99}Tc is oxidised to pertechnetate form (TcO_4^-). After the acidification the seawater was allowed to stand for few days. The seawater was then pumped through an ion exchange column: 2.5x12cm Bio Rad AG 1-X4 -nitrate form. The nitrate form was used to lower the selectivity of the resin towards various transition metal anions such as Mo and Ru oxianions which are possible interferences in the ICP-MS measurement. Because of the high selectivity of the pertechnat anion towards the active group of the resin the chemical yield is unaffected when the nitrate form is used instead of original chloride form. The column is then washed with 450mL of 1M HNO_3 and a spot test of chloride and sulphate performed with AgNO_3 and BaCl_2 . The column is then washed with 50mL of distilled water and the Tc is then eluted with 200mL of 8M HNO_3 . The washing with 1M HNO_3 removes various anions retained by the resin. Of special concern are Mo oxianions. The spot test of chloride and sulphate is simply to check if any traces of Cl^- and SO_4^{2-} persist in the column. Most transition metal chloro-complexes should be removed in the washing and when sulphate is finally removed from the column metals like Ti, Zr and Th (which form anionic complexes with sulphate) are probably eluted. 8M HNO_3 is used for elution to minimise the amount of organic residue washed out from the column when eluting the Tc. We found that the amount of organic residue increases when stronger solutions of HNO_3 are used indicating that the nitric acid oxidises the resin. Some organic material coming from the seawater is also retained on the column. The elute is then evaporated to 0.5mL and organic residue is then oxidised with hydrogen peroxide in ammonia solution. The organic material needs to be removed for the sample to be suitable for ICP-MS measurement. Oxidation with hydrogen peroxide in ammonia solution is quite rapid and effective and both reagents are extremely clean. After oxidation excess ammonia is simply evaporated resulting in final sample volume around 1-2mL which was then filtered through 0.45 μm filter. The final solution was then made slightly acid with HNO_3 . During the process only Supra pure HNO_3 was used. A flow chart of the sample preparation is shown below.

The time taken to prepare the sample after the seawater has been pumped through the column is around 5-6 hours, the evaporation of the Tc eluent being the most time consuming. The whole process takes about 26-28 hours. A flow chart of the separation process is shown below

Unlike most of the sample preparation methods for Tc reported in the literature we decided not to estimate chemical recovery using $^{99\text{m}}\text{Tc}$. The reason was that the samples were thought to have such low concentration of Tc that spiking the sample could lead to measuring mostly contamination from the $^{99\text{m}}\text{Tc}$ if the samples would fall below limit of detection. When developing the method three 50L seawater samples were spiked with $^{99\text{m}}\text{Tc}$ and the estimated recovery was about 90%.

We also estimated the chemical recovery by spiking 5L of tap water with 1mL of 10ppb solution of ^{99}Tc and running the solution through the process and measure the sample on ICP-MS. The estimated recovery was 96%.

Few samples were spiked with 1mL of 10ppb solution of Re. This was done to check the suitability of Re as an internal standard for the ICP-MS measurement and as an estimate of chemical recovery.



Mass spectroscopy measurements

Multicollector double focusing magnetic sector ICP-MS instrument (Nu-Instruments, Wrexham, Wales) was used for the analytical part of the project. This instrument has 12 Faraday detectors and 3 Ion collectors (22 dynodes) arranged at fixed positions along the focal plane of the instrument. Ion beams are collimated onto the detectors by two electrostatic lenses (Friedman lenses) to produce flat peaks that are counted simultaneously in the selected detectors. This instrument can be used in a static mode within a narrow range of the mass spectrum or in a dynamic mode where different counting cycles can be performed sequentially over the entire mass range.

Sample introduction is one of the most critical operations in ICP-MS work. In this study we used an Aridus microconcentric nebuliser due to its low sample consumption of 45 microlitres/min which allows about 20 minutes working time with a sample of only one mL. Nebulisation of solutions containing the volatile pertechnetate follows similar paths as the well known nebulisation of perosmate solution. The Aridus nebuliser which contains a desolvation unit has to be used at a very low desolvation efficiency when analysing these elements in acid solution. A combination of high nebuliser gas flow mixed with a trace of nitrogen and low desolvation gas flow gives the highest long term signal stability at some expense of sensitivity.

When developing a method for measuring the Tc it became evident that Mo had been insufficiently removed so it was not possible to use the ion counters for measuring the Tc. The reason was that the ion counters trip on mass 100 due to high molybdenum concentration. The molybdenum concentration in the samples were ranging from 110 to 450ppb but need to be below 100ppb for using the ion counter. We therefore measured the Tc on the Faraday detectors monitoring masses 98, 99, 100 101 and 102. Using the Faraday detectors instead of the ion counters lowers the sensitivity by factor 10.

Standard solution of technetium were made from the NIST SRM 4288A reference solution. A set of standards in the interval 0.001-50ppb Tc were used for calibration of the instrument. The lowest reproducible standard on the Faraday detector was 0.01ppb (10ppt). Standard solutions for Mo, Ru and Re were made from NIST- traceable 1000 ppm solutions produced by High Purity Standards, Inc. All standards and a blank solution were made in acid washed polyethylene bottles. Standards were diluted with deionised water of 18,3 mOhm resistivity and made 1% vol in fuming nitric acid produced from analytical grade nitric acid by sub-boiling distillation in Teflon apparatus. Preparation of sub ppb reference solutions is a dedicated clean-lab task and all standards and blanks were made fresh for the day.

Re concentration in the seawater was so high that the amount of standard we added to the sample was not enough to mask the natural Re concentration. Re in the seawater samples could therefore not be used as an internal standard. To check the stability of the nebuliser calibration lines for standard Tc solution and standard Tc/Re solution were made. The results from the two calibration lines were virtually similar which indicated that there was no need to use internal standard with this instrumental setup.

The analysis of the seawater samples were performed in a sequence of 20 measurements consisting of 10 one second counting cycles, in all 200 sec counting time. This gives standard deviation of $3,5 \times 10^{-6}$ for the blank measurements in all the analytical routines.

Analysis of unknown samples were made by sequential counting of a blank, standard, unknown, blank etc. The analysis of the blank were sometimes repeated for testing of the 3 min washing time used was adequate

Results and Discussion

^{99}Ru is a major isobaric interference at mass 99 and because of high Mo concentration in the samples the isobaric interference from $^{98}\text{Mo}^1\text{H}$ could not be excluded. We found by measuring Mo solutions of the same strength as in the samples (110-460ppb) that the ratio of Mo/MoH was 0.002. It was confirmed that this ratio was insensitive to the addition of ammonia and acid strength.

Other interferences such as $^{59}\text{Co}^{40}\text{Ar}$, $^{49}\text{Ti}^{50}\text{Ti}$ and $^{64}\text{Zn}^{35}\text{Cl}$ were found to have sub ppb concentration and therefore excluded.

Estimate of the corrected signal for ^{99}Tc therefore involves, in addition to the background subtraction, correction for contribution from Ru and $^{98}\text{Mo}^1\text{H}$. Correction factors were derived from actual analysis of Ru and Mo solution which eliminates the need for special detector calibration for all channels used and mass fractionation correction. The masses ^{101}Ru and ^{98}Mo are used for this correction including reduction of mass 98 due to minor contribution from Ru. Analysis of Ru and Mo were based on the intensity of ^{101}Ru and ^{98}Mo . Limit of detection was estimated 3σ for the blank count at mass 99 or 0.04Bq/m^3 . Results for the estimated ^{99}Tc concentration are shown in figure 3.

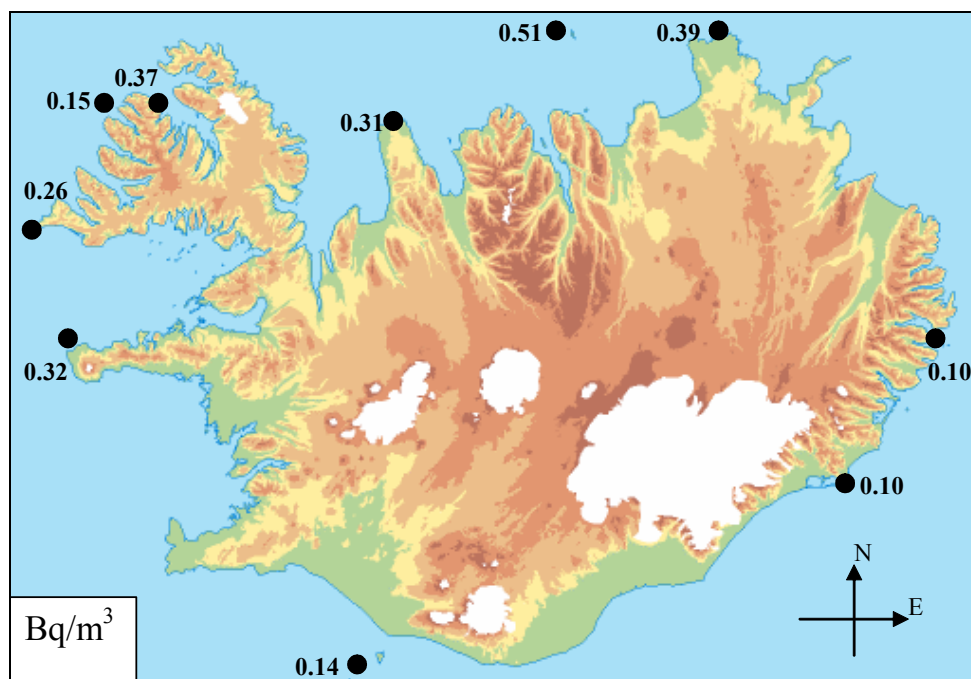


Figure 3. ^{99}Tc concentration as measured in Icelandic Coastal waters 2001. Estimated uncertainty is $\pm 0.04 \text{ Bq/m}^3$

These results are considerably higher than earlier results from the East Greenland current (approx. 0.08 Bq/m^3) and the Faeroe Island (approx. 0.02 Bq/m^3) measured at the Risø National Laboratory Denmark 1999. These results were obtained by radio counting method [4].

The observed values are similar to values reported from the Atlantic (west) coast of Ireland in 1998 [5]. Our estimate of ^{99}Tc concentration in the intercalibration seawater sample was also approximately five times higher than measured by radio counting at other Nordic laboratories.

The difference in ^{99}Tc concentration measured here from the south to the north coast is similar to what we have observed in Seaweed measurements using radio counting. That indicates that if these values are truly too high it is probably so by a constant amount at all sample locations and the samples are not just randomly too high.

Conclusions

Even though these results are considerably higher than those reported by radio counting methods we have not found any interferences that could count for such high signal apart from those discussed above and have been corrected for. The values measured show the same pattern as seaweed measurements with higher concentration on the north coast and lower concentration on the south coast which means we are not measuring a random signal.

The separation process used was relatively fast and simple and the samples were well acceptable for ICP-MS measurements. The relatively short sample preparation time and simplicity of the process is a property which is essential when large number of samples need to be measured in a short period of time for instance in emergency situation. High Mo concentration was disappointing and slight modification of the process to reduce Mo concentration would be helpful and increase the measurements sensitivity.

It is our belief that further work is necessary to work out those problems encountered in our measurements. They should however be solvable.

Acknowledgements

In addition to support from the NKS BOK 2.2 project special thanks goes to Gylfi Sigurdsson at the Nordic Volcanological Institute

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Methodology for Defining Exemption Levels of Radionuclides in Timber and Wood Products

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Introduction

Timber and wood products are major forestry products in many countries where they are used both for needs of the local population and for export in other countries. Many countries import wood from neighbouring countries. As a result of radiation emergencies, nuclear weapons tests and the long term activities of a range of nuclear installations, radionuclides have been deposited in regions where there is an intensive forest industry. The best known example is the forests that were contaminated with long-lived radionuclides such as ^{137}Cs , ^{90}Sr and ^{134}Cs after the Chernobyl accident.

Another source of contamination is from global radioactive fallout after nuclear weapons tests, but the level of contamination is much lower than that from the Chernobyl accident. For instance, the level of ^{137}Cs in wood in Sweden is about $2\text{--}5\text{ Bq kg}^{-1}$ from fall-out. Global values are very similar to the Swedish levels. In contrast, the level of ^{137}Cs in Swedish wood due to Chernobyl is around 50 Bq kg^{-1} . Levels in wood from the countries of the Former Soviet Union (FSU) are about an order of magnitude higher. Forests in the Ural region of the Russian Federation were also contaminated during industrial Kyshtim accident in 1957 with radionuclide mixture including long-lived ^{90}Sr .

There is concern about radiation exposure of people from radionuclides in wood, especially after the Chernobyl accident. In January 1996 a system of international certification of radiation safety for commodities was implemented whereby the activity concentration of ^{137}Cs and ^{90}Sr in timber to be exported has to be specified. However, specification of the level of radionuclides in wood does not necessarily address the potential radiological exposure from such wood. Such exposure would depend on the specific radionuclide and also the use of the wood. There is no uniform radiological standard that is universally applied in all countries. There is a concern that the levels of exposure due to the concentration of, for instance, ^{137}Cs in wood are such that it may limit both the domestic use and the export of wood to certain countries, depending on the radiological standard in a country. This has important economic implications to the exporting countries in terms of loss in revenue and employment opportunities, but also to the importing countries who must seek raw material elsewhere at greater expense.

There is therefore a need to increase the confidence of both the regulators and end-users in the radiological safety of wood produced in radioactively contaminated forested areas. It is in the interest of both exporting and importing countries to optimise the trade of wood, taking the eventual radiological consequences into consideration.

It is therefore proposed that a generic methodology be developed that could be used to determine the long-lived radionuclide level in wood that would result in public exposure not exceeding the radiological standards in the country under consideration. The development of such a methodology is the aim of this document. The approach to develop the methodology is

based on a previously developed safety assessment approach [IAEA, 1999] to ensure that the end result is transparent, understandable and credible. As the exposure would be a function of the use of the wood, the methodology is based on a number of scenarios that were used to develop appropriate dosimetric models and derive Dose Conversion Coefficients (DCC). Thus the ultimate aim of this document is to provide a tool that can be used to calculate the dose that would arise from a certain long-lived radionuclide (mainly, ^{137}Cs and ^{90}Sr) level in wood, or, *vice versa*, the radionuclide level in wood that would correspond to a certain dose standard.

Generic methodology

In this study a methodology of determination of the exemption levels of long-lived radionuclide concentrations (mainly, ^{137}Cs and ^{90}Sr) in locally produced or imported wood and its products is developed in the following way. For major technological sorts of wood and wood products, the main technologies of industrial processing in different branches of the wood industry are selected. It is assumed that the industrial processes under consideration would be carried out in the country where wood was harvested but outside the area contaminated with radionuclides or in countries importing wood, which are expected to be technologically advanced. Operations implied by the selected technologies which would lead to the exposure, both external and internal, of industrial workers by radionuclides contained in the wood, are selected and used for description of exposure scenarios based on the treatment of the wood or wood products.

The scenarios are then used to define the mathematical models of human exposure, provided with site-specific parameters and used for calculation of the annual effective dose in workers. For the assessment of workers exposure, concentration of radionuclides in wood, geometrical conditions of the external exposure and volume of inhaled air for calculating internal exposure as well as time spent in conditions under consideration are used. Total annual dose is determined as the sum of the external dose received during the year and committed internal dose caused by the inhalation of radionuclides during the same period. Ratio of the total annual dose to radionuclide concentration in the wood is defined as the Dose Conversion Coefficient (DCC). The unit of DCC is $\text{mSv}\cdot\text{Bq}^{-1}\cdot\text{kg}$.

For members of the public different scenarios of utilisation of the products made from locally produced or imported wood are considered (living in a timber house, in-house furniture, etc.). These scenarios are used for development of mathematical models of human exposure, both external and internal, and calculation of the dose in members of the public considered in each particular scenario of the wood utilisation. Total annual effective dose originating from different kinds of utilisation of the wood products by the general public and relevant DCC values are calculated for further determination of exemption levels of ^{137}Cs and ^{90}Sr concentration in wood.

The values of DCC are calculated for wood and major wood products and technologies used in the hypothetical wood producing or wood importing country, separately for workers and for members of the public. For each kind i of wood product the largest value of DCC is selected and used for calculation of Exemption Concentrations (EC) of particular radionuclide r activity, separately for workers and members of the public:

$$\text{EC}_{ir} = E_e / \text{DCC}_{ir}, \text{ Bq/kg},$$

where: E_e , mSv, is the national annual dose level for exemption from regulatory radiological control for practice or intervention exemption level in the case of intervention, accordingly.

If exemption dose levels for non-radiological workers and for members of the public are equal only one value of EC_{ir} is considered for particular imported product and radionuclide. If these dose criteria and appropriate values of EC_{ir} are different then the lowest value is selected for radiological control of wood import.

In some countries, especially in countries of the former USSR severely contaminated with ^{137}Cs and ^{90}Sr due to the Chernobyl and Kyshtim radiation accidents, different permissible levels of wood contamination with radionuclides were established for different kinds of wood utilization (dwelling construction, general construction, paper production, etc). This approach is applicable if portions of contaminated wood are certified and thus traced during their movement and processing within the country with strict system of radiological control.

But similar approach is hardly applicable for wood imported in other countries where its unrestricted use can be assumed. For the later, generic exemption levels of radionuclide concentration in wood EC_{ir} should be derived as described above under conservative assumption of unrestricted use in the range of scenarios considered in the present document. For conditions of wood import it is important to take into account that the bulk of imported wood is usually harvested in different forest areas, most frequently non-contaminated ones, and only small fractions of imported wood are contaminated at the EC_{ir} level and above.

As only generic models and best estimates of their parameters are used for assessment of human doses and derived parameters as Dose Conversion Coefficients and Exemption Concentrations these should be mainly applied for screening purposes. Thus, if the measured radionuclide concentration in wood exceeds EC it is recommended to undertake more detailed assessment. In this case, it is necessary to specify local model parameters, include them in dose calculations instead of generic parameters and perform site-specific calculations to be used for decision making on the possible use of contaminated wood.

SYSTEM DESCRIPTION

The system under consideration, ie. the utilisation of timber and wood products contaminated with the long-lived radionuclides ^{137}Cs and ^{90}Sr , can be defined as shown in Figure 1. For the purpose of this document 'utilisation' includes transportation, storage, processing, production of commodities, usage of these commodities and disposal of used commodities and relevant by-products. Figure 1 indicates that timber and wood products can be transported from the forest or from the site of primary processing to the site where they are further treated and/or utilised (including export to other countries) in four forms, as follows.

1. Raw timber (round wood with and without bark)
2. Sawn wood
3. Wood pulp for paper manufacture
4. Waste wood in the form of both wood and bark chips.

Following delivery of timber and wood products these four basic products can be used in a variety of ways, in both industrial and domestic situations. Five primary industrial uses can be identified, as follows.

1. Processing of raw timber in a saw mill
2. Use of raw timber and /or sawn wood in construction
3. Use of sawn wood in furniture manufacture
4. Use of raw timber and wood pulp in paper and cardboard manufacture
5. Use of waste wood and bark chips in biofuel plants.

An important by-product of the use of waste wood and bark chips for biofuel energy generation is wood/bark ash in which radionuclides (such as ^{137}Cs) can become significantly concentrated. The disposal of ash, usually to landfill, is a problem which can also be considered within the industrial context. Hence, there are six distinct radiological exposure scenarios which can be identified under the broad heading of 'Industrial Exposures', as shown in Figure 1.

Following industrial processing of wood and associated products such as wood pulp, a variety of finished products is produced, each of which has the potential to cause radiological exposures to end-users in the domestic situation, as described below.

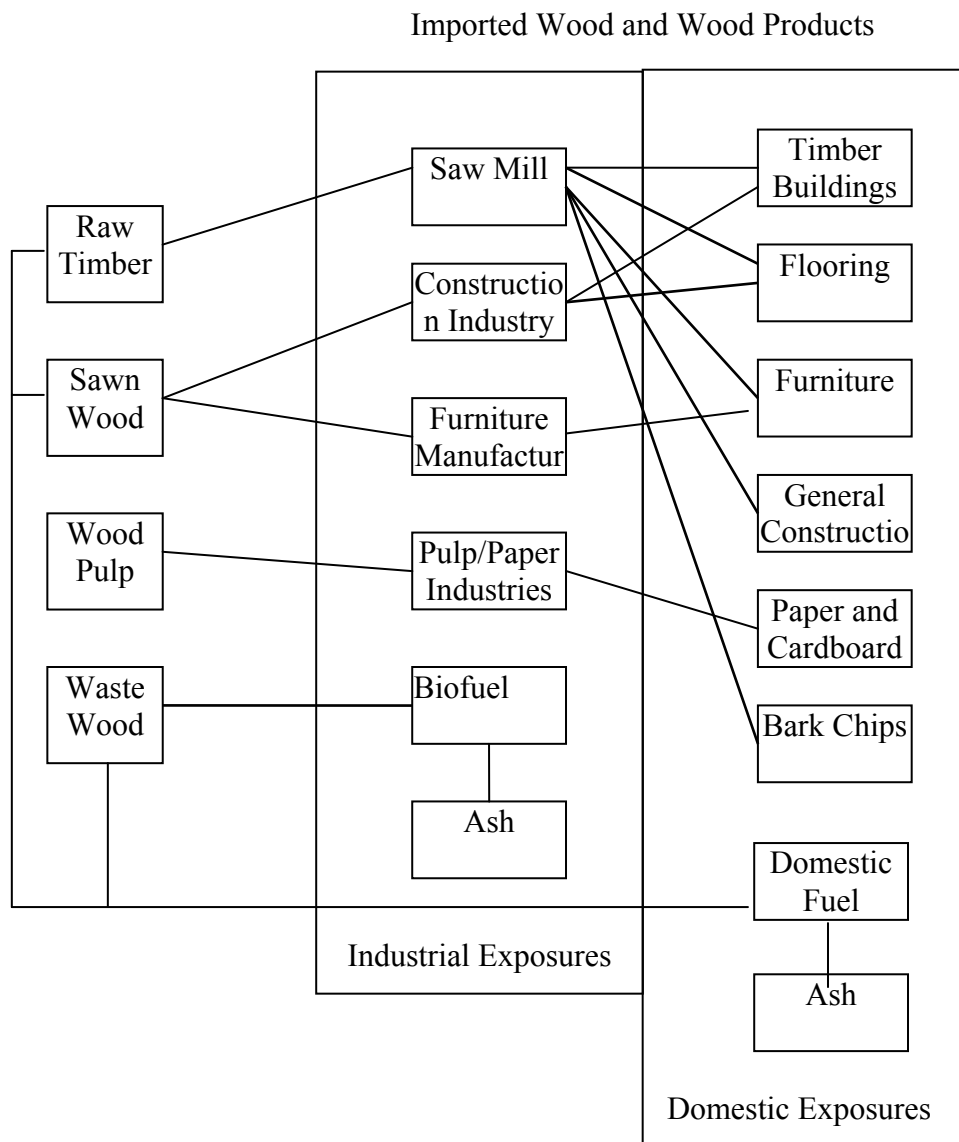
1. Timber houses and other buildings constructed from sawn wood and manufactured construction materials such as plywood and chipboard.
2. Wooden floors constructed from sawn wood and manufactured materials.
3. Furniture constructed from sawn wood and manufactured materials such as plywood and chipboard.
4. Paper and cardboard manufactured from wood pulp.
5. Bark chips, used in domestic gardens and children's play areas.
6. Processed wood chips manufactured from waste wood and used for domestic heating.

Some of these products, particularly the relatively high value products such as furniture, pulp and paper, may even be exported to a third country, thus extending the potential geographical range over which radiological exposure may potentially occur.

Hence, the system to be considered in any generic methodology to calculate *ex situ* doses deriving from contaminated wood encompasses its industrial and domestic utilisation in the country of origin and/or in a country importing contaminated wood. One feature of the system to be considered is that all potentially exposed people, whether in an industrial or domestic situation, are not registered radiation workers and, in radiological protection terms, are to be considered as members of the public.

The exposure that a person receives is a function of the use of the wood or wood product. The radiological significance of each technology and relevant exposure pathways, whether industrial or domestic, needs to be considered individually. To achieve this, detailed scenarios of human exposure must be defined for each technology and relevant exposure pathways.

Figure 1



Results and Discussion

Calculation and Derivation of Reference Levels

A set of reference values for wood from a contaminated forest was calculated, based on the scenarios. The results are listed in Table 1 and 2 below

Table 1. Example Reference Levels for ^{137}Cs in Dry Wood

Scenario	1 mSv Dose Level		
	Example reference levels for ^{137}Cs in dry wood (Bq kg^{-1})		Occupational exposure factor (h)
	With bark	Without bark	
Resident in timber house	Not relevant	$3.4\text{E}+7$	$4.0\text{E}+3$
Paper and cardboard	Not relevant	$1.4\text{E}+8$	$2.0\text{E}+3$
Worker in pulp industry External exposure from: ash pile*	$1.0\text{E}+3$	$3.9\text{E}+3$	$1.0\text{E}+2$
liquor holding tank	$4.0\text{E}+3$	$4.0\text{E}+3$	$1.0\text{E}+2$
Worker in saw mill industry stock piled wood	$5.3\text{E}+6$	$7.5\text{E}+6$	$2.0\text{E}+3$
dust inhalation	$2.4\text{E}+7$	$3.33\text{E}+7$	$2.0\text{E}+3$
Worker in forest fertilised with ash External Inhalation	$9.3\text{E}+3$	$1.3\text{E}+4$	$1.5\text{E}+3$
Garden fertilised with ash External Inhalation Ingestion	$2.4\text{E}+4$	$3.3\text{E}+4$	$1.0\text{E}+3$

* This also applies to ash generated during bio-fuel heating

There is a linear relationship between the reference levels, the dose and the occupational factor listed in Table 1. Thus, in the case of dose limits or constraints different from that in Table 1, or different occupational factors, a different set of reference levels can be calculated by using an appropriate scaling factor.

Approach is that all the wood being processed is from a contaminated forest. However, blending would result in a (linear) reduction in exposure. Similarly, for timber house.

Limiting factor is the exposure from ash produced during pulping, or bio-fuel heating.

Exposure from ash were calculated assuming 'normal' practices. Exposure can be decreased by remedial actions to, for instance, reduce the level of resuspension, or by limiting the time a single person is working with the ash.

Table 1. Areal Activity of ^{137}Cs in Soil Estimated from Example Reference Levels for ^{137}Cs

Scenario	1 mSv Dose Level	
	Example areal activity of ^{137}Cs in soil (Bq m^{-2})	
	Based on wood with bark	Based on wood without bark
Resident in timber house	Not relevant	(0.7 to 3.4)E+10
Paper and cardboard	Not relevant	(0.3 to 1.4)E+11
Worker in pulp industry External exposure from: ash pile* liquor holding tank	(0.2 to 1.0)E+6 (0.8 to 4.0)E+6	(0.8 to 4)E+6 (0.8 to 4.0)E+6
Worker in saw mill industry stock piled wood dust inhalation	(1.1 to 5.3)E+9 (0.5 to 2.4)E+10	(1.5 to 7.5)E+9 (0.7 to 3.3)E+10
Worker in forest fertilized with ash External Inhalation	(1.9 to 9.3)E+6	(0.3 to 1.3)E+7
Garden fertilized with ash External Inhalation Ingestion	(0.5 to 2.4)E+7	(0.7 to 3.3)E+7

- This also applies to ash generated during bio-fuel heating

The ranges in the above table were derived assuming a transfer factor from soil to wood of $1.0\text{E-}3$ to $5.0\text{E-}3 \text{ Bq kg}^{-1}$ per Bq m^{-2} .

Acknowledgements

This study was supported by Nordic Nuclear Safety Research Program and is a part of an IAEA investigation. The collaboration in this context with M. Balonov, G. Shaw and A. Venter is greatly acknowledged.

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Appendix: Related paper from the EKO-2 project

During the process of publishing and printing the final report of the EKO-2 project, one of the project papers was left out by mistake. It was to have been included in NKS(97)FR5

Ecological half-lives of radioactive elements in semi-natural systems. This missing paper is included in the compilation report published now, since it is relevant for other work being reported and the missing paper has been referred to in other papers. This was decided in consultation with the leader of the former EKO-2 project. It was also decided to place this paper in an appendix to the report, since it has not been updated since being written in 1997 and its style is different from the other papers.

Transfer of Radiocaesium from Soil to Vegetation and to Grazing Lambs in Iceland

Icelandic part of the NKS/EKO-2.1 project

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1 INTRODUCTION

The Cs-137 in the Icelandic terrestrial ecosystem is almost entirely from the nuclear weapons tests carried out in the atmosphere until the early sixties. Less changes have therefore been observed in radiocaesium concentration during the RAD-3 and EKO-2.1 project periods (1990-1997) than in those countries which received a significant amount of fallout from the Chernobyl accident. Due to the relatively stable concentrations, more emphasis was moved from studying changes in concentration with time, to studies of the variability of the concentration and how the variability could be explained.

The Icelandic part of the EKO-2.1 project has been conducted at two different locations during the current NKS project period. In 1994, the same location was used as in the previous period (RAD-3), the experimental farm at Hestur in the Borgarfjörður area. In 1995 - 1997 the project was carried out at another experimental farm, Stóra Ármót in the Selfoss area. These two sites differ in many respects as can be seen in the description of the experimental sites and Table 1. One of the aims in the Icelandic team has been to cover as broad spectrum of types of land as possible, within the project framework. This is mainly due to the fact that in Iceland, sheep are mostly free ranging during the summer and thus have access to abundant types of uncultivated land. They tend therefore to vary their diet considerably during each grazing season as they select species with the highest nutritional content and quality at any given point of time. It is thus obvious that grazing research, conducted in small experimental pasture cannot be considered as representative for conditions in a larger area in a diverse environment. Therefore the decision was made to move the experimental plot to another location, as the budget did not allow the project to be conducted at two sites at the same time. In the previous period, the RAD-3 experiment was also being conducted at Auðkúluheiði, a common highland grazing district in north central Iceland. Thereby it was felt that a wide range of some typical land types used for Icelandic

sheep grazing was covered. It is our opinion that more information is gathered by changing the sampling place, rather than continue on the same through the EKO-2 period. This will also give broader spectrum of data for interpretation.

1.1 SHEEP FARMING IN ICELAND

The Icelandic farm animal population, including the sheep, was brought to the island some 1100 years ago during the time of settlement. Very little has been imported since then. The farm animals derives from Norway and do belong to the North European short tailed group of sheep (Adalsteinsson 1981). This same sheep breed is still the only to be found, producing both meat and wool. Winterfed sheep are about 450.000 (1996).

Since the time of settlement sheep have been released into the highlands. Depending on vegetation, this usually happens in mid June - late July. They are gathered in September and kept on lowland pastures until the lambs are slaughtered in September and October. During the winter all sheep are housed, fed almost solely dry hay. The ewes give birth in May thus the lambs are about 4-5 months old when slaughtered.

In the summer the sheep are free ranging. The highlands are uncultivated, vast parts are eroded but pastures are also covering considerable areas where the sheep can choose from wide variety of mountainous vegetation species. Being eager browser and highly selective in its diet, they tend to select the species at a given point of time that makes it possible to optimize their energy and nutritional requirements. The Auðkúluheiði experimental site, used in the previous RAD NKS experimental period can be looked at as a typical highland pasture. Keeping sheep in their free ranging highland pastures is laborious in the autumn when gathering takes place. This has forced more and more farmers to consider lowland summer grazing. Lowland pastures will thus become more important for sheep farming than they used to be. Both the Hestur site and the Stóra Ármót site are typical for lowland grazing areas, Hestur as wetland and Stóra Ármót as dryland.

2 DESCRIPTION OF EXPERIMENTAL SITES

2.1 HESTUR

The study area is a 12 ha uncultivated lowland mire. It slopes a little from east towards west, and in a small part of the area there is an elevated gravel ridge with well drained soil. The uppermost and the gravel parts are the driest areas. The surface is hummocky, except for the gravel ridge. Wetland vegetation is dominant, most common plant species are *Carex nigra* (common sedge), *Equisetum palustre* (marsh horsetail), *Calamagrostis stricta* (smallreed) and *Carex panicea* (carnation sedge). In the uppermost parts of the pasture, grasses like *Deschampsia caespitosa* (tufted hair-grass), *Agrostis capillaris* (common bent-grass) and *Poa pratensis* (common meadow-grass) are frequent. The most common species on the gravel

ridge are *Festuca richardsonii* (creeping fescue), *Festuca vivipara* (northern fescue) and *Carex bigelowii* (stiff sedge). Due to differences within the pasture, it dealt into five sub areas, area three being the wettest, two and four intermediate and one the driest. The fifth area was dry gravel ridge, sparsely vegetated. Soil and vegetation sampling was done within these sub areas and treated separately.

2.2 STÓRA ÁRMÓT

The study area is a 12 ha uncultivated lowland pasture. The surface is level, but considerably hummocky. With respect to vegetation, the whole area is homogenous in appearance. Dominating species are *Racomitrium hypnoides* (moss), low growing shrubs like *Salix lanata* (woolly willow) and *Vaccinium uliginosum* (bog bilberry), grasses like *Festuca vivipara* (northern fescue) and *Poa pratensis* (common meadow-grass). Other abundant species are *Calluna vulgaris* (ling, heather), *Carex bigelowii* (stiff sedge) and *Carex nigra* (common sedge). The pasture has been divided into three very similar looking sub-areas, where soil and vegetation has been sampled.

The pasture was fenced in 1995 when the experiment was moved from Hestur. Before it was used as common rangeland, mostly for horses but also for sheep. The soil depth at the site varies. The bedrock below is an 8000 years old lava, and it can be seen in some parts of the pasture. In other parts the soil is at least 2 m thick.

Figures 1 and 2 (see Appendix) show location of the experimental sites and vegetation map of Iceland. The experimental sites are located in the two main agricultural production areas in Iceland. Some site statistics can be seen in Table 1 and meteorological data can be found in Figures 11 and 12.

Table 1. Site statistics for Hestur and Stóra Ármót.

	Hestur	Stóra Ármót
location (lat.&long.)	64°25'N 21°38'W	63°60'N 20°55'N
size (km ²)	0.12	0.12
altitude (m.a.s.)	20	20
dist. to ocean (km)	5.5	19.5
type of climate	coastal	coastal
soil type	peat, gravelly	freely drained mineral soil
vegetation type	lowland mire	lowland heath type

Meteorological data for Hestur research station was collected at Hvanneyri (64°34' N 21°46' W), 6,5 km away. Data for Stóra Ármót was collected at Eyrarbakki (63°52' N 21°09' W) 17 km away (Figures 9 – 10).

3 MATERIALS AND METHODS

3.1 VEGETATION AND SOIL

Three different sampling schemes were used:

- a) individual plant species
- b) biomass, composite plant samples collected from microplots evenly distributed over all of the experimental area
- c) composite plant samples collected from each microplot used for soil sampling

3.1.1 INDIVIDUAL PLANT SPECIES

Individual plant species were taken as follows:

At Hestur: Common bent-grass (*Agrostis capillaris*), tufted hair-grass (*Deschampsia caespitosa*), fescue (*Festuca* sp.), common sedge (*Carex nigra*), lyngbye's sedge (*Carex lyngbyei*), stiff sedge (*Carex bigelowii*) and common cottongrass (*Eriophorum angustifolium*).

At Stóra Ármót: Common bent-grass (*Agrostis capillaris*), tufted hair-grass (*Deschampsia caespitosa*), fescue (*Festuca* sp.), stiff sedge (*Carex bigelowii*), woody willow (*Salix lanata*) and heather (*Calluna vulgaris*).

These samples were chosen to get better samples of what is known to be preferred by sheep, as the biomass samples do not represent the actual feed. Sheep are highly selective in their grazing, both with respect to plant species, but also to plant parts (Arnold, 1966a&b; Arnold et.al, 1966; Bazely, 1990, Thorhallsdottir and Thorsteinsson, 1993). They can also be used to explain deviations from the estimated intake of radiocaesium, especially in the case of species with relatively high uptake of radiocaesium and which are eaten by (at least some of) the sheep.

3.1.2 BIOMASS

The “Double sampling” method (Brown 1954) was used. The method is based on eye estimate of biomass, combined with clipping and weighing, or visual weight estimate, of the herbage. Quadrants were placed on the ground at approximately equal intervals on already fixed transects. Biomass was estimated in the quadrant by eye estimate and every third or fourth quadrant was clipped at ground level. All plant species within the quadrant, except northern crowberries (*Empetrum nigrum*) were clipped. Mosses were also excluded. Each sample was dried at 60°C and weight correlated to estimated biomass. The resulting linear formula was then applied to convert data from the estimated quadrants into biomass. To minimise observational bias, every estimation was done by at least two persons. R^2 found for each estimator was usually 0.8 or higher.

At Hestur the estimation was based on an approximate number of fifty 0,1 m² quadrants. Similarly the number at Stóra Ármót was hundred and fifty and the quadrant size was 0,05 m².

Additionally adjacent pasture, ungrazed but similar in all other respects, was harvested to get an estimation on how much might have been removed by grazing during the summer. Clipped frames (0,05 m²) in both pastures were compared with Mann-Whitney U/Wilcoxon rank sum test and Kolmogorov-Smirnov test. These samples give the best estimate of the average concentration in the whole area, but do not represent what is preferred by the sheep.

The vegetation collected from the total biomass estimation was used for nutritional analyses, after grinding through a 1 mm sieve.

3.1.3 SOIL AND COMPOSITE PLANT SAMPLES

Samples were collected and analysed for radiocaesium using the method agreed upon during the NKS / RAD-3 project (1990-1993). Frames, 0,5 m each side, were thrown at random in each plot to be sampled. First the vegetation above the grass mat in the selected microplot was cut and collected (composite plant samples). Then three soil cores were collected within the frame to a minimum depth of 10 cm. Each core was divided into 5 cm thick slices and the three slices corresponding to the same depth layer mixed together. During part of the sampling the random throwing of the frame was abandoned and the frame placed on selected spots. This is due to how inhomogeneous the soil is (Figure 3).

All vegetation was dried at 60°C, ground and passed through 1 mm sieve before radiocaesium analysis.

3.2 OTHER SAMPLES

3.2.1 HAY

Samples were taken randomly in the barn. It represents the same hay fed to the sheep while housed.

3.2.2 FAECES

Faecal samples were taken directly from the rectum or collected by placing collection bags and harnesses on the sheep for at least 6 hrs. This eliminates the risk of getting the samples contaminated by soil or other particles and also makes it possible to relate every sample to an individual animal.

All samples were dried at 60°C, ground and passed through 1 mm sieve before radiocaesium analysis.

3.2.3 MILK

Milk was collected during some of the sampling days. This was done by dosing the ewes intramuscularly with the hormone ocytocin and milking two minutes later.

3.2.4 Lamb meat

Right foreleg was taken the time of slaughter, usually at the end of September or in the beginning of October. Samples were kept at -18°C until they could be analysed for radiocaesium.

3.2.5 FUNGI

Fungi in diet was determined by counting spores in faeces, based on method given by Morten Strandberg (Strandberg & Knudsen 1994). Mushroom species and corresponding spores were identified from specimens collected within the experimental site.

The faecal samples were taken as described previously, without risk of contamination by earth particles or spores not ingested by the animal. The screening work was done on dried and milled samples (60°C and milled through 1 mm sieve) by dr. G.G. Eyjolfssdottir at the Icelandic Institute of Natural History (Eyjolfssdottir, 1997).

3.3 ANIMAL PERFORMANCE

Animals were weighed four times during each summer: When the sheep were released (approx. 20th June), at end of July, at end of August (or beginning of September) the day before slaughtering (late September or beginning of October).

3.4 GRAZING INTENSITY

The number of animals used has varied between years. In 1994, when the experiment was conducted at Hestur, the available herbage was considerably higher than at Stóra Ármót. Hestur could thus support more animals per ha than Stóra Ármót as became apparent in 1996 (Figure 17). Therefore the number was reduced as can be seen in Table 2.

3.5 RADIOCAESIUM AND NUTRITIONAL ANALYSES

3.5.1 RADIOCAESIUM MEASUREMENTS

Radiocaesium measurements were carried out at *Geislavarnir ríkisins* using gamma spectrometry (HPGe detectors). Some measurements were also carried out at other laboratories participating in the EKO-2.1 sub-project.

3.5.2 NUTRITIONAL AND CHEMICAL ANALYSIS

All nutritional and chemical analysis was done at *Rannsóknastofnun landbúnaðarins* on the samples taken for total biomass estimation. They have been collected four times during the grazing period: When the sheep were released (approx 20th June), at end of July, at beginning of September and the day before slaughtering (last day of grazing, see Table 2).

Each sample was analysed for crude protein, crude fat, crude fibre and ash.

Protein was measured by modified Kjeldahl method, based on Hach (Hach et.al, 1987), by digestion at 340°C in conc. H₂SO₄ with continuous addition of 30% H₂O₂ (analytical grade) for 45 min. Automatic steam distillation and titration was according to Tecator Application Note 46/82¹. Fat was measured by modified soxhlet extraction according to Tecator Application Note 23/80¹. Fibre was measured by modified methods of Van Soest (1963) and ash was measured by ignition at 550°C.

¹Tecator AB Box 70. S-26321, Höganäs, Sweden.

Table 2. General data on animals and performance.

Year			Lambs				Grazing period (days)
	No. ¹	No. ²	Day of birth (avg)	Birth-weight (kg (avg))	Carcass weight (kg)	Meat dressing %	
1994, Hestur	8	9	18 May	4,4	11,6	34,8	20 June-28 Sep (100)
1995, Stóra Ármót	8	12	31 May	NA	11,0	37,5	21 June-1 Oct (102)
1996, Stóra Ármót	7	9	28 May	NA	11,2	37,7	10 June-6 Oct (118)
1997, Stóra Ármót	7	7	24 May	NA	12,4	37,5	10 June-22 Sep (104)

¹Total number of ewes.

²Total number of lambs.

NA= Not available

4 RESULTS AND DISCUSSION

There is a great diversity in the environmental conditions of the free ranging Icelandic sheep during the summer and beginning of autumn. This is e.g. apparent in the vegetation map in Figure 2. Another related factor is the variability of soil types. The diversity is also apparent on a small scale, e.g. within a pasture field. The terrain is typically uneven, e.g. with hummocks, and includes some dry and wet areas. Added to this is the fact that the sheep graze selectively selecting plants (and parts of plants) according to their own preference and plant availability. Two sheep grazing in exactly the same area can consume very different levels of radiocaesium. All this is reflected in the variable radiocaesium concentration that can be found in lamb meat in the autumn. The concentration has been monitored by collecting samples from all major slaughterhouses in Iceland annually since 1989. The concentration has ranged from a few Bq/kg to almost one hundred. The aim of Geislavarnir ríkisins and Rannsóknastofnun landbúnaðarins when starting participation in the NKS projects was to obtain a better understanding of the sources of this variability. The aim has of course also been to estimate the long term trend of radiocaesium concentration in Icelandic lamb meat, as in the other Nordic countries. The studies RAD-3 and EKO-2.1 projects indicate that the

change with time is very slow. Studying such a slow trend is very difficult if it is buried in variability created by other sources. It was therefore decided to try to

study more factors than just those necessary for studying the long term trend in lamb meat, vegetation and soil. The results of these studies are at the time of writing this report being prepared for publication in a scientific journal. The amount of detail has therefore to be reduced, but the authors can be contacted for further information until the time of publication.

Summary of the main results for soil, vegetation and lamb meat can be found in the modelling section of the EKO-2.1 report and those results will not be repeated here. Main emphasis here is put on introducing the supplementary studies and some of the obtained results.

Unlike many of the other Nordic countries, the radiocaesium found in the Icelandic terrestrial environment is almost entirely old fallout from the early sixties. The Icelandic data in the EKO-2.1 project shows very little change within sites over time as can be seen in Figures 9 and 10.

4.1 ANIMAL PERFORMANCE

Data on animal performance is presented in Figures 15 – 17.

4.2 SOIL AND VEGETATION

Results from the biomass estimation and comparison between grazed and ungrazed adjacent pasture can be seen in Figures 13 and 14.

4.2.1 SOIL

Samples were collected every year throughout the summer in different parts of the area of study. Soil samples generally show a great variability in Cs-137 concentration and the Icelandic samples are no exception. The average values for the two study areas are fairly similar. No clear difference between years or different parts of each area could be seen at Stóra Ármót. Typical values would be 4 - 5 kBq/m², but the range spanned 2 - 8 kBq/m².

Since the area at Stóra Ármót is considerably more hummocky than the area at Hestur, it was decided to carry out a special study on the effects of these hummocks on the concentration of Cs-137 in the soil and thus possibly account for one source of variation. Instead of selecting the sampling spots at random, adjacent top and bottom spots were selected. The results are displayed in Figure 3. The results were striking. The concentration in a bottom area was in all cases much higher than the concentration in the adjacent top part. The same pattern was also visible in other samples not displayed here.

4.2.2 Vegetation

The radiocaesium concentration in the composite plant samples from the experimental farm Hestur is an order of magnitude higher than at Stóra Ármót (Figures 4 and 5). The radiocaesium concentration in the soil is similar at both

sites. This great difference is due to much higher rates of transfer of radiocaesium from soil to plants at Hestur. The concentration seems generally to be similar throughout each summer (most often decreasing towards the end of summer). The meteorological data for the period May-September during the last year at Stóra Ármót was considerably different from the two previous years. The precipitation was more and the average temperature was less. Table 3 show total precipitation and average temperature. See also Figures 11 and 12.

Table 3. *Total precipitation during May – September and average temperatures during the same period for the meteorological stations nearest to Hestur research station (Hvanneyri – 1994 data) and Stóra Ármót research station (Eyrarbakki).*

year	meteorological station – experimental site	precipitation (mm)	temperature (°C)
1994	Hvanneyri – Hestur	131	9,1
1995	Eyrarbakki – Stóra Ármót	441	9,1
1996	Eyrarbakki – Stóra Ármót	490	9,9
1997	Eyrarbakki – Stóra Ármót	598	6,1

Table 4 shows values for composite plant samples, hay and individual species found on both sites. The hay samples represent the same hay as was fed to the animals while housed. The hay values are included here for use when interpreting the concentration in milk.

Radiocaesium concentration in individual species are also shown in Figure 6. The values for the experimental farm Hestur are generally higher than those from Stóra Ármót as for the composite plant samples. There is however considerable variability of values for each species as a function of time and no common pattern clearly visible. This variability could be partly due to that these samples were generally smaller than the composite plant samples and consequently less averaging effect in the sampling. Another possible source of the variation could be that the samples were picked at different spots in the area at different times and that these spots have different characteristics.

4.2.3 NUTRITION IN VEGETATION

Data on nutrition in vegetation (biomass samples) is presented in Table 5.

4.3 Other results

4.3.1 MEAT

The radiocaesium concentration in lamb meat at the two experimental farms is summarised in Figure 9. The values show the same relative difference in concentration as the composite vegetation samples. The level at the experimental farm Hestur is approximately an order of magnitude higher than the level at

Stóra-Ármót. No clear time trend can be seen in the values for Stóra Ármót during the three years the experiment was conducted there.

Table 4. Average concentration of Cs-137 in hay and vegetation.

Year and site	Sample type or plant species	date collected	habitat	Bq/kg dry weight
1994, Hestur	hay	-	-	32
	composite plant samples 1	summer avg.	wet	88
	composite plant samples 2	summer avg.	dry	2,6
	<i>Carex lyngbyei</i>	28 July	wet	66
	<i>Carex nigra</i>	28 July	wet	129
	<i>Carex bigelowii</i>	28 July	dry	2
	<i>Deschampsia caespitosa</i>	6 September	dry	38,4
	<i>Festuca</i> spp.	6 September	dry	3,8
	<i>Agrostis capillaris</i>	6 September	dry	5,1
1995, Stóra Ármót	hay	-	-	1,33
	composite plant samples	summer avg.	dry	7,5
	<i>Carex nigra</i>	2 August	wet	50,4
	<i>Carex bigelowii</i>	2 August	dry	2
	<i>Deschampsia caespitosa</i>	2 August	dry	1,9
	<i>Festuca</i> spp.	2 August	dry	6,1
	<i>Agrostis capillaris</i>	2 August	dry	4,3
	<i>Calluna vulgaris</i>	2 August	dry	15,9
	<i>Salix lanata</i>	2 August	dry	1,8
1996, Stóra Ármót	hay	-	-	18
	composite plant samples ¹			
	<i>Carex nigra</i>	1 August	wet	22
	<i>Carex bigelowii</i>	1 August	dry	2,5
	<i>Deschampsia caespitosa</i>	1 August	dry	0,8
	<i>Festuca</i> spp.	1 August	dry	3,3
	<i>Agrostis capillaris</i>	1 August	dry	5,3
	<i>Calluna vulgaris</i>	1 August	dry	11,4
	<i>Salix lanata</i>	1 August	dry	5,7
1997, Stóra Ármót	composite plant samples			
	<i>Carex nigra</i>	4 September	wet	22,4
	<i>Carex bigelowii</i>	4 September	dry	3,44
	<i>Deschampsia caespitosa</i>	4 September	dry	0,7
	<i>Festuca</i> spp.	4 September	dry	
	<i>Agrostis capillaris</i>	4 September	dry	
	<i>Calluna vulgaris</i>	4 September	dry	20
	<i>Salix lanata</i>	4 September	dry	2,31

¹Composite plant samples taken in 1996 are not comparable due to different method used as described previously. See also Figure 3.

Table 5. Chemical composition and energy. Average for each summer (n=4).

Table 5: Chemical composition and energy: Average for each summer (n = 7).												
		Crude		Crude			<i>in vitro</i> ³			ME ⁴	AAT ⁵	PBV ⁶
Year	Ash	prot.	fat	fibre	NDF ¹	ADF ²						
Site	% DM	% DM	% DM	% DM	% DM	% DM	% DM	% DM	MJ	g/kg	g/kg	
Hestur	1994	8,3	10,5	1,5	22,3	60,2	30,7	51,8	8,1	67,2	-11,8	
Stóra Ármót	1995	6,0	9,0	3,7	23,2	48,8	31,4	41,9	8,4	55,8	-0,2	
Stóra Ármót	1996	5,8	9,1	2,5	20,9	47,6	27,8	52,0	8,4	68,9	-2,7	
Stóra Ármót	1997	7,2	9,8	3,2	23,7	54,0	30,3	47,5	8,2	61,7	-10,1	

¹Neutral Detergent Fibre

⁴Metabolic energy (ME)

²Acid Detergent Fibre

⁵Aminoacid absorb.in the small intestines

³In vitro Dry matter digestibility

⁶Protein balance in the rumen

Note: AAT and PBV calculated using data obtained from regular hay.

Coefficient of degradation =0,60. DM=Dry Matter

Metabolic energy calculated from chemical data..

4.3.2 FAECES

Figure 10 shows the radiocaesium concentration in faeces from ewes and lambs. Similar difference is in the levels between Hestur and Stóra Ármót as was seen for vegetation and meat, the values at Hestur being approximately an order of magnitude higher. No clear difference is seen between lambs and ewes and the concentration follows similar pattern as individual species (Figure 6) and composite samples (Figures 4 and 5).

4.3.3 MILK

Figures 7 and 8 show the radiocaesium concentration in ewes milk at Hestur (in 1994) and Stóra Ármót (in 1995). The sheep were kept indoors and fed with hay until being released in late June. This caused a dramatic increase in the radiocaesium concentration of milk at Hestur due to the higher concentration in the vegetation in the field than in the hay used for indoor feeding.

4.3.4 CONSUMPTION OF FUNGI

Only faecal samples from 1995 and 1996 were screened for mushroom spores as no mushrooms did appear in 1997. In samples from 1995, spores were found in 18 of the 28 samples and in 1996 spores were found in 12 of the 28 samples.

Spores were both in samples from ewes and lambs, but there seemed to be difference between time of summer. In August, both years, fewer ewes seemed to have ingested spores than in September.

Not all of the spores could be identified, some some or them were believed to belong to the genus *Inocybe* but others to *Cortinarius*. The identified spores belonged to the species *Stropharia semiglobata*, *Psilocybe* sp., *Panaeolus* sp.,

Coprinus sp. and *Cortinarius* sp. Seasonal variation was found in spore content, as frequency of spores did get higher in late summer (September) (Eyjolfsdottir, 1997).

It is well known that sheep vary their diet, and factors like daylength, availability and quality have been found to affect their intake, as well as prior experience. Ewes also "teach" their offsprings what to eat (Arnold et. al, 1966; Thorhallsdottir et. al, 1987; Thorhallsdottir and Thorsteinsson, 1993).

Spores in faeces cannot be looked at as evidence for mushroom ingestion. Spores are easily transmitted by winds, water or by other means. They can sit on grass or other eaten vegetation, they can be inhaled and they can be transmitted to the sheep by drinking water. They can also be digested, resulting in possible underestimation of the consumption. The samples from Stóra Ármót did contain fragments of fungal tissue, some with spores attached. This must be considered as clear evidence for mushroom ingestion, but the data does not allow any estimation on how much might have been ingested.

4.3.5 TRANSFER FACTORS

In all cases there is considerable variability in the data but a rough estimate can be made of transfer factors by using the following values as representative for Stóra Ármót:

$$\begin{aligned} \text{Soil: } & 4.5 \text{ kBq/m}^2 \\ \text{Vegetation: } & 5 \text{ Bq/kg (estimate for September values)} \\ \text{Lamb meat: } & 4 \text{ Bq/kg fresh weight} \end{aligned}$$

This produces the following transfer factors:

$$\begin{aligned} TF (\text{soil-plant}): & 1 \cdot 10^{-3} \text{ m}^2 \text{ kg}^{-1} \\ CF (\text{vegetation-meat}): & 0.8 \\ T_{ag} (\text{soil-meat}): & 0.9 \cdot 10^{-3} \text{ m}^2 \text{ kg}^{-1} \end{aligned}$$

These factors should only be taken as indicative.

No attempt is made here to estimate transfer factors for Hestur research station. The field is very inhomogeneous and relating concentration in meat to deposition density in soil would require a good estimate of the grazing behaviour of each lamb and comprehensive data for the whole area. The current data indicates however that the CF (vegetation-meat) is similar as for Stóra Ármót.

5 CONCLUSIONS

The vegetation map of Iceland shown in Figure 2 displays well the diversity of areas where the free ranging Icelandic sheep are grazing during the summer. The two sampling sites, Hestur and Stóra Ármót, show very different properties with respect to soil to plant transfer of radiocaesium. These two places offer an opportunity to study further the characteristics of the soil which are responsible for this difference. Such a study would have to involve a soil specialist. Traditional simple soil classification schemes might easily be misleading for the young volcanic Icelandic soil due to its unusual properties. The ultimate goal would be to be able to assess the vulnerability of different areas to radionuclide fallout, based on a better understanding of the diversity in soil to plant transfer of radiocaesium in Iceland.

The Icelandic results for Hestur and Stóra Ármót show insignificant changes over time. It would be of interest to try to monitor how the levels at both places change over a longer time period.

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7 Appendix

7.1 FIGURES

Figure 1. Experimental sites 1994 - 1997.

Figure 2. Vegetation map of Iceland.

Figure 3. Variations in Cs-137 concentration in soil at Stóra Ármót.

Figure 4. Cs-137 concentration in composite plant samples from Hestur.

Figure 5. Cs-137 concentration in composite plant samples from Stóra Ármót.

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Figure 14. Herbage density (biomass) at ungrazed and grazed adjacent pastures.

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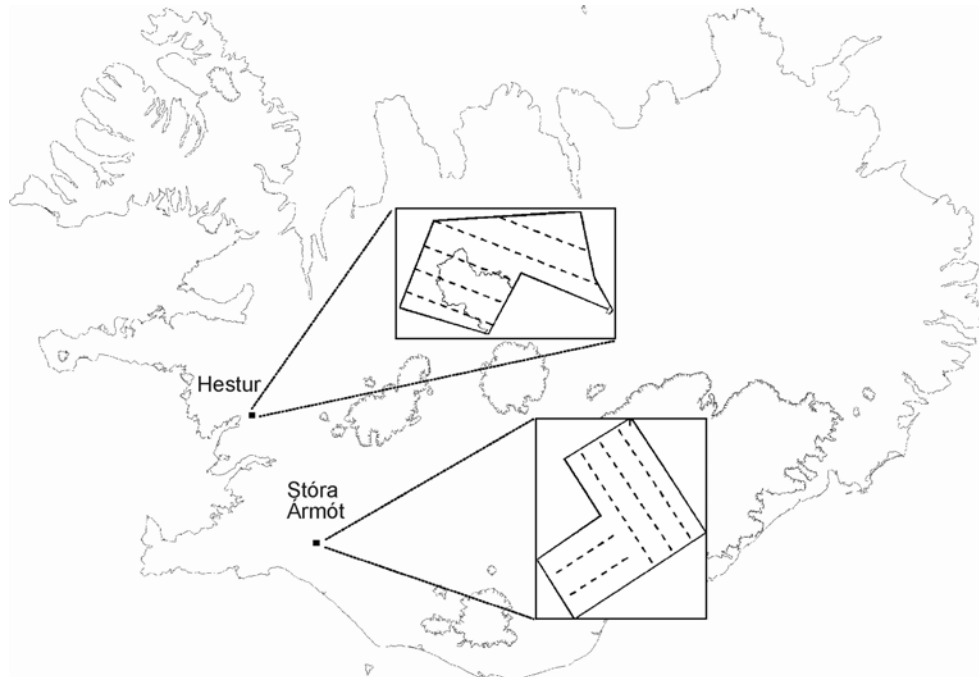


Figure 1. Map of the experimental sites. Dotted lines indicate transects used for biomass estimation (see Materials and methods).

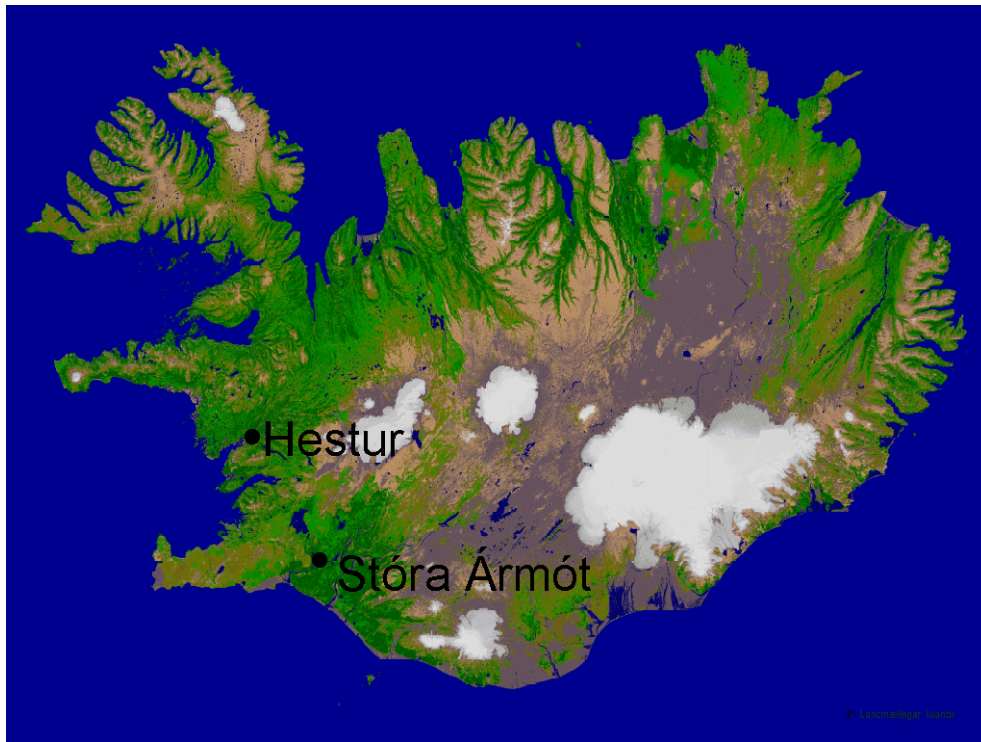
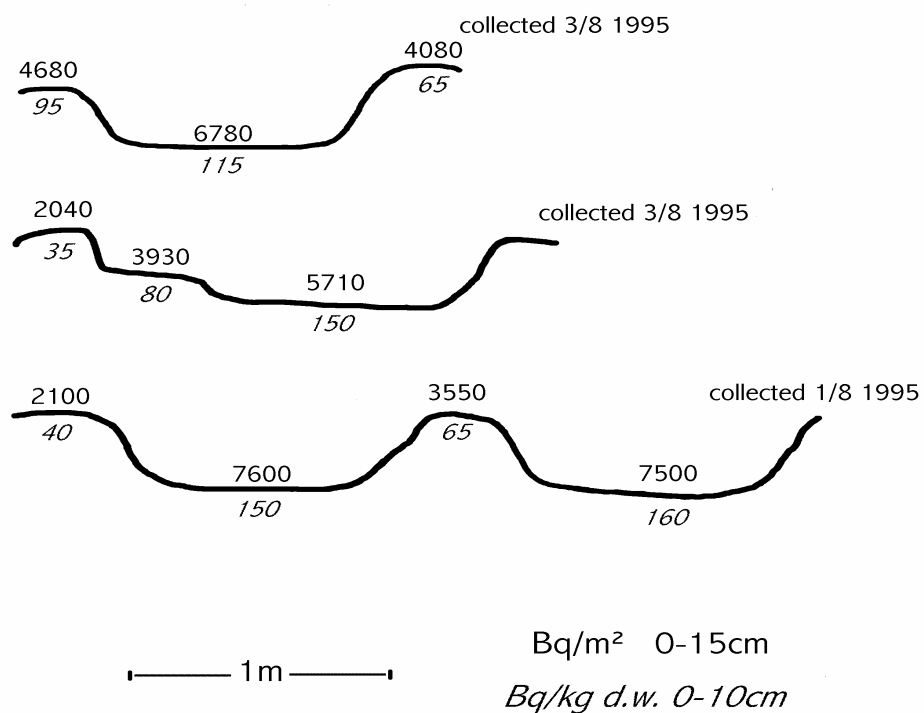


Figure 2. Vegetation map of Iceland. © LMÍ.



SEP/jp 1997

Figure 3. Variations in Cs-137 concentration in soil samples between top of hummocks and adjacent low areas. Above the line is concentration per unit area down to a depth of 15 cm. Below the line is concentration per unit mass (dry weight) down to a depth of 10 cm.

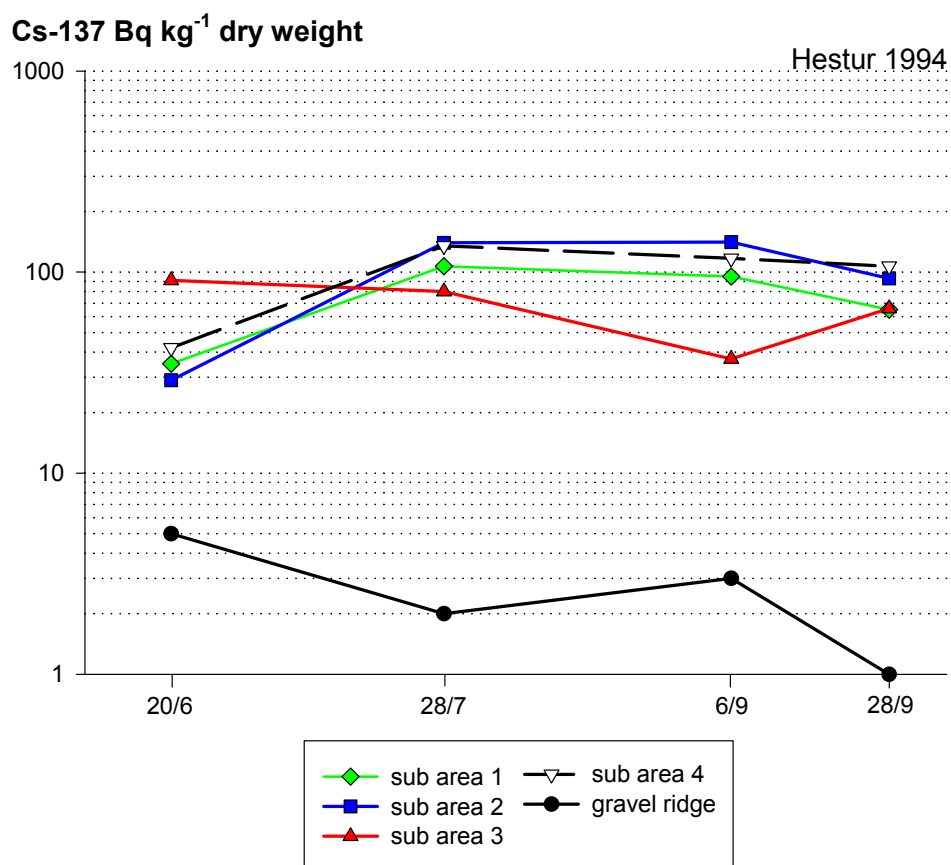


Figure 4. Cs-137 concentration in composite plant samples from Hestur 1994.

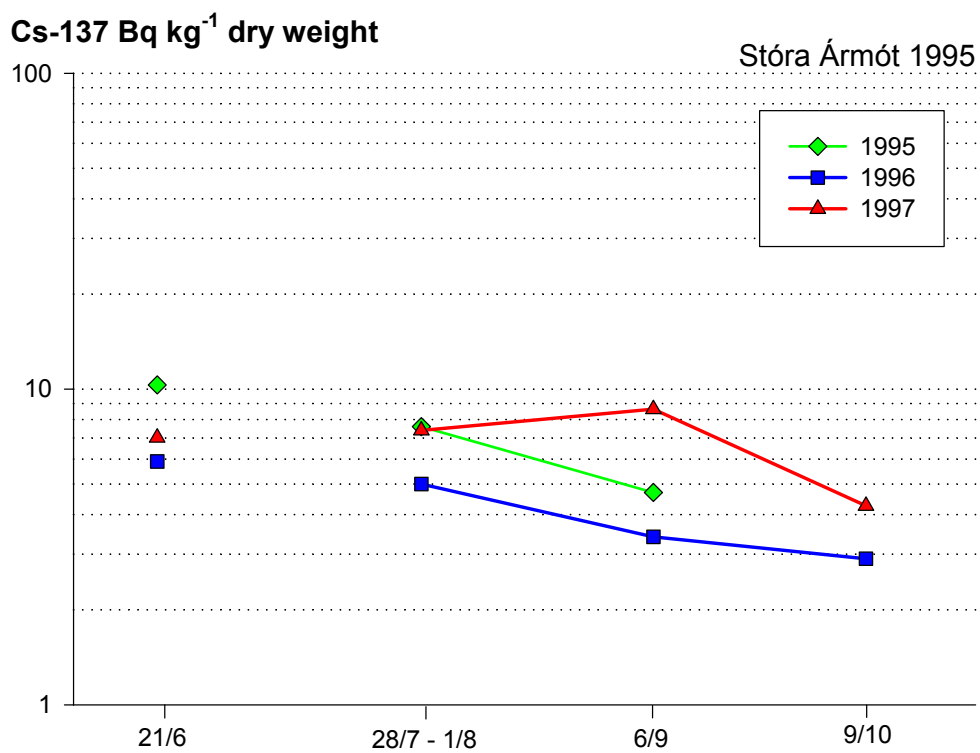


Figure 5. Cs-137 concentration in composite plant samples from Stóra Ármót 1995 – 1997.

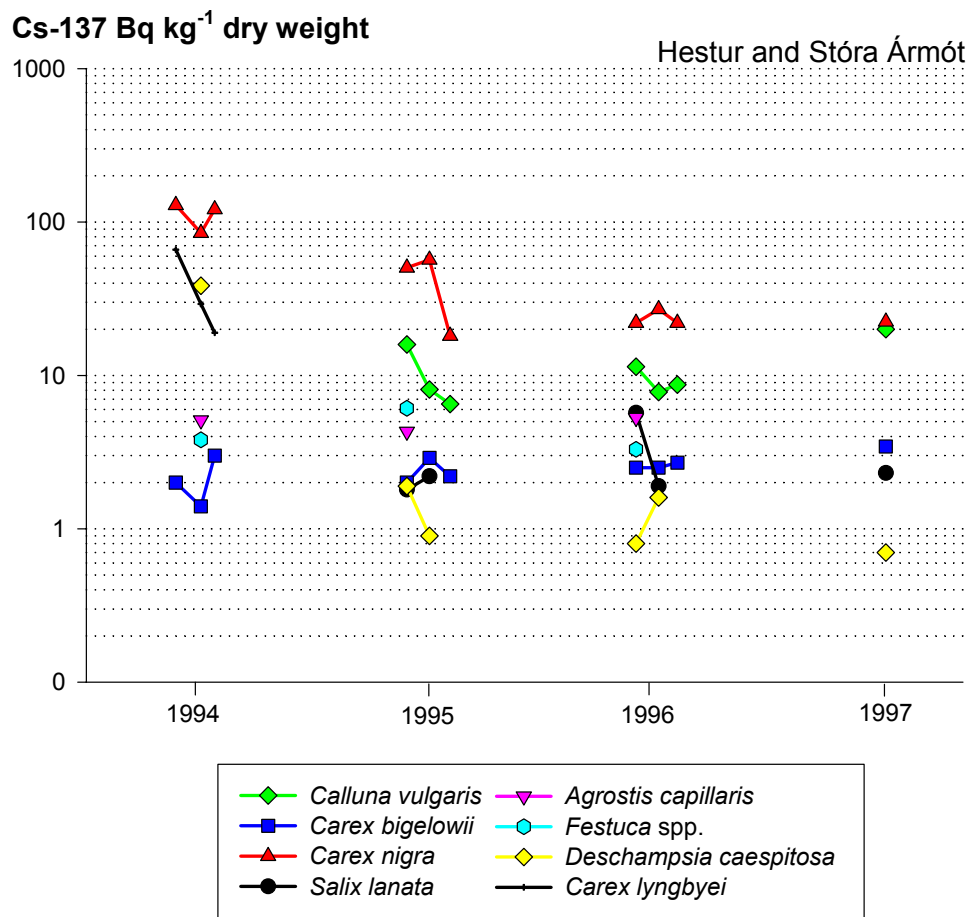


Figure 6. Cs-137 concentration in individual plant species from Hestur and Stóra Ármót. Samples were taken late July, beginning of September and end of September / beginning of October every year. The 1997 samples are taken in September.

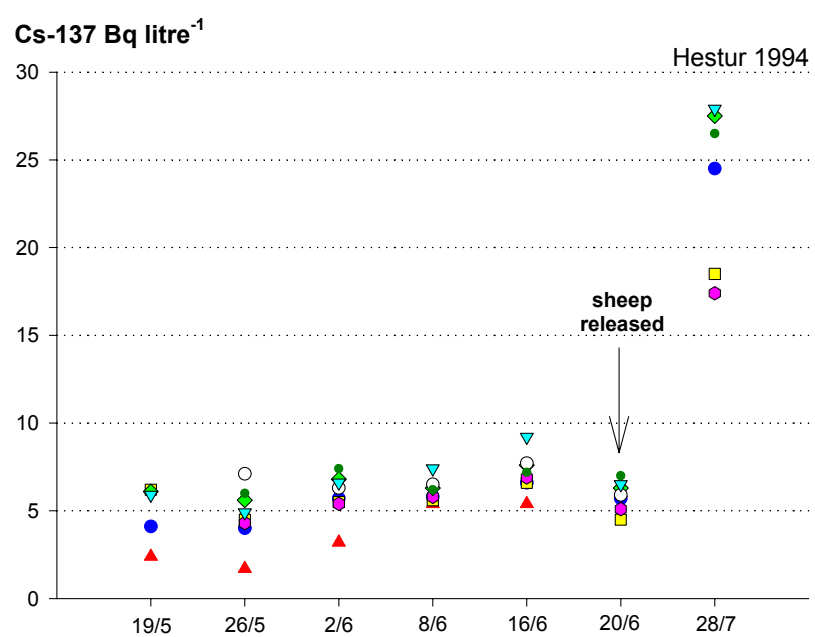


Figure 7. Cs137 in sheep milk, Hestur 1994.

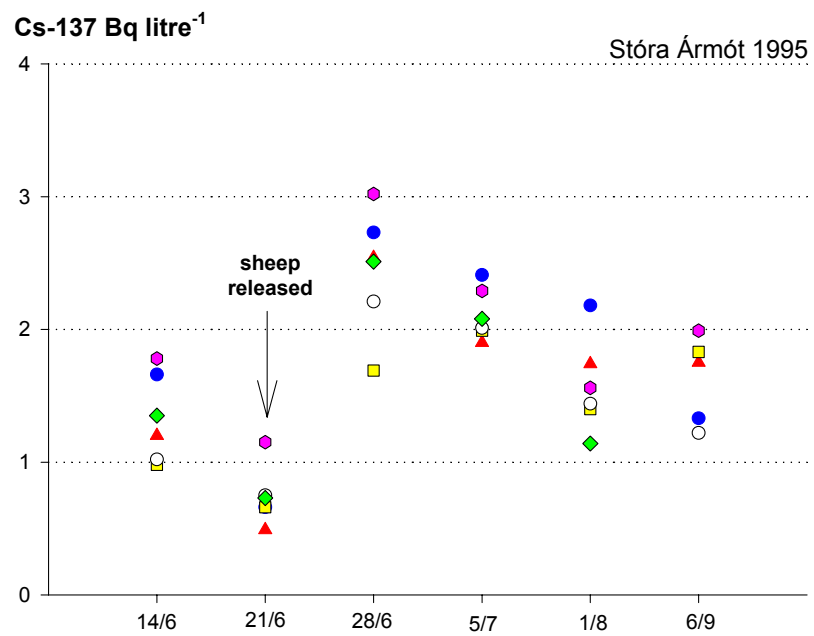


Figure 8. Cs137 in sheep milk, Stóra Ármót 1995.

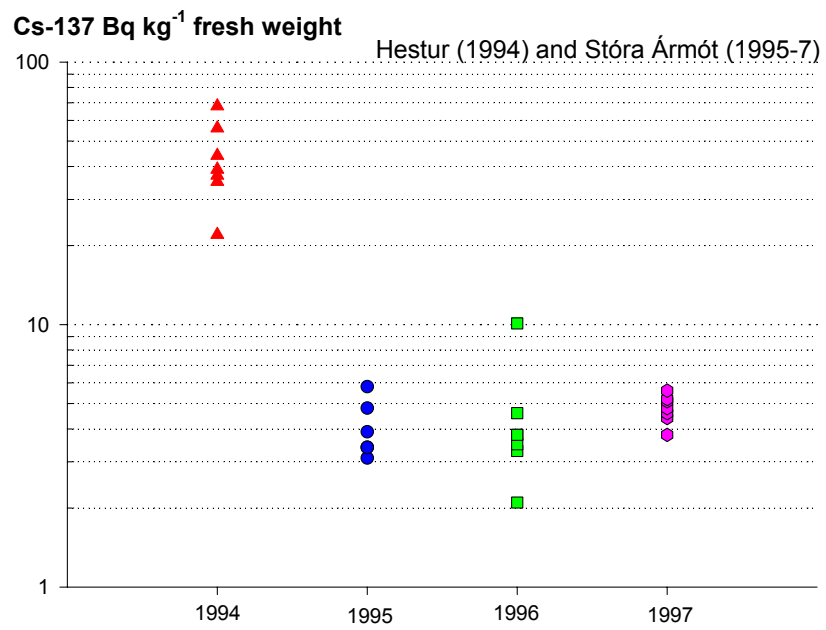


Figure 9. Cs137 in lamb meat, Hestur 1994 and Stóra Ármót 1995-7.

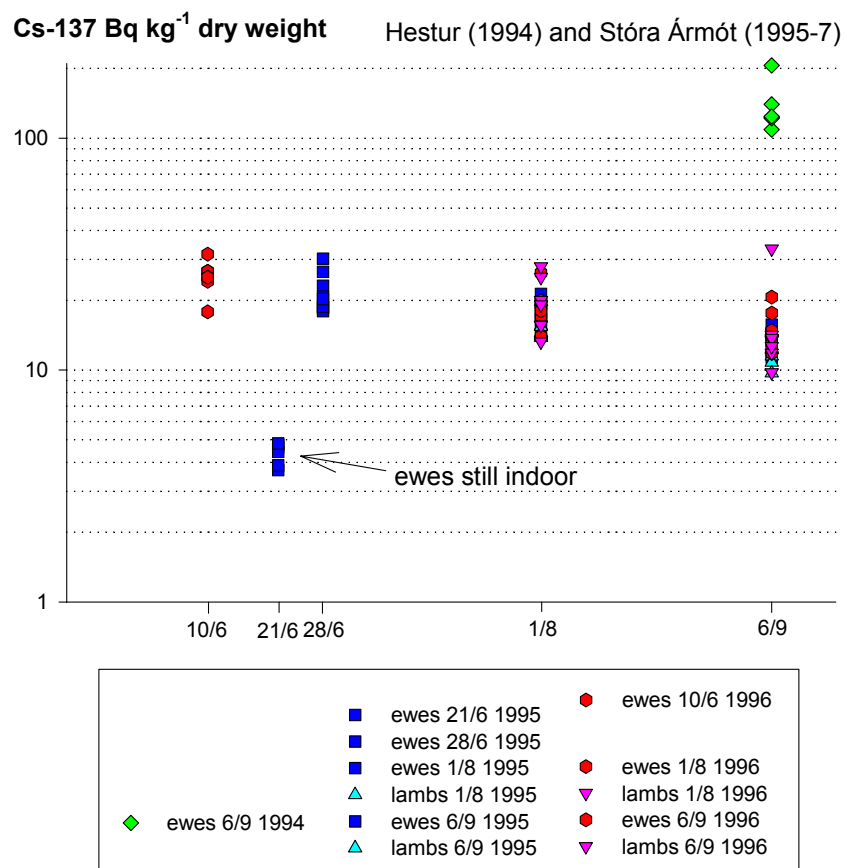


Figure 10. Cs137 in faeces, Hestur 1994 and Stóra Ármót 1995-7.

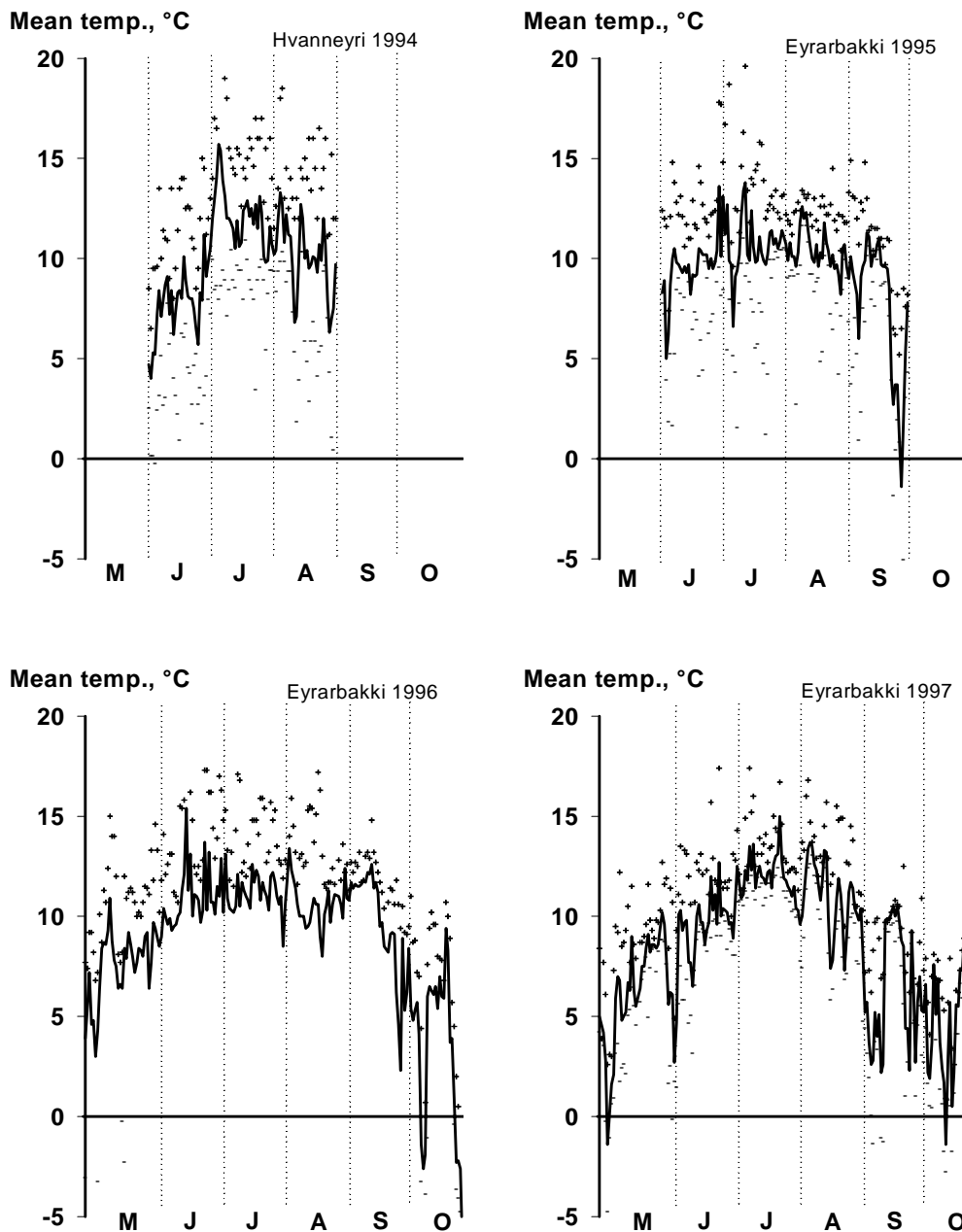


Figure 11. Mean daily temperature (line), maximum and minimum temperature (+/-) 1994-97. Hvanneyri is the meteorological station nearest to Hestur research station and Eyrarbakki represents Stóra Ármót research station.

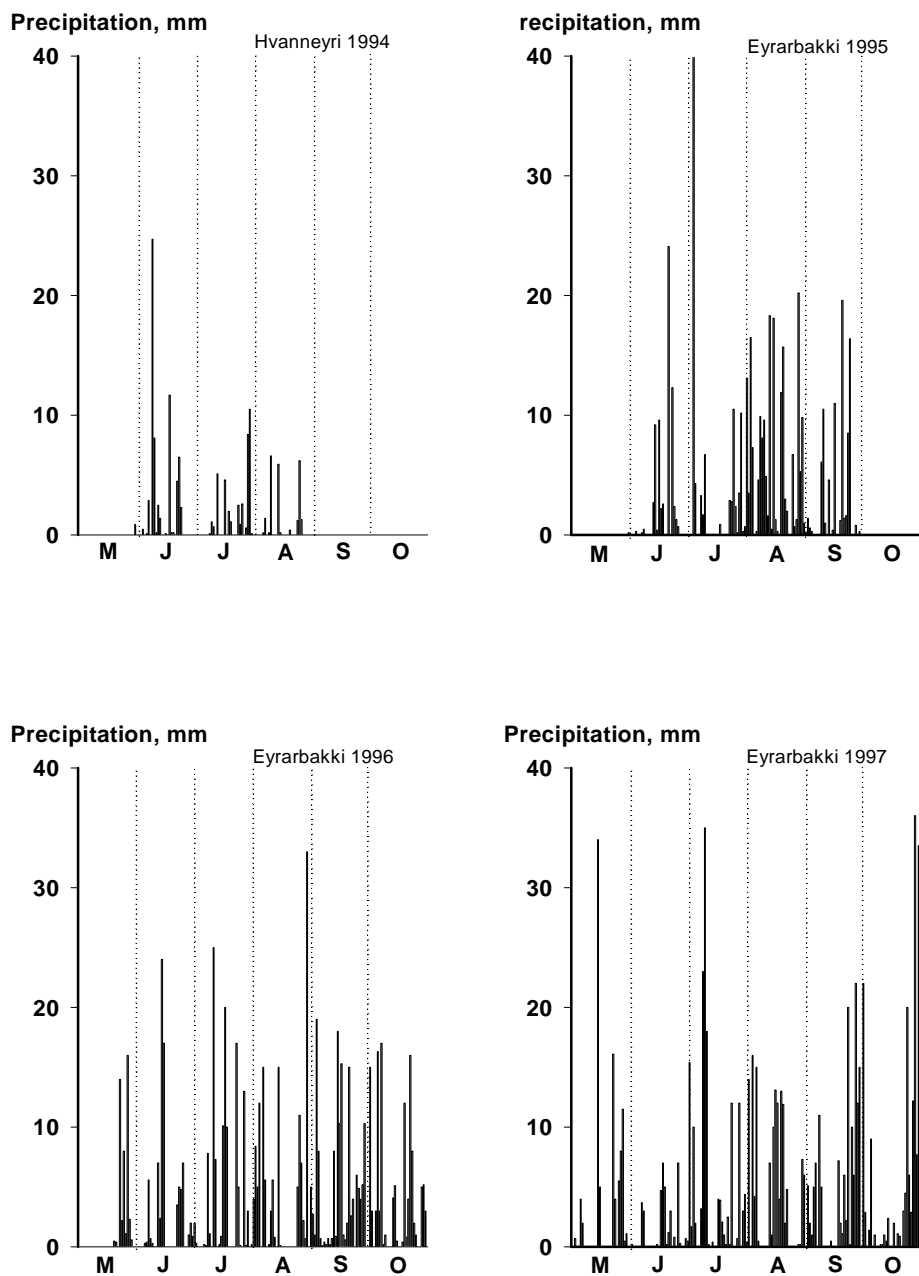


Figure 12. Daily precipitation 1994-97. Hvanneyri is the meteorological station nearest to Hestur research station and Eyrarbakki represents Stóra Ármót research station.

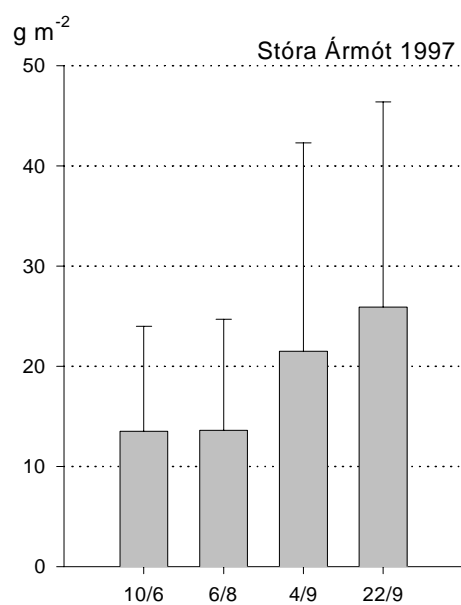
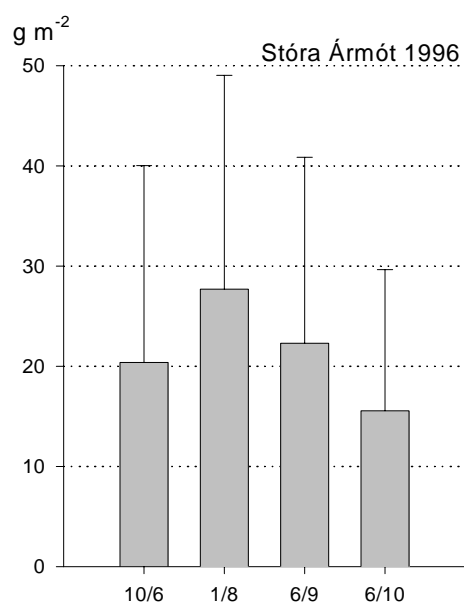
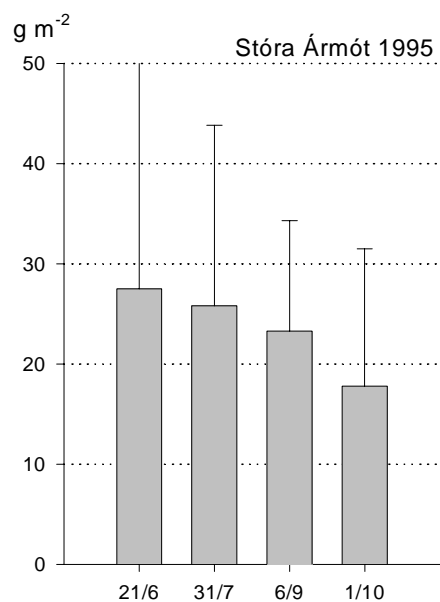
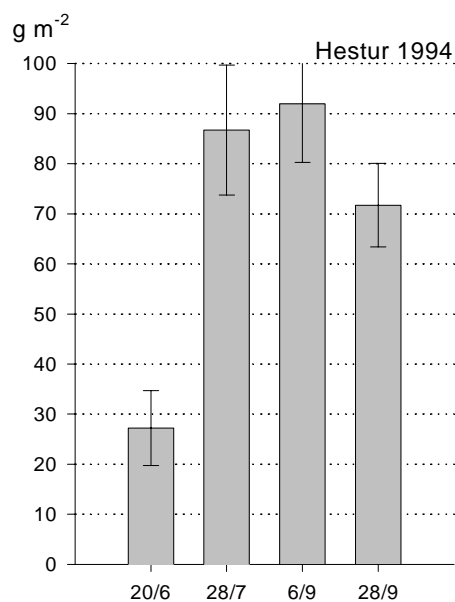


Figure 13. Herbage density (biomass) at Hestur and Stóra Ármót. Bars indicate SD

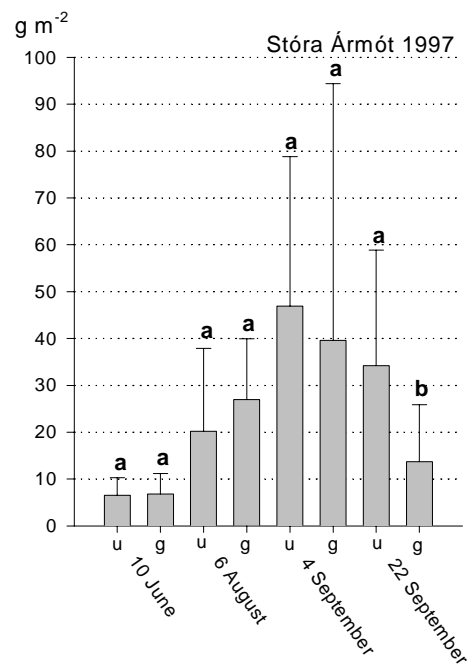


Figure 14. Herbage density (biomass) at ungrazed (u) and the grazed (g) adjacent identical pastures at the experimental site. Bars indicate SD and different letters indicate statistical difference within dates.

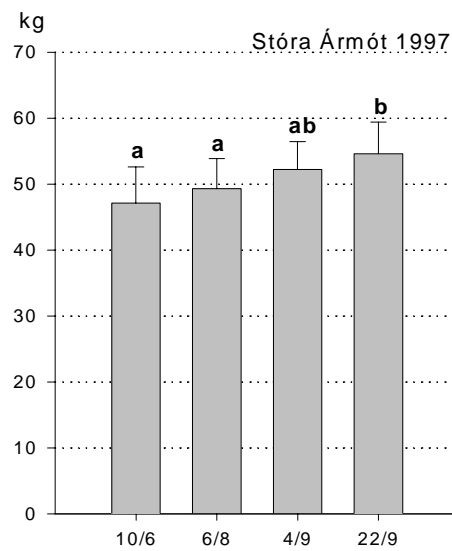
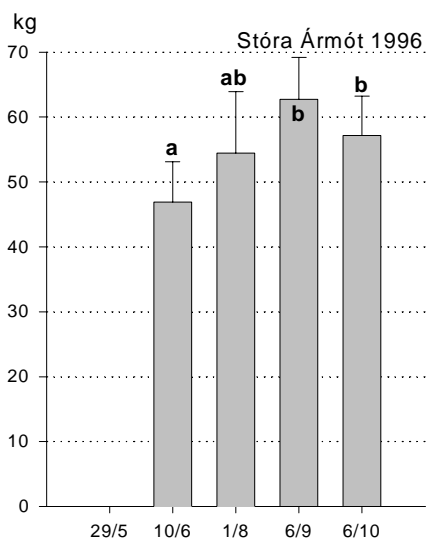
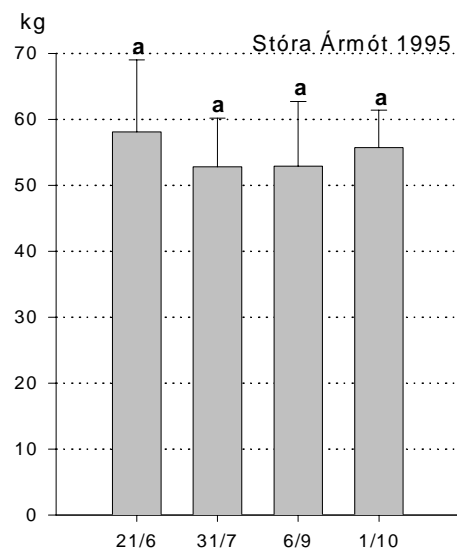
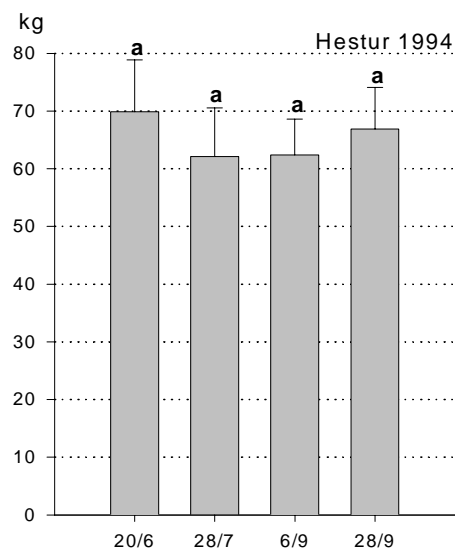


Figure 15. Live weight of ewes. Bars indicate SD and different letters indicate statistical difference.

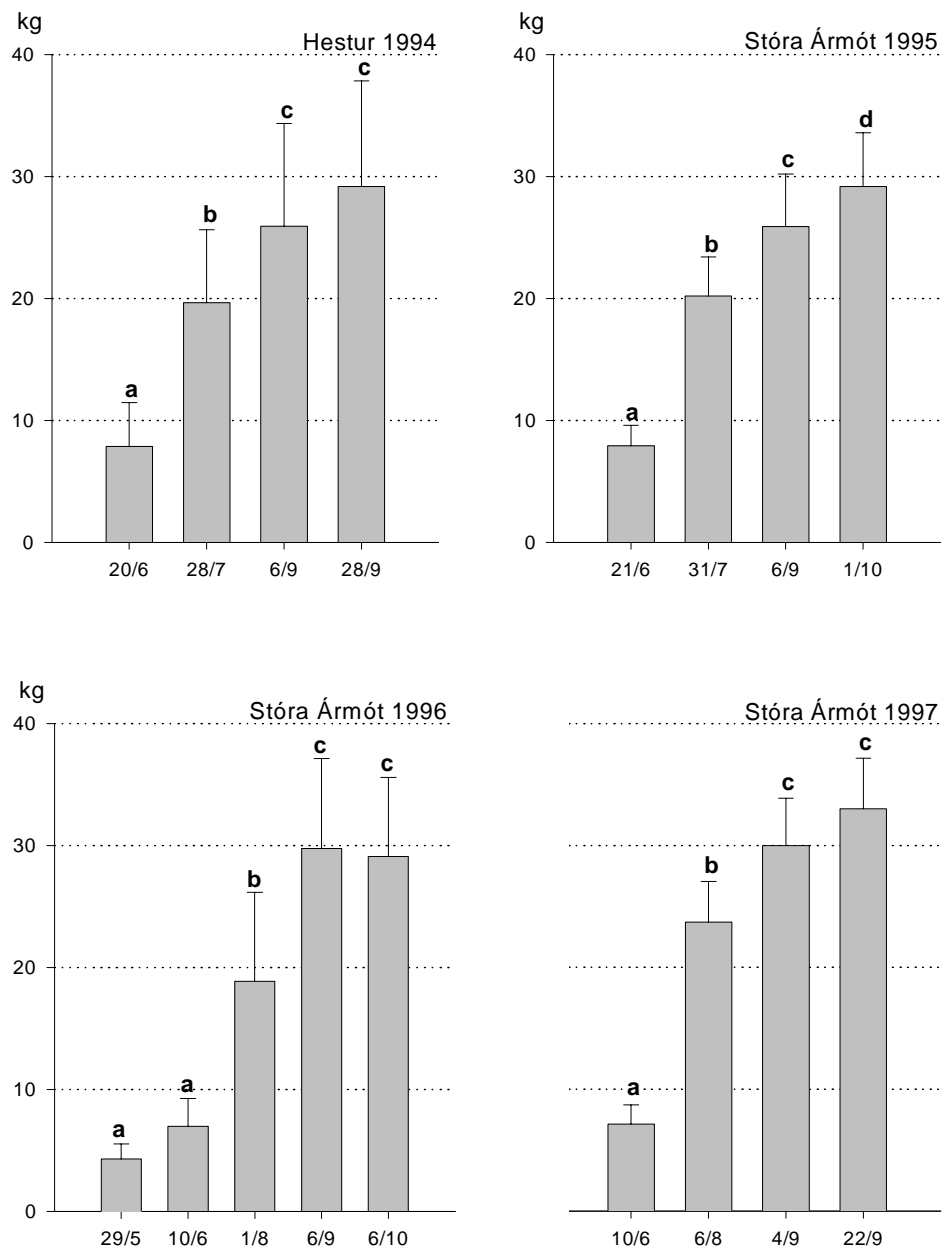


Figure 16. Live weight of lambs. Bars indicate SD and different letters indicate statistical difference

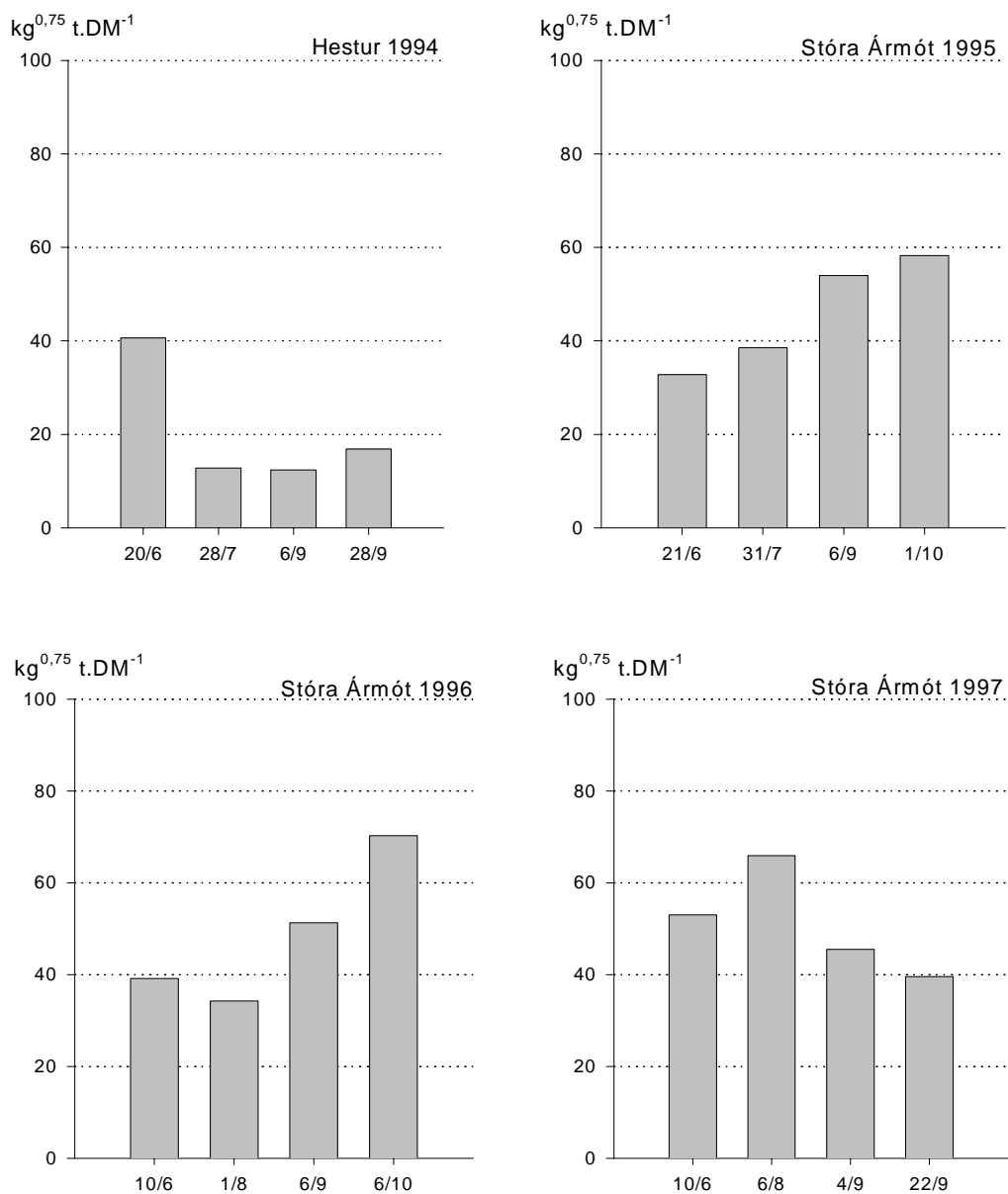


Figure 17. Grazing pressure (availability of herbage vs. live weight $\text{kg}^{0.75}$ at Hestur and Stóra Ármót.

Title	Summaries of studies carried out in the NKS/BOK-2 project – Technical report
Author(s)	Sigurður Emil Pálsson (editor)
Affiliation(s)	Geislavarnir ríkisins (Icelandic Radiation Protection Institute)
ISBN	87-7893-086-3
Date	December 2002
Project	NKS/BOK-2
No. of pages	238
No. of tables	61
No. of illustrations	112
No. of references	254
Abstract	<p>Summaries of studies carried out in the NKS/BOK-2 project, <i>Radiological and Environmental Consequences</i>. The structure of the project as such is described in NKS-64, <i>Radiological and Environmental Consequences – Final Report of the Nordic Nuclear Safety Research Project BOK-2</i>. That report also includes compilations based on the summaries presented in this report. The project was carried out 1998-2001 with participants from all the Nordic countries. Representatives from the Baltic States were also invited to some of the meetings and seminars. The project consisted of work on terrestrial and marine radioecology and had a broad scope in order to enable participation of research groups with various fields of interest. The topics included improving assessment of old and recent fallout, use of radionuclides as tracers in Nordic marine areas, improving assessment of internal doses and use of mass spectrometry in radioecology. This report is a compilation of summaries from each research group, 32 papers in all, and gives references to papers published in scientific journals. Some of the studies have been described previously, at least to some degree, in NKS-70, <i>Proceedings of the 8th Nordic Seminar on Radioecology, 25-28 February 2001, Rovaniemi, Finland</i>.</p>
Key words	Radioecology, radioactive fallout, sheep, radiocaesium, radiostrontium, technetium, seaweed, oceanic tracers, mass spectrometry, internal doses, doses to man