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within the NKS-B Programme 2002-2005.
24-25 October 2005, Tartu, Estonia

Erkki Ilus (editor)
Radiation and Nuclear Safety Authority, STUK, Finland

August 2006

Abstract

This report contains the proceedings of the NKS-B Summary Seminar held on 24-25 October 2005 in Tartu, Estonia. The aim of the seminar was to provide a forum for presenting and discussing the results obtained in the NKS-B programme during the project period 2002 - 2005. The main attention was focused on radioecology and measurement techniques including presentations on the work done in the Projects INDOFERN (New Indicator Organisms for Environmental Radioactivity), LABINCO (Intercomparison of Laboratory Analyses of Radionuclides in Environmental Samples) and ECODOSES (Improving Radiological Assessments of Doses to Humans from Terrestrial Ecosystems). The total number of presentations in the seminar was 27.

The seminar was also the final seminar of the four-year INDOFERN Project. The objective of the project was to identify new indicator organisms and biomarkers for assessment of environmental radioactivity in normal and emergency situations. The goal was to search new useful organisms accumulating effectively and specifically certain radionuclides in various Nordic ecosystems (forest, fresh water, marine), and to compare their indicator value to those of the earlier known indicators. The project yielded new data on the occurrence and transport of radionuclides in a wide scale of Nordic ecosystems. A summary of the whole project, and summaries of the work done in all the participating laboratories were presented in 13 presentations in the seminar.

Key words

Radioecology, terrestrial radioecology, aquatic radioecology, indicator organisms, laboratory analyses of radionuclides, modelling, sampling, doses to man

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NKS Secretariat
NKS-775
P.O. Box 49
DK - 4000 Roskilde, Denmark

Phone +45 4677 4045
Fax +45 4677 4046
www.nks.org
e-mail nks@nks.org



Participants at the NKS-B seminar in Tartu, Estonia, 24-25 October 2005

Introduction

The structure of the NKS co-operation

The NKS has for decades supported Nordic co-operation in research and development within

- radiological and nuclear emergency preparedness
- radioecology
- nuclear technology and safety

The formal co-operation began in 1957 and has had different forms. This is vividly described in the book “*Half a Century of Nordic Nuclear Co-operation*” by Franz R. Marcus, which is available from the NKS secretariat. More information on NKS can be found at the NKS web site:

<http://www.nks.org/>

Traditionally the NKS work was organised in programme periods, each usually lasting for 4 years. A different structure was adopted in 2002. The concept of fixed programme periods was dropped and the time frame for proposed activities could be short or long, but funding would only be given for one year at a time. Two main programmes were defined:

- NKS-B: Emergency Preparedness
- NKS-R: Reactor safety

The aim with the NKS-B programme (as stated in 2002) is to ***strengthen radiological emergency preparedness in the Nordic countries***. *Apart from activities directly targeted on emergency preparedness this also includes activities in related areas such as **radioecology and effective communication and information management**. Also these activities must, however, be focused on emergency preparedness related questions.* Later increased emphasis has been put on maintaining competence in radioecology.

NKS-B activities 2002 - 2005

A variety of different activities have been supported with NKS-B. The following are concluded:

- UrbContSem: *Urban contamination seminar*
- EccoMags: *Nordic-EU collaboration on design and evaluation of the Resume 2002 exercise*
- MGS-ModMeth: *Co-ordination and modernisation of methods for AGS and CGS measurements of multi-nuclide contamination*
- MGS-Course: *Course in advanced methods for processing AGS and CGS data and similar sets of spectral data*
- ASS-1: *Area specific stripping for CGS and AGS*
- Nova Course: *Additional funding of Ph.D. course in radioecology*
- NucVess: *Impact assessment of accidents with nuclear powered vessels - analysis of release mechanisms and source term composition*
- Knowledgebase: *Nuclear threats in the vicinity of the Nordic countries - A base of knowledge*
- RadSem: *Radioecology and measurement techniques*
- ASSb: *Area Specific Stripping of lower energy windows for AGS and CGS NaI systems. PART 2*
- CommTech: *Communication technology and emergency preparedness*
- Cskinetic: *Human metabolism of caesium*
- NorCMass: *Nordic collaboration on the use of mass-spectrometers for the analysis of radioisotopes*
- RadChem: *Radiochemical analysis in emergency and routine situations*
- UrbHand: *Decision Support Handbook for remediation of contaminated inhabited areas*
- INDOFERN: *New indicator organisms for environmental radioactivity*
- SAMPSTRAT: *Sampling strategy and sample preparation in emergency situations*

The following activities are continuing in 2006, along with some new ones in accordance with the decision of the NKS Board in November 2006:

- Metnet: *Nordic network of meteorological services engaged in nuclear emergency preparedness*
- EMARAD: *Emergency management & radiation monitoring in nuclear and radiological accidents*
- ECODOSES: *Improving radiological assessment of doses to man from terrestrial ecosystems*
- Labinco: *Intercomparison of laboratory analyses of radionuclides in environmental samples*
- Irades: *Internal Radiation Doses in Emergency Situations*
- Rein: *Regional differences in reindeer radiocaesium contamination*
- NORDRISK: *Nuclear risk from atmospheric dispersion in Northern Europe.*
- FOREST: *Guidance for sampling in forests for radionuclide analysis and update of the Nordic forest radioecology network*

Since the NKS-B has been running since 2002, it was assumed appropriate to hold a seminar giving participants in NKS-B work a chance to present and discuss their work amongst

colleagues. The activities focusing on emergency preparedness have been regularly presented and discussed at meetings of a contact group with representatives from Nordic authorities dealing with nuclear and radiological emergency preparedness (the NEP group). It was thus decided to let the NKS-B Tartu seminar focus on radioecology and related measurement technologies.

Concluding remarks

I have been active in NKS work since December 1990, when I first attended a meeting in an NKS project group (this was at STUK in Helsinki). The NKS work soon became very important to me. It provided a stimulating forum, not only for co-operation on projects but also for exchanging views and ideas which could later lead to new co-operation projects. What impressed me right away was the dedication and commitment of those participating in NKS work. They were not there because of the funding, but because this co-operation was of such importance for them. Radioecology and related measurement technologies consist of highly specialised fields, often with a very limited number of experts within each country. The NKS has helped to create and nourish networks in this field, networks which are important for creating a stimulating environment, especially for young scientists entering the field.

It has been a special pleasure to be able to host this seminar in Tartu, Sweden's second oldest university town (founded by Gustaf II Adolf in 1632) as I like to put it. My first contact with Estonian experts in our field was made during the summer of 1991 (with Enn and Küllike Realo), and it has been a pleasure to follow the rapid growth of expertise in this field in Estonia and the other Baltic States. Although the Baltic States are not a part of the NKS co-operation, representatives from them have been able to participate in NKS seminars and intercomparison exercises, including now LABINCO. It was thus an interesting option to hold the seminar in Estonia. Efficient planning by the local organisers and their hospitality has enabled us to hold the seminar on a very low budget and yet provide very interesting and pleasant surroundings. I would especially like to thank Merle Lust, director of the Estonian Radiation Protection Centre for her work in preparing the seminar and making sure that all things ran smoothly.

The editing of these proceedings is one example of the dedication people have been willing to show to the NKS. I would like to thank Erkki Ilus for the excellent work he has done. It has been a rewarding privilege to work with him as well as all those who have contributed to the NKS-B Tartu seminar. My thanks to all of you and may the NKS work continue in the same spirit.

Sigurður Emil Pálsson,
Programme Manager, NKS-B

NKS-B Summary Seminar, Tartu, Estonia - Programme

Monday, 24 October 2005

9:00-11:00

Welcome – practical arrangements during the seminar

Sigurður Emil Pálsson, NKS

The NKS and the NKS-B programme 2002 – 2005

Elis Holm, Lund University, Sweden

Concentration factors and aggregated transfer factors in selected bioindicators from Sweden

Vesa-Pekka Varti, STUK - Radiation and Nuclear Safety Authority, Finland

Cs-137 in terrestrial environment in the Loviisa, Olkiluoto and Mänttä areas (Finland)

Runhild Gjelsvik, Norwegian Radiation Protection Authority, Norway

Evaluation of different mushroom species and lobster as indicator organisms

Sven P. Nielsen, Risø National Laboratory, Denmark

Transfer of radionuclides to plants

11:00 – 11:20 Coffee break

11:20 – 12:40

Elisabeth Strålberg, Institute for Energy Technology, Norway

Indicator organisms for marine and terrestrial environmental radioactivity

Magnús Sigurgeirsson, Icelandic Radiation Protection Institute, Iceland

New indicator organisms for environmental radioactivity in Iceland 2002-2005

Hans Pauli Joenssen, University of the Faroe Islands

Bioindicators for environmental radioactivity. Studies in the Faroe Islands

Erkki Ilus, STUK - Radiation and Nuclear Safety Authority, Finland

Indicator value of some aquatic organisms for radioactive substances in the sea areas off the Loviisa and Olkiluoto nuclear power plants (Finland)

12:40 – 13:40 Lunch - Restaurant Püssirohukelder ("Gunpowder Cellar")¹

13:40 – 15:00

Vesa-Pekka Varti, STUK - Radiation and Nuclear Safety Authority, Finland

¹³⁷Cs in aquatic organisms in the southern Lake Keuruselkä (Finland)

Tarja K. Ikäheimonen, STUK - Radiation and Nuclear Safety Authority, Finland

Behaviour of Sr-90 and transuranic elements in three areas in Finland

Sven P. Nielsen, Risø National Laboratory, Denmark

Radionuclides in seal

Anicke Jerpetjøn, Norwegian University of Life Sciences, Norway

Seaweed, fish and Crustaceans as bioindicators for ⁹⁹Tc released to marine environment

15:00 – 15:20 Coffee break

15:20 – 17:00

Erkki Ilus, STUK - Radiation and Nuclear Safety Authority, Finland

¹ Restaurant Püssirohukelder, Lossi 28 - tel +372 7303 555; www.pyss.ee

Summary Report of the NKS-B / INDOFERN Project (New indicator organisms for environmental radioactivity)

Sven P. Nielsen, Risø National Laboratory, Denmark

Intercomparison of laboratory analyses of radionuclides in environmental samples

Tarja K. Ikäheimonen, STUK - Radiation and Nuclear Safety Authority, Finland

Preliminary results of intercomparison on natural radioactivity in ground water arranged within the NKS-RADCHEM project

Galina Lujanienė, Institute of Physics, Lithuania

Determination of Cs, Am and Pu isotopes in environmental samples

Mark Dowdall, Norwegian Radiation Protection Authority, Norway

Practicable Monte Carlo Calibration of HPGe Detectors for Environmental Measurements

19:00 – Dinner - Restaurant Püssirohukelder ("Gunpowder Cellar")

Tuesday, 25 October 2005

9:00 – 11:00

Merle Lust, Estonian Radiation Protection Centre, Estonia

Radioecology in Estonia - developments

Per Roos, Risø National Laboratory, Denmark

NorCMass - Nordic collaboration on the use of mass-spectrometers for the analysis of radioisotopes,

Ulrika Nygren, Swedish Defence Research Agency, Sweden

Age determination of Pu using ICP-SFMS

Rajdeep Singh Sidhu, Institute for Energy Technology, Norway

RadChem - Radiochemical analysis in emergency and routine situations

Sigurður Emil Pálsson, Icelandic Radiation Protection Institute, Iceland

On use of precipitation data for estimation of global fallout

Håvard Thørring, Norwegian Radiation Protection Authority, Norway

Radioactive contamination of milk from the Nordic countries

11:00 – 11:20 Coffee break

11:20 – 13:00

Kasper Andersson, Risø National Laboratory, Denmark

Adaptation of ECOSYS for Nordic food chain modelling

Jon Bering, Department of Radiation Physics, Lund University, Malmö University Hospital, Sweden

Cs kinetic in humans

Elisabeth Strålberg, Institute for Energy Technology, Norway

Guidance for sampling in forests for radionuclide analysis and update of the Nordic forest radioecology network

Closing discussions:

How can NKS best support Nordic co-operation in radioecology and related measurement techniques?

(13:00 - ... Lunch, optional)

List of participants

	Name	Affiliation	Country
1	Alan Birchall	HPA-NRPB	UK
2	Anicke Jerpetjón	UMB	NO
3	Anita Skujina	RSC	LV
4	Anne Lene Brungot	NRPA	NO
5	Anne Liv Rudjord	NRPA	NO
6	Antons Lapenas	Metrc. centre	LV
7	Bredo Møller	NRPA	NO
8	Christina Greis	Örebro U.	SE
9	Daina Riekstina	RAPA	LV
10	Eia Jakobson	ERPC	EE
11	Elis Holm	Lund U	SE
12	Elisabeth Strålberg	IFE	NO
13	Erkki Ilus	STUK	FI
14	Galina Lujaniene	IoP	LT
15	Hans Pauli Joensen	Univ Faroe Isl.	FO
16	Håvard Thørring	NRPA	NO
17	Hilde Elise Heldal	IMR	NO
18	Inger Margrethe H. Eikermann	NRPA	NO
19	Janis Rudzitis	State Vet.	LV
20	Jon Bering	MAS	SE
21	Kadri Isakar	UoT (student)	EE
22	Kasper Andersson	Risø	DK
23	Küllike Realo	UoT / IoP	EE
24	Lars Frøsig Østergaard	Risø	DK
25	Maarit Muikku	STUK	FI
26	Madis Kiisk	UoT / IoP	EE
27	Magnús Sigurgeirsson	Geislavarnir R	IS
28	Mark Dowdall	NRPA	NO
29	Merle Lust	ERPC	EE
30	Per Roos	Risø	DK
31	Rajdeep Sidhu	IFE	NO
32	Rasa Gvozdaite	IoP	LT
33	Rein Koch	UoT / IoP	EE
34	Runhild Gjelsvik	NRPA	NO
35	Sigurður Emil Pálsson	NKS / Gr.	IS
36	Susanna Salminen	Univ. Helsinki	FI
37	Sven P. Nielsen	Risø	DK
38	Tarja K. Ikäheimonen	STUK	FI
39	Tua Rahola	STUK	FI
40	Ulrika Nygren	FOI	SE
41	Vesa-Pekka Vartti	STUK	FI
42	Øyvind G. Selnæs	NRPA	NO

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Summary Report of the NKS-B / INDOFERN Project

(New indicator organisms for environmental radioactivity)

Erkki Ilus

STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Preface: Bioindicator or indicator organism?

At the beginning of the INDOFERN project, it was discussed in the project group, which term (*bioindicator* or *indicator organism*) should be used when studying the ability of organisms to exhibit presence and quantity of radionuclides in the environment. At the request of the group, the following essay was written by Drs Elis Holm, Henning Dahlgaard and Christer Samuelsson.

Bioindicators are commonly grouped into accumulation indicators and response indicators. Accumulation indicators store pollutants without any evident change in their metabolism. Response indicators react with cell changes or visible symptoms of damage when taking up even small amounts of harmful substances.

A response bioindicator is an anthropogenically-induced effect in biomolecular, biochemical, or physiological parameter that has been causally linked to biological effects in one or more of the organisms, population, community, or ecosystem. Within radioecology, the term bioindicator is normally synonymous with accumulation bioindicator, i.e. organisms or organism communities that accumulate pollutants without any visible effects. The most important reasons for using bioindicators are:

- the direct determination of biological effects*
- the determination of synergetic and antagonistic effects of multiple pollutants*
- the early recognition of pollutant damage to plants, as well as toxic dangers*
- accumulation and time integration*
- focus on bioavailable physicochemical species*
- relatively low costs compared to technical measurement methods.*

Bioindicators have been used in radioecological studies to assess the environmental levels of radioactivity. Such indicators have been lichens, mosses, mussels, macroalgae etc. This has been done with rather a great success with some of these though the actual uptake processes have not always been described or studied.

For heavy metals animals have been used as so called biomarkers, i.e. they are reflecting the local pollution of the environment.

According to the Finnish Dictionary of Biology (2001), *Bioindicator* indicates the state of the environment, i.e. it is an organism which displays the state of the environment as unambiguously as possible. Required qualifications of a bioindicator are, e.g., that their

fundamental biology is sufficiently known, their mobility is relatively small, they are widely distributed, they should have clear tolerance limits for the environmental factor to be studied, and they react to that factor quickly and always in the same way. Examples of bioindicators are plants that suffer from impurities in the air, first of all lichens, and carnivores at the top of the food chain (e.g. white-tailed eagle, seals).

Furthermore, according to the Dictionary of Biology, a *Biomarker* is an organism which reacts easily to environmental changes, such as pollutants, and thus reveals their appearance.

Consequently, both terms might be usable, but *indicator organism* might be more orthodox in this case.

Introduction

The objective of the project was to identify new indicator organisms and biomarkers for assessment of environmental radioactivity in normal and emergency situations. The mechanisms of uptake and accumulation were also to be studied. Indicator organisms are valuable monitoring objects in emergency situations because they accumulate effectively (and often very rapidly) radioactive substances from the medium. Thus, they can give information about the nuclide composition and dispersion of radionuclides in early stages of emergency situations.

Traditionally, many organisms have been successfully used in radioecology (such as lichens, mosses, mushrooms, mussels and macro algae, especially the bladder-wrack *Fucus vesiculosus*). The objective of the project was to search new useful organisms accumulating effectively and specifically certain radionuclides in various Nordic ecosystems (terrestrial, fresh water, marine), and to compare their indicator value with those of the earlier known indicators. Since the Chernobyl accident, there was plenty of data on ^{137}Cs in biota but other radionuclides had been left to less attention. The aim of the project was to get more information on other long-lived nuclides (^{90}Sr , Pu and Am) and the most abundant discharge nuclides from the nuclear power plants (e.g. ^{60}Co). In addition, the usability of different organs and tissues of the organisms as indicators were to be studied.

The study was expected to yield new data on the occurrence, transport and concentrations of many important radionuclides in potential candidates of indicator organisms concerning a wide scale of Nordic ecosystems. The choice of candidates should be based on references to literature concerning accumulation of radionuclides and stable elements to certain species or groups of organisms.

The project was participated by eight laboratories representing all the Nordic countries: Risø National Laboratory (Denmark), University of Faroe Islands, STUK (Finland), Icelandic Radiation Protection Institute, Norwegian Radiation Protection Authority, Institute for Energy Technology (Norway), Norwegian University of Life Sciences and Lund University (Sweden).

The duration of the project was 3.5 years (from June 2002 to December 2005). The results of the project will be presented via publications in open literature (some already published, some in print). In addition, the Project Group has decided to prepare a joint scientific publication based on the results. Each laboratory should contribute to this publication. This report should

give a common overview about the use of indicator organisms in the Nordic ecosystems, not only national results from various countries.

This report summarizes the results of the whole project. More detailed results are presented in the papers published in these Proceedings by all the participating laboratories.

Collection of data

The idea in sampling was to take the samples from relatively small areas where the environmental factors (type of soil etc.) and the amount of radioactive deposition are likely homogenous, which makes it possible to compare the indicator value of various organisms.

Data were collected from about 180 organisms (species, family or group). The list of organisms consists of 60 mushrooms, 7 lichens, 5 mosses, 13 spore-bearing plants (*Pteridophytes*), 38 seed plants, 12 algae (including plankton and periphyton), 12 benthic animals, 15 fish species, 11 birds, 4 seals, one whale, a lynx and a Polar Bear. A majority of the organisms (109 species) represent terrestrial environment, 58 of them are from the marine or brackish-water environment and 19 from the fresh water environment. The most extensively studied individual organisms were from the marine/brackish-water environment, such as *Fucus vesiculosus* and *Mytilus edulis*.

Studies carried out in the participating laboratories within the project

1. Denmark: Risø National Laboratory

The contribution of the Risø National Laboratory consisted of five separate studies:

- 1) Polonium-210 and lead-210 in Seal Flesh and Liver
- 2) Transfer of ^{99}Tc from seawater to biota, data from 1993-2003
- 3) Transfer of radionuclides to plants
- 4) Iodine-129 in the marine environment
- 5) Radiocaesium uptake in mushrooms

These studies produced valuable information about the accumulation of ^{210}Po , ^{210}Pb , ^{226}Ra , ^{232}Th , ^{129}I and ^{127}I in biota. The earlier data on these nuclides have been relatively few.

2. University of Faroe Islands

The Faroese contribution to the INDOFERN project included measurements of activity concentrations of ^{137}Cs in selected indicator organisms from terrestrial, fresh water and marine environments typical of the Faroe Islands. Results from earlier measurements during the last four decades were also considered. The selected indicator organisms were *Empetrum sp.*, moss, lichen, *Calluna vulgaris* and *Erica cinerea* from the terrestrial environment, trout from the fresh water environment and haddock from the marine environment.

3. Finland: STUK-Radiation and Nuclear Safety Authority

The contribution of the NPP Environmental Laboratory at STUK was focused on three subprojects:

- 1) Study of indicator organisms in the terrestrial and marine environments of the Loviisa Nuclear Power Plant (South coast of Finland) in 2000
- 2) Study of indicator organisms in the terrestrial and marine environments of the Olkiluoto Nuclear Power Plant (West coast of Finland) in 2001
- 3) Study of indicator organisms in the terrestrial and freshwater environments in the Finnish Lake District, in an area (Mänttä) that was fairly strongly contaminated with the Chernobyl fallout in 1986. This expedition was carried out in 2003.

In total 112 (68 terrestrial and 44 aquatic) samples were taken from the Loviisa area, 144 (71+73) from the Olkiluoto area (+ those of the regular monitoring programmes going on in the areas) and 99 (51+48) from the Mänttä area. All the samples were analysed for gamma nuclides and a notable number of samples also for strontium and transuranics.

4. Icelandic Radiation Protection Institute

In Iceland, the study was focused on selected indicator organisms in Icelandic terrestrial, fresh water and marine environments. In the terrestrial environment the organisms were *Racomitrium* sp., *Equisetum fluviatile*, *Equisetum palustre*, *Empetrum nigrum*, *Calluna vulgaris*, *Eleocharis palustris*, *Carex bigelowii* and *Festuca richardsonii*, in the fresh water environment the trout, *Salvelinus alpinus*, and in the marine environment the seaweeds *Fucus vesiculosus* and *Ascophyllum nodosum*, and the minke whale *Balanopetra acutorostrata*.

5. Norwegian Radiation Protection Authority (NRPA)

The contribution of the NRPA consisted of five studies:

- 1) Transfer factors of ^{137}Cs in different mushrooms species
- 2) Aggregated transfer factor of ^{137}Cs in lynx (*Lynx lynx*)
- 3) Concentration factors of $^{239+240}\text{Pu}/^{241}\text{Am}$ in mussels (*Mytilus edulis*)
- 4) Concentration factors of ^{99}Tc in lobsters (*Homarus gammarus*)
- 5) Concentration factors of ^{99}Tc in seaweed (*Fucus vesiculosus*).

The studies were based on a noteworthy number of data. 1283 samples were analysed in the mushroom study and muscles of 748 animals were analysed for ^{137}Cs in the lynx study. In both studies the samples were collected from different parts of Norway. In the lobster study the number of samples was 63 (40 males, 23 females).

6. Norway: Institute for Energy Technology (IFE)

The studies carried out by the Institute for Energy Technology were divided into three subprojects:

- 1) A vegetation-square study in the Ringeby-Koppang region that was fairly strongly contaminated with the Chernobyl fallout in 1986

- 2) Seasonal variation in activity concentrations of ^{137}Cs , ^{40}K , ^7Be , ^{228}Ra , ^{99}Tc , ^{90}Sr and $^{239,240}\text{Pu}$ in *Fucus vesiculosus* and *Ascophyllum nodosum* from the south-eastern coast of Norway
- 3) ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ in *Potamogeton perfoliatus* from the River Nitelva.

The Ringeby-Koppang study was focused on comparison of six species (*Rozites caperata*, *Boletus sp.*, *Cladina stellaris*, *Athyrium distentifolium*, *Betula nana* and *Empetrum hermaphroditum*) as indicators of ^{40}K , ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ in the environment.

7. Norwegian University of Life Sciences (UMB)

The contribution of the Norwegian University of Life Sciences consisted of six studies:

- 1) Changes in ^{99}Tc activity in algae from the Norwegian coast
- 2) Concentration factors in seaweed
- 3) Measurement of ^{99}Tc in lobster, fish and winkle
- 4) Uptake studies in fish
- 5) Archive data on transfer from soil to vegetation
- 6) Uptake studies of ^{99}Tc in reindeer.

The contribution of the UMB involved the studies of uptake mechanisms mentioned in the project proposal of INDOFERN.

8. Sweden: Lund University

The studies of the Lund University were focused on:

- 1) Ferns, other plants and fresh water mussels as indicator organisms for ^{40}K , ^{137}Cs and $^{239,240}\text{Pu}$
- 2) *Fucus* as an indicator organism for ^{99}Tc , ^{137}Cs , $^{239+240}\text{Pu}$ and ^{237}Np
- 3) Lichens as indicator organisms for Pu and ^{237}Np
- 4) Seals and their inner organs as indicators of ^{137}Cs , ^{210}Po , ^{90}Sr and other nuclides
- 5) Peat bogs as indicators of ^{137}Cs and ^{210}Po .

The study summarised data on concentration factors and aggregated transfer factors in old samples from the 1960s obtained from a sample bank and from samples collected in 2002-2004.

The results obtained in the project

Terrestrial environment

Mushrooms

Mushrooms are known to be efficient organisms in accumulating radionuclides (especially caesium) from the surrounding environment. In the INDOFERN project, mushrooms were studied in Norway (NRPA and IFE), Finland (STUK) and Denmark (Strandberg, 2004). In particular, the extensive study carried out by the NRPA in Norway (1283 samples) should be mentioned. In addition to the data collected by the STUK in the INDOFERN project, also

other data reported by Kostiainen (2005) from Finland, have been taken into account in this consideration. Most of the data were focused on ^{137}Cs , but certain results reported by STUK also considered ^{90}Sr and transuranic elements.

From the nearly 50 mushroom species studied, the best indicators for caesium were:

- certain *Cortinarius* species (esp. *Cortinarius armillatus* (NOR), *C. brunneus* (NOR) and *C. traganus* (NOR+FIN))
- certain Hedgehog mushrooms (esp. *Hydnum rufescens* (FIN) and *H. repandum* (NOR+FIN))
- certain *Tricholoma* species (esp. *Tricholoma album* (NOR))
- certain *Hygrophorus* species (esp. *Hygrophorus camarophyllus* (FIN))
- the Gypsy, *Rozites caperata* (NOR+FIN)
- certain *Amanita* species (*Amanita fulva* (NOR)); however the Fly agaric *A. muscaria*, accumulates only moderately caesium
- certain Milk cap species (esp. *Lactarius deterrimus* (NOR), *L. torminosus* (NOR), *Lactarius trivialis* (FIN) and the Red-brown lacteous agaric, *L. rufus* (FIN+NOR))
- *Boletus badius* (NOR)
- *Paxillus involutus* (NOR)
- *Leccinum scabrum* (NOR)
- *Suillus variegatus* (FIN+NOR)
- *Russula decolorans* (NOR+FIN)
- *Sacrodon imbricatus* (FIN)

The aggregated transfer factors of ^{137}Cs for these mushrooms were generally > 1 , (max. 7) on dry weight basis.

The Orange-cap boletus (*Leccinum vesipelle*), *Hygrophorus camarophyllus* and *Cortinarius traganus* were good indicators of ^{90}Sr , too.

The seasonal occurrence of mushrooms diminishes their usability in environmental monitoring especially in emergency situations.

Lichens and mosses

Lichens and mosses have traditionally been considered as good indicators of radionuclides in the environment. However, in comparison with the mushrooms, they seem to be less efficient. In Finland, the highest transfer factors of ^{137}Cs for Hair moss (*Polytrichum* sp.) and Reindeer lichen (*Cladina alpestris*) were 0.16 and 0.05 on dry wt. basis, respectively. In fact, it is not correct to calculate the transfer factors for lichens based on the amounts of radionuclides in soil, because they take most of the contamination from the air, but the motive in the comparison was to bring the different organisms to the same starting line. In some cases, the concentrations of ^{137}Cs in *Hypogymnia physodes*, growing on trunks and branches of trees, were fairly high (Ikäheimonen et al., 2003).

In the vegetation-square study carried out in the Ringeby-Koppang region in Norway, *Cladina stellaris* (= *Cladina alpestris*) was considered to be the best indicator for ^{137}Cs , although the highest concentrations and transfer coefficients of ^{137}Cs were found in Gypsy, *Rozites caperata*. In Iceland and Faroe Islands, the mosses *Racomitrium* sp., *Racomitrium lanuginosum* and Glittering feather moss (*Hylocomnium splendens*) were successfully used as indicators of caesium, but the low concentrations of ^{137}Cs in those countries made the

comparison difficult with the results from Norway and Finland. Nevertheless, *Hylocomnium splendens*, *Racomitrium laniginosum* and *Parmelia saxatilis* were found to be suitable indicators of radionuclides in these environments.

In the study at the Ringeby-Koppang region, *Cladina stellaris* (= *Cladina alpestris*) was the best indicator for Pu, out of the species measured for Pu. In Finland, *Hypogymnia physodes* showed in general higher ^{90}Sr and Pu concentrations than the Reindeer lichen. The studies carried out in Sweden showed that lichens (*Cladina stellaris*=*C.alpestris*) are very good indicators for ^{237}Np , too.

Spore-bearing plants (Pteridophytes)

Ferns have also been considered to be good indicators of radionuclides. In Finland, the transfer factors of ^{137}Cs in Common polypody (*Polypodium vulgare*) and Tall shield fern (*Dryopteris carthusiana*) were in general higher (about 0.2 on dry wt. basis) than in lichens and mosses, but lower than the highest values in mushrooms. In the Swedish data reported by the Lund University, the transfer factors were up to 0.35 for Bracken (*Pteridium aquilinum*) and 0.04 for Wood horsetail (*Equisetum sylvaticum*). In the Ringeby-Koppang region, Norway, the transfer factor for Alpine fern (*Athyrium alpestre* = *A. distentifolium*) was 0.01. In Iceland, the highest transfer factor of ^{137}Cs among the terrestrial plants studied was obtained in Horsetail (*Equisetum* sp.). In Denmark, the mean transfer factors of ^{137}Cs , ^{40}K , ^{226}Ra and ^{232}Th for Bracken and Water horsetail (*Equisetum fluviatile*) were 3.3, 19, 1.9 and 2.0, respectively.

Seed plants (Magnoliophytes)

Needles of coniferous trees are generally used as indicators in air quality control. Both in Finland and Sweden, the ranking list of the transfer factors for ^{137}Cs was the same: spruce (*Picea abies*) > pine (*Pinus sylvestris*) > juniper (*Juniperus communis*); the maximum TF values being 0.07, 0.04 and 0.002, respectively. In Finland, the transfer factors of ^{137}Cs for the leaves of birch (*Betula pendula*) and alder (*Alnus incana*) were lower than those of spruce and pine needles. In Norway, the aggregated transfer coefficient of ^{137}Cs for the Dwarf birch (*Betula nana*) was 0.02-0.03 and those of ^{90}Sr and $^{239,240}\text{Pu}$, 0.12-0.50 and 0.001, respectively.

Twigs, especially those of Heather ling (*Calluna vulgaris*) and crowberry (*Empetrum* spp.) were widely used in this study as indicators of radioactive substances in the Nordic environments. In Iceland and Faroe Islands, the Heather ling has proved to be one of the best indicators in local terrestrial ecosystems. In the Mänttä area in Finland (which was fairly strongly contaminated with Chernobyl fall-out) the transfer factor of ^{137}Cs for the twigs of blueberry (*Vaccinium myrtillus*), Heather ling and crowberry (*Empetrum nigrum*) were 0.15, 0.09 and 0.03, respectively. The concentrations of ^{137}Cs in berries of *Vaccinium myrtillus*, *V. vitis-idaea* and *Empetrum nigrum* were usually slightly higher than those in the twigs.

From the plants with herbaceous stem, e.g. Rosebay Willowherb (*Epilobium angustifolium*) and some sedge species (*Carex* spp.) have been used as indicators of radionuclides. In this study, also samples of Tormentil (*Potentilla erecta*) and Arctic festuca (*Festuca richardsonii*) were analysed for ^{137}Cs in Faroe Islands and Iceland, but the concentrations were very low. In the Mänttä area in Finland, the mean transfer factor of ^{137}Cs for Rosebay Willowherb was 0.05 and in Sweden that for Bottle sedge (*Carex rostrata*) 0.04.

Mammals

The mean aggregated transfer factor of ^{137}Cs for lynx in Norway was about 1 (0-18), which means that this big felid uptakes efficiently caesium, but it might not be an ideal indicator organism in emergency situations because of difficult availability of samples.

Fresh water environment

The share of data from fresh water environments remained rather small in the INDOFERN project. More widely data were collected only from the Mänttä area in Finland and from Sweden. In Faroe Islands and Iceland, recurrent data have been collected on ^{137}Cs and ^{90}Sr in trout (*Salmo trutta*), Rainbow trout (*Oncorhynchus mykiss*) and Arctic charr (*Salvelinus alpinus*). Pond weed (*Potamogeton perfoliatus*) has been used as an indicator of ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ in River Nitelva in Norway.

In the Mänttä area in the Finnish Lake District, the clearly highest concentration of ^{137}Cs was measured in a sample of Water horsetail (*Equisetum fluviatile*), 1 430 Bq kg⁻¹ dry wt; CF 29 200. In addition, Spiked water millfoil (*Myriophyllum spicatum*) and Broad-leaved pondweed (*Potamogeton natans*) seemed to be good indicators of ^{137}Cs . The stems and leaves, and the rootstocks of Water lilies (*Nymphaea candida* and *Nuphar lutea*) seemed to accumulate ^{137}Cs quite equally. The Freshwater clam (*Anodonta sp.*) was only a modest accumulator of ^{137}Cs . The concentration factors of ^{137}Cs for small perches (*Perca fluviatilis*) and roaches (*Rutilus rutilus*) were also relatively high (17 200 and 12 900 d.w.), and clearly higher than the CFs for these species in brackish-water on the Finnish coast.

Also in the Swedish data, the highest concentration factor (14 700) was obtained in Water horsetail (*Equisetum fluviatile*). A relatively large number of Freshwater clams (*Anodonta sp.*) were collected in Lake Åsnen in Sweden. The samples were analysed for radiocaesium and ^{210}Pb . The mussels seemed to be good indicators for polonium but not for caesium. The activity concentrations of ^{210}Pb in the soft parts and in the shells of *Anodonta* were 56.4 and 6.4 Bq kg⁻¹ dry wt., respectively. The ^{137}Cs concentration in the soft parts of the mussels was 7.4 Bq kg⁻¹ dry wt., being below the detection limit in the shells.

Marine environment

Marine ecosystems studies in brackish-water circumstances on the Finnish coast

Intensive ecosystem studies were carried out in the sea areas off Loviisa (2000) and Olkiluoto (2001), in brackish-water circumstances of the Finnish coast. Samples were taken from 27 different species (80 samples) throughout the ecosystems, and from different tissues and organs of fish and birds (64 samples).

Discharge nuclides from the local nuclear power plants (^{51}Cr , ^{54}Mn , ^{58}Co , ^{60}Co , ^{95}Zr , ^{95}Nb , $^{110\text{m}}\text{Ag}$, $^{123\text{m}}\text{Te}$ and ^{124}Sb) were only detected at the lower trophic levels of the ecosystem: in periphyton, zooplankton, phytoplankton, macroalgae, vascular plants and benthic animals.

The best indicators of ^{137}Cs among the lower organisms were periphyton, zooplankton, the Bladder-wrack (*Fucus vesiculosus*), phytoplankton, filamentous green algae (*Cladophora glomerata*) and Spiked water milfoil (*Myriophyllum spicatum*). Among the vertebrates, the

best indicators of ^{137}Cs were the predatory fish, perch (*Perca fluviatilis*) and pike (*Esox lucius*). The concentration factors were clearly lower than in fresh water (see above): 3 500 for periphyton, 2 100 for perch and 1 800 for pike (on dry weight basis). In the inner organs and milt and spawn of fish the ^{137}Cs concentrations were lower than in fish flesh. But on the contrary, in some fish-eating birds (Goosander, *Mergus merganser* and Great black-backed gull, *Larus marinus*), the highest concentrations were not in the muscles but in liver and entrails. In bird's eggs the concentrations were generally very low; especially in egg shells. The benthic animals, *Saduria entomon* and *Macoma balthica* were slightly more efficient than *Fucus* in accumulating ^{90}Sr .

Macroalgae in real marine environments

The brown algae *Laminaria hyperborea*, *L. digitata*, *Ascophyllum nodosum*, *Pelvetia canaliculata*, *Fucus spiralis*, *F. vesiculosus*, *F. serratus*, *F. distichus* and *Halidrys siliquosa* have been widely, traditionally and successfully used as indicator organisms of radioactive substances in the Nordic sea areas.

The studies carried out in 2002-2003 nearby Tromøya in south-eastern Norway show that the concentrations of all the measured radionuclides, except those of ^{99}Tc , were higher in Bladder-wrack (*Fucus vesiculosus*) than in Egg Wrack (*Ascophyllum nodosum*). The activity concentrations of ^{137}Cs were on an average 30% higher and those of ^{40}K about 10% higher in *Fucus* than in *Ascophyllum*. On the contrary, the activity concentrations of ^{99}Tc were on an average 60% higher in *Ascophyllum* than in *Fucus*. For ^{90}Sr , the results did not show any clear difference between the two species. The ^7Be concentrations were significantly higher in *Fucus* than in *Ascophyllum* and those of ^{228}Ra were in *Fucus* on an average twice as high as in *Ascophyllum*. The $^{239,240}\text{Pu}$ concentrations were very low in both species. The concentration ratios of ^{137}Cs , ^{90}Sr , ^{99}Tc and $^{239,240}\text{Pu}$ between the algae and seawater were 130, 260, 90 000 and 11 000 for *Fucus vesiculosus* and 100, 150, 120 000 and 5 900 for *Ascophyllum nodosum*, respectively.

The data show that there were great seasonal variations in the concentrations of the measured radionuclides. For ^{137}Cs , ^{90}Sr , ^{228}Ra and ^7Be , the seasonal variations showed the same pattern for the two species, whereas for ^{99}Tc the pattern was different. The difference indicates different dynamic processes in uptake and retention of technetium in the two species. Because *Ascophyllum nodosum* grows more slowly and lives longer than *Fucus vesiculosus*, technetium might be integrated over a longer time span in *Ascophyllum*, and also the effect of growth dilution should be less pronounced in *Ascophyllum* than in *Fucus*. This study confirms that *Fucus vesiculosus* is a very good indicator for radioactivity in marine environments. *Ascophyllum nodosum* is also a good indicator, but has slightly lower concentration factors for all the radionuclides considered in this study, except for technetium.

The concentration factors of ^{99}Tc in *Fucus vesiculosus* at Hillesøy in northern Norway varied between 60 000 and 450 000 (mean 230 000) Bq/kg d.w. per Bq/L. The activity concentrations in seaweed responded rapidly with increased ^{99}Tc concentrations in water.

At Danish coastal sites the concentration ratios (Bq/kg d.w. per Bq/L) of ^{137}Cs and ^{99}Tc in *Fucus vesiculosus* varied between 130 - 400 and 47 000 - 190 000. The concentration ratios for ^{137}Cs showed a direct correlation to water salinity with higher values in low salinity water in Bornholm than in high salinity water on the west coast of Jutland.

On the Swedish west coast the concentration factor of ^{99}Tc for *Fucus vesiculosus* varied between 28 000 and 150 000. In Skagerrak, the concentration factor of $^{239+240}\text{Pu}$ was estimated to be about 7000 and that of ^{237}Np about 400.

Norwegian studies showed that the concentration of ^{99}Tc is higher in the stipes than in the blades of Cuvie (*Laminaria hyperborea*), which probably reflects a long-term retention in this part of the plant. Results from Kvitsøy between 1997 and 2000 indicated an increase in ^{99}Tc activities in both stipes and blades.

The Bladder-wrack, *Fucus vesiculosus*, has proved to be a suitable indicator of radioactivity also in the Faroese marine environment. The shorter half-life of Chernobyl-derived ^{137}Cs compared to that from the 1960s, which was demonstrated in the Faroese studies, was explained by the fact that the input from Chernobyl came as a short pulse, while the input to the atmosphere in the 1960s was distributed over a longer time scale. The results show that *Ascophyllum nodosum* and *Fucus vesiculosus* contain practically the same activity concentrations of ^{137}Cs in the Faroe Islands.

In Iceland, samples of *Fucus vesiculosus*, *Ascophyllum nodosum*, mussels, crustaceans, aquatic birds, seals, a whale and a polar bear have been included in the indicator studies of the marine environment, but the radiocaesium concentrations have been very low or below the detection limit of the gamma spectrometer.

Table 1. Concentration factors for ^{99}Tc found from literature (Salbu and Holm, 2005) as reported by the UMB (Annual report 2005 of UMB).

Species	CF	Reference
Lobster (tail muscle)	6 850	(Smith et al., 2001)
Lobster	8 000	(Brown et al., 1999)
Lobster	1 000	(IAEA, 2004)
Prawns	2 800	(Smith et al., 2001)
Mussels (<i>Mytilus</i>)	970	(Smith et al., 2001)
Mussels (<i>Mytilus</i>)	486	(Brown et al., 1999)
Oysters	280	(Smith et al., 2001)
Molluscs	500	(IAEA, 2004)
Fish (flesh)	12	(Smith et al., 2001)
Fish	80	(IAEA, 2004)
Zooplankton	100	(IAEA, 2004)
Phytoplankton	40	(IAEA, 2004)
Macroalgae	30 000	(IAEA, 2004)
<i>Fucus vesiculosus</i>	132 000	(Smith et al., 2001)
<i>Fucus vesiculosus</i>	85 000	(Holm et al., 1984)
<i>Fucus serratus</i>	48 000	(Holm et al., 1984)
<i>Fucus spiralis</i>	16 000	(Holm et al., 1984)
<i>Fucus distichus</i>	102 000	(Holm et al., 1984)
<i>Ascophyllum nodosum</i>	178 000	(Holm et al., 1984)
<i>Laminaria digitata</i>	22 000	(Holm et al., 1984)
<i>Halidrys siliquosa</i>	63 000	(Holm et al., 1984)
<i>Pelvetia canaliculata</i>	102 000	(Holm et al., 1984)

Shellfish

In 2001-2003, 63 samples of lobsters were collected in four different areas along the southern and western coasts of Norway. Large variations in activity concentrations of ^{99}Tc in tail muscles of the lobsters were observed. The concentrations ranged from 2.2 to 62 Bq kg⁻¹ fresh wt. and the concentration factors varied between 1 400 and 43 600 (on fresh weight basis). The ^{99}Tc concentrations and the concentration factors were clearly higher in female lobsters compared to male lobsters.

Concentration factors of ^{99}Tc for some crustaceans from the Norwegian coast, as reported by the UMB, are given in Table 2.

Table 2. ^{99}Tc in crustaceans with Concentration Factors for Norwegian coastal areas using average seawater concentration of 0.5 Bq/m³ (=0.0005 Bq/l) for ^{99}Tc (Annual Report 2005 of UMB).

Crustacean	Bq/kg dry weight average + range	CF (dry wt.)
Crab	0.121 ± 0.005	240
Crab	0.61 ± 0.01	1 220
Lobster	34 ± 12 (25 – 42)	68 000
Lobster	79 ± 13 (69 – 88)	158 000

Fish

Concentration factors of ^{99}Tc for certain fish species from the Norwegian coast, as reported by the UMB, are given in Table 3.

Table 3. ^{99}Tc in fish from the Norwegian coastal areas with Concentration Factors using average sea water concentration of 0.5 Bq/m³ (=0.0005 Bq/l) for ^{99}Tc (Annual Report 2005 of UMB).

Fish	Bq/kg dry weight	CF
Halibut (Kveite)	< 0.04	-
Halibut (Kveite)	0.132 ± 0.003	260
Cod (Torsk)	0.04 ± 0.01	80
Mackerel (Makrell)	0.19 ± 0.01	380
Lumpfish (Steinbit)	< 0.04	-
Salmon (Laks)	0.12 ± 0.01	240
Salmon (Laks)	0.16 ± 0.01	320
Herring (Nordsjø Sild)	0.23 ± 0.05	460
Salmon (Laks)	0.15 ± 0.01	300
Frog fish (Breiflabb)	0.12 ± 0.06	240
Frog fish (Breiflabb)	< 0.04	-
Salmon (Laks)	0.25 ± 0.01	500
Halibut (Kveite)	0.40 ± 0.01	800
Salmon (Laks)	0.17 ± 0.01	340
Cod (Torsk)	< 0.04	-
Herring (Sild nvg)	0.19 ± 0.01	380

Seals and other mammals

Different organs of seals (Grey seal, *Halichoerus grypus* and Common seal, *Phoca vitulina*) from the west and south coast of Sweden and from the Baltic Sea were analysed by the Lund University for ^{90}Sr , ^{99}Tc , ^{137}Cs , ^{210}Po and $^{239+240}\text{Pu}$. The levels of ^{210}Po were high compared to average global values of ^{210}Po in fish, molluscs and crustaceans. The ^{210}Po concentrations were higher in liver and kidney than in meat, whereas radiocaesium is rather homogeneously distributed in different organs. A clear salinity effect could be seen in the levels of radiocaesium; the levels were on the west coast of Sweden much lower than in the Baltic Sea. The ^{210}Pb concentrations were very low, i.e. generally not detectable or 100 times lower than those of ^{210}Po . ^{90}Sr could be detected in teeth and skeletons of the seals. Low concentrations of ^{99}Tc were found while plutonium was not detectable. Ranges of activity concentration ratios are given in Table 4.

Table 4. Ranges of activity concentration ratios (CR = soft tissue (dry weight)/water) for different organs of seals from west and south coast of Sweden (Annual Report 2004 of Lund University).

Organ	CR, ^{210}Po	CR, ^{137}Cs (east coast)	CR, ^{137}Cs (west coast)
muscle	5 000 - 40 000	375-1 250	15 - 600
liver	12 000 - 125 000	250-750	15 - 360
kidney	10 000 – 130 000	250-875	15 - 500
genitals	1 000 – 3 500	375-1 000	15 - 360

In Finland, 25 samples of seals (*Phoca hispida* and *Halichoerus grypus*) and their inner organs, collected in 1986-1992 from the south and southwest coast of Finland were analysed for gamma-emitting nuclides (Ilus et al., 2005). The concentration ratios of ^{137}Cs between seal flesh and seawater, and between seal flesh and Baltic herring were clearly higher for the grey seal (*Halichoerus grypus*) than for the ringed seal (*Phoca hispida*). The concentration ratios between seal flesh and seawater were 2 730 - 4 080 (average 3 430) for the grey seal, and 1 080 - 1 830 (average 1 460) for the ringed seal on dry weight basis. The concentration ratios between seal flesh and Baltic herring were 8.8 and 4.8 on fresh weight basis.

Five subsamples of flesh and liver from a Harp seal (*Phoca groenlandica*) were taken at Thule, Greenland, in August 2003. The concentrations of ^{210}Po were about 8 Bq kg⁻¹ f.w. (28 Bq kg⁻¹ d.w.) in flesh and 43 Bq kg⁻¹ f.w. (150 Bq kg⁻¹ d.w.) in liver of the seal. The concentration ratios of ^{210}Pb between seal flesh and seawater and between seal liver and seawater were 28 000 and 151 000 Bq/kg d.w. per Bq/L, respectively. The concentration ratios of ^{210}Pb , ^{137}Cs and $^{239,240}\text{Pu}$ between seal flesh and seawater were 700, 100 and 20, respectively.

In 2004, ten samples of whale meat from Minke whales (*Balaenoptera acutorostrata*) were measured in Iceland. The results indicated a low radiocaesium uptake; the activity concentrations of ^{137}Cs were less than 0.6 Bq kg⁻¹ fresh wt.

Uptake studies

Injection tests carried out in Norway show that almost all the injected $^{99\text{m}}\text{TcO}_4^-$ was found in the spleen (52%) and kidney (40%) of rainbow trout, while small traces were found in the

brain and the eyes. Preliminary results from a pilot study show a surprisingly low retention of Pu with the highest fractions being found in head, skin, bone and muscle/filet.

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Concentration factors and aggregated transfer factors in selected bioindicators from Sweden

E. Holm, C. Samuelsson, M. Holstensson

Lund University, SE-22185 Lund, Sweden

Introduction

Bioindicators are used as monitors of environmental pollution. By definition there are two aspects a) when real effects on population are seen or b) when they show high transfer and concentration factors without any effect. The latter is at present levels the case for radioactive elements. Previously most common has been to use lichens and mosses in the terrestrial environment and mussels and algae in the marine environment. Several plants are promising and less commonly used such as ferns (e.g. Bracken, *Pteridium aquilinum*) and the fresh water plant, Water horsetail (*Equisetum fluviatile*). These show high uptake of radiocaesium but also accumulate plutonium and americium. Even if the uptake of transuranic elements is low, this behaviour is rather unique for plants. The work will be directed to understand mechanisms and how they can be used for mapping deposited radioactivity in an emergency situation.

Results

On bases of available data we have calculated concentration factors C_f and aggregated transfer factors, ATF according to their definitions i.e. $\text{Bq kg}^{-1}(\text{biota})/\text{Bq kg}^{-1}(\text{water})$ and $\text{Bq kg}^{-1}(\text{biota})/\text{Bq m}^{-2}$ respectively. For earlier samples the water concentrations can only roughly be estimated while deposition data are rather well known. For fresh water samples it is a little awkward to use concentration factors since the uptake generally is not from water. Therefore a kind of Aggregated Transfer Factor has also been calculated on basis of using the same areal deposition in sediments as on land. We have used dry weight concentrations for biota. For the older samples (1966-69) an areal deposition of 2500 Bq m^{-2} of ^{137}Cs and 40 Bq m^{-2} for $^{239+240}\text{Pu}$ have been used. In 2002-2003 the ^{137}Cs concentration is estimated to 2000 Bq m^{-2} taking into account the Chernobyl fallout and physical decay. The plutonium deposition, especially in this area, was very small (about 20 mBq m^{-2}) following the Chernobyl accident. Our data are compiled for various bioindicators and are shown in the following two tables.

Table 1. Results for various samples from the sample bank

Type of sample	Collection dates	^{137}Cs , C_f	$^{239+240}\text{Pu}$, C_f	^{137}Cs , ATF	$^{239+240}\text{Pu}$, ATF
Bracken	1968-69			$(4.3-35)\times 10^{-2}$	$(1-3)\times 10^{-3}$
Wood horsetail	1962			4×10^{-2}	2.1×10^{-2}
Bottle sedge	1966			4×10^{-2}	5×10^{-3}
Water horsetail	1962-69	3000-18000	7000-22000	$(4.7-45)\times 10^{-2}$	$(0.7-2.2)\times 10^{-2}$
Bulrush	1965	260-300	4000	$(4.1-4.7)\times 10^{-3}$	4×10^{-3}

Table 2. Samples collected 2002-2004

Type of sample	Collection dates	^{137}Cs , C_f	$^{239+240}\text{Pu}$, C_f	^{137}Cs , ATF	$^{239+240}\text{Pu}$, ATF
Bracken, plants	2002			$(2.5-14)\times 10^{-2}$	
Bracken, roots	2002			$(2.5-16)\times 10^{-2}$	
Rosebay Willowherb, plants	2002			$(3-10)\times 10^{-3}$	
Rosebay Willowherb, roots	2002			$(3-8)\times 10^{-3}$	
Common polypody	2003			4×10^{-3}	
Common Juniper needles	2003			4×10^{-4}	2×10^{-4}
Scots Pine, needles	2003			$(2-3)\times 10^{-3}$	3×10^{-4}
Norway Spruce, needles	2003			$(4-18)\times 10^{-3}$	3×10^{-4}
Water horsetail	2003	14700		$(2.2)\times 10^{-2}$	
Bulrush	2003	1100		1.6×10^{-3}	
Common reed	2003	2200		3.2×10^{-3}	
White water lily	2003	5000		7.5×10^{-3}	
Broad-leaved pondweed	2003	7000		1.1×10^{-2}	
Common club-rush	2003	600-1300		$(0.9-1.8)\times 10^{-4}$	
Fresh water mussel	2004	2500			

Lichens

The fallout and isotopic ratios of plutonium, $^{238}\text{Pu}/^{239+240}\text{Pu}$ and $^{240}\text{Pu}/^{239}\text{Pu}$, from the Chernobyl accident has been studied and Np-237 was studied in lichen (*Cladina stellaris*) at 18 sites in Sweden using post Chernobyl samples from our sample bank. The activity concentrations ranged from 0.1 to 2 mBq/kg with the highest concentrations where Chernobyl fallout was highest. Data show that at the Gävle area 20 % of Np-237 originated from Chernobyl while the corresponding fraction for ^{239}Pu was 70%. The atomic ratio Np/Pu in Chernobyl fallout is lower than in nuclear test fallout. Neptunium is more rapidly than plutonium transported downwards in the lichen plant to underlying soil. In spite of this lichens are very good bioindicators also for neptunium especially for assessing source terms.

Peat bogs

Large areas of Nordic countries consist of different types of peat bogs. These act as traps for fallout radionuclides and other pollutants. They are ecologically important from especially hydrologic point of view. In addition they are and may become more important as energy source and the waste products must be considered from radiological point of view.

In order to establish the fallout levels for radiocaesium and plutonium cores from a peat bog were collected during the summer 2004. Such a peat bog, i.e. the plants on the surface and the upper layer might constitute an effective bioindicator. These peat bogs are normally rather undisturbed.

The growth rate can be determined by the so called ^{210}Pb method.

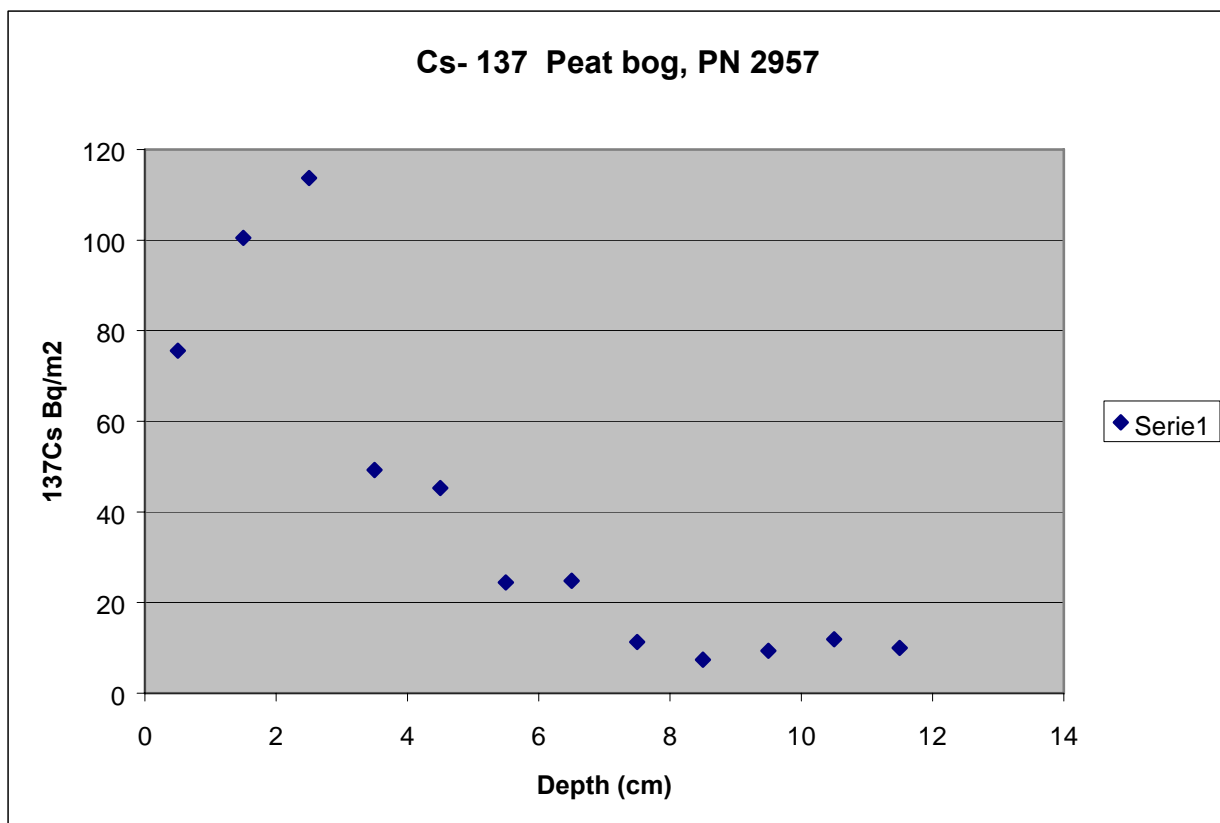


Figure 1. Activity concentration of ^{137}Cs in a peat bog as a function of depth.

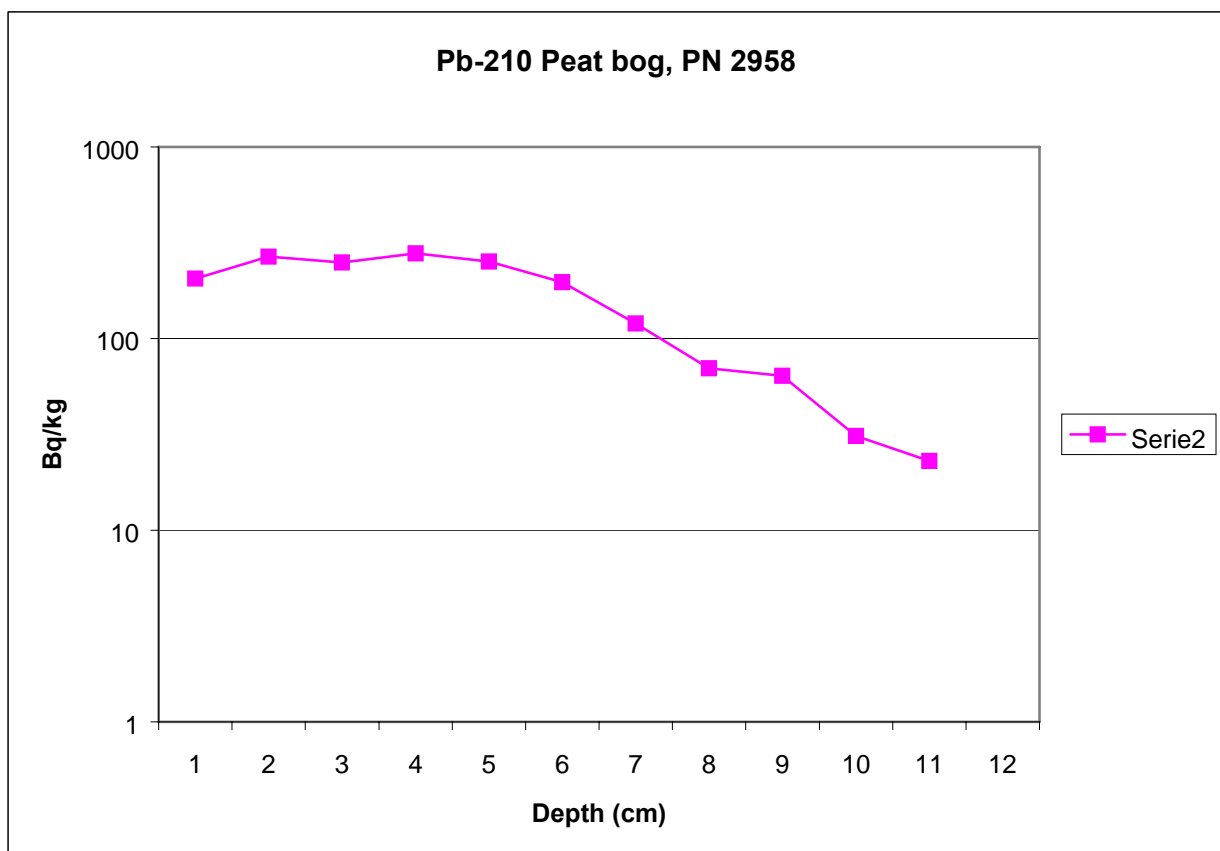


Figure 2. Activity concentration of ^{210}Pb in a peat bog as a function of depth.

Fucus sp.

Technetium. The studies of ^{99}Tc at the Swedish west coast using *Fucus* sp. as bioindicator have been terminated. The ^{99}Tc activity concentrations were measured in samples from the Swedish west coast collected in 1991, 1995 and 2001 over a latitude from 55.40 °N to 58.87 °N. Time series from Särö, 56.76 °N, are available from 1967 – 2001. The data were published in Journal of Environmental Radioactivity (Lindahl *et al.*, 2003). Data are available from 1967 – 2001. As well known the major source is releases from the Sellafield reprocessing plant. The data show a transit time of 4-5 years to the Swedish west coast.

It is clearly shown that the CF for Tc versus salinity has an opposite behaviour compared to caesium. The CF is for example 28 000 at a salinity of 9.2 ‰ and 115 000 at salinity of 21.5 ‰.

Table 3. Estimated ^{237}Np , $^{239+240}\text{Pu}$ and ^{99}Tc concentration factors in *Fucus vesiculosus* from the Swedish west coast collected 1995-2001.

Latitude °N	Salinity ‰	^{237}Np , mBq kg ⁻¹	$^{239+240}\text{Pu}$ mBq kg ⁻¹	^{99}Tc Bq kg ⁻¹	^{137}Cs Bq kg ⁻¹	^{237}Np C _f	$^{239+240}\text{Pu}$ C _f	^{99}Tc C _f	^{137}Cs C _f
55-59	5-22	0.2-1.0	20-120	10-130	5-25	270 -800	8000-10000	30000-120000	120-550

Seals

The grey seal (*Halichoerus grypus*) is the largest seal in the Baltic Sea and exists also at the Norwegian west coast, Iceland and along the British Islands. A male grey seal can reach a length of 3 m and a weight of 300 kg. Harbour seal (*Phoca vitulina*) is more common in the North Sea and can reach a length of 2 m and a weight of 130 kg. Their main food is herring but also other fish species, shellfish and crustaceans are consumed.

Relatively few studies have been done regarding radioactivity in seals, especially following the Chernobyl accident. Within the EU-project Framework for Assessment of Environmental Impact of Ionizing Radiation (FASSET) it was recently concluded that particularly radiological data on marine mammals are lacking .

Our study of radioactivity in seals shows that radiocaesium as expected has higher concentrations in seals where water salinities are lower. The whole body content of ^{137}Cs in seals from the Baltic Sea is often 10 000–20 000 Bq and whole body counting could easily be accomplished. Polonium is accumulated in the food chain in particular to internal organs such as liver and kidneys. The activity ratio $^{210}\text{Po}/^{210}\text{Pb}$ is very high in this top predator with high concentration factors, which is in agreement with other marine studies. The doses from polonium can exceed 1-2 mGy per year and if a quality factor had been applied this would correspond to 20-40 mSv per year. It is of interest to compare doses from natural radionuclides with those of anthropogenic in order to assess the radiological impact.

^{90}Sr could be detected in teeth and skeleton but no trend with time was found. Low concentrations of ^{99}Tc were found while plutonium was not detectable.

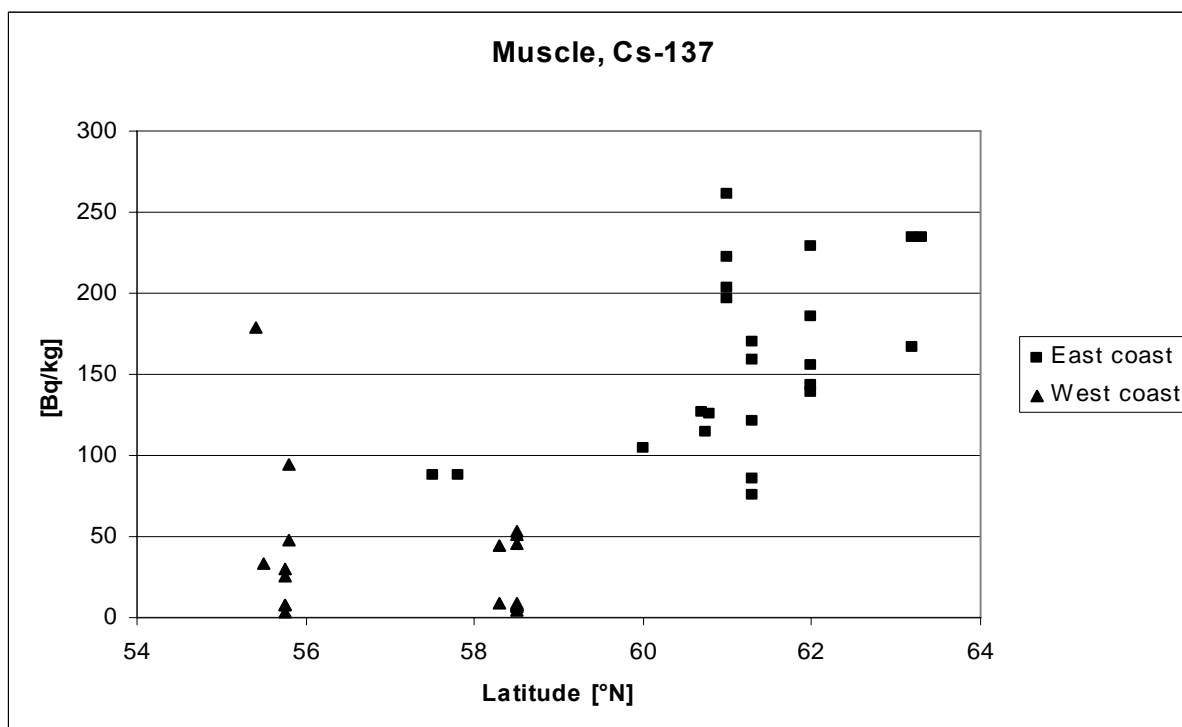


Figure 2. Activity concentration of ^{137}Cs in muscle tissue from seals from the Swedish coast.

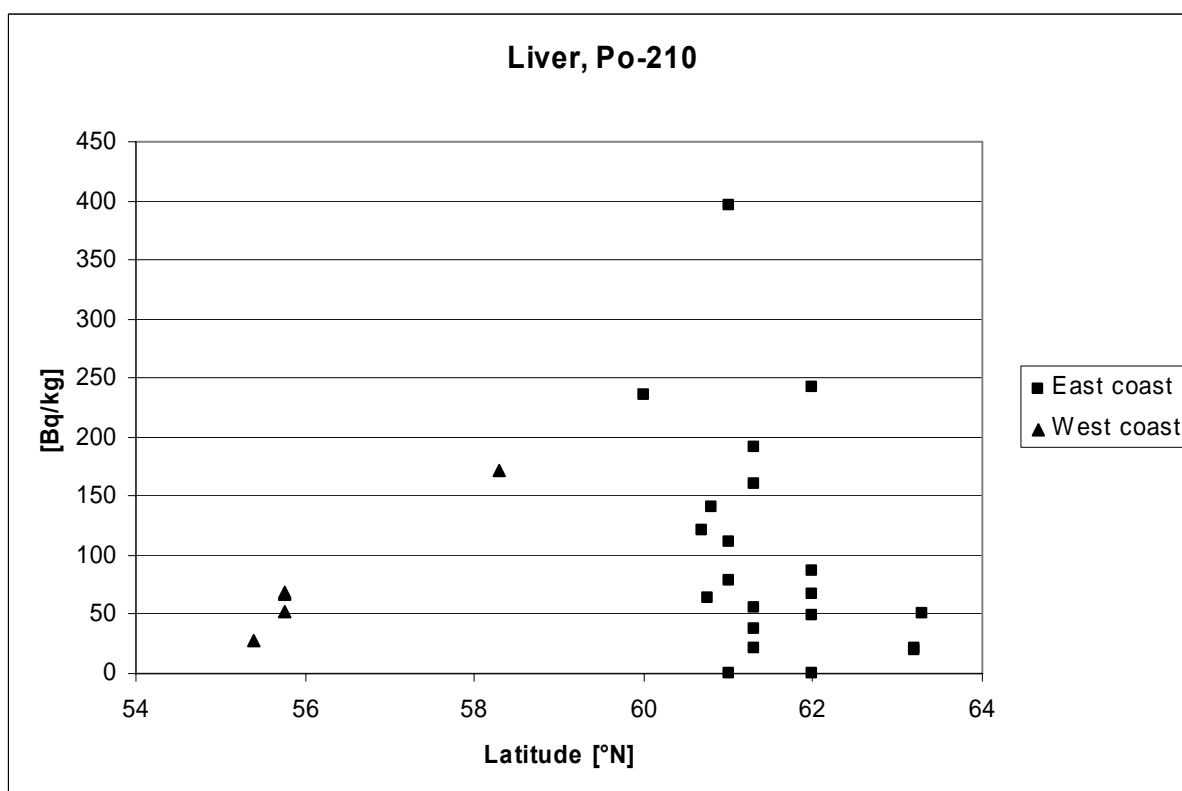


Figure 3. Activity concentration of ^{210}Po in liver from seals from the Baltic Sea.

Cs-137 in terrestrial environment in Loviisa, Olkiluoto and Mänttä areas (Finland)

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

STUK- Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Introduction

The aim of the study was to clarify how Cs-137 is accumulated in different biota species in Finnish terrestrial environment. The study was carried out in three different areas. The Mänttä area was one of the areas in Finland with the highest contamination by the Chernobyl fallout. The other areas (Loviisa and Olkiluoto) represent the environs of the Finnish NPPs located in the South and West coast of Finland, respectively (Fig. 1). The sampling was carried out in 2000 in Loviisa, in 2001 in Olkiluoto and in 2003 in Mänttä. Altogether over 120 samples from over 50 species were taken including soil profiles, mushrooms, lichens, mosses, ferns, twigs, needles, leaves and berries. All the samples were dried and homogenized and Cs-137 was measured with HPGe gamma spectrometers.

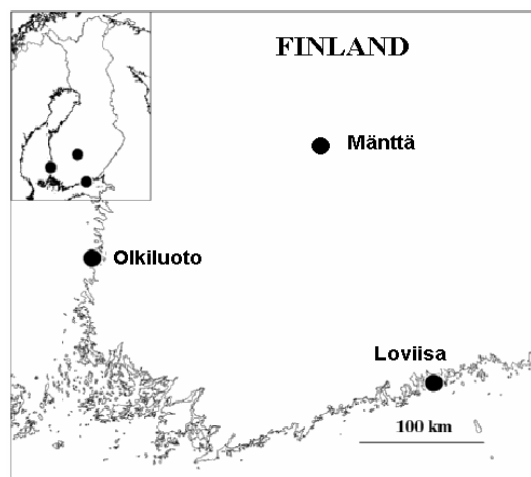


Figure 1. Location of the sampling areas.

Results and discussion

Vertical distributions of Cs-137 in the soil samples taken from the three areas are presented in Fig. 2. The total amount of Cs-137 (Bq/m^2) was clearly highest in Mänttä, about 39 kBq/m^2 . In Loviisa it was 18 kBq/m^2 and in Olkiluoto 13 kBq/m^2 . The highest Cs-137 concentrations were found in the uppermost layers in all the sampling areas. The total amount of caesium was used in the calculation of aggregated transfer factors (T_{agg}) for the sampled organisms. All T_{agg} values were calculated according to the following definition: Cs-137 concentration in the sample (Bq/kg dry weight) / total caesium in soil (Bq/m^2).

The highest Cs-137 concentrations and T_{agg} values were found in mushrooms (Table 1). The highest concentration was found in *Cortinarius traganus* sampled in Mänttä (112 kBq/kg d.w.). Other high concentrations were found in *Hydnum rufescens* (95 kBq/kg d.w., in Loviisa) and in *Hygrophorus camarophyllus* (34 kBq/kg d.w., in Mänttä). Some *Lactarius* species (e.g. *Lactarius rufus*, *Lactarius torminosus*, *Lactarius trivialis*, *Lactarius necator*) and *Russula* species (e.g. *Russula paludosa*, *Russula claroflava*, *Russula aeruginea*, *Russula decolorans*) were collected from various sampling areas, and they seemed to be quite good indicator organisms for Cs-137. The T_{agg} s in mushrooms varied from 0.04 to $3.6 \text{ m}^2/\text{kg}$ d.w.

The Cs-137 concentrations in needles, twigs and ferns are presented in Table 2, where the concentrations were clearly lower than in mushrooms. The highest concentrations were found in ferns (*Dryopteridaceae* and *Polypodium vulgare*) from 3 to 7 kBq/kg d.w. In Mänttä, the concentrations in twigs and leaves of *Vaccinium myrtillus* were also at the same level (5.6 kBq/kg d.w.). In needles of *Juniperus communis* the concentrations were quite small (about 30 Bq/kg d.w.), but in needles of *Pinus sylvestris* and *Picea abies* the concentrations were somewhat higher and close to each other (100 - 300 Bq/kg d.w. in Loviisa and Olkiluoto, 1400-2300 Bq/kg d.w. in Mänttä).

Aggregated transfer factors in ferns varied from 0.09 to 0.25 m²/kg d.w., in twigs and leaves from 0.022 to 0.15 m²/kg d.w. and in needles from 0.001 to 0.058 m²/kg d.w.

The activity concentration of Cs-137 in lichens, mosses and leaves were at the same level as in needles and ferns and the results are presented in Table 3. In *Hypogymnia physodes* samples collected from the Loviisa area in 2000, the concentrations of Cs-137 varied from 310 to 2640 Bq/kg d.w. The samples (altogether about 20 samples) were collected from a relatively small area (within 20 kms) around the Loviisa NPP and the results show that concentrations can vary greatly within a small distance. The concentrations in leaves of *Betula pendula* were small, from 50 Bq/kg d.w. in Loviisa to 470 Bq/kg d.w. in Mänttä.

The concentrations of Cs-137 in berries are presented in Table 4. The concentrations in berries of *Vaccinium vitis-idaea*, *Vaccinium myrtillus* and *Empetrum nigrum* varied from 160 to 1200 Bq/kg d.w. In other berries the concentrations were very low. The T_{agg}s for the berries of *Vaccinium vitis-idaea*, *Vaccinium myrtillus* and *Empetrum nigrum* varied from 0.009 to 0.030 m²/kg d.w.

The best indicator species seemed to be mushrooms. The results of *Hypogymnia physodes* sampled in Loviisa in 2000 showed that Cs-137 concentrations can vary greatly within a small area.

Table 1. Cs-137 concentrations and T_{agg}s for mushrooms.

	Bq/kg d.w.			T _{agg} m ² /kg d.w. (averages)		
	Loviisa 2000	Olkiluoto 2001	Mänttä 2003	Loviisa 2000	Olkiluoto 2001	Mänttä 2003
<i>Lactarius sp.</i>	3500-9500	1550-13200	20600 (avg.)	3,59E-01	6,33E-01	5,33E-01
<i>Leccinum sp.</i>	2100	520-570		1,20E-01	4,32E-02	
<i>Russula sp.</i>	2700 (avg.)	970	10400-16700	1,57E-01	7,67E-02	4,34E-01
<i>Hydnum rufescens</i>	29400-95000			3,55E+00		
<i>Rozites caperata</i>	18000			1,03E+00		
<i>Cantharellus tubaeformis</i>		2100			1,63E-01	
<i>Cantharellus cibarius</i>		680	6200		5,36E-02	1,61E-01
<i>Boletus edulis</i>	1420	870	7300	8,11E-02	6,83E-02	1,89E-01
<i>Hygrophorus camarophyllus</i>			33900			8,77E-01
<i>Cortinarius traganus</i>			93200-112400			2,66E+00

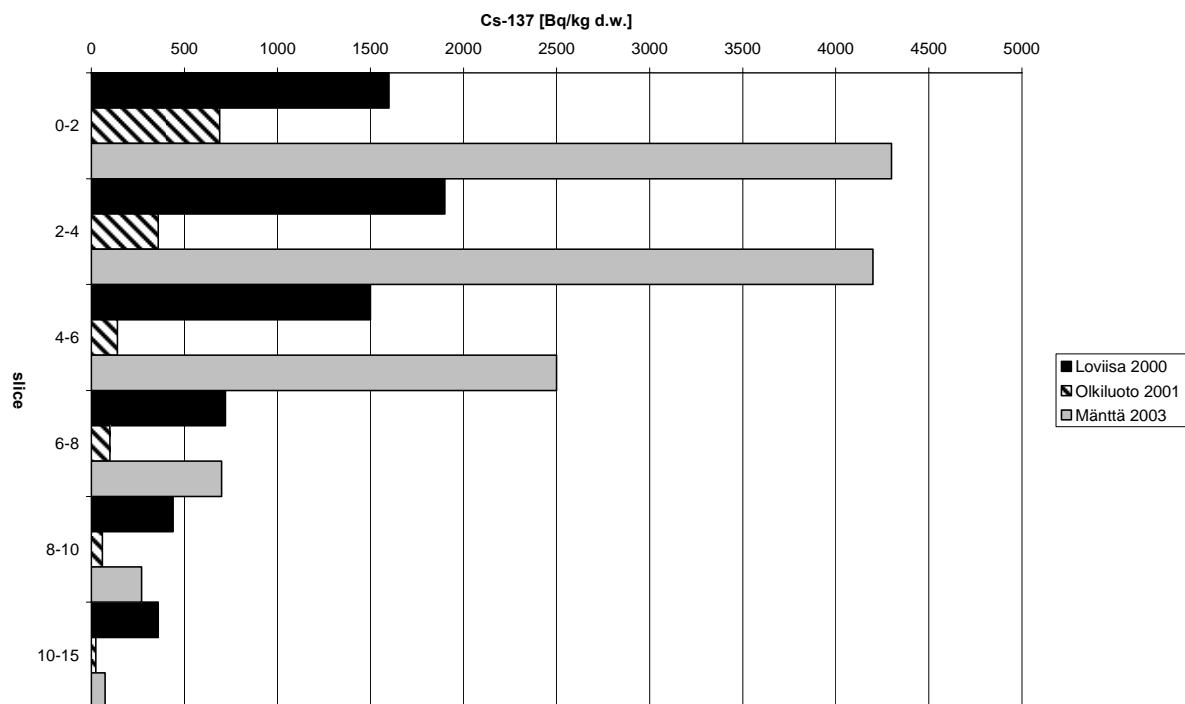


Figure 2. Soil profiles of Cs-137 (Bq/kg d.w.) in Loviisa, Olkiluoto and Mänttä.

Table 2. Cs-137 concentrations and T_{agg} s for needles, twigs and ferns.

	Bq/kg d.w.			T_{agg} m ² /kg d.w. (averages)		
	Loviisa 2000	Olkiluoto 2001	Mänttä 2003	Loviisa 2000	Olkiluoto 2001	Mänttä 2003
Needles						
<i>Pinus sylvestris</i>	170	80	1440	9,71E-03	6,30E-03	3,73E-02
<i>Picea abies</i>	210	270	2250	1,20E-02	2,13E-02	5,83E-02
<i>Juniperus communis</i>	20	30		1,14E-03	2,36E-03	
Twigs and leaves						
<i>Vaccinium myrtillus</i>	610	280	5640	3,49E-02	2,20E-02	1,46E-01
<i>Empetrum nigrum</i>		370	1150		2,91E-02	2,98E-02
Ferns						
<i>Dryopteridaceae</i>		1180	7000		9,29E-02	1,81E-01
<i>Polypodium vulgare</i>	4300	2750		2,46E-01	2,17E-01	

Table 3. Cs-137 concentrations and T_{agg} s for lichens, moss and leaves.

Plant	Bq/kg d.w.			aggr. TF m ² /kg d.w.		
	Loviisa 2000	Olkiluoto 2001	Mänttä 2003	Loviisa 2000	Olkiluoto 2001	Mänttä 2003
Lichens						
<i>Cladina spp.</i>	750	580	1940			
<i>Hypogymnia physodes</i>	310 - 2640	1050	2230			
Moss						
<i>Polytrichum sp.</i>	1100	470	5990	6,29E-02	3,70E-02	1,55E-01
Leaves						
<i>Betula pendula</i>	52	125	470	2,97E-03	9,84E-03	1,22E-02

Table 4. Cs-137 concentrations and T_{agg} s for berries.

	Bq/kg d.w.			aggr. TF m ² /kg d.w.		
	Loviisa 2000	Olkiluoto 2001	Mänttä 2003	Loviisa 2000	Olkiluoto 2001	Mänttä 2003
<i>Vaccinium vitis-idaea</i>	160	320	1160	8,92E-03	2,50E-02	2,99E-02
<i>Vaccinium myrtillus</i>	310	310		1,79E-02	2,44E-02	
<i>Empetrum nigrum</i>		350-720			4,19E-02	
<i>Rubus idaeus</i>		11			8,54E-04	
<i>Sorbus aucuparia</i>	11			6,55E-04		
<i>Hippophae rhamnoides</i>		1,7			1,32E-04	
<i>Rosa sp.</i>		1,9			1,47E-04	

Evaluation of different mushroom species as indicator organisms

Runhild Gjelsvik and Helene Stensrud

Norwegian Radiation Protection Authority, P.O.Box 55, NO-1332 Østerås, Norway

Introduction

The fallout deposition in Norway after the Chernobyl accident was highest in the highlands of central and southern parts of Norway. In 1988 and 1991 the activity levels in meat and milk increased due to ingestion of mushrooms by grazing animals. Numerous of studies have documented the mushrooms capacity to accumulate radiocaesium. In the past, the fungi have been classified as plants, but in the latest decade they are placed in their own kingdom. The kingdom fungi are mostly multicellular, parasitic and scavenging organisms and include the club fungi including mushrooms, sac fungi and the bread molds. The aim of this study was to evaluate different mushroom species as indicator organism.

Methods

To investigate the differences between accumulation capacity and transfer factor from soil to different mushroom species, 25 species were collected at 9 locations in south and central parts of Norway. Yearly sampling has been carried since 1988 and a total of 1283 samples analysed for ^{137}Cs . Entire, fresh fruit bodies were collected, homogenized and measured fresh weight. Each sample contained a mixture of fruit bodies of a single species collected within one area, enough to fill up a 200 ml box used for measurement. Levels of ground deposition of ^{137}Cs in Norway were taken from a nationwide sampling program carried out by National Institute of Radiation Hygiene in 1986 following the Chernobyl accident. The estimated ground deposition of ^{137}Cs (Bq m^{-2}) and the corresponding activity concentrations of ^{137}Cs in mushrooms were used to calculate the ratio between activity concentration in mushroom and ground deposition (transfer factor, TF). Both the mushroom and the soil data are decay corrected to 2004.

Results

The highest levels of ^{137}Cs were found in the White Knight (*Tricholoma album*) with a mean activity level above $14\,200\text{ Bq kg}^{-1}$ fresh weight. The White Knight was followed by the Red Banded Web cap (*Cortinarius armillatus*), the Gypsy (*Rozites caperata*), Brown Web cap suede (*Cortinarius brunneus*) and the Gassy Web cap (*Cortinarius traganus*). The same pattern is found for the calculated transfer factor. *Cortinarius armillatus* and *Cortinarius brunneus* had the most efficient accumulation with a transfer factor of 0.5 and $0.4\text{ m}^2/\text{kg}$. The species *Tricholoma album* and *Rozites caperata* also had high transfer factors with values around $0.3\text{ m}^2/\text{kg}$ (Figure 2).

Conclusion

As expected, considerable differences in accumulation of ^{137}Cs in different mushroom species were found. The *Tricholoma album*, *Cortinarius armillatus*, and *Rozites caperata* were found to have the highest levels. Followed by two *Cortinarius* species, *C. brunneus* and *C. traganus*. The highest transfer factors were found in the *Cortinarius armillatus* and *C. brunneus*, but also *Tricholoma album* and *Rozites caperata* had high transfer factors. Other mushroom species, e.g. *Leccinum versipelle* (Orange Birch Bolete), *Amanita muscaria* (Fly Agaric), *Boletus subtomentosus* (Suede Bolete), *Collybia butyracea* (Butter Cap) generally show a low radiocaesium uptake and are therefore not considered as good indicators. Even though *Tricholoma album*, *Cortinarius armillatus*, *C. brunneus*, *C. traganus*, and *Rozites caperata* accumulate high levels of ^{137}Cs , their seasonality and local occurrence should be evaluated before they are considered as good indicator organisms.

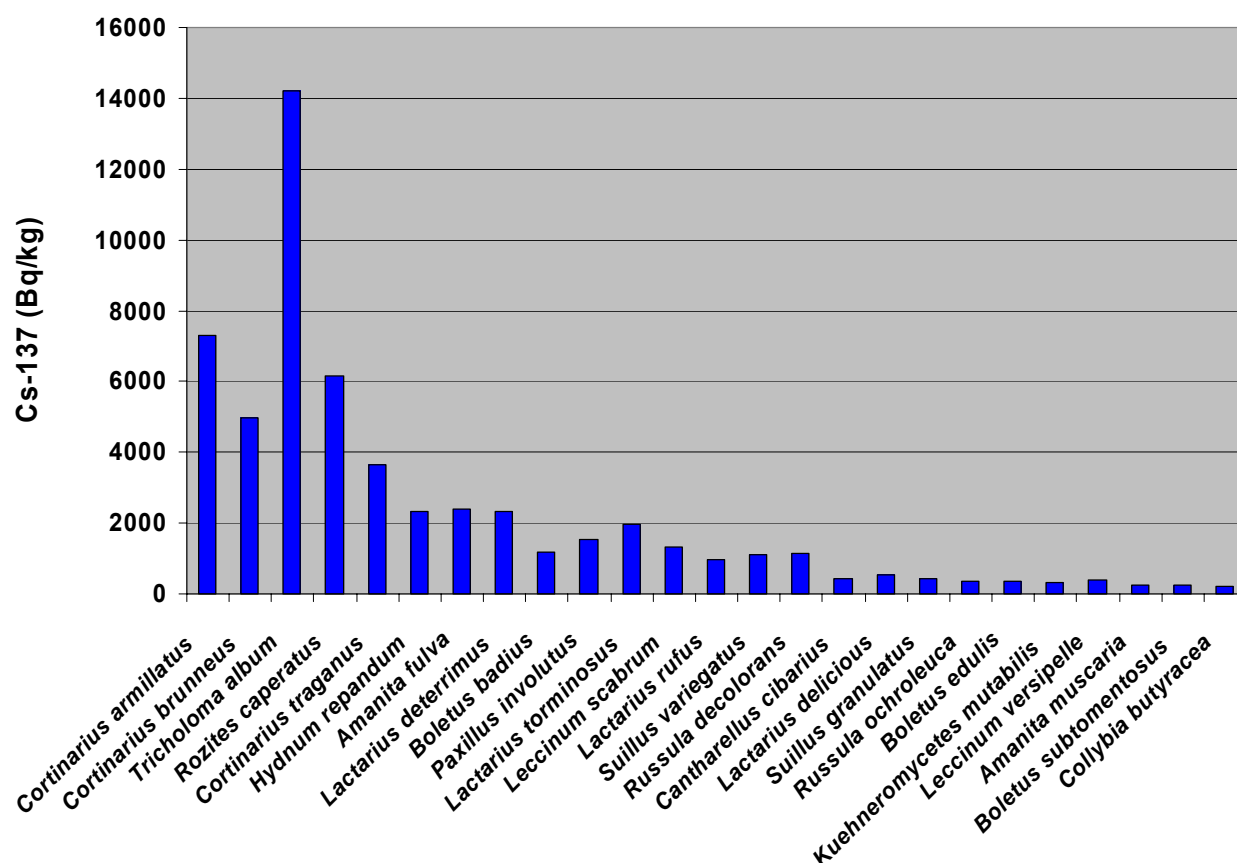


Figure 1. Mean activity levels of ^{137}Cs (Bq/kg dry weight) in different mushroom species collected in 9 different locations in Norway during the years 1988-2002. All data are decay corrected to 2004.

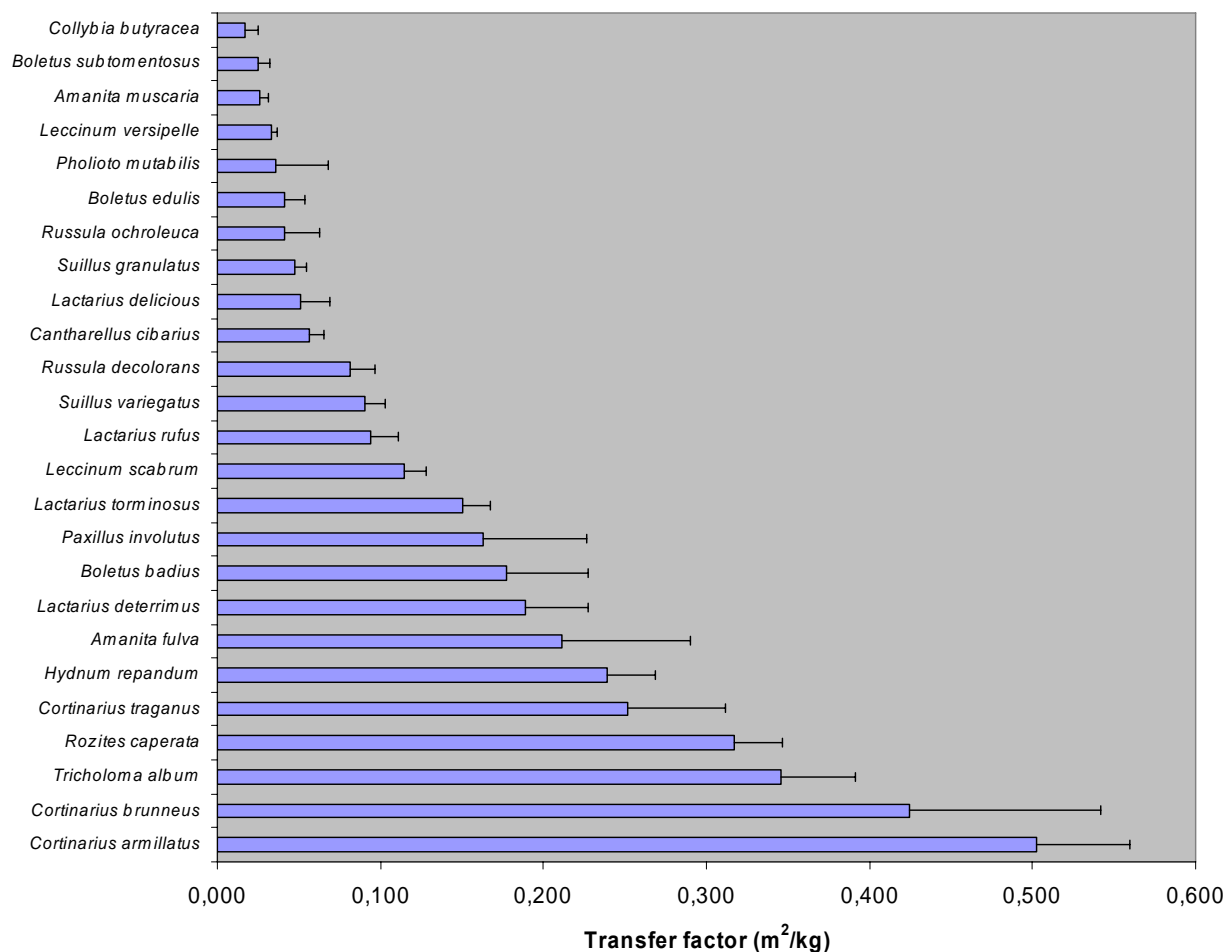


Figure 2. Mean transfer factor of ^{137}Cs ($\text{m}^2 \text{kg}^{-1}$ fresh wt.) with standard deviation in different fungi species (n=1283) in Norway in 1988-2002.

Since only wet weight of mushrooms are analysed in Norway, the dry/wet weight ratios of some mushroom species are given in table 2 (data collected in Finland and analysed by STUK).

Table 2. Dry/wet weight ratios of different mushroom species collected in Finland and analysed by STUK.

Species	No of samples	Variation of dry/wet w.	Mean	St.dev. of mean
<i>Boletus edulis</i>	5	0.048 - 0.0108	0.076	0.022
<i>Hygrophorus sp.</i>	1	0.071	0.071	
<i>Russula sp.</i>	6	0.067 - 0.135	0.092	0.025
<i>Cantharellus sp.</i>	6	0.072 - 0.09	0.082	0.0075
<i>Cortinarius sp.</i>	3	0.047 - 0.09	0.066	0.022
<i>Paxillus atrotomentosus</i>	1	0.055	0.055	
<i>Boletus sp.</i> (other than <i>B.edulis</i>)	7	0.053 - 0.101	0.075	0.019
<i>Lactarius sp.</i>	13	0.064 - 0.113	0.081	0.014
<i>Lycoperdon perlatum</i>	1	0.076	0.076	
<i>Hydnum sp.</i>	3	0.063 - 0.089	0.075	0.013
<i>Rozites caperata</i>	1	0.054	0.054	
<i>Clitocybe nebularis</i>	1	0.079	0.079	

Evaluation of a top predator from Norway as indicator organism

Runhild Gjelsvik and Helene Stensrud

Norwegian Radiation Protection Authority, P.O.Box 55, NO-1332 Østerås, Norway

Introduction

The European lynx (*Lynx lynx*), a middle sized felid, is found in most parts of the Scandinavian Peninsula (Johnsson 1983). By eradication of the wolf (*Canis lupus*) in the first half of the 20th century, the lynx is today the only remaining large carnivore occurring in widespread viable populations in the forested areas in Norway that effectively predate upon middle sized cervids. Population estimates are uncertain, but it is believed that there are 400-600 lynxes in Norway. Almost reaching the point of extinction in Norway during the 1930's, lynx populations have slowly increased in numbers and are believed to have spread to all parts of the country during the 1960's and 70's (Sunde 1996, Heggberget and Myrberget 1980). In the 1960s, lynx were so common that it was the only species among large predators not protected and hunting was allowed at all times of the year, without restrictions. Hence, no compensation for lynx kills was paid. In 1993 the number of sheep verified as lynx kills was equal to the sum of bear and wolverine kills. At present, compensation is given for sheep and reindeer killed by lynx. Now the lynx has become an interesting species and knowledge of its biology is required in order to manage the population and determine optimum hunting quotas.

After the Chernobyl accident in 1986, mid and northern part of Norway was contaminated by radioactive fallout. As an example the total fallout of ¹³⁷Cs in Norway was estimated at 2300 ± 200 TBq (Bache et al. 1987), and about 10 % of the territory received more than 20 kBq/m² of this nuclide. The region in Norway affected by the radioactive fallout was largely grazing land during the summer months for sheep, goat and cow, and all through the year for domestic reindeer. Reindeer are particularly subject to fallout since their winter diet consists of lichens (Skuterud 1999). Lichens have a high ability to absorb radionuclides directly from precipitation (Tuominen and Jaakkola 1973). A slow growing rate in the lichens, are resulting in a relatively long biological half-life of radionuclides. During the wintertime, the domestic reindeer is heavily preyed upon by lynx if present in the area. The domestic reindeer is reported to be an important pathway of radiocaesium to lynx (Mohn and Teige 1968, Åhman et al. 2002). Lynx from high contaminated areas after the Chernobyl accident are therefore expected to have a high intake of radiocaesium and thus high activity concentrations in their bodies.

The diet of lynx in Norway is reported by Sunde in 1996 to compose of a very limited number of species. Stomach analyses of 380 Norwegian lynxes showed that 67 % of the diet composed of cervides, 25 % of small games and 8% of other species i.e. fox *Vulpes vulpes*, rodents and waste/carrion. The "small game" group composed of mountain hare *Lepus timidus* and different birds like capercaillie *Tetrao urogallus*, black grouse *Tetrao tetrix*, grouse *Lagopus* sp. and hazel hen *Bonasa* sp. The "cervid" group mainly composed of roe deer *Capreolus capreolus* and semi-domestic reindeer *Rangifer tarandus*. These two species differ in their regional distribution with the roe deer occurring mainly in the southern half of

the country, and semi-domestic husbandry being widespread in the northern half (Norwegian animal, mammals 1990).

By investigation of stomach samples of Norwegian lynxes Sunde and Kvam (1997) found that sex was the only factor that clearly affected prey choice with males killing slightly more cervids than females. For females they found a tendency towards a body weight effect of female food choice, however may indicate that the smallest individuals of the smallest sex may have problems handling the largest prey types. Males, on the other hand were reported to show no sign of any additive effects of prey size and body weight. In addition, they concluded that the most likely explanation for the sexual prey size segregation is that male lynx kill proportionately more cervids due to differences in habitat choice and/or area utilisation. Males of the European lynx range areas of 1-3 times the size of female home ranges (Breitenmoser et al. 1993, Sunde 1996).

The main purpose of this study was to find out to what extent radiocaesium is transferred from fallout deposition after the Chernobyl accident to lynx. Muscle samples were taken from killed animals for monitoring of radiocaesium, and the ^{137}Cs activity concentration in lynx muscle has been compared to ^{137}Cs ground deposition data and reindeer herding areas. In addition, the data were used to evaluate geographical variations and long-term decline of ^{137}Cs in lynx.

Material and methods

Lynx sampling

From 1945 to 1980, state bounties were paid for lynxes killed in Norway. From 1960 to 1980 the condition for receiving a bounty was that the skinned carcass was sent to the Division for Game Research at the Directorate for Nature Management. From 1988 to 1993 the bounty was replaced by a payment for carcasses donated for scientific use. From 1994 to 1996, all lynx shot or found dead anywhere in the country had to be sent to the Norwegian Institute for Nature Research (NINA). The legal hunting season was all through the year until 1980. Since 1981 the hunting season is from 1. February through 31. March or until the regional quotas are filled. Thus the available material from the last years is mainly restricted to the late winter period. Majority of the lynx received had been shot during the ordinary hunting season. Rest of the remaining causes of mortality, was due to car and train accidents and trapping. During the years 1986-2001, 748 lynxes have been measured for ^{137}Cs activity concentrations from dry muscle samples (Figure 2 and 3). Samples were stored frozen until analyses. The activity concentrations of ^{137}Cs in the samples have been measured at Norwegian Institute for Nature Research (NINA). Geographical location has been recorded for all samples and in 707 cases the date of death has been recorded. In 41 cases no exact date of death were reported. Considering the legal hunting season, the dates of death were set to 15. March in the respectively killing year. For all lynxes the ^{137}Cs concentrations were decay corrected to date of death. The municipality, in which the lynx had been killed, was considering as the lynx habitat.

Relation to ground deposition

Levels of ground deposition of ^{137}Cs in Norway were taken from a nationwide sampling program carried out by Norwegian University of Science and Technology in 1995 and from an earlier nationwide survey of radiocaesium in topsoil carried out by the National Institute of Radiation Hygiene in 1986 following the Chernobyl accident. In 1995 soil humus samples at 0-3 cm depth were collected at 455 sites in 425 different municipalities (Figure 1). Total number of samples in each municipality varied from 1 to 8. In 1986, soil samples were taken to a depth of 4 cm in all 454 municipalities of Norway. In each municipality two samples were taken in each of four localities. One set of samples were mixed and subjected to gamma spectrometry. The sampling method and method of measuring ground deposition in 1986 and 1995 has been described by Bach et al. (1987) and Bjerk et al. (1999), respectively. Since the sampling procedure in 1995 was reasonably similar to that employed in 1986, the average deposition data from 1986 has been used for 29 municipalities lacking sampling sites in 1995. During the years 1986-2001, 96 lynxes were killed in these 29 municipalities. All samples collected in 1986 and 1995 were decay corrected to 1 July 1995, and average deposition of ^{137}Cs in each municipality and county calculated.

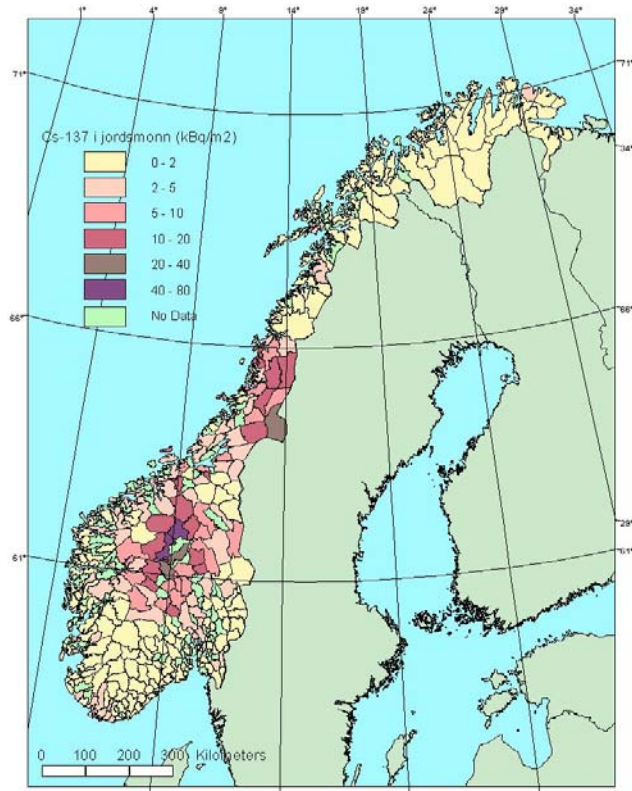


Figure 1. Ground deposition of ^{137}Cs in different municipalities in Norway, 1. July 1995.

The estimated ground deposition of ^{137}Cs (Bq m^{-2}) in each municipality and the corresponding activity concentrations of ^{137}Cs in lynx were used to calculate the ratio between activity concentration in lynx and ground deposition (aggregated transfer coefficient, T_{ag}) for each year, county and municipality. For grazing animals in natural ecosystems, the use of aggregated transfer coefficient (T_{ag} = activity concentration in animal tissue, Bq kg^{-1} , relative to soil contamination, Bq m^{-2}) has been suggested as a better measure to quantify transfer of radionuclides from fallout to animals (Howard et al. 1991) than a transfer coefficient based on daily intake (d kg^{-1}).

Seasonal variation and long-term decline

Because of the timing of the hunting season, 691 lynxes were from the months January-April, and 57 lynxes were from May-December. Comparisons between means were used to obtain any seasonal variation of radiocaesium in lynx. To test any seasonal variation of radiocaesium in lynx and T_{ag} -value, general linear models with univariate analyses were used. To calculate

the decline of radiocaesium in lynx over the years, the logarithmic values on activity concentration of ^{137}Cs in lynx muscle were used. Each mean value, $\log(\text{Bq kg}^{-1})$, from each county was used as a single observation in the statistical analyses. In addition, only data from the mid and late winter season (Jan.-April) were included. Data from counties with small sample numbers were excluded. The effect of year on ^{137}Cs activity concentration in lynx was tested in a linear regression for each county. The slope values were used to calculate effective ecological half-lives (T_{Eff}) for each county.

Results and discussion

Caesium-137 in lynx in relations to ground deposition

The deposition of ^{137}Cs (Bq m^{-2}) in the ground measured in 1995 varied from 221 Bq m^{-2} in a soil sample from Byggland, Aust-Agder to 73100 and 70089 Bq m^{-2} from Vågå in Oppland and Lierne in Nord-Trøndelag, respectively. The average ground deposition for all municipalities varied from 316 Bq m^{-2} to 27 447 Bq m^{-2} .

The activity concentration of ^{137}Cs in lynx muscle samples ranged from 90 to 124 614 Bq kg^{-1} dry weight. The lowest value was observed in a male cub (less than 1 year) killed 15 March in 1999 in Alta, Finnmark. Taking into account the 100 lowest obtained values, 45 were shot in Troms, 18 in Hedmark, 9 in Nordland, 9 in Akershus and 6 in Finnmark. Maximum caesium concentration was observed in a two year old male lynx killed in March 1987 in Alvdal, Hedmark. Taking into account 100 lynxes with highest level of radiocaesium, 55 were killed in Nord-Trøndelag and 34 in Nordland.

Both the caesium concentration in lynx and the aggregated transfer coefficient varied between years (^{137}Cs activity in lynx, logarithmic values, One-Way ANOVA, $df=15$, $F=6.385$, $p<.000$, Tag: logarithmic values, One-Way ANOVA, $df=15$, $F=11.018$, $p<.000$, Figure 2 and 3). The average activity level in lynx increased the first three years after the Chernobyl accident, and reached a maximum in 1989. The high variation in ^{137}Cs concentration from 1986 to 1993 is probably due to a lower sampling number in these years. After 1989 the average concentration activity were reduced in the two following years before reaching a new average maximum in 1993. Probably due to physical decay of ^{137}Cs , a decreasing tendency was observed for the whole period.

Like the caesium concentration in lynx, the aggregated transfer coefficient increased from 1986 to 1989. Then a reduction in average Tag-values occurred, reaching an average minimum in 1993. The T_{ag} -values for the years 1986-1993 follows the curve for ^{137}Cs activity in lynx. For the years 1993-2001, a slight increasing tendency in T_{ag} -values was observed in spite of a decreasing tendency of ^{137}Cs in lynx. Since the ground deposition is decay corrected to 1995, the increasing tendency of T_{ag} -values in the late period may be due to differences in number of lynx killed in different municipalities and counties (Appendix I). Another problem that should be considered is the non homogeny contamination of radionuclides after the Chernobyl accident, and thus the estimated ground deposition may vary a lot within an area. In addition, the lynx hunting area or main habitat, may be in another municipality or region and not be identical with the place of dead.

T_{ag} values for lynx appear to dependent on whether there are reindeer present within the county or not (Table 1). Differences in T_{ag} values between areas with and without reindeer may be explained by considerably higher ^{137}Cs activity concentration in reindeer than in other

prey, e.g. roe deer (Johnson and Bergström 1994). Roe deer is probably the main prey in most of the counties outside reindeer herding areas.

Before statistical analyses were performed, the variations between years were removed since the “between years” variations have a large impact of the regression model. Performing linear regressing analyses, the standardized residuals² for the dependent variable “year of death” was calculated. The ¹³⁷Cs activity concentration in lynx varied between municipalities (One-Way ANOVA, df=152, F=2.613, p<.000, Figure 4) and between counties (One-Way ANOVA, df=14, F=7.694, p<.000). The same pattern was found for aggregated transfer coefficient (municipality: One-Way ANOVA, df=152, F=2.379, p<.000, Figure 5. County: One-Way ANOVA, df=152, F=5.122, p<.000).

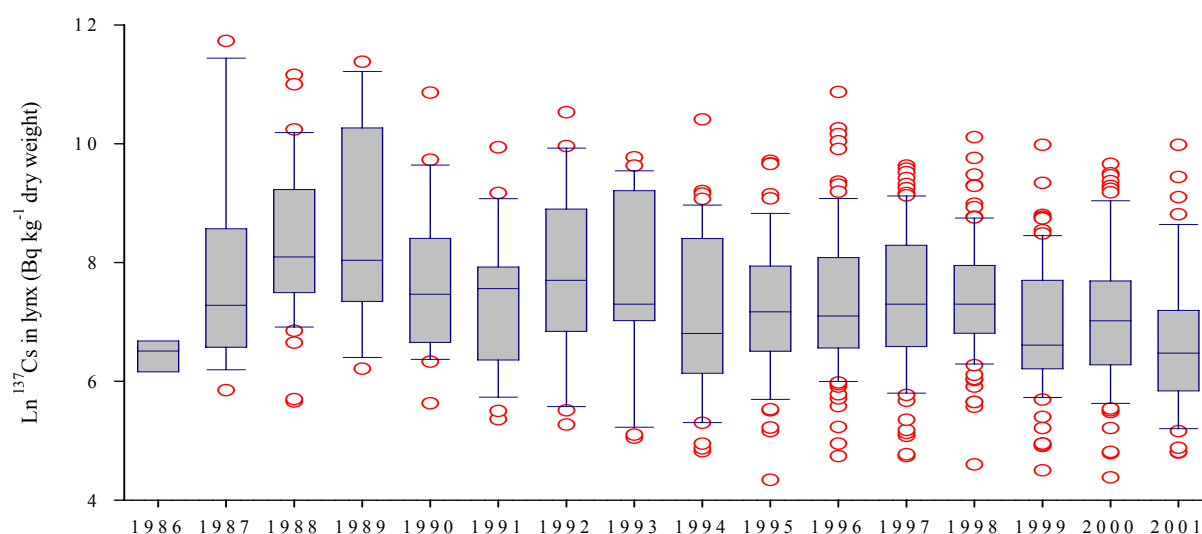


Figure 2. ¹³⁷Cs (Bq kg⁻¹ dry weight, ln-values) in Lynx (n=748) in Norway during 1986-2001. Box plots with horizontal lines for the median and 90th percentiles are shown as vertical boxes with error bars. Open red circles are outliers.

² **Residuals:** the actual value of the dependent variable minus the value predicted by the regression equation.

Standardized Residuals: the residual divided by an estimate of its standard error. Standardized residuals, which are also known as Pearson residuals, have a mean of 0 and a standard deviation of 1.

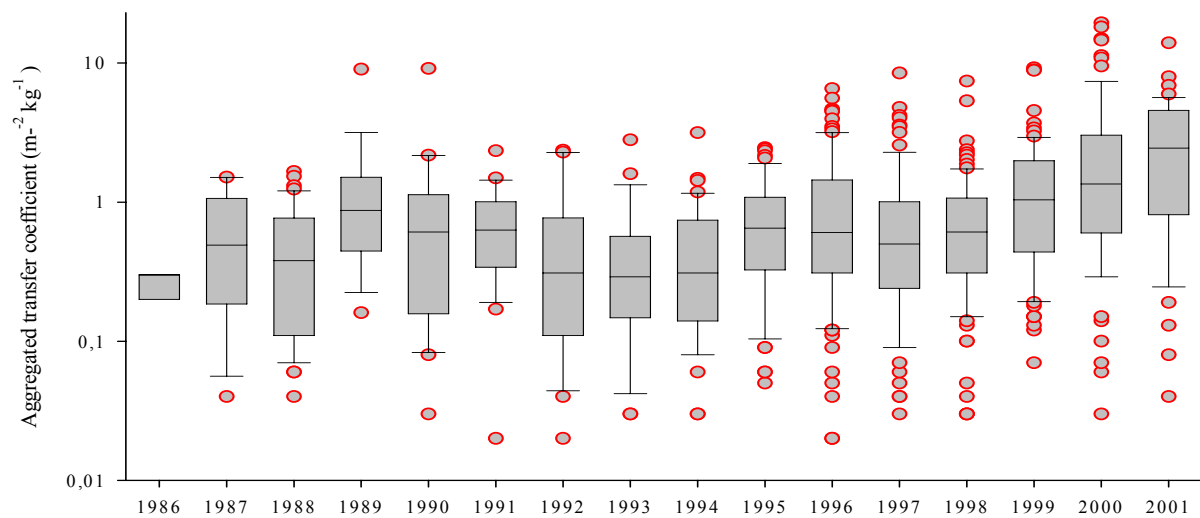


Figure 3. Aggregated transfer coefficient ($\text{m}^2 \text{kg}^{-1}$) in Lynx ($n=748$) in Norway during 1986-2001. Box plots with horizontal lines for the median and 90th percentiles are shown as vertical boxes with error bars. Open red circles are outlayers.

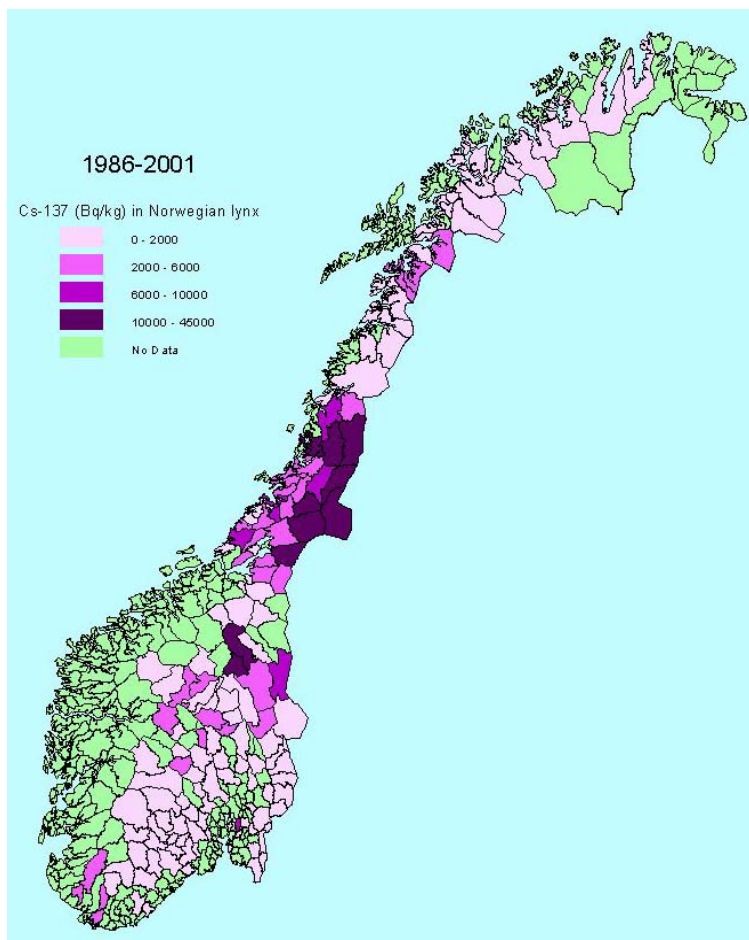


Figure 4. Average ^{137}Cs activity concentration in lynx from different municipalities in Norway from 1986 to 2001.

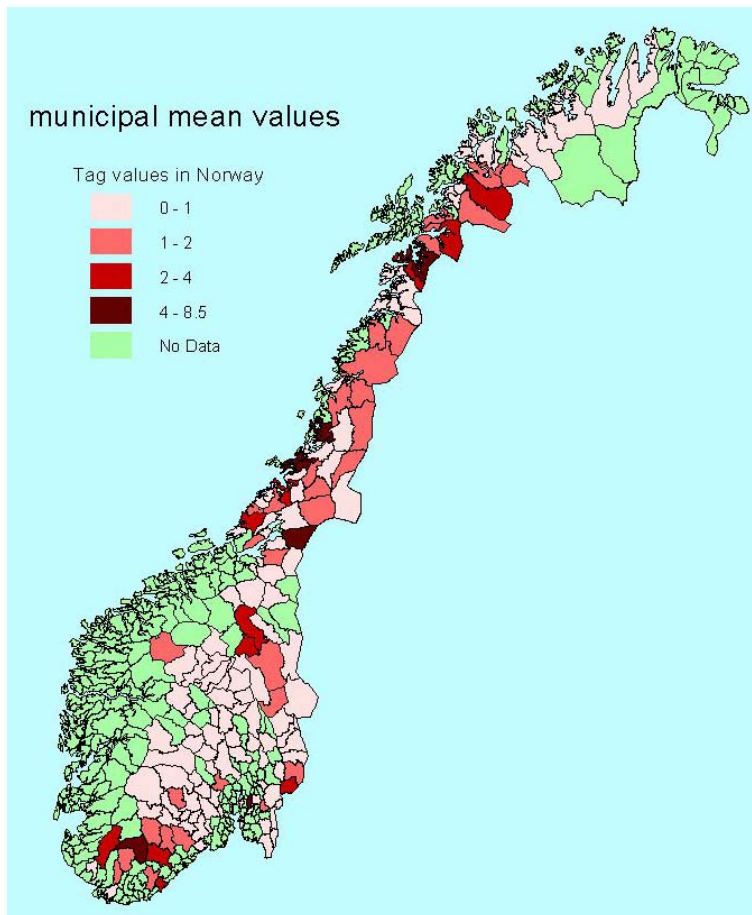


Figure 5. Average aggregated transfer coefficient ($\text{m}^2 \text{kg}^{-1}$) in different municipalities in Norway from 1986-2001.

Table 1. Average ground deposition of ^{137}Cs and activity concentration of ^{137}Cs during January-April in 1986 to 2001 in muscle samples from lynx (dry weight) from different counties in Norway.

County	Average ground deposition (Bq m ²)	Min. ground deposition (Bq m ²)	Max. ground deposition (Bq m ²)	No of lynx	Reindeer present	Average in lynx (Bqkg ⁻¹)	Min. in lynx (Bqkg ⁻¹)	Max. in lynx (Bq kg ⁻¹)	SD in lynx (Bq g ⁻¹)	Average T _{ag} in lynx (m ² kg ⁻¹)	Min. T _{ag} in lynx (m ² kg ⁻¹)	Max. T _{ag} in lynx (m ² kg ⁻¹)	SD, T _{ag} in lynx (m ² kg ⁻¹)
Aust-Agder	316	221	763	14	No reindeer	1290	503	2396	574	4,81	1,83	10,84	2,73
Nordland	4641	598	17370	148	Reindeer	7216	156	87542	13554	2,08	0,14	18,16	2,79
Vest-Agder	1735	1145	3531	5	No reindeer	2299	1049	3356	978	1,49	0,71	2,32	0,72
Troms	761	545	1887	76	Reindeer	896	100	3953	823	1,35	0,07	4,98	1,23
Nord-Trøndelag	9364	598	26463	137	Reindeer	6979	396	86263	9456	1,27	0,04	9,11	1,60
Hedmark	3450	316	9791	72	Few reindeer	3664	186	124614	15049	0,98	0,06	13,96	1,86
Akershus	1201	879	2759	21	No reindeer	1206	142	15278	3234	0,88	0,09	9,21	1,92
Telemark	1033	638	2481	62	No reindeer	853	234	2555	435	0,88	0,21	2,90	0,50
Sør-Trøndelag	2891	1518	11693	26	Few reindeer	2328	526	16990	3739	0,88	0,05	5,49	1,20
Østfold	1932	625	3652	10	No reindeer	887	184	1842	520	0,52	0,08	0,85	0,28
Rogaland	6027	6027	6027	3	Few reindeer	3138	2183	4424	1156	0,52	0,36	0,73	0,19
Finnmark	1059	823	1769	8	Reindeer	441	90	1692	549	0,37	0,11	0,96	0,35
Buskerud	6461	883	19082	38	No reindeer	1504	413	9628	1561	0,33	0,04	1,68	0,31
Oppland	27447	721	73100	54	No reindeer	1628	200	9396	1526	0,14	0,00	1,60	0,23

Sex dimorphism, seasonal variation and long-term decline

Both male and female lynx can be considered as fully grown from the age of 2 years and female lynx normally give birth for the first time at the age of 2 years. (Kvam 1991). From that age, no significant increase in body weight with age occurs (Sunde and Kvam 1997). Average body weights were 19.8 kg for male and 15.1 for female. Adult males are approximately 24% heavier than adult females, but the weight ranges of the two sexes overlap considerably (Figure 6).

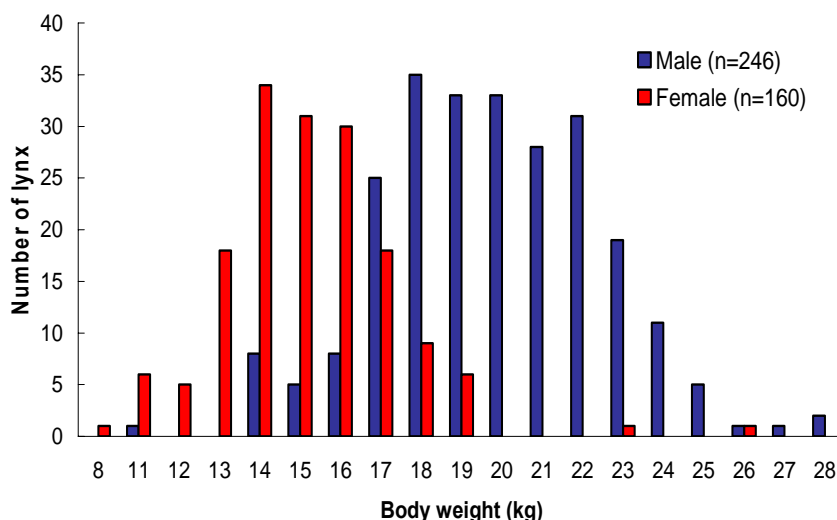


Figure 6. Distribution of sex by body weight of independent lynxes ≥1 year old. Numbers of male and female lynxes are given in brackets.

An ongoing project of the ecology of the large predators in Norway, has found by investigation of lynx carcasses and excrements that lynx takes more deer in winter than at other times of the year. Differences in ^{137}Cs activity concentrations in lynx depending on season of year is therefore expected, since reindeer have been reported to have much lower ^{137}Cs activity concentrations during summer due to seasonal variation in their diet (Åhman and Åhman 1994). No such seasonality was observed in this study (Table 1 and 2). It should be considered that lack of seasonality in this study might also be a result of small sampling number during the summer period ($n=57$), differences between counties, habitat or seasonal division. In this study the winter season has been defined as 1.January to 30.April and summer season as 1.May to 31.December. Another study of the Norwegian lynx (Sunde and Kvam 1997) have adopted the seasonal division employed by Birkeland and Myrberget (1980), with winter from November through April, and summer from May through October, determined by the average periods of snow cover in Norway.

When males roam much larger areas and have far longer daily roaming distances than females, sexual prey size segregation may indicate differences in habitat use and hunting pattern. Locating small game and ungulates may require different behaviour tactics. While walking with juveniles, females may spent more time locally searching fore hares, for instance, whereas males may encounter prey along their routes or search out potential prey habitats in the vicinity. A large prey is likely to be easier to detect at a distance than a small one (Sunde and Kvam 1997). However, no differences between the sexes in ^{137}Cs activity concentration or T_{ag} -values were found (Table 2 and Table 3).

Bagshaw and Brisbin (1985) defined ecological half-life as the half-life of a contaminant in a free-living organism, resulting from the decline in levels or availability of the contaminant in

the habitat of the organism. In 1993, Howard *et al.* described effective half-life as “the time required for the activity concentration of a radionuclide in a food product to be reduced to one half of the origin concentration in a specific system.” When applied to ^{137}Cs in lynx, this would be the decline or decrease in availability of the nuclide in soil or by forage plants utilized for prey species of the lynx inhabiting a specific system. In this work, the term “effective ecological half-life” is used (T_{Eff}) and observed half-lives involve ecological factors as well as physical decay.

Table 2. General linear model (GLM), Univariate analyse with ^{137}Cs activity concentration in lynx (log transformed values) as dependent variable against county, season and sex.

	SS	Df	MS	F	P
Total	722	729			
County	77	14	5,47	8,96	,000
Season	,001	1	,001	,002	,968
Sex	,970	1	,970	1,59	,208
Error	416	681	,611		

Table 3. General linear model (GLM), Univariate analyse with aggregated transfer coefficient (log transformed) as dependent variable against county, season and sex.

	SS	Df	MS	F	P
Total	736	729			
County	93	14	6,68	9,86	,000
Season	,002	1	,002	,003	,957
Sex	,082	1	,082	,121	,728
Error	461	681	,677		

A difference in aggregated transfer factor has been found for different counties, and therefore different ecological half-lives may be found for different counties. Much of the observed variation between counties could be explained by large variations in ground deposition (Figure 7). In the present study T_{Eff} values for lynx calculated for different counties varied from 2.3-4.9 years (for counties with R-square better than 0.5, Table 4). No other studies has reported T_{Eff} for lynx, but looking at one of the mayor prey species, the T_{Eff} values obtained this study, are in good agreement with those obtained for domestic reindeer in Sweden (Åhman and Åhman, 1994) and in Norway after the Chernobyl accident. Hove et al. (1992) calculated T_{Eff} for ^{137}Cs in domestic reindeer to be 3.5 y for the period 1987-1991. Eikermann (1992) found T_{Eff} values between 2.5 and 3 y for radiocaesium ($^{134}\text{Cs} + ^{137}\text{Cs}$) in domestic reindeer in Norway during the period 1986-1990.

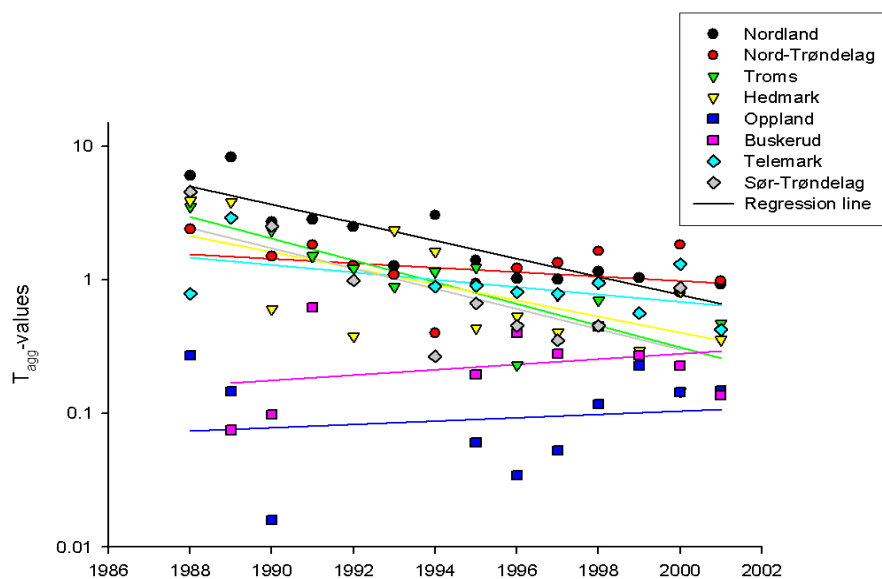


Figure 7. The relation between ground deposition of ^{137}Cs (Bq m^{-2}) and the activity concentration of ^{137}Cs in lynx muscle (Bq kg^{-1}) in 1988-2001. Calculated aggregated transfer coefficient (T_{agg} , $\text{m}^2 \text{kg}^{-1}$) for different counties in Norway.

Table 4. Effective ecological half-lives, T_{Eff} for ^{137}Cs in lynx (mean \pm SE, years) for different periods and counties. Estimates made on the basis of the average values each year for different counties. R-square for the regressions, number of years observed and n=total number of animals.

County	Period	T_{Eff} (y)	T_{Eff} Min. $T_{\text{Eff}} - \text{SE}$	T_{Eff} Max $T_{\text{Eff}} + \text{SE}$	R^2	Number of years	n
Troms	1987-2001	4.9	4.0	5.8	0.64	13	76
Nordland	1988-2001	4.8	3.6	6.0	0.53	14	145
Nord-Trøndelag	1987-2001	7.1	6.9	7.3	0.48	14	136
Sør-Trøndelag	1988-2000	3.1	2.4	3.8	0.58	9	26
Hedmark	1988-2001	2.3	1.9	2.7	0.69	14	72
Telemark	1988-2001	9.4	6.7	12.1	0.46	10	62
Oppland	1988-2001	18.7	10.3	27.1	0.16	10	54
Buskerud	1989-2001	17.0	7.9	26.1	0.01	10	38

Conclusions

^{137}Cs concentrations in muscle tissue of 748 Norwegian lynx were collected from different parts of Norway during 1986-2001. The activity concentration of ^{137}Cs in lynx muscle samples ranged from 90 to 124 614 Bq kg^{-1} dry weights. The highest average activity concentrations were found in lynxes from Nordland and Nord-Trøndelag. These counties had also quite high ^{137}Cs ground deposition with respectively 4641 and 9364 Bq m^{-2}

The activity concentrations of ^{137}Cs in lynx were fairly well correlated to ground deposition in different counties. The relatively high values seem also to be connected with consumption of caesium-137 contaminated reindeer, which periodically represents an essential part of the lynx diet. 67% of the lynx diet in Norway is mainly composed of roe deer and semi-domestic reindeer. These two species differ in their regional distribution with the roe deer occurring mainly in the southern half of the country, and semi-domestic husbandry being widespread in the northern part of Norway.

The aggregated transfer factor of ^{137}Cs in lynx varied from 0.001 to 18.16 ($\text{m}^{-2} \text{kg}^{-1}$) and differed for different counties. Similar to ^{137}Cs activity concentration, T_{ag} values for lynx appear to dependent on whether there are reindeer present within the county or not. Differences in T_{ag} values between areas with and without reindeer may be explained by considerably higher ^{137}Cs activity concentration in reindeer than in other prey, e.g. roe deer and mountain hare. Roe deer is probably the main prey in most of the counties outside reindeer herding areas.

The ^{137}Cs decline was generally more rapid in high-contaminated counties, corresponding to an effective ecological half-life (T_{eff}) of 3-5 years. The results of the present investigation show that also predators on reindeer in contaminated areas may reach high levels of radiocaesium in their bodies but are not considered as a good indicator organism immediately after an emergency situation since it takes time to reach the top of the food chain.

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Transfer of radionuclides to plants

Sven P. Nielsen

Risø National Laboratory, DK-4000 Roskilde, Denmark

Sampling of Water Horsetail and Bracken Fern including upper soil layer (0-10 cm) and water was carried out in Torshult, Sweden, in Almindingen on Bornholm, in Asserbo and Arresø on Zealand and in Sdr. Hostrup and Nydam mose in Jutland. Analyses of these samples were carried out for gamma emitters. Furthermore, sampling was carried out in 2004 for seawater, seaweed and shrimps at locations in Danish waters at Bornholm (Svenskehavn), at Zealand (Klint), at Lolland/Falster (Guldborgsund) and on the west coast of Jutland (Hirtshals, Agger, Hvide Sande, Rømø). The sampling locations in Denmark are shown in Fig. 1.

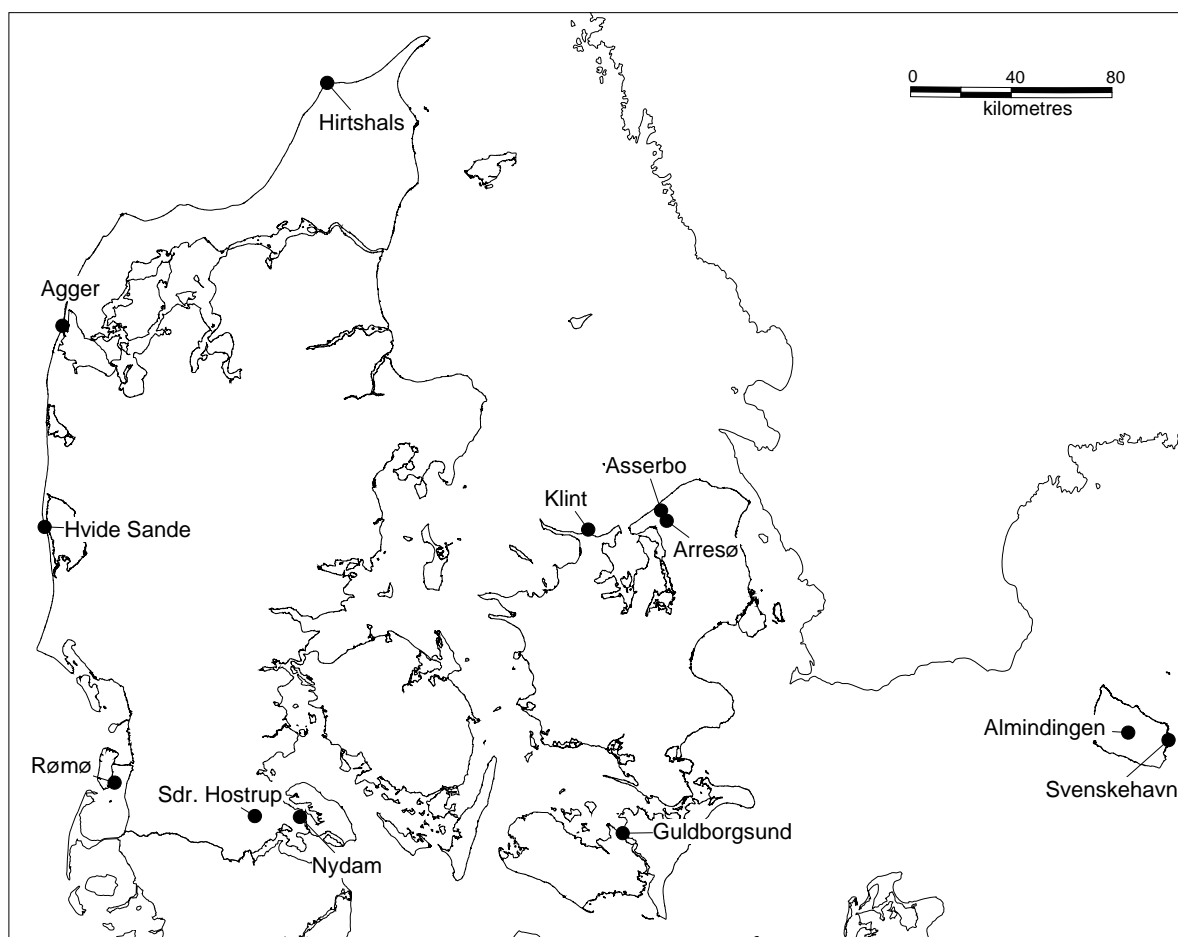


Figure 1. Sampling locations.

Results for terrestrial plants

Concentrations of gamma-emitting radionuclides and uranium were determined in Bracken Fern, Water Horsetail and soil samples. The results are summarised in terms of concentration ratios (Bq kg^{-1} dw plant per Bq kg^{-1} dw soil) in the graphs below.

The concentration ratios (CR) are highest for ^{40}K in both plant species and show the lowest variability across locations. The CR's for ^{40}K range from 1 to 2, while the CR's for the other radionuclides range one to three orders of magnitude lower. The CR's for ^{137}Cs show particularly high variability across locations. The CR's were analysed in a two-way ANOVA on the log-transformed values to test differences between plant species and radionuclides. The difference between radionuclides was highly significant, $p < 0.01$, whereas the difference between plant species was not significant. The concentration ratios are listed by radionuclide in Table 1 showing geometric mean values and geometric standard deviations.

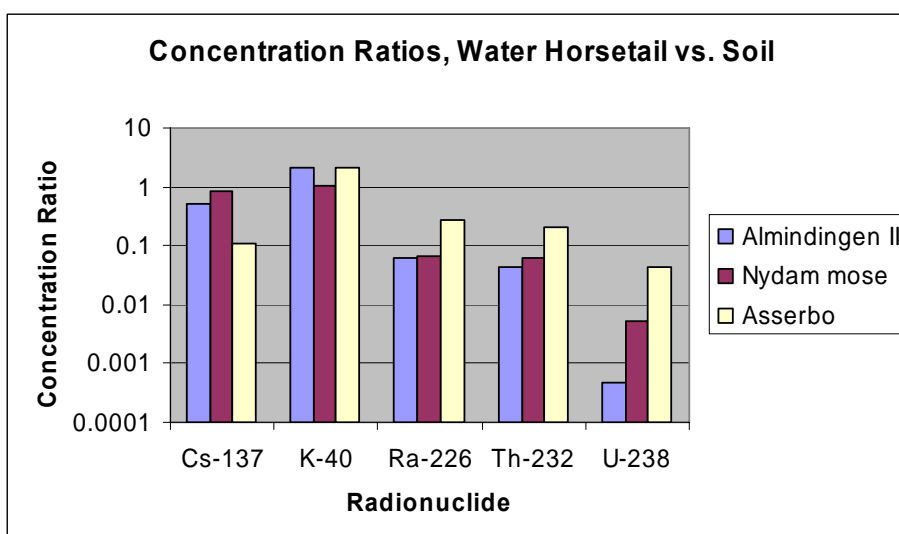
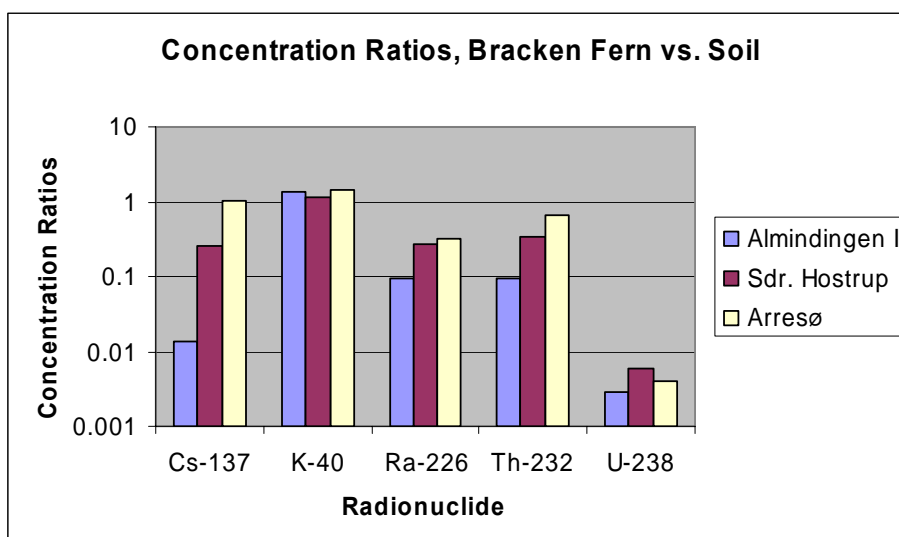


Table 1. Geometric means and geometric standard deviations of concentration ratios (CR) for Bracken Fern and Water Horsetail for transfer of radionuclides from soil (Bq kg⁻¹ dw plant per Bq kg⁻¹ dw soil).

	¹³⁷ Cs	⁴⁰ K	²²⁶ Ra	²³² Th	²³⁸ U
CR, geometric mean	0.24	1.5	0.14	0.15	0.004
CR, geometric standard deviation	5.1	1.3	2.2	2.8	4.2

The observed concentration ratios for ¹³⁷Cs are compared to literature values of the transfer from soil to crops (IAEA, 1994) as shown in Fig. 2. The graph shows expected values with 95% confidence ranges. Values without range are observed values from this study. Corresponding comparisons for ²²⁶Ra, ²³²Th and ²³⁸U are shown in Figs. 3-5.

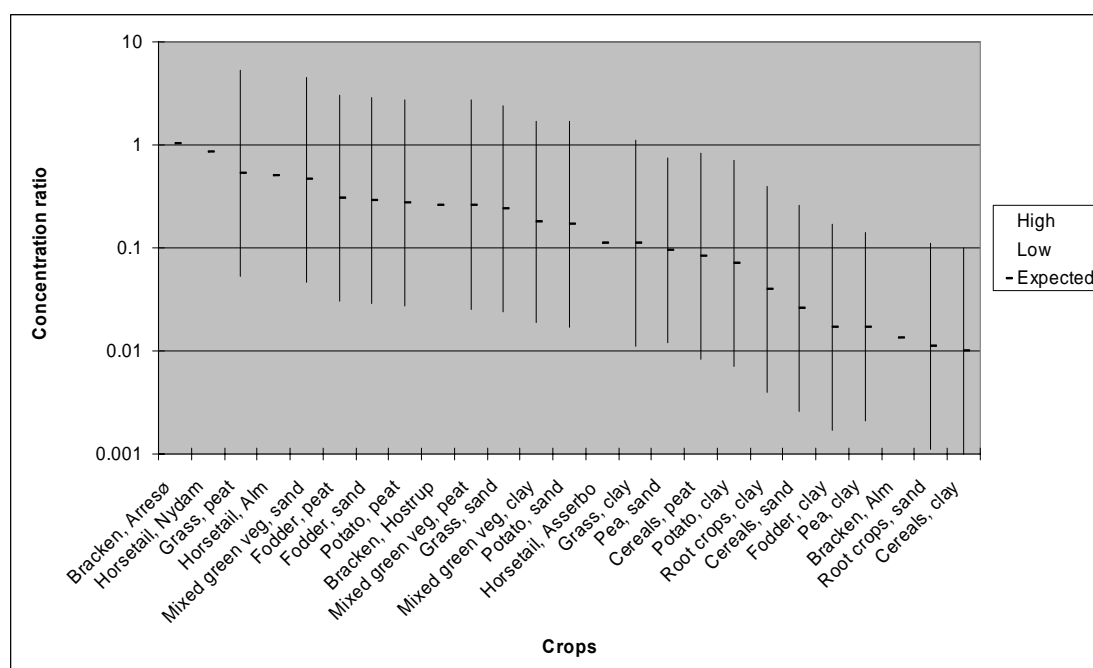


Figure 2. Comparison of concentration ratios for ¹³⁷Cs from this study with literature values (IAEA, 1994). Literature values include 95% confidence ranges while observed values are without range.

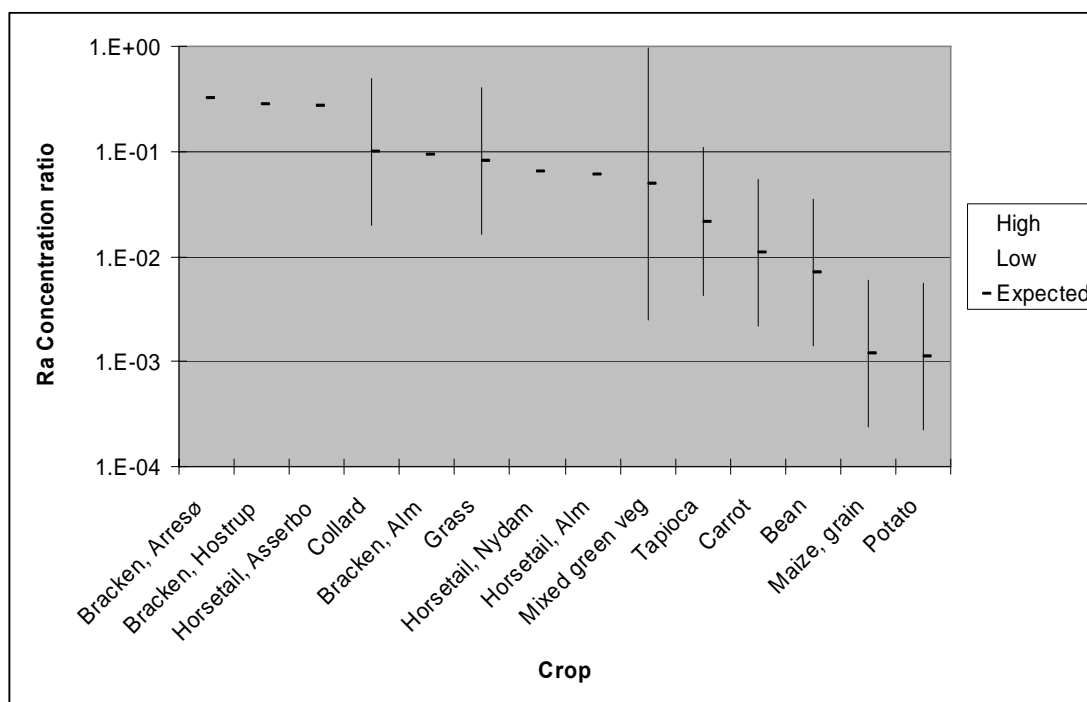


Figure 3. Comparison of concentration ratios for ^{226}Ra from this study with literature values (IAEA, 1994). Literature values include 95% confidence ranges while observed values are without range.

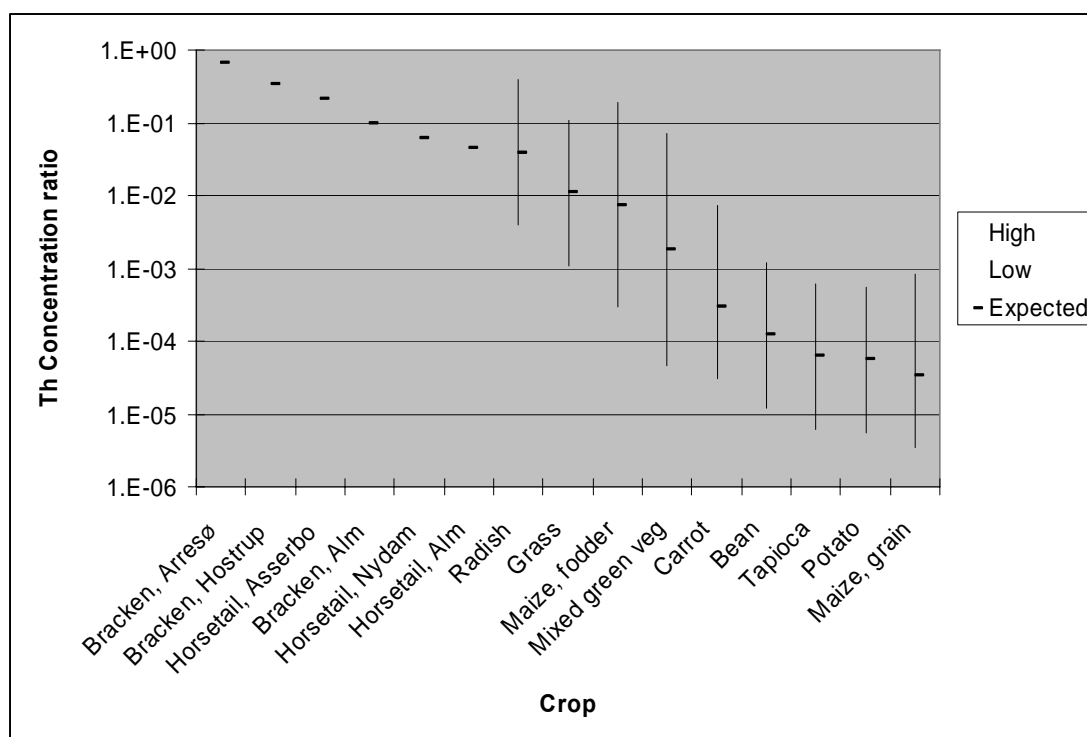


Figure 4. Comparison of concentration ratios for ^{232}Th from this study with literature values (IAEA, 1994). Literature values include 95% confidence ranges while observed values are without range.

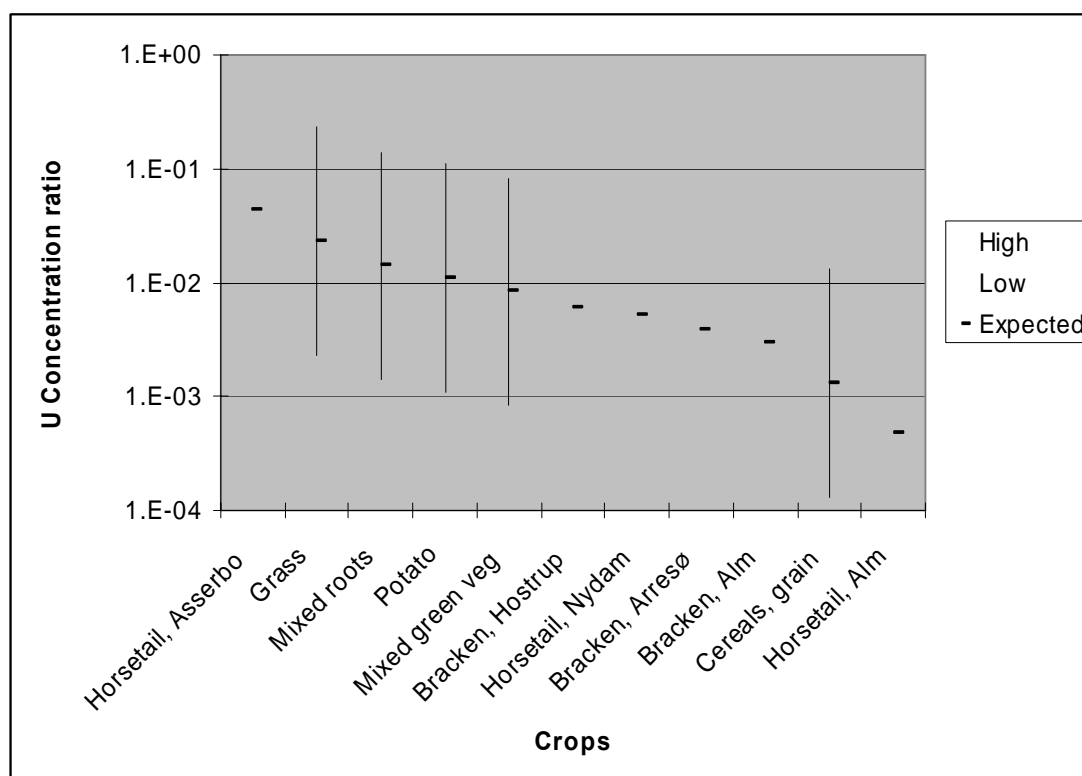


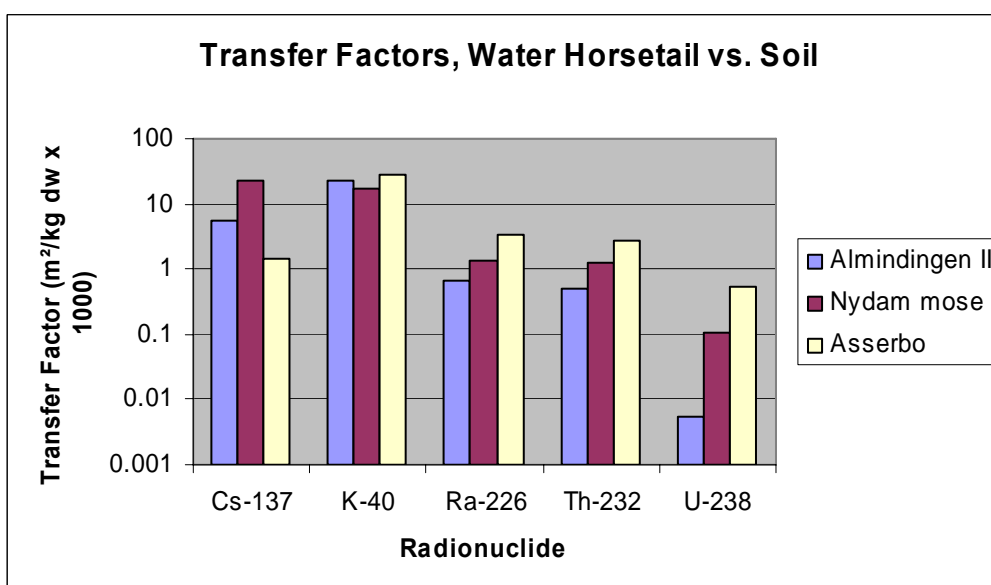
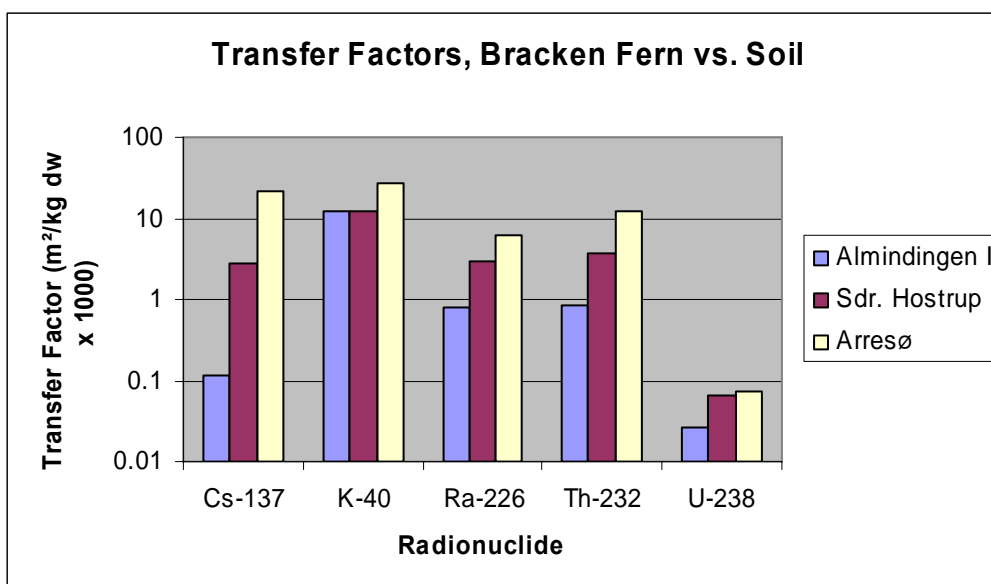
Figure 5. Comparison of concentration ratios for ^{238}U from this study with literature values (IAEA, 1994). Literature values include 95% confidence ranges while observed values are without range.

These comparisons show that the observed transfer of ^{137}Cs , ^{226}Ra , ^{232}Th and ^{238}U from soil to Bracken fern and Water horsetail from this study generally falls within reported ranges of transfer to crops. However, the observed values of the transfer of ^{232}Th rank high compared to literature values.

Furthermore, the transfer of radiocaesium from soil to plant was evaluated in terms of the concentration in plant relative to the total concentration by area (Bq kg^{-1} per Bq m^{-2} , or $\text{m}^2 \text{kg}^{-1}$). The transfer factors (TF) are highest for ^{40}K in both plant species and show the lowest variability across locations. The TF's for ^{40}K range from 12 to 22, while the TF's for the other radionuclides range one to two orders of magnitude lower. The TF's for ^{137}Cs show high variability across locations. The TF's were analysed in a two-way ANOVA on the log-transformed values to test differences between plant species and radionuclides. The difference between radionuclides was highly significant, $p < 0.001$, whereas the difference between plant species was not significant. The transfer factors are listed by radionuclide in Table 2 showing geometric mean values and geometric standard deviations.

Table 2. Geometric means and geometric standard deviations of transfer factors (TF) for Bracken Fern and Water Horsetail for transfer of radionuclides from soil (Bq kg^{-1} dw plant per Bq m^{-2} soil).

	^{137}Cs	^{40}K	^{226}Ra	^{232}Th	^{238}U
TF, geometric mean	3.3	19	1.9	2.0	0.06
TF, geometric standard deviation	7.1	1.4	2.4	3.2	4.6



Results for marine plants

Analyses were made of ^{137}Cs and ^{99}Tc in marine samples collected at coastal locations. The results are shown in Table 3 giving sample type, sampling location, date of sampling, concentrations of ^{137}Cs and ^{99}Tc (Bq/kg dw for biota and Bq/m³ for water) and salinity at sampling location. Seaweed samples were all *Fucus vesiculosus*.

Table 3. Caesium-137 and ⁹⁹Tc in samples collected at coastal sites.

Sample type	Location	Date	Cs-137 (Bq/kg)	Tc-99 (Bq/kg)	Salinity (PSU)
Seaweed	Svenskehavn	apr-04	22	12	8
Seaweed	Guldborgsund	aug-04	12		
Seaweed	Klint	mar-04	4.6	160	21
Seaweed	Klint	jun-04	7.3	110	19
Seaweed	Klint	sep-04	7.5	75	17
Seaweed	Klint	dec-04	4.6		22
Seaweed	Rømø	mar-04	0.47	33	26
Seaweed	Rømø	jun-04	0.45		31
Seaweed	Rømø	sep-04	0.92		28
Seaweed	Rømø	dec-04	0.46		25
Seaweed	Agger	mar-04	0.29		31
Seaweed	Agger	jun-04	0.78		33
Seaweed	Agger	sep-04	0.46		33
Seaweed	Agger	dec-04	0.32		33
Seaweed	Hvide sande	mar-04	0.54		10
Seaweed	Hvide sande	sep-04	1.0		24
Shrimp	Guldborgsund	aug-04	11		
Shrimp	North Sea	sep-04	0.58		
Lobster	Kattegat	maj-04	1.2		
Lobster	Kattegat	sep-04	1.4		
Seawater	Svenskehavn	apr-04	55	0.11	8
Seawater	Guldborgsund	jun-04	46		11
Seawater	Klint	jun-04	28	0.6	21
Seawater	North Sea	jun-04	3.5	0.7	32

Concentration ratios calculated from the data in Table 3 are shown in Table 4. The concentration ratios for ⁹⁹Tc agree with those reported elsewhere in the Indofern Project. The concentration ratios for ¹³⁷Cs in Fucus show a correlation to salinity with higher values in low salinity water at Bornholm than in high salinity water on the west coast of Jutland.

Table 4. Concentration ratios (Bq/kg dw per Bq/L) of ¹³⁷Cs and ⁹⁹Tc in seaweed, shrimp and lobster relative to seawater.

Sample	Location	Cs-137	Tc-99
Seaweed	Svenskehavn	4.0E+02	1.1E+05
Seaweed	Guldborgsund	2.6E+02	
Seaweed	Klint	2.1E+02	1.9E+05
Seaweed	Rømø	1.6E+02	4.7E+04
Seaweed	Agger	1.3E+02	
Seaweed	Hvide sande	2.2E+02	
Shrimp	Guldborgsund	2.3E+02	
Shrimp	North Sea	1.7E+02	
Lobster	Kattegat	4.6E+01	

Indicator Organisms for Marine and Terrestrial Environmental Radioactivity

A. Raaum, G.C. Christensen, A.M.B. Ruud and E. Strålberg

Institute for Energy Technology, IFE, P.O.Box 40, NO-2027 Kjeller, Norway

Abstract

The aim of this work was to study different terrestrial and marine indicator organisms for environmental radioactivity. An area in central Norway, consisting of forests, mountain forests and high mountains without forests was subject to the study for terrestrial indicator organisms. This area received fairly high contamination of Chernobyl fallout. In 1990 samples of several species of biota were collected from the same area as a part of a NKS project. For marine indicator organisms, samples of the two species of brown algae *Fucus vesiculosus* and *Ascophyllum nodosum* were collected from Tromøya in southern Norway. Seasonal variations in activity concentrations and indicator properties of *Fucus vesiculosus* and *Ascophyllum nodosum* were compared for several radionuclides, including some radionuclides that have not been focused on previously.

Terrestrial environment

Introduction

The area subject to this study is the stretch from Ringebu to Koppang, which includes forests, mountain forests and high mountains without forests. This area received fairly high contamination of Chernobyl fallout, with area activity densities ranging from 20 to 80 kBq ^{137}Cs per m^2 in June 1986 [1]. In 1990 several species of biota were sampled from these same locations as a part of the NKS RAD 4 project [2].

Sampling and analyses

Soil profiles and different plants were collected from three locations between Ringebu and Koppang. Location 1 and 2 consist of mountain forests, and location 3 is in a high mountain without forest. Each location covered an area of about 100 x 100 meters. Sampling was performed four times during the period early summer 2002 to fall 2003. Except for the *Athyrium distentifolium*, which was only found within a very small area, the vegetation samples were pooled samples of sub samples from the entire area. In 2003, 3 or more sub samples from separate areas within each location were collected in order to look at the variations within each location. Soil profiles (10-17 cm diameter) were collected from each location. The profiles were collected reasonably scattered within each area. The soil profiles from 2002 were divided into layers at the sampling site. In 2003 the whole profiles were measured without dividing into layers.

All samples were dried to constant weight at 105°C, and soil samples were sieved through a 2 mm sieve. All samples were measured by gamma spectrometry using HPGe detectors, and selected samples were analysed for ^{90}Sr and $^{239,240}\text{Pu}$. ^{90}Sr was analysed by means of chemical separation and measurement of the beta activity of the daughter product ^{90}Y . The method is based on a modified HASL 300 procedure [3]. Following chemical separation of strontium, the samples were left for ingrowth of ^{90}Y . Yttrium was then precipitated as oxalate, collected on a filter, and measured by low level anticoincidence beta counters, model Risø GM-25-5. Chemical yield was determined by ^{85}Sr tracer and titration of Y with EDTA [4]. Analyses of $^{239,240}\text{Pu}$ were performed by chemical separation and electro deposition of Pu on steel plates. The activity of $^{239,240}\text{Pu}$ was measured by alpha spectrometry using PIPS detectors, and the yield was determined by ^{242}Pu tracer. The method is based on Chen [5]. The laboratory has an internal quality control system, and participates in intercomparison exercises for these types of analyses at a regular basis.

Results and discussions

The area activity density for ^{137}Cs was higher in location 3 than at the other two locations (Table 1). This is consistent with what was found in 1990 [2]. For ^{90}Sr the area activity density was about 4 times higher at loc. 2 than at the other two locations. The area activity densities of ^{137}Cs in 2002 and 2003 were about 30-40 % of the concentrations reported in 1990 [2]. The results from the measurements of the individual layers of the samples from 2002 showed that the highest mass activity densities of ^{137}Cs was found in the top 2 cm layer, and generally more than 80 % of the total ^{137}Cs activity was found in the uppermost 4 cm.

Table 1. Mean area activity densities in soil (reported uncertainties represent two standard deviations of the mean, number of samples in brackets).

Nuclide	Area activity densities in Bq/m ²		
	Loc. 1	Loc. 2	Loc. 3
^{137}Cs	2680 ± 230 (10)	5100 ± 1500 (11)	7780 ± 690 (10)
^{90}Sr	124 ± 4 (2)	580 ± 210 (3)	150 ± 120 (2)
$^{239,240}\text{Pu}$	33 ± 10 (2)	31 ± 8 (2)	23.7 ± 1.0 (2)

^{137}Cs concentrations were highest in *Rozites caperata* (5100 Bq/kg dry weight). The levels were also high in *Cladonia stellaris*, varying from 200-1800 Bq/kg. The ^{137}Cs concentrations were similar in plant and roots of *Athyrium distentifolium*, *Betula nana* and twigs and berries of *Empetrum hermaphroditum*, ranging from 20-230 Bq/kg. For *Cladonia stellaris* the highest concentration was in the samples from loc. 3, which is consistent with the measured levels in soil. For the other species there were no clear differences in the ^{137}Cs -concentrations between the different locations.

The ^{90}Sr activity concentrations were higher in *Betula nana* and twigs of *Empetrum hermaphroditum* (35-150 Bq/kg), than in *Cladonia stellaris* and berries of *Empetrum hermaphroditum* (5-20 Bq/kg). There was no correlation between plant activity and soil activity for ^{90}Sr . For *Empetrum hermaphroditum* the highest ^{90}Sr activity concentrations were measured in the samples from loc. 3 whereas for soil the highest activities were in the samples from loc. 2.

Cladonia stellaris had the highest concentrations of $^{239,240}\text{Pu}$, ranging from 0.03 to 0.11 Bq/kg. The $^{239,240}\text{Pu}$ activity concentrations were similar in *Betula nana*, plants of *Empetrum hermaphroditum* and berries of *Empetrum hermaphroditum*, ranging from 0.001 - 0.008 Bq/kg, except for one outlier for *Betula nana* of 0.059 Bq/kg.

The mass activity densities of *Cladonia stellaris* found in 2002 and 2003 were compared with the values found at the same locations in 1990 [2]. The decrease in activity is due to growth of new low-activity material (diluting material directly contaminated by fallout, wash out by precipitation and radioactive decay). Ecological half-lives of ^{137}Cs in *Cladonia* were calculated assuming first order kinetics and ranges from 4.3 years to 6.2 years with an average of 5.3 years. The average time for one year growth to reach the bottom zone with dead material has been reported to be about 7-8 years in sub alpine areas of central Norway [6], which gives an expected ecological half life of 3.5-4 years. This indicates that the major contribution to the ecological half-life of *Cladonia stellaris* is due to growth of new low-activity material. Varskog *et al.* [7] reported shorter half-lives for lichens in the time period from 1987-89, ranging from 2 to 3.5 years. However it is not unlikely that radiocaesium adsorbed on the outer surfaces may have been washed out more efficiently during the first years after the fallout from Chernobyl in 1986, thus leading to shorter ecological half-lives in this time period.

The aggregated transfer coefficient, C_{ag} , is defined as the mass activity density, A_m , in a specified object per unit area activity density in the soil [8]. C_{ag} 's were calculated using the average activity concentrations for each species and the average area activity density for the soil sample within each location. The ranges of the calculated C_{ag} 's were 0.0065-1.0 for ^{137}Cs , 0.008-0.8 for ^{90}Sr and 0.00006-0.0020 for $^{239,240}\text{Pu}$.

Conclusions

The best indicator for ^{137}Cs in this study is probably *Cladonia stellaris*, although the highest concentrations and transfer coefficients of ^{137}Cs was found in *Rozites caperata*. *Rozites caperata* is however only available in a limited time period of the year, and may be difficult to find in some environments. *Cladonia stellaris* is very common in all alpine and sub-alpine ecosystems in Norway. *Betula nana* (leaves) and *Empetrum hermaphroditum* are both good indicator organisms for ^{90}Sr . *Cladonia stellaris* is the best indicator for Pu among the species that were measured for Pu.

Marine environment

Introduction

The seawater near the island Tromøya at the southeastern coast of Norway, consists of a mixture of high salinity Atlantic Ocean water and low salinity Baltic Sea water. There are great temporal variations in the mixing ratios, leading to relatively large variations in the salinity. The Atlantic Ocean water has a high concentration of ^{99}Tc compared to the Baltic Sea water due to discharges from Sellafield reprocessing plant, while the concentrations of ^{137}Cs , ^{90}Sr and Pu are highest in the Baltic Sea water due to the Chernobyl accident. The

concentration of these nuclides in the seawater at the sampling location is therefore assumed to vary with the salinity.

Fucus vesiculosus and *Ascophyllum nodosum* are both commonly found along the coast of Norway, and they often occur together on the middle shore. While the fronds of *Fucus vesiculosus* grow relatively fast and live for only 3-5 years, *Ascophyllum nodosum* is very slow growing and individual fronds become up to 15 years before breakage.

Sampling and analyses

Samples of these algae species were collected every month in 2002 and 2003 from one location nearby the island Tromøya. The samples consisted of several individual plants, with a mixture of young and old plants, collected along a stretch of 10-20 m. Only plants growing on rocks or big stones were collected. Salinity was measured in water samples collected at the same location at a depth of about 20 cm.

All samples were dried to constant weight at 105 °C, milled and homogenized before being measured by gamma spectrometry. Samples from 2002 were also analysed for ^{99}Tc , but only samples from every second month in 2002 were analysed for ^{90}Sr and $^{239,240}\text{Pu}$.

Gamma analysis and radiochemical analysis of ^{90}Sr and $^{239,240}\text{Pu}$ were performed at Institute for Energy Technology. The procedures used are described above. The ^{99}Tc -analyses were performed by the Department of Radiation Physics at University of Lund, which also participates in intercomparison exercises at a regular basis. Technetium was extracted by TBP from sulphuric acid-hydrogen fluoride media. Back-extraction was done from sodium hydroxide solution from which the technetium was electrodeposited onto stainless steel discs. $^{99\text{m}}\text{Tc}$ was used as chemical yield determinant. After decay of the yield determinant, ^{99}Tc was measured by an anticoincidence-shielded GM counter [9].

Results and discussion

The salinity at the sampling location varied from 18.0-32.6 ‰. The salinity was lowest in April, May and August 2002, and March and July 2003, which should indicate high concentrations of ^{137}Cs and ^{90}Sr , and low concentrations of ^{99}Tc in these periods. The radionuclide concentrations were in average higher in *Fucus vesiculosus* than in *Ascophyllum nodosum* for all the measured radionuclides except ^{99}Tc . The data show that there were great seasonal variations in the concentrations of the measured radionuclides. For ^{137}Cs , ^{90}Sr , ^{228}Ra and ^7Be the temporal variations in activity concentrations showed the same pattern for the two species, whereas for ^{99}Tc the temporal pattern was completely different for the two species.

The ^{137}Cs -concentrations varied from 0.86-7.3 Bq/kg d.w. in *Fucus vesiculosus*, and from 0.73-3.9 Bq/kg d.w. in *Ascophyllum nodosum*. The concentrations were in average 30 % higher in *Fucus vesiculosus* than in *Ascophyllum nodosum*. The difference is statistical significant ($p < 0.01$). The temporal pattern was similar for the two species, with the highest ^{137}Cs concentrations during April-June and September 2002. The highest concentrations in 2003 were also in the spring-summer, but the variations were less pronounced than in 2002. The variations were larger for *Fucus vesiculosus* than for *Ascophyllum nodosum*. There was a significant negative correlation between salinity and concentrations of ^{137}Cs for both species (Pearson correlations significant at a 0.01 level). The negative correlation may at least partly

be explained by higher water concentrations of ^{137}Cs due to high Baltic outflow in the periods with low salinities. The salinity has also been shown to influence the uptake of ^{137}Cs in *Fucus vesiculosus* [10], with a higher uptake in low salinity water. This may also contribute to the negative correlation between salinity and ^{137}Cs -concentrations seen in our data. Significant effects of temperature and light on the initial rate of uptake of ^{137}Cs in *Fucus vesiculosus* have been reported [11], high temperatures and light giving higher uptake. Increasing water temperatures and improving light conditions in spring and early summer might therefore also explain the increased concentrations in this period. Part of the decrease in ^{137}Cs concentrations in the fall may be explained by growth dilution because of rapid growth in this period as well as lower ^{137}Cs concentrations in the seawater (high salinity). The actual loss of caesium from *Fucus vesiculosus* has been reported to be in the same order of magnitude as the growth dilution [11].

The accumulation of ^{137}Cs has been shown to be highest in the receptacles and the new vegetative tissue of *Fucus vesiculosus* [12]. Because *Fucus vesiculosus* grows more rapidly and lives for a shorter time than *Ascophyllum nodosum* the ratio of the biomass of new vegetative tissue to the biomass of the whole plant is greater for *Fucus vesiculosus* than for *Ascophyllum nodosum*. This may explain why the concentration of ^{137}Cs is higher in *Fucus vesiculosus* than in *Ascophyllum nodosum*.

The ^{90}Sr -concentrations varied from 0.3-1.9 Bq/kg d.w. in *Fucus vesiculosus* and from 0.5-1.5 Bq/kg d.w. in *Ascophyllum nodosum*. The highest concentrations were measured in the samples from April. There was a significant negative correlation between ^{90}Sr concentration in *Fucus vesiculosus* and salinity (Pearson correlations significant at a 0.01 level (*Fucus*) and 0.05 level (*Ascophyllum*)). The $^{239,240}\text{Pu}$ -concentrations were very low in both species, ranging from 0.03-0.18 Bq/kg d.w. For *Ascophyllum nodosum* there were apparently no seasonal variations, and for *Fucus vesiculosus* the concentrations were highest during winter and early spring.

The ^{99}Tc -concentrations varied from 90-270 Bq/kg d.w. in *Fucus vesiculosus*, and from 140-390 Bq/kg d.w. in *Ascophyllum nodosum*. The concentrations were in average 60 % higher for *Ascophyllum* than for *Fucus* ($p < 0.01$), which indicates a more efficient uptake of ^{99}Tc in *Ascophyllum* than in *Fucus*. The higher concentrations of ^{99}Tc in *Ascophyllum nodosum* compared to *Fucus vesiculosus* found in this study are in agreement with earlier measurements of seaweed from the Norwegian coast [13]. There were no correlations between salinity and ^{99}Tc concentration for either of the two species. This may indicate that there is a slower dynamic in the mechanisms of uptake and loss of technetium than for caesium and strontium. The difference in temporal pattern for the two species indicates different dynamic processes for uptake and retention of technetium. Because *Ascophyllum nodosum* grows more slowly and lives longer than *Fucus vesiculosus*, ^{99}Tc might be integrated over a longer time span in *Ascophyllum nodosum* than in *Fucus vesiculosus*, and also the effect of growth dilution should be less pronounced in *Ascophyllum nodosum* than in *Fucus vesiculosus*. This could give a greater memory effect in *Ascophyllum nodosum* than in *Fucus vesiculosus* when pooled samples of individual plants of different age are measured.

The concentrations of ^{228}Ra varied from 5.1-14.4 Bq/kg d.w. in *Fucus vesiculosus*, and from below the detection limit (0.5 Bq/kg) to 6.1 Bq/kg d.w. in *Ascophyllum nodosum*. The concentrations in *Fucus vesiculosus* were in average twice as high as in *Ascophyllum nodosum*. The difference is statistical significant ($p < 0.01$). This is in agreement with previous results reported on naturally occurring radionuclides in the marine environment [14],

showing higher concentration factors for radium in *Fucus vesiculosus* than in *Ascophyllum nodosum*.

The ^7Be -concentrations were significantly higher in *Fucus vesiculosus* than in *Ascophyllum nodosum*, varying from 18-100 Bq/kg d.w. for *Fucus vesiculosus*, and from below the detection limit of about 1 Bq/kg to 30 Bq/kg in *Ascophyllum nodosum*. ^7Be is formed in the atmosphere, and is mainly released to ground level by precipitation. The observed variations might be due to variations in precipitation and other atmospheric conditions, although at the moment we do not have any weather data to support this.

Concentration ratios

The concentration ratio, C_r , is defined as the ratio of the activity concentration of a radionuclide in the receptor compartment to that of the donor compartment. Activity concentrations in seawater has not been measured in this study, but the Norwegian Radiation Protection Authority (NRPA) reports activity concentrations of ^{90}Sr , ^{99}Tc and $^{239,240}\text{Pu}$ in seawater at Narestø [15], which is very close to our sampling location. The seawater was collected in October 2002. The reported activity concentrations are $4.0 \pm 0.5 \text{ Bq/m}^3$ of ^{90}Sr , $1.72 \pm 0.18 \text{ Bq/m}^3$ of ^{99}Tc and $5.0 \pm 1.3 \text{ mBq/m}^3$ of $^{239,240}\text{Pu}$. Activity concentrations of ^{137}Cs in surface water from Narestø in 2001 are reported by NRPA [16] to 19 Bq/m^3 . Date of sampling is not given in the report. Estimated concentration ratios for *Fucus vesiculosus* and *Ascophyllum nodosum*, based on the seawater concentrations reported by NRPA, are given in Table 2. Activity concentrations in seaweed from October 2002 are used in the calculations.

Table 2. Estimated concentration ratios for *Fucus vesiculosus* and *Ascophyllum nodosum* (references in brackets).

Species		C_r [l/kg d.w.]			
		^{137}Cs	^{90}Sr	^{99}Tc	$^{239,240}\text{Pu}$
<i>Fucus vesiculosus</i>	This study	130	260	90 000	11 000
	Literature data	136-549 [17,18]	50 [20*]	35 000- 93 000 [13,19]	20000 [20*]
<i>Ascophyllum nodosum</i>	This study	100	150	120 000	5 900
	Literature data		40 [20*]	31 000-132 000 [13,17]	16000 [20*]

* IAEA [20] reports concentration factors based on fresh weight of sample. The numbers in Table 2 has been calculated using a w.w. to d.w. ratio of 5 for *Fucus vesiculosus* and 4 for *Ascophyllum nodosum*.

Taking into account the relatively rapid shifts in salinity, and the corresponding great variation of the radionuclide concentration in the seawater, the resulting concentration ratios – based on only one seawater dataset – are in good agreement with literature data. It is worth noting that the IAEA data [20] “have been deliberately biased towards the red and green algae data” found in the literature, and they may therefore be somewhat inadequate in comparison with brown algae results.

Modelling of ^{137}Cs -concentrations in *Fucus vesiculosus* from Tromøya by SENSI

The concentration of ^{137}Cs in seawater at the sampling site varies with the salinity because of varying mixing of high salinity Atlantic Ocean water with low ^{137}Cs concentration and low salinity Baltic Sea water with higher ^{137}Cs concentration. The ^{137}Cs concentration in the seawater can therefore be expected to depend linearly on the salinity. The SENSI model [11]

is developed to predict monthly activity concentrations of some radionuclides in *Fucus vesiculosus*, knowing the average monthly discharge in the same and previous months from a Swedish nuclear power plant (Ringhals). The model uses a matrix of sensitivity factors taking into account variations in uptake and biological decay due to variations in light, temperature and growth rates during the year. The sensitivity matrix is multiplied by monthly discharge values and a site-specific calibration factor. A sensitivity matrix for ^{137}Cs was developed in [11], but the model was not tested for ^{137}Cs because of to high background level compared to the discharge of ^{137}Cs .

Replacing monthly discharge values and the site specific calibration factor with a linear function of salinity (best fit to measured concentrations) the SENSI model was used to calculate predicted monthly ^{137}Cs concentrations in *Fucus vesiculosus* from Tromøy for the years 2000-2003. The calculated values are shown together with the measured values in Figure 10. There is a good agreement between the calculated and measured concentrations, although the model seems to underestimate the highest ^{137}Cs concentrations. An explanation for this might be that the model does not take into account that the salinity itself influences the uptake of ^{137}Cs in *Fucus vesiculosus*, with a higher uptake in low salinity water [10].

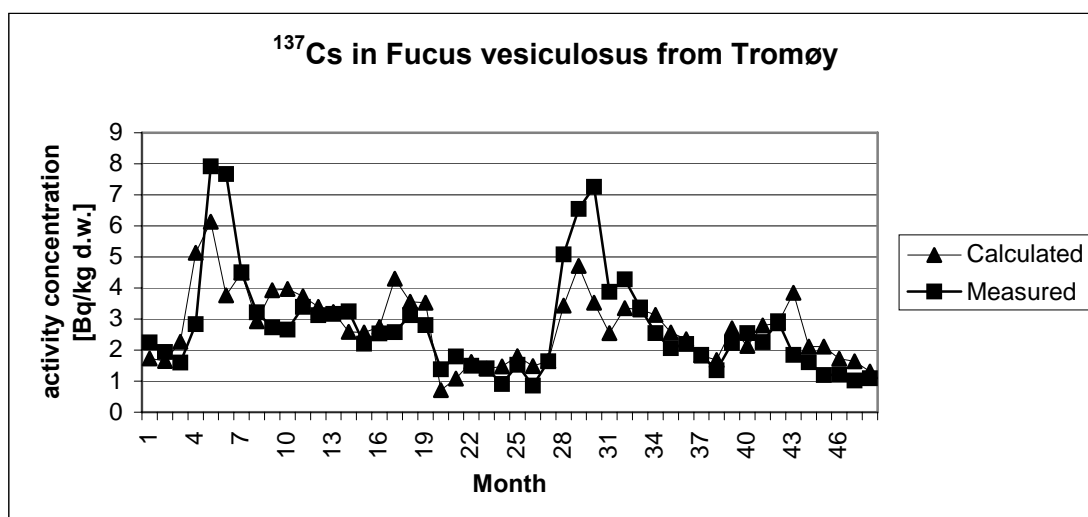


Figure 1. Measured and calculated concentrations of ^{137}Cs in *Fucus vesiculosus* from Tromøy using the SENSI model (starting January 2000).

Conclusions

This study confirms that *Fucus vesiculosus* is a very good indicator organism for radioactivity in marine environments. *Ascophyllum nodosum* is also a good indicator organism, but have slightly lower concentration factors for all the radionuclides in this study except for technetium. *Ascophyllum nodosum* seems to have a slower response to change in seawater concentrations of technetium than *Fucus vesiculosus*, which gives a lower time resolution for *Ascophyllum nodosum*. This might be an advantage when sampling is less frequent.

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New indicator organisms for environmental radioactivity in Iceland 2002-2005

Magnús Á. Sigurgeirsson, Kjartan Guðnason

Icelandic Radiation Protection Institute, Raudarárstíg 10, IS-150 Reykjavík

Terrestrial environment

Within the NKS-B/INDOFERN project the main emphasis of the Geislavarnir laboratory has been on terrestrial bioindicators, mainly plants. Since summer 2002 about one-hundred samples from fifteen different plant species have been collected for radioanalytical studies (table 1). The sampling sites selected, which are 45 in total, are distributed evenly throughout the country (figure 1). All the species sampled are widespread in Iceland and rather easily identified in the field. Until now only the Cs-137 activity concentrations of the samples have been measured.

Soil cores were collected at each sampling site down to 20 cm depth. The total radiocaesium inventory of the soils measured 750-4800 Bq/m², correlating closely to total annual precipitation. The vertical distribution of radiocaesium in volcanic soils, which are characteristic for Iceland, is markedly influenced by soil erosion and aeolian activities. On average about 83 % of total radiocaesium is retained in the uppermost 5 cm of the soil (Sigurgeirsson *et al.* 2005).

Table 1. Plant species sampled 2002-2005, range of values.

Plant species	n	Range of values Bq/kg (d.w.)
<i>Calluna vulgaris</i>	22	1,0-23,1
<i>Racomitrium sp.^a</i>	21	1,5-23,0
<i>Empetrum nigrum</i>	20	0,3-5,4
<i>Salix phylicifolia</i>	12	0,3-19,6
<i>Equisetum palustre</i>	4	0,5-73,3
<i>Eleocharis palustris</i>	3	3,4-7,6
<i>Equisetum fluviatile</i>	3	4,1-48,2
<i>Equisetum variegatum</i>	3	17,7-35
<i>Hippuris vulgaris</i>	3	0,3-89,9
<i>Salix callicarpaea</i>	3	0,7-2,2
<i>Cladonia arbuscula</i>	2	1,9-4,3
<i>Juniperus communis</i>	2	0,6
<i>Carex bigelowii</i>	1	1,7
<i>Carex rostrata</i>	1	3,5
<i>Festuca richardsonii</i>	1	1,5

^a only measurements from the green part (annual growth) are included

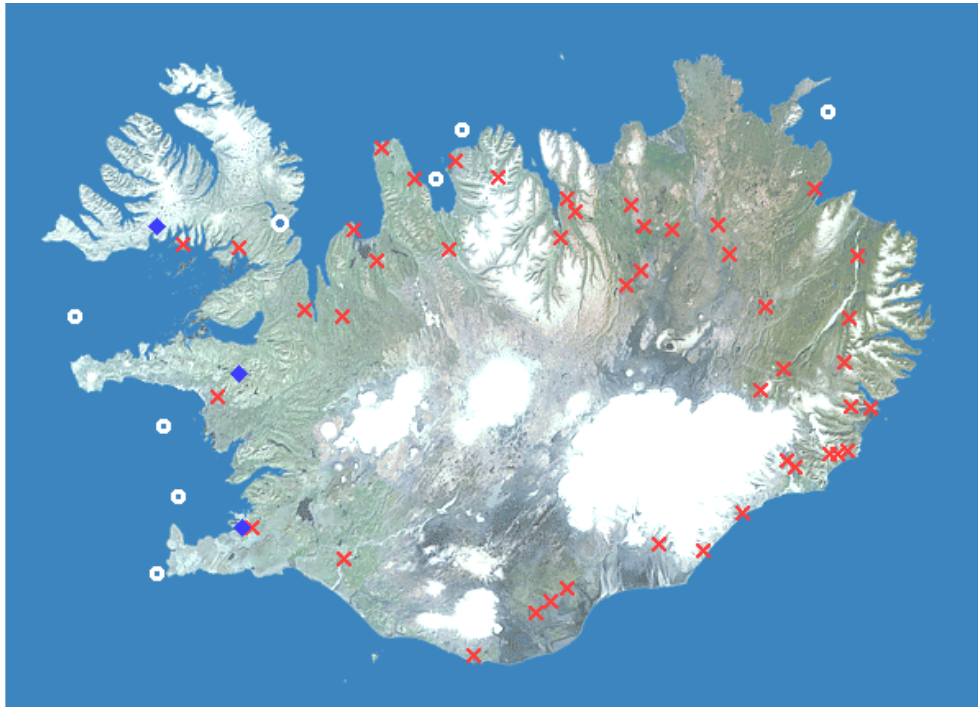


Figure 1. Sampling sites 2002-2005, plants (x), fresh water fish (♦) and whales (⊙).

Two types of willow (i.e. *Salix phylicifolia* and *Salix callicarpaea*), were extensively sampled in the first season of the INDOFERN project. The results indicated a very low radiocaesium activity in willow, mostly giving values below the detection limit. Those species have not been sampled further. Also the gamma activity of crowberry (i.e. *Empetrum nigrum*) appeared to be very low, commonly below the detection limits.

The highest individual values were obtained from the *Equisetum* sp. and *Hippuris vulgaris*, about 90 Bq/kg. Moderately high values were obtained for *Calluna vulgaris* (heather), a common plant in the Icelandic heaths along with *Empetrum nigrum*. Of these two species generally higher levels of radiocaesium were shown by the former. *Calluna vulgaris* is ingested by sheep which range freely in the heaths during the summertime. A high radiocaesium level of ruminants has been related to the grazing of *Calluna vulgaris* and other ericaceous species (Howard *et al.* 1991). The plants stiff sedge (*Carex bigelowii*) and arctic festuca (*Festuca richardsonii*), important constituents of the regular reindeer diet, show low radiocaesium activities.

The measurements indicate that mosses, in general, may be regarded as good bioindicators. In the first season of INDOFERN whole-plant samples were collected, including both the roots and the annual growth of the mosses. The results showed a large scatter in radiocaesium activity concentrations of the samples, most probably controlled by soil contamination of its basal part. In later seasons of the project, a new sampling technique was applied, where only the annual growths were sampled for analysis. Considerably more uniform and in general lower values are obtained with this method. At one sampling site in S-Iceland samples of both types were collected for comparison, i.e. of cut (green part) and uncut mosses (whole-plant sample). The difference in radiocaesium concentration of these samples is noteworthy, the cut moss gave 1,5 Bq/kg (dw) while the uncut moss gave about 80 Bq/kg (dw).

Transfer factors (TFs) for the most suitable terrestrial bioindicators, collected in 2002-2004, are summarised in table 2. As indicated the highest TFs are obtained for horsetails (i.e.

Equisetum sp.) but somewhat lower for mosses (i.e. *Racomitrium sp.*) and *Calluna vulgaris*. Transfer factors for crowberry (i.e. *Empetrum nigrum*) are considerably lower. The large scatter of TFs obtained may be explained mostly by variability in radiocaesium inventory of the soils and soil types at the sampling sites.

Table 2. Transfer factors for plants, results from 2002-2004.

Species	TFs, range of values $\text{m}^2(\text{kg d.w.})^{-1}$	TFs, averages $\text{m}^2(\text{kg d.w.})^{-1}$
<i>Equisetum sp.</i>	0,0001-0,0816	0,0254
<i>Racomitrium sp.</i>	0,0004-0,0285	0,0074
<i>Calluna vulgaris</i>	0,0007-0,0231	0,0066
<i>Eleocharis palustris</i>	0,0015-0,0083	0,0044
<i>Empetrum nigrum</i>	0,0002-0,0020	0,0009

Other samples

Two samples of bird meat have been analysed. One sample of puffin meat (*Fratercula arctica*), a common sea bird in Iceland, has been measured for radiocaesium. The radiocaesium activity concentration was below the detection limit, i.e. $< 0,19 \text{ Bq/kg (f.w.)}$. One measurement from a wild goose indicates a considerable radiocaesium uptake of this bird, a value of $5,6 \text{ Bq/kg (f.w.)}$ was obtained.

Aquatic environments

Fresh water fish

Samples of fresh water fish, i.e. trout, have been sampled from three different lakes in S- and W-Iceland (figure 1, table 3). Water samples from the lakes have not been collected yet.

Table 3. Fresh water fish sampled in 2003-2004.

Fish species	Lake	Sampling date	n (of indiv.)	Bq/kg (f.w.)
Trout (<i>Salvelinus alpinus</i>)	Vatnsdalsvatn (NW-Iceland)	July 2003	~10	2,03
	Vatnsdalsvatn (NW-Iceland)	May 2004	2	5,84
	Vatnsdalsvatn (NW-Iceland)	May 2004	3	9,58
	Vífilsstadavatn (SW-Iceland)	May 2004	1	1,96
	Vífilsstadavatn (SW-Iceland) ^b	April 2004	2	6,90
Trout (<i>Salmo trutta</i>)	Hítarvatn (W-Iceland)	August 2003	2	4,32

^b dried fish sample

Marine samples

One sample of Norway lobster (*Nephrops norvegicus*) has been measured and one of scallop (*Chlamys islandica*). Number of individuals in each sample was c. 30. Both the samples measured below detection limits, giving values of $< 0,1 \text{ Bq/kg (f.w.)}$ and $< 0,3 \text{ Bq/kg (f.w.)}$, respectively.

The radiocaesium activity of ten samples from minke whale (*Balanoptera acutorostrata*), have been measured. The results are listed in table 4. The results indicate a low radiocaesium concentration of this whale species, i.e. < 0,6 Bq/kg.

Table 4. Cs-137 in whale meat, minke whale.

Species (id. number in parenthesis)	Hunting location	Cs-137, Bq/kg (f.w.)
Minke whale (A0303)	Skagafjörður	0,19
Minke whale (A0304)	Skagafjörður	<0,20
Minke whale (A0307)	Bakkaflói	0,29
Minke whale (A0313)	Steingrímsfjörður	0,21
Minke whale (B0303)	West of Reykjanes	0,20
Minke whale (B0304)	Faxaflói	0,17
Minke whale (B0307)	Faxaflói	0,22
Minke whale (B0311)	Breidafjörður	0,18
Minke whale	Meat from a store	0,33
Minke whale	Meat from a store	0,59

So far only one sample of seal meat (*Phoca vitulina*) has been measured at IRPI, giving a value of 0,20 Bq/kg (f.w.).

Samples of seaweed are collected from six different sites at the Icelandic shores (figure 2, table 5). From each site four samples are obtained annually. Regular measurements of radiocaesium in seaweed have been ongoing at IRPI since 1989.

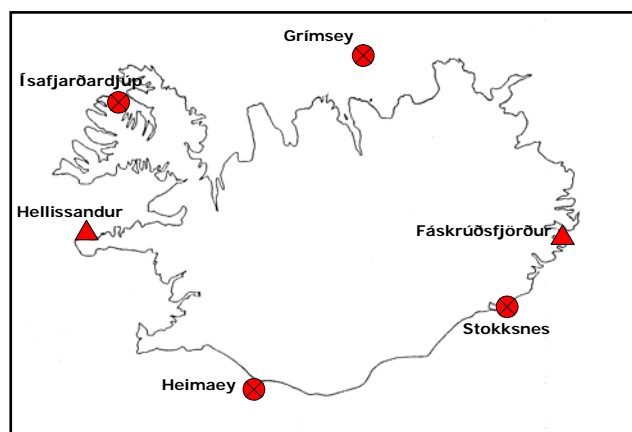


Figure 2. Seaweed sampling sites at the Icelandic coast (Circles: *Fucus vesiculosus*; triangles: *Ascophyllum nodosum*).

Table 5. Concentration ratios for seaweed.

Sampling site	Species	¹³⁷ Cs, Bq/kg (avr.)	CF (liter kg ⁻¹) ^c
Grímsey	<i>Fucus vesiculosus</i>	0,23	74,2
Fákrúðsfjörður	<i>Ascoph. nodosum</i>	0,20	64,5
Stokksnes	<i>Fucus vesiculosus</i>	0,22	71,0
Heimaey	<i>Fucus vesiculosus</i>	0,18	58,1
Hellissandur	<i>Ascoph. nodosum</i>	0,17	54,8
Ísafjarðardjúp	<i>Fucus vesiculosus</i>	0,19	61,3

^c Radiocaesium concentration activity of Icelandic coastal waters is about 3 Bq/m³ (=0,003 Bq/liter) on average.

Conclusions

- For all the plant species sampled the highest indicator values are obtained for horsetails and mosses, giving the highest transfer factors (m² (kg d.w.)⁻¹), 0,0254 and 0,0074, respectively.
- For heath vegetation, considerably higher transfer factors were obtained for *Calluna vulgaris* than *Empetrum nigrum*, 0,0066 and 0,0009, respectively. This indicates a higher bioindicator value of the former species.
- Measurements of two different types of birds, i.e. a puffin and a goose, indicate considerably higher radiocaesium levels of birds living and grazing in the interior of Iceland than seabirds.
- Measurements of fresh water fish (i.e. trout) indicate a considerable scatter of values, i.e. between 2 and 10 Bq/kg (f.w.).
- Ten samples of whale meat from minke whale (*Balanoptera acutorostrata*) have been measured. The results indicate a low radiocaesium uptake of this whale species. Values of less than 0,6 Bq/kg (f.w.) were obtained.
- Measurements of marine samples, i.e. Norway lobster and scallop (one sample of each) indicate a low Cs-137 uptake of these species. Both samples measured below the detection limit, giving values of less than 0,3 Bq/kg (f.w.).
- Concentration factors for seaweed, i.e. *Ascophyllum nodosum* and *Fucus vesiculosus*, are in the range of 54 and 74 (liter kg⁻¹).

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Indicator organisms for environmental radioactivity. Studies in the Faroe Islands.

Hans Pauli Joensen

Faculty of Science and Technology, Nóatún 3, FO 100 Tórshavn, University of the Faroe Islands

Introduction

Radioactive contamination of the Faroese environment derives mainly from nuclear weapons tests in the 1950's and 1960's and from the Chernobyl nuclear accident in April 1986. Radioactive discharges from Sellafield and La Hague are potential sources for anthropogenic radionuclides in the Faroese marine environment.

This paper presents results from the Faroese part of the INDOFERN project. The ^{137}Cs activity concentration has been measured in selected indicator organisms from the terrestrial, freshwater and marine environments in the years 2002-2004. Results from earlier measurements during the last four decades are also presented.

The indicator value of the organisms is discussed, and a ranking list is suggested in some cases.

Material and methods

Indicator organisms have been sampled for the INDOFERN project in the years 2002, 2003 and 2004. The radioactivity measurements have been carried out with a lead shielded Germanium detector. Data from previous measurements of relevance to INDOFERN have become available from earlier data archives and annual reports from Risø National Laboratory, Roskilde, Denmark (Sven P. Nielsen, Risø, personal communication; Risø Reports, 1962-93).

The indicator organisms include *Empetrum sp.*, moss, lichen, *Calluna vulgaris*, *Erica cinerea* from the terrestrial environment, trout sp. and water from the freshwater lakes, and *Fucus sp.*, haddock and seawater from the marine environment.

Results and Discussion

Terrestrial environment

Tables 1 and 2 present results from terrestrial samplings in the years 2002-2004. It should be noted that the sampling stations were not exactly the same every year. It is wellknown from other studies (Joensen, 1999) that there is large variability in the ^{137}Cs activity concentration in grass, both within and between pastures, and this is presumably also the case for other vegetation.

Measurements prior to INDOFERN are presented in Tables 3 and 4. Measurements of ^{137}Cs and ^{90}Sr from April 1967 indicate a significant geographical variation of the activity concentrations in moss and lichen with lower activity in the southern part of the country (Tvøroyri) as compared with the central area (Tórshavn). The moss and lichen species are, however, not identified in the old data files, and there is neither any information about soil characteristics from the respective sampling sites that could assist to explain the observed differences. The ^{137}Cs activity concentration in moss at Lake Toftavatn decreased from 600 Bq/kg dw in July 1987 to 270 Bq/kg dw in September 1999 and further to 77.5 Bq/kg dw in September 2002 and 58.9 Bq/kg dw in August 2003 (cf. Table 1).

Cs-134 was measured in the moss and lichen samples from Tórshavn in 1987. The $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was 0.532 and 0.585 for moss and lichen, respectively, as date corrected to 26 April 1986, indicating that practically all the radiocaesium activity in 1987 derived from the Chernobyl accident.

Table 1. Cs-137 in terrestrial samples taken on the shores of lake Toftavatn and Leitisvatn on the islands Eysturoy and Streymoy, respectively. Values are given with 1 counting standard deviation. *: *Empetrum hermaphroditum*. **: *Empetrum nigrum*. NS: No Sample.

	Sampling date	Soil		Mixed Grass Bq/kg dw	<i>Empetrum sp.</i>	<i>Hylocomium splendens</i>
		0-5cm Bq/m ²	5-10cm Bq/m ²		Bq/kg dw	Bq/kg dw
Toftavatn	9 Sept. 2002	1514±74	1619±66	97.8±9.49	17.8±1.66 *	77.5±4.37
Leitisvatn	6 Sept. 2002	2106±99	1940±112	75.4±4.90	NS	NS
Toftavatn	30 Aug. 2003	NS	NS	NS	26.3±2.31 **	58.9 ± 4.16

Table 2. Cs-137 in samples from 2003 and 2004. All activity concentrations are given as Bq/kg dry weight, together with 1 counting standard deviation. The samples were collected at the site Norðoyri on the island Borðoy 4 August both years, except *Parmelia saxatilis* which was sampled 17 July 2004 at the site Trøllanes on the island Kalsoy. (NS: Not Sampled).

Year	<i>Empetrum herma-phroditum</i> (Northern crowberry)	<i>Empetrum nigrum</i> (Crowberry)	<i>Erica cinerea</i> (Bell heather)	<i>Calluna vulgaris</i> (Heather)	<i>Racomitrium lanuginosum</i>	<i>Potentilla erecta</i> (Common tormentil)	<i>Parmelia saxatilis</i>
2003	NS	11.8 ± 2.21	11.2 ± 2.25	91.3 ± 5.33	31.9 ± 1.90	NS	NS
2004	6.13 ± 0.34	NS	10.2 ± 0.86	29.3 ± 0.82	46.1 ± 0.78	14.1 ± .76	49.5 ± 0.74

Table 3. Measurements of ^{137}Cs , ^{134}Cs and ^{90}Sr in moss from 1967, 1987 and 1999 (unspecified species). The unit for all data is Bq/kg fw.

1 April 1967			1 July 1987		1 Sept. 1999
Tvøroyri	Tórshavn		Tórshavn		Toftavatn
^{137}Cs	^{137}Cs	^{90}Sr	^{134}Cs	^{137}Cs	^{137}Cs
363	2479	220	219	600	270

Table 4. Measurements of ^{137}Cs and ^{90}Sr in lichen (unspecified species) sampled in Tórshavn and Tvøroyri 1 April 1967 and “4-5km from Tórshavn” 1 July 1987. (NS: No Sample)

Year	Tórshavn			Tvøroyri	
	^{134}Cs Bq/kg fw	^{137}Cs Bq/kg fw	^{90}Sr Bq/kg fw	^{137}Cs Bq/kg fw	^{90}Sr Bq/kg fw
1967	NS	3352	816	1221	89.0
1987	262	654	3.44	NS	NS

Freshwater environment

Samples have been taken from the three freshwater lakes Leitisvatn, Stórávatn and Toftavatn, in the islands Vágoy, Sandoy and Eysturoy, respectively. All three lakes have non-migratory stocks of brown trout (*Salmo trutta*). The trophic level of the lakes is decreasing in the order Stórávatn, Toftavatn, Leitisvatn. Some earlier samplings exist from lake Leynavatn in the island Streymoy.

Leitisvatn is an oblong relatively deep lake gradually descending towards a center line. It has a surface area of 3.42km² and a maximum depth of 59m. Stórávatn is a shallow lake with an almost uniform depth. It has a surface area of 0.160km² and a maximum depth of 1.8m. Toftavatn consists of a shallow northern part with maximum depth of 3.5m and a southern deeper part with almost circular isobaths and maximum depth 22m. The surface area of lake Toftavatn is 0.509km². Leynavatn is an oblong lake with a surface area of 0.18km² and maximum depth of 33m. A description of the lakes can be found in Christoffersen *et.al.* (2002).

The results from 2002 are presented in the Table 5. Only two trouts were sampled in 2003, one from Leitisvatn (23 July) and one from Stórávatn (1 June) with the ¹³⁷Cs activity concentrations 1.26 Bq/kg fw and 16.2 Bq/kg fw, respectively. The significantly higher activity concentration in the trout from Stórávatn as compared to Leitisvatn may be a reflection of feeding habit of the trout, as trout in Stórávatn would be expected to feed more on benthic fauna than trout in Leitisvatn. The trout stomach contents were, however, not studied. But studies in July 1987 and July 1988 showed that trout from Stórávatn feed mainly on benthic fauna followed by diptera, while trout from Leitisvatn feed almost equally on diptera and benthic animals (Joensen and Vestergaard, 1992).

The observed ¹³⁷Cs activity concentration in lake water increased from 2002 to 1999 in Toftavatn. This should not be expected, but one reason may be that the samples have been collected from different parts of the lake in these years, as it consists of a deeper and a shallower relatively separated parts. The higher ¹³⁷Cs concentration in water from Toftavatn as compared to Leitisvatn (Table 5) may be explained by higher resuspension from sediments in Toftavatn, as this lake is much shallower and smaller than Leitisvatn.

Table 5. ¹³⁷Cs in Brown trout flesh (*Salmo trutta*) and lake water from Toftavatn and Leitisvatn in 2002. Trout were caught 15 August in both lakes. Water samples (each 200 liter) were collected 3 September from Toftavatn and 6 September from Leitisvatn.

Lakes in 2002	<i>Salmo trutta</i>					Water Bq/m ³
	Number of trout	Mean Bq/kg fw	St. dev Bq/kg fw	Min Bq/kg fw	Max Bq/kg fw	
Toftavatn	9	9.88	4.20	5.85	19.8	5.51±0.32
Leitisvatn	10	5.31	1.99	4.02	10.7	3.45±0.27

Table 6. Measurements of radiocaesium in lake water after the Chernobyl reactor accident. All values are given in Bq/m³. (NS: No Samples).

	July 1987		July 1989		August 1991	June 1993	Sept. 1999
	¹³⁴ Cs	¹³⁷ Cs	¹³⁴ Cs	¹³⁷ Cs	¹³⁷ Cs	¹³⁷ Cs	¹³⁷ Cs
Leitisvatn	1.60	9.94	0.63	6.23	6.24	6.24	NS
Leynavatn	0.90	3.74	NS	1.84	2.60	2.30	1.26
Toftavatn	NS	NS	NS	NS	NS	NS	4.17

Table 7. Measurements of ¹³⁷Cs and ⁹⁰Sr in rainbow trout flesh (*Salmo irideus*) in the Faroe Islands (no site specification). One trout from each of the years 1975 and 1976. In 1987, the data represent averages in flesh from 18 Brown trout (*Salmo trutta*) from the lake Leitisvatn.

1 August 1975				1 June 1976				July 1987	
¹³⁷ Cs		⁹⁰ Sr		¹³⁷ Cs		¹³⁷ Cs		⁹⁰ Sr	¹³⁷ Cs
Bq/kg fw	Bq/kg K	Bq/kg fw	Bq/kg Ca	Bq/kg fw	Bq/kg K	Bq/kg fw	Bq/kg Ca	Bq/kg fw	Bq/kg fw
0.78	196	0.17	207	0.45	126	0.22	207	42.4	115.4

Measurements from the 1970's, 1980's and 1990's are presented in Tables 6 and 7. Date correcting to 26 April 1986, the ¹³⁴Cs/¹³⁷Cs ratio for lake water in 1987 was 0.235 and 0.351 in Leitisvatn and Leynavatn, respectively, and 0.281 in Leitisvatn in 1989. The trouts from Leitisvatn in 1987 contained practically only radiocesium from Chernobyl, as the average ¹³⁴Cs/¹³⁷Cs ratio was 0.536 (date corrected to 26 April 1986).

Marine environment

Seawater and *Fucus sp.* were sampled in 2002, 2003 and 2004. Measurements of ¹³⁷Cs are presented in Tables 8. The ⁹⁹Tc activity concentration was measured to 1.68±0.08 Bq/kg dw in the *Fucus vesiculosus* sample from 20 February 2003.

Table 8. Cs-137 in marine samples from Kirkjubøur. Concentrations are given with 1 counting std. deviation. (BDL: Below Detection Limit. NA: Not Analyzed. NS: No Sample).

Sampling date	Seawater Bq/m ³	<i>Fucus vesiculosus</i> Bq/kg dw	<i>Ascophyllum nodosum</i> Bq/kg dw
27.06.2002	1.85 ± 0.14	BDL	NS
20.02.2003	1.70 ± 0.23	<0.3	NS
12.05.2003	2.04 ± 0.16	BDL	NS
25.09.2003	1.92 ± 0.34	0.262 ± 0.059	NS
05.03.2004	NA	0.175 ± 0.053	0.220 ± 0.050
26.05.2004	NA	*) 0.300 ± 0.063	NS
24.09.2004	NA	NA	NS

*) *Fucus vesiculosus* was sampled 15 June 2004

Ascophyllum nodosum and *Fucus vesiculosus* were both sampled at the same site in 2004 in order to compare their ¹³⁷Cs activity concentrations. The activity concentrations in the two species were the same within one standard deviation.

Results from 2003 indicate that haddock (*Melanogrammus aeglefinus*) is a rather poor indicator organism. The ^{137}Cs activity concentration in a sample of haddock flesh from 15 March 2003 was 0.071 ± 0.007 Bq/kg fw, i.e. about 27% of the dry weight concentration in *Fucus vesiculosus*.

Long-term measurements exist for ^{137}Cs and ^{90}Sr in *Fucus* since 1963. The samples have been collected at different sites in the Faroe Islands, but they do all refer to the same water mass. The particular *Fucus* species have been recorded as *Fucus vesiculosus* since 1972, except for 1986 (*Fucus serratus*) and for September 1972 and April 1987 (*Fucus distichus*). The species are recorded as just *Fucus sp.* until 1968. The ^{137}Cs activity concentration in *Fucus sp.* decreased with short effective ecological half-life during the first three years of measurements in the 1960's. The level was constant from the mid 1970's until 1986 when input from the Chernobyl nuclear accident caused the activity concentration to exceed the level from the mid 1960's. Exponential curve fitting to the ^{137}Cs data from March 1963 to August 1966 and from August 1986 to April 1988 resulted in half-lives of 271 days ($R^2=0.986$) and 142 days ($R^2=0.883$), respectively. The shorter post-Chernobyl half-life is explained by the fact that the input from Chernobyl came as a short pulse, while the input to the atmosphere in the 1960's was distributed over a longer time scale. The ^{90}Sr data do not show trends similar to ^{137}Cs in the early 1960's and after the Chernobyl nuclear accident, although the general decreasing pattern is similar (except for early 1960's and a maximum ^{90}Sr value in March 1968). There was no ^{90}Sr input from Chernobyl.

Cs-134 was measured in four *Fucus* samples after the Chernobyl accident, showing date corrected (to 26 April 1986) $^{134}\text{Cs}/^{137}\text{Cs}$ ratios of 0.491, 0.465, 0.681, 0.497 for samples collected 1 August 1986, 1 April 1987, 1 June 1987 and 17 July 1989, respectively. It shows that *Fucus* is an effective marine indicator organism for ^{137}Cs .

Transfer factors and concentration ratios

Estimates of transfer factors in the terrestrial environment are presented in Table 9. The value for mixed grass at Toftavatn in 2002 was calculated as the activity concentration (Bq/kg dw) in grass divided by the measured deposition (3133 Bq/m^2) in the uppermost 10cm of the soil. Grass and soil samples were collected at the same place. The transfer factors for other species than grass must be considered as more rough estimates, as they were collected some 100 meters away from the soil sampling station. The estimates at Toftavatn in 2003 were calculated from the deposition in 2002, and the values at Norðoyri in 2003 and 2004 were both years based on the deposition measured in 2003 (5455 Bq/m^2). The results show differences between species regarding uptake of ^{137}Cs , as well as a geographical variability.

Table 9. Transfer factors ($\text{m}^2/10^3 \text{ kg dw}$) for ^{137}Cs .

	Mixed grass	<i>Empetrum hermaphroditum</i>	<i>Empetrum nigrum</i>	<i>Hylocomium splendens</i>	<i>Erica cinerea</i>	<i>Calluna vulgaris</i>	<i>Racomitrium lanuginosum</i>	<i>Potentilla erecta</i>
Toftavatn 9 Sep. 2002	31.2	5.68		24.7				
Toftavatn 30 Aug 2003			8.39	18.8				
Norðoyri 4 Aug. 2003			2.16		2.05	16.7	5.85	
Norðoyri 4 Aug. 2004		1.12			1.86	5.37	8.44	2.58

Apart from mixed grass, the results indicate that *Hylocomium splendens* and *Calluna vulgaris* are the most suitable indicator organisms among the selected species. The transfer factors for *Racomitrium lanuginosum* and *Calluna vulgaris* turn out in opposite order in 2003 and 2004. *Parmelia saxatilis* is also a suitable indicator organism (cf. Table 2).

The concentration ratio $[(\text{Bq/kg dw})/(\text{Bq/m}^3)]$ between ^{137}Cs activity in *Fucus vesiculosus* and seawater from Kirkjubøur 25 September 2003 was $0.136 \text{ m}^3/\text{kg dw}$.

The concentration ratio $[(\text{Bq/kg ww})/(\text{Bq/m}^3)]$ between ^{137}Cs activity in haddock flesh (*Melanogrammus aeglefinus*) and seawater was $0.038 \text{ m}^3/\text{kg ww}$. The value is calculated from the concentration in the haddock sample from 15 March 2003 and the average seawater concentration in the samples from 20 February and 12 May 2003.

Conclusion

Hylocomium splendens and *Calluna vulgaris* are found to be good indicator organisms. *Racomitrium lanuginosum* and *Parmelia saxatilis* are also found to be suitable indicator organisms. The results indicate, however, that mixed grass is one of the best terrestrial indicators. The highest transfer factor for ^{137}Cs was found for mixed grass.

Salmo trutta is a suitable indicator organism for the freshwater environment. It showed a ^{137}Cs signal soon after the Chernobyl accident.

Fucus vesiculosus is a good indicator organism for the marine environment. It showed a soon reaction to ^{137}Cs input from the Chernobyl accident. It was observed that the halflife for ^{137}Cs in *Fucus vesiculosus* was shorter after the Chernobyl accident as compared to the situation in the early 1960's.

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Indicator value of certain aquatic organisms for radioactive substances in the sea areas off the Loviisa and Olkiluoto nuclear power plants (Finland)

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

STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Introduction

The results of the marine radioecology studies carried out in 2000-2001 in the sea areas off the Loviisa and Olkiluoto Nuclear Power Plants (South and West coast of Finland; see Fig 1) are reported. Extensive regular monitoring programmes of environmental radioactivity have been carried out already for about 30 years in these areas. The aim of the present study was to compare the indicator value of the various members of the aquatic ecosystem with respect to environmental monitoring. Samples were taken from 27 species including phytoplankton (9 samples), zooplankton (9 samples), periphyton (12 samples), macroalgae and vascular plants (16 samples), benthic animals (8 samples), fish (20 samples) and birds (6). Special attention was paid to different tissues and organs of fish and birds, such as flesh, liver, entrails, bones, milt, spawn, eggs, egg shells etc. (in total 64 samples), because there has been a lot of debate among the opponents of nuclear power in the course of time about the role these objects in the environmental monitoring of the power plants. The samples were taken from relatively small areas both in Loviisa and Olkiluoto, which makes the results well comparable inside each of the sites.

The study areas

The study areas are typical brackish water environs of the Finnish coast. In the Loviisa area, the salinity of surface water varies from almost 0‰ in winter to nearly 6‰ in late autumn (in general, about 3.5-5‰ during the growing period). In the Olkiluoto area, the average salinity of surface water is ca 6‰ during the growing period. The Loviisa area received slightly more ^{137}Cs (on an average about 23 kBq m^{-2}) as a consequence of the Chernobyl fallout in 1986 than the Olkiluoto area (on an average about 20 kBq m^{-2}), both belonging, however, to the same deposition category (3) in the Finnish classification (Arvela et al., 1987). During this study the average activity concentration of ^{137}Cs in surface water was 0.0475 Bq l^{-1} (47.5 Bq m^{-3}) in the Loviisa area and 0.069 Bq l^{-1} (69 Bq m^{-3}) in Olkiluoto. The total amounts of ^{137}Cs in the bottom sediments varied from 23 to 49 kBq m^{-2} in the Loviisa area in 1998, and from 10 to 43 kBq m^{-2} in the Olkiluoto area in 1999.



Figure 1. Location of Loviisa and Olkiluoto NPPs.

Material and methods

The plankton samples were taken by drawing horizontal plankton nets behind a slowly moving motor boat in the surface water for about one hour. The mesh size of the phytoplankton net was 25 μm and that of the zooplankton net 100 μm . Both nets were drawn parallelly at the same time, and they were emptied every 15-20 minutes to wide-mouthed glass bottles. The target size of the sample was 25 g dry weight. The periphyton samples were collected with 50 x 100 cm plywood boards (total collection area 1 m^2) that were installed with buoys and anchors at a depth of 0.5-1 m. Periphyton was scraped out from the plates four times during the growing season. The sample was let flow into a large collection vessel and was then transferred to plastic boxes. The plankton and periphyton samples were kept in refrigerator temperature and carried as soon as possible to the lab for drying. The algae, the vascular plants and the common mussels were collected by SCUBA diving and the other benthic animals with benthos crabs, except *Saduria entomon* that was collected using bait nets. The fish samples were caught with nets and the birds were shot with the permission of authorities.

The activity concentrations of gamma-emitting nuclides were measured by low-background, high-resolution spectrometer systems. The relative efficiencies of the HPGe detectors range from 21% to 78% and the energy resolution from 1.8 keV to 2.1 keV at 1.33 MeV. The measurements were performed in 15-cm thick cylindrical background shields lined with cadmium and copper in the inside. The spectra were analysed using the GAMMA-99-computer code developed at STUK especially for environmental sample measurements.

Results and discussion

In both areas, the activity concentration of ^{40}K was clearly highest in a filamentous green alga *Cladophora glomerata* and in other aquatic plants. The lowest ^{40}K concentrations were detected in the egg shells of birds, in the mussels and in the bones of birds and fish (Tables 1 and 2, Fig. 2).

Discharge nuclides from the local nuclear power plants (^{51}Cr , ^{54}Mn , ^{58}Co , ^{60}Co , ^{95}Zr , ^{95}Nb , $^{110\text{m}}\text{Ag}$, $^{123\text{m}}\text{Te}$ and ^{124}Sb) were only detected at the lower trophic levels of the ecosystem. ^{60}Co was detected in phytoplankton, zooplankton, periphyton (maximum), macroalgae and other submerged aquatic plants (especially Spiked water milfoil, *Myriophyllum spicatum*), and in two mussel species (Common mussel, *Mytilus edulis*, and Baltic Tellin, *Macoma balthica*) and in a relict crustacean *Saduria entomon*. ^{54}Mn , ^{58}Co , $^{110\text{m}}\text{Ag}$ and $^{123\text{m}}\text{Te}$ were most abundantly detected in periphyton, in submerged aquatic plants (e.g. Hair pondweed, *Potamogeton pectinatus* and *Myriophyllum spicatum*), and in the bladder-wrack (*Fucus vesiculosus*). ^{124}Sb was most abundant in *Myriophyllum spicatum* and *Potamogeton pectinatus*. Local discharge nuclides were not detected in fish or birds, or in their inner organs or reproductive products (Tables 1 and 2). This was in agreement with the results of the study carried out in the Loviisa area in 1988-1989 (Ilus et al., 1992, Ilus, 1995).

The activity concentrations of ^{137}Cs per dry weight were highest in periphyton in the both areas (mean values 240 and 310 Bq kg^{-1} dry weight). However, if the results are given on fresh weight basis, the ^{137}Cs contents were highest in fish flesh (23-38 Bq kg^{-1} fresh weight in perch, *Perca fluviatilis* and 18-26 Bq kg^{-1} in pike, *Esox lucius*). In the muscle tissues of aquatic birds, the ^{137}Cs concentrations were generally lower than in fish flesh. Likewise, the

^{137}Cs concentrations were lower in the inner organs, milt and spawn of fish than in fish flesh. But on the contrary, in certain fish-eating birds (Goosander, *Mergus merganser* and Great black-backed gull, *Larus marinus*), the highest concentrations were not in the muscles but in liver and entrails. The concentrations were generally very low in the birds' eggs; especially in egg shells. In benthic animals, the ^{137}Cs concentrations were equal or slightly lower than in aquatic plants.

The concentration factors of ^{137}Cs (on dry weight basis) were calculated using the average ^{137}Cs concentrations in sea water given earlier. For the sake of uniformity, the CF values were calculated in the same way for the aquatic birds, their organs and eggs, although their living habits differ drastically from the others (not living in water). The highest concentration factors were those of periphyton (3500-6500), perch (1900-2100), pike (1800), zooplankton (1000-1100) and bladder-wrack (720-1100). In some cases (e.g. periphyton and bladder-wrack), the CF values in the Loviisa area were clearly higher than those in Olkiluoto, presumably because of the lower salinity of water in the area. In 2000-2001, most of the Chernobyl-derived ^{134}Cs had already disappeared from the environment; only traces were detected in some fish samples, in zooplankton, periphyton and the bladder-wrack.

The best indicators for the local discharge nuclides were organisms from the lower level of the ecosystem (e.g. periphyton, *Myriophyllum spicatum*, *Fucus vesiculosus*, zooplankton, *Potamogeton pectinatus*, *Cladophora glomerata*, *Saduria entomon* and phytoplankton for ^{60}Co). The best indicators of ^{137}Cs among the lower organisms were periphyton, zooplankton, *Fucus vesiculosus*, phytoplankton, *Cladophora glomerata*, and *Myriophyllum*, and in vertebrates the predatory fish perch (*Perca fluviatilis*) and pike (*Esox lucius*).

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Table 1. Concentrations of gamma-emitting radionuclides and ^{90}Sr in indicator samples taken from the sea area off the Loviisa Nuclear Power Plant in 2000.

Sample		Bq/kg dry wt.												Bq/kg f.w.	CF d.w.	
Organism	Tissue	⁴⁰ K*	⁵¹ Cr	⁵⁴ Mn	⁵⁸ Co	⁶⁰ Co	⁹⁰ Sr	⁹⁵ Zr	⁹⁵ Nb	^{110m} Ag	^{123m} Te	¹²⁴ Sb	¹³⁴ Cs	¹³⁷ Cs*	¹³⁷ Cs*	¹³⁷ Cs*
Phytoplankton		325												34	0.19	720
Zooplankton		420				0-0.49								49	0.37	1000
Periphyton		600	0-161	1.46-10.2	0-25.8	3.9-14.7		0-5.0	0-9.6	0-58	0-1.10	0-8.9	0-3.1	310	10	6500
Macroalgae and other aquatic plants																
Cladophora glomerata		1000				0.68								28	2.94	590
Fucus vesiculosus		770		0-0.95	0-1.69	0.76-2.40	12			0-4.6			0.27-0.68	54	10	1100
Potamogeton pectinatus		730		3.7	14.8	3.6				23.4	0.53	2.2 (6)		8.9	1.05	190
Myriophyllum spicatum		490		3.0	6.5	6.1				16.8	0.31	2.4 (15)	0.35	24	1.93	510
Benthic animals																
Marenzelleria viridis		51												30	5.32	630
Saduria entomon		230				0.7	15			0.46				25	5.5	530
Macoma balthica		70												2.0	0.86	42
Fish																
Clupea harengus memb.	whole**	110					0.054						0.06	31	7.4	650
Esox lucius	fillets	110											0.14	84	18	1800
Rutilus rutilus	whole**	110											0.029	25	5.9	530
Perca fluviatilis	whole**	100					0.15						0.15	88	23	1900
Birds																
Anas platyrhynchos	egg white	390												9.8	1.16	210
	egg yolk	70												1.6	0.73	34
	egg shells													<	<	
Larus canus	muscle	400												43	12	910
	liver	280												29	9.2	610
	heart+kidney+lung	330												31	6.6	650
	stomach+intestine	430												28	7.2	590
	egg entrails	160												8.8	2.17	190
	embryos													<	<	
	egg shells	61												2.9	2.17	61
Sterna hirudo	egg entrails	200												4.5	1.11	95
	embryos	230												8.4	1.21	180
	egg shells													<	<	

** = flesh and bones analysed in one

* = mean value

Table 2. Concentrations of gamma-emitting radionuclides in indicator samples taken from the sea area off the Olkiluoto Nuclear Power Plant in 2001.

Sample		Bq/kg dry wt.					Bq/kg	CF
Organism	Tissue	⁴⁰ K*	⁵⁴ Mn	⁶⁰ Co	¹³⁴ Cs	¹³⁷ Cs*	f.w. ¹³⁷ Cs*	(d.w.) ¹³⁷ Cs*
Phytoplankton		340		2.9 - 4.3		63	0.33	910
Zooplankton		570	0 - 0.47	2.0 - 11	0 - 0.53	73	0.81	1100
Periphyton		620	0 - 0.58	0 - 19	0 - 1.2	240	12	3500
Macroalgae								
<i>Cladophora glomerata</i>		1770		3.5		59	6.3	860
<i>Fucus vesiculosus</i>		760	0 - 0.47	1.7 - 5.8	0 - 0.47	50	9.2	720
Vascular plants								
<i>Ranunculus baudotii</i>		750	0.79	3.1		17	1.1	240
<i>Potamogeton perfoliatus</i>		350		1.7		7.7	0.7	110
<i>Potamogeton pectinatus</i>		630		1.8		25	3.2	360
<i>Myriophyllum spicatum</i>		600		8.5		24	2.0	350
Benthic animals								
<i>Saduria entomon</i>						24	8.6	350
<i>Mytilus edulis</i>		54		1.7		4.5	1.2	65
<i>Macoma balthica</i>		65		0.5		11	3.6	150
<i>Cardium edule</i>		50				2.5	0.91	36
Fish								
<i>Clupea harengus memb.</i>	whole**					48	11	690
	milt	350				28	5.8	400
	spawn	260				17	4.6	250
<i>Esox lucius</i>	fillets	570			0.54 - 1.2	125	26	1800
	liver	390				55	13	800
	entrails	270			0 - 0.91	100	24	1400
	bones	63				10.7	4.6	150
	spawn	290				34	11	490
<i>Abramis brama</i>	whole**				0 - 0.31	24	5.7	350
	spawn	180				9.2	2.4	130
<i>Rutilus rutilus</i>	whole**				0 - 0.06	33	7.6	480
<i>Perca fluviatilis</i>	whole**					144	38	2100
	liver	360				118	24	1800
	spawn	200				76	16	1100
Birds								
<i>Somateria mollissima</i>	muscle	330				13	2.7	190
	heart	400				12	2.1	170
	liver	330				8.5	2.0	120
	entrails	120				3.5	1.8	51
	egg entrails	110				2.0	0.86	29
	egg shells	27				0.63	0.43	9.1
<i>Mergus merganser</i>	muscle	380				23	6.7	330
	heart	300				19	5.3	280
	liver	310				27	7.9	390
	entrails	410				16	7.2	230
	bones	41				1.5	0.79	22
	feathers					3.0	3.0	43
<i>Cygnus olor</i>	egg white	330				17	2.0	250
	egg yolk	66				0.80	0.42	12
	egg shells	10				0.50	0.41	7.2
<i>Larus marinus</i>	muscle	320				13	4.3	190
	heart	240				12	4.0	170
	liver	310				16	5.2	230
	entrails	160				10	4.9	150
	fat					<	<	
	bones	26				1.1	0.55	16
	egg entrails	160				5.1	1.2	74
	egg shells	15				<	<	
<i>Larus argentatus</i>	egg entrails	160				2.0	0.49	29
	egg shells					<	<	

** = flesh and bones analysed in one

* = mean value

K-40

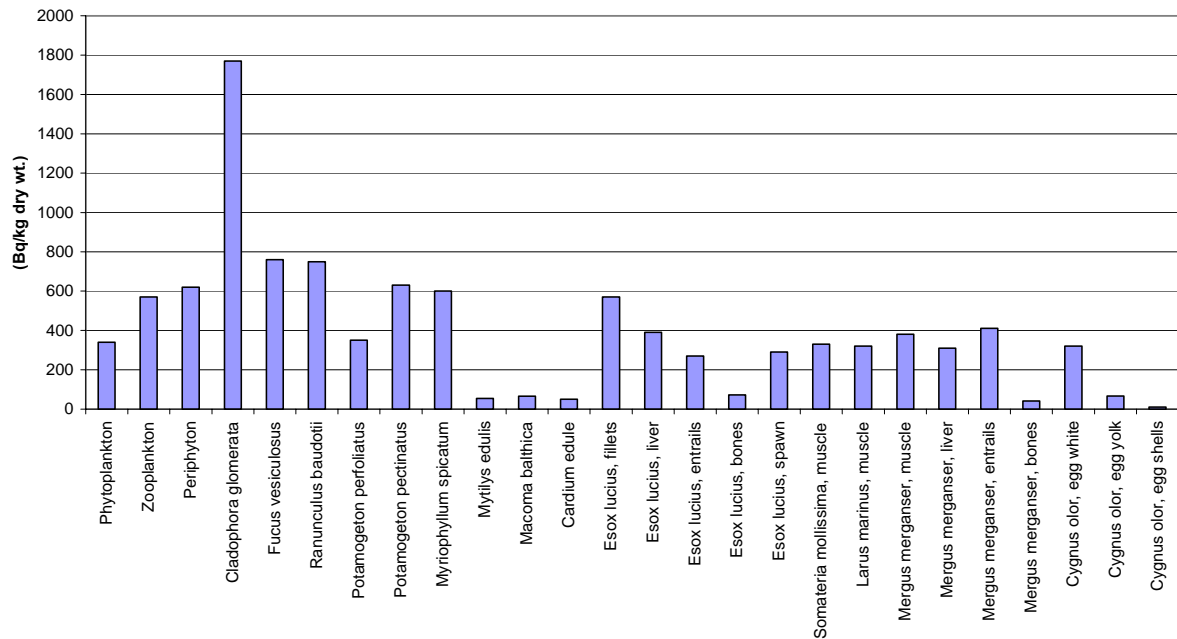


Figure 2. Mean values for ^{40}K (Bq kg^{-1} dry weight) in certain indicator samples in the sea area off Olkiluoto in 2001.

Co-60

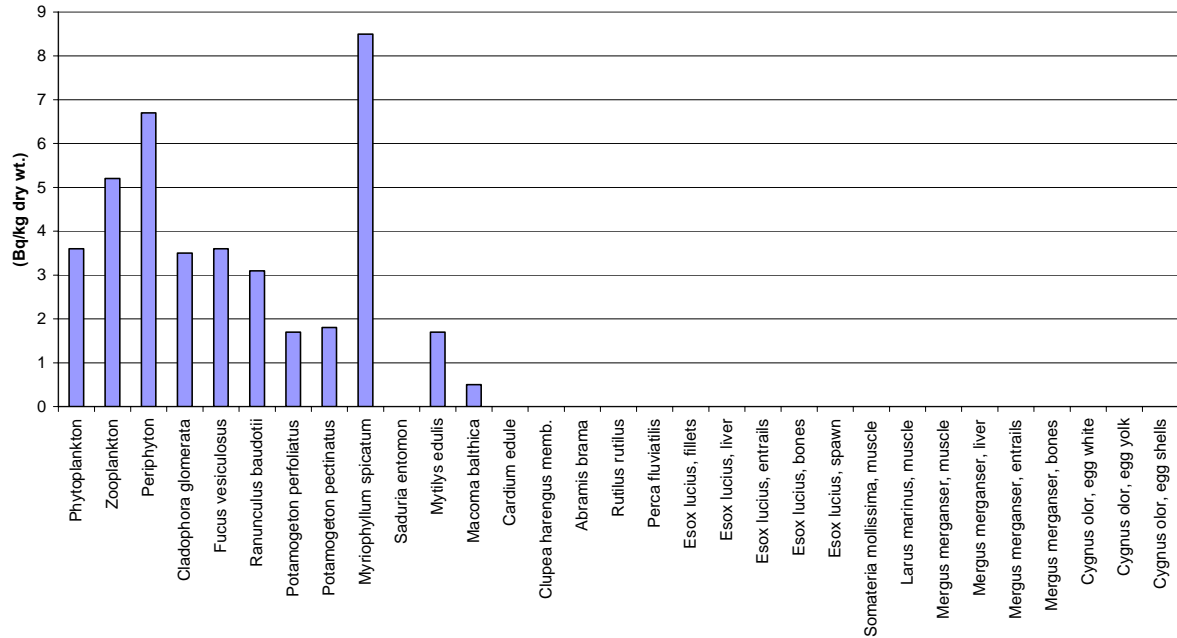


Figure 3. Mean values for ^{60}Co (Bq kg^{-1} dry weight) in certain indicator samples in the sea area off Olkiluoto in 2001.

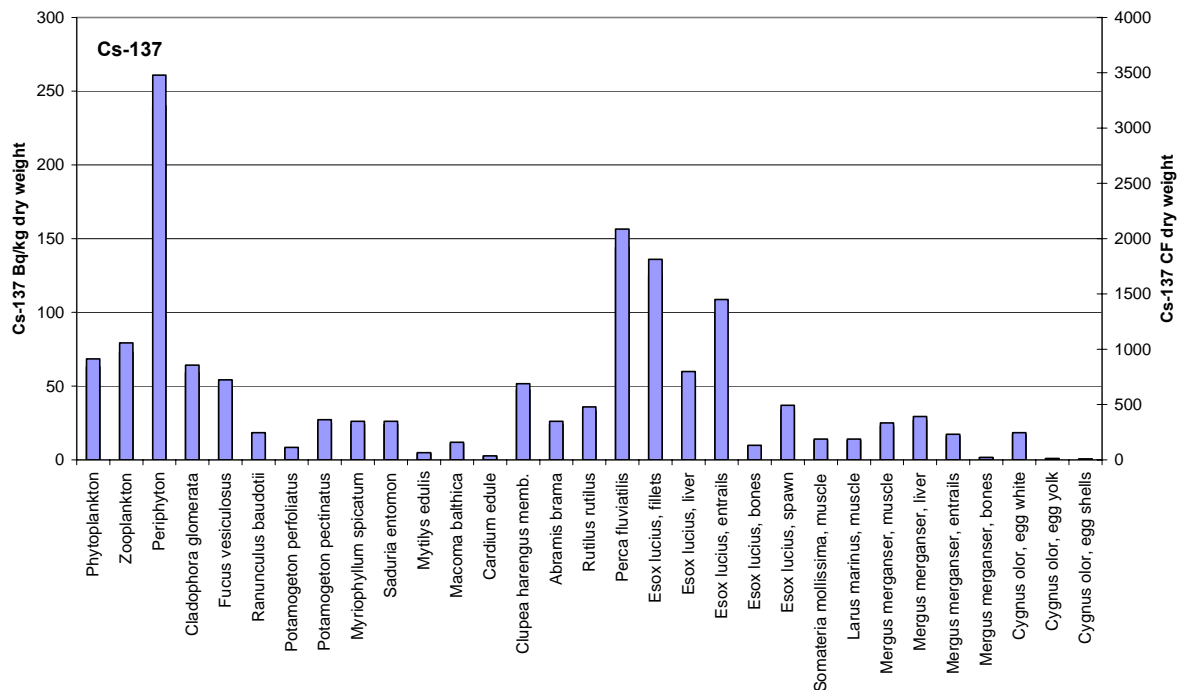


Figure 4. Mean values for ¹³⁷Cs (Bq kg⁻¹ dry weight) and mean concentration factors (on dry weight basis) in certain indicator samples in the sea area off Olkiluoto in 2001.

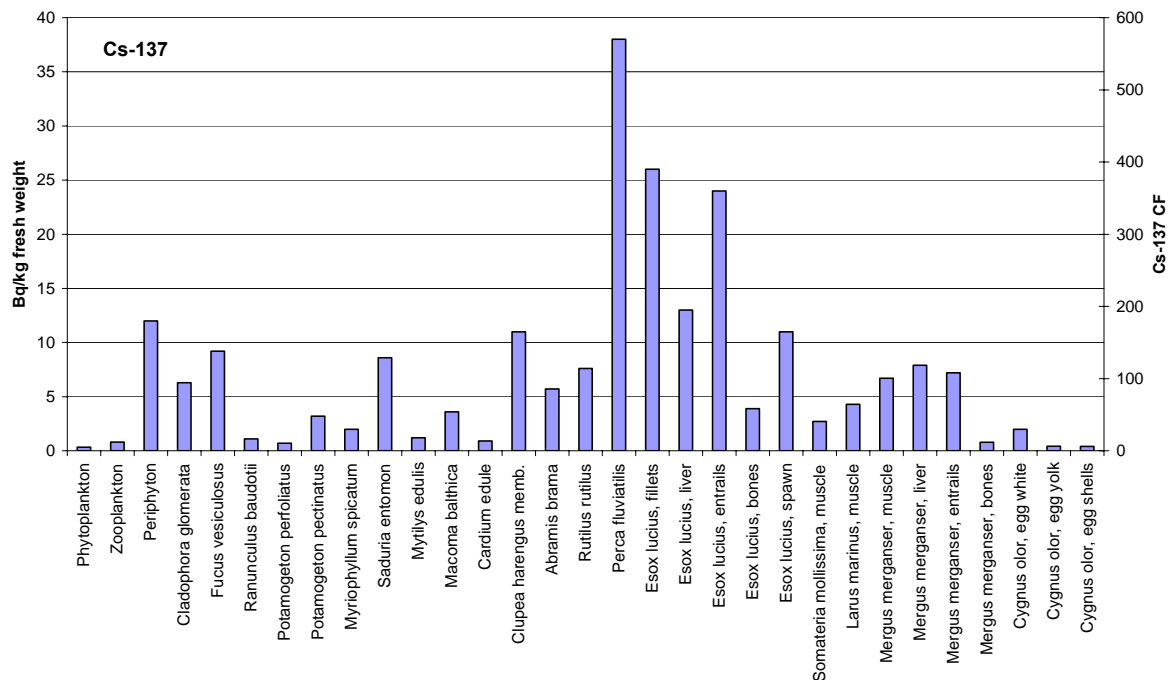


Figure 5. Mean values for ¹³⁷Cs (Bq kg⁻¹ fresh weight) and mean concentration factors (on fresh weight basis) in certain indicator samples in the sea area off Olkiluoto in 2001.

Cs-137 in aquatic organisms in the southern Lake Keuruselkä (Finland)

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

STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Introduction

The results of a study carried out in Lake Keuruselkä, in the Finnish Lake District, are reported. The aim of the study was to collect biota samples for the INDOFERN Project from an area that was rather highly contaminated (70 kBq m^{-2} of ^{137}Cs in 1986) with the Chernobyl fallout in Finland (Fig. 1). The samples were taken both from the terrestrial and aquatic environment in August 2003. The results of the terrestrial samples are presented elsewhere in this Proceedings publication. This paper is focused on the results of the aquatic environment. The samples were taken from a relatively small area surrounding the island of Iso Riihisaari in the southern part of the Keuruselkä water course. In total 15 samples of aquatic plants, 6 samples of aquatic animals, 1 water sample and 2 sediment cores were taken.

Study area

Lake Keuruselkä is part of the Kokemäenjoki River drainage basin. The area of the lake is 119 km^2 and the length is about 30 km but the mean depth is only 6.4 m. Humic substances and local sewage weaken the water quality in the northern parts of Lake Keuruselkä. The colour number of water, the sight depth and the average concentration of total phosphorus in water are 90 mg Pt l^{-1} , 1.8 m and $15 \text{ } \mu\text{g l}^{-1}$, respectively. The central part of the lake is in almost natural condition. The settlement in the town of Mänttä has had a slight impact on the waters of the southern part of the lake, where the study area is located. However, the quality of water is good in the Lake Keuruselkä according to the usability classification of the Finnish Environment Institute.

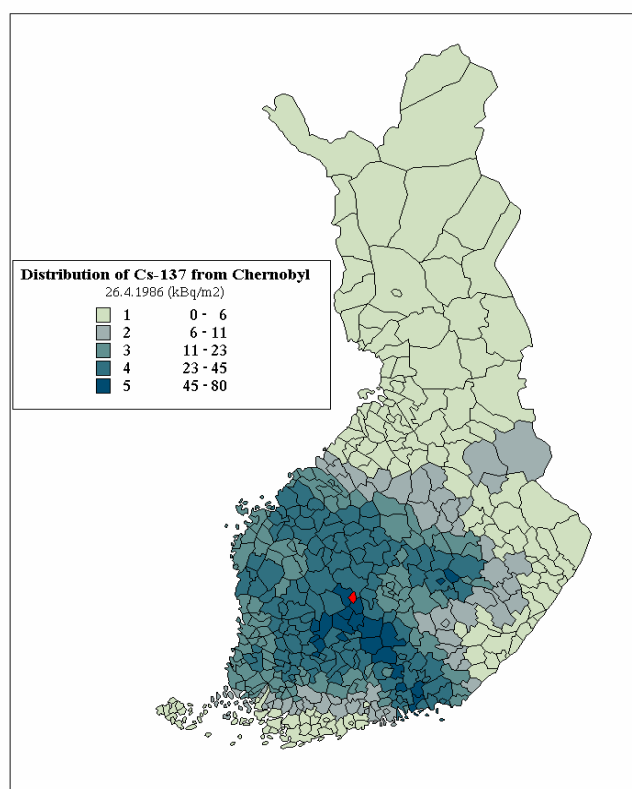
Material and methods

The samples of *Equisetum*, *Eleocharis* and *Typha* were collected by hand (or using scissors) from the geolittoral or shallow water. The plant samples (*Nymphaea*, *Nuphar*, *Myriophyllum*, *Potamogeton* and *Phragmites*) from sublittoral and the clam (*Anodonta*) sample were collected by skin diving. All the plant samples were taken by cutting the stems from the base. The rootstocks of *Nymphaea*, *Nuphar* and *Phragmites*, which are very strong and fast stuck in the mud, were tugged as whole from the bottom (Fig 2). The *Anodonta* sample consisted of 54 animals (58-98 mm, 10-59 g), and the total fresh weight of the sample was 2010 g. The perch and roach samples consisted of small fish caught with hook and line.

The activity concentrations of gamma-emitting nuclides were measured by low-background, high-resolution spectrometer systems. The relative efficiencies of the HPGe detectors range from 21% to 78% and the energy resolution from 1.8 keV to 2.1 keV at 1.33 MeV. The measurements were performed in 15-cm thick cylindrical background shields lined with cadmium and copper in the inside. The spectra were analysed using the GAMMA-99-computer code developed at STUK especially for environmental sample measurements.

Results and discussion

In August 2003, the activity concentration of ^{137}Cs in the surface water of the southern Lake Keurusselkä was 49 Bq m^{-3} , whereas it was 310 Bq m^{-3} in 1988, two year after the Chernobyl accident (Ilus et al., 1993). In the relatively shallow area surrounding the island of Iso Riihisaari, the total amount of ^{137}Cs in sediments was $32\text{--}37 \text{ kBq m}^{-2}$ in 2003, but in a deeper basin close to this area the total amount of ^{137}Cs was 130 kBq m^{-2} in 1990 (Ilus and Saxén, 2005). The peak concentrations were now at the depth of 2–4 cm (Fig. 3) or 4–6 cm indicating a relatively slow sedimentation rate. The concentration of ^{137}Cs in the surface layers of the sediment (0–8 cm), where most of the benthic animals live, was $1\ 100\text{--}4\ 500 \text{ Bq kg}^{-1}$ dry wt.



Ref. Arvela H, Markkanen M, Lemmelä H, Radiation Protection Dosimetry 1990;32:177-184

Figure 1. The five Deposition Zones of Chernobyl-derived ^{137}Cs in Finland (Arvela et al., 1987) and the location of Mänttä (red).



Figure 2. Sampling rootstocks of water lilies in Mänttä.

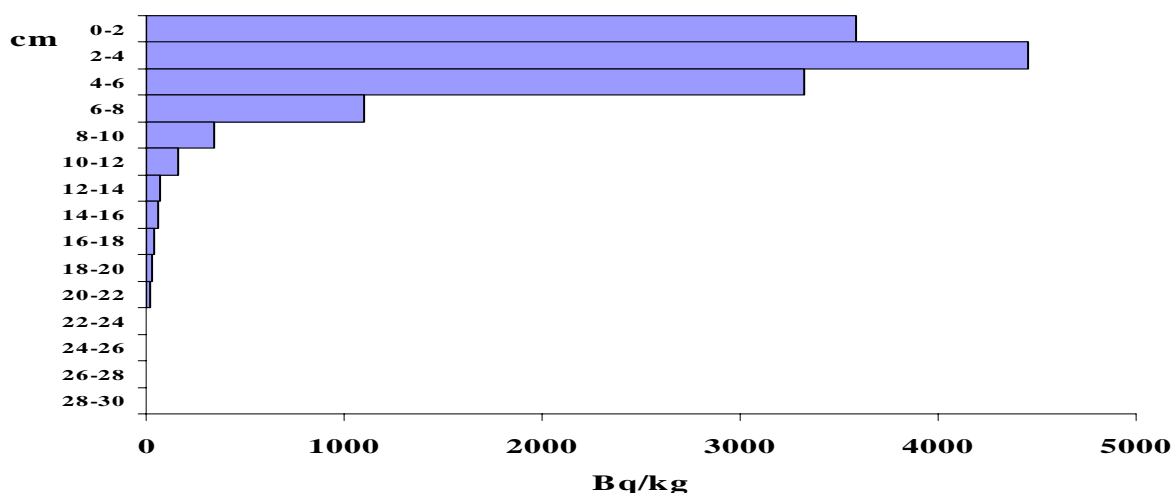


Figure 3. Vertical distribution of ^{137}Cs (Bq kg^{-1} dry weight) in a sediment core taken at a depth of 3.7 m east of Iso Riihisaari.

The clearly highest activity concentration and concentration factor of ^{137}Cs was found in one sample of Water horsetail (*Equisetum fluviatile*), $1\,430\text{ Bq kg}^{-1}$ dry wt; CF 29 200, whereas in another sample of the same species the concentration was only 174 Bq kg^{-1} dry wt (Table 1, Fig. 4). In addition, the Water lily (*Nymphaea candida*), Spiked water millfoil (*Myriophyllum spicatum*), Broad-leaved pondweed (*Potamogeton natans*) and Yellow water lily (*Nuphar lutea*) seemed to be good indicators for ^{137}Cs . The stems and leaves and the rootstocks of water lilies (*Nymphaea candida* and *Nuphar lutea*) seemed to accumulate quite equally ^{137}Cs . The tall freshwater clam (*Anodonta sp.*) seemed to be a modest accumulator of ^{137}Cs . On the contrary to our results from the coastal areas of the Baltic Sea, many aquatic plants demonstrated in fresh water similar accumulation capacity of ^{137}Cs as fish (perch and roach), while in the sea the uptake of ^{137}Cs in fish seemed to be more efficient than in aquatic plants.

Besides ^{137}Cs , the samples contained also naturally occurring ^{40}K and ^7Be and traces of the Chernobyl-derived ^{134}Cs .

The concentration factors of ^{137}Cs were much higher in the Mänttä area than in the coastal areas at Loviisa and Olkiluoto. This is due to the difference between the uptake of ^{137}Cs and certain other radionuclides in fresh water and brackish or saline water. The uptake is much more efficient in freshwater. The difference in fish was almost 10-20 fold and even larger in aquatic plants. The concentration factors of ^{137}Cs for *Myriophyllum* were 350 and 510 in the brackish-water areas of Olkiluoto and Loviisa while it was 12 200 in fresh water at Mänttä. The CF values for perch and roach were 1900-2100 and 480-530 in Loviisa and Olkiluoto, but 17200 and 12900 in Mänttä. In conclusion, different concentration factors are needed for marine, brackish water and freshwater ecosystems.

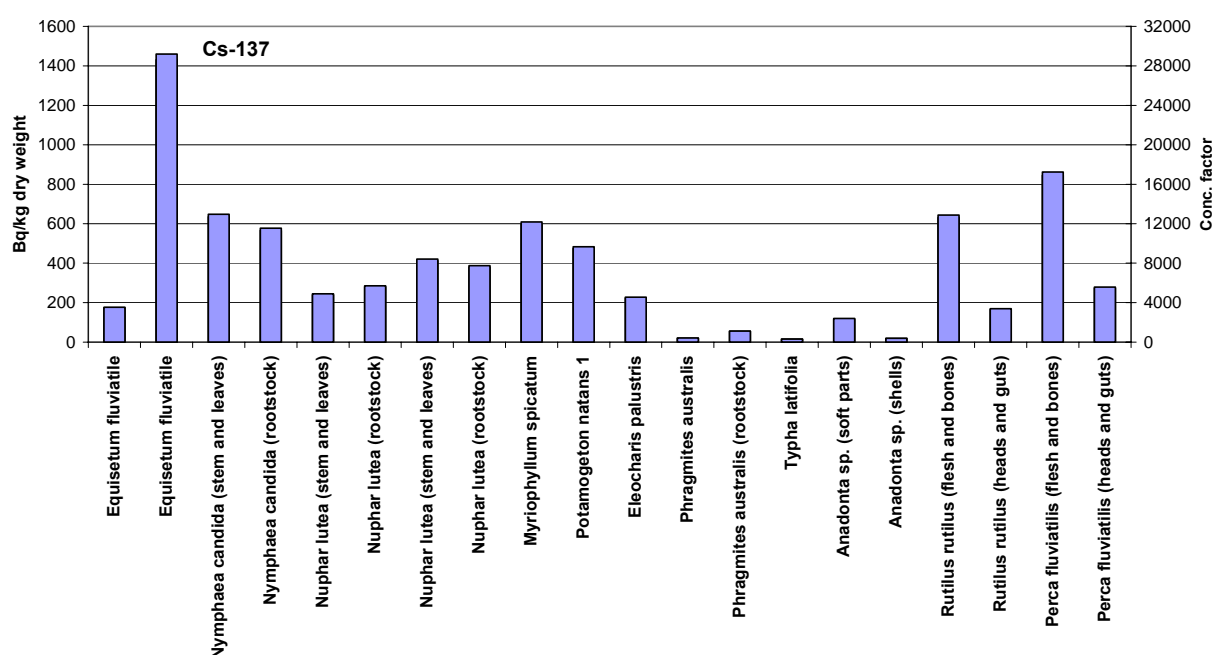


Figure 4. Activity concentrations (Bq kg^{-1} dry weight) and concentration factors of ^{137}Cs (Bq kg^{-1} d.w. / Bq kg^{-1} water) in aquatic indicator organisms collected from Lake Keuruselkä (Mänttä) in August 2003.

Table 1. Activity concentrations and concentration factors of ^{137}Cs in aquatic samples collected from Lake Keuruselkä (Mänttä) in August 2003. Uncertainties include statistical and calibration uncertainty

Sample	¹³⁷ Cs concentration				CF
	Bq kg ⁻¹ f.w.	unc. 1σ	Bq kg ⁻¹ d.w.	unc. 1σ	Bq kg ⁻¹ d.w. / Bq kg ⁻¹ water
Aquatic plants					
<i>Equisetum fluviatile</i> 1	38	2	174	9	3 600
<i>Equisetum fluviatile</i> 2	320	16	1 430	72	29 200
<i>Nymphaea candida</i> (stems and leaves)	62	3	635	25	13 000
<i>Nymphaea candida</i> (rootstock)	71	3	566	23	11 600
<i>Nuphar lutea</i> 1 (stems and leaves)	27	1	240	12	4 900
<i>Nuphar lutea</i> 1 (rootstock)	31	1	280	11	5 700
<i>Nuphar lutea</i> 2 (stems and leaves)	52	2	413	17	8 400
<i>Nuphar lutea</i> 2 (rootstock)	27	1	380	15	7 800
<i>Myriophyllum spicatum</i>	410	16	597	24	12 200
<i>Potamogeton natans</i> 1	47	2	474	19	9 700
<i>Potamogeton natans</i> 2 (27.6.2004)	27	1	267	11	5 400
<i>Eleocharis palustris</i>	35	2	224	11	4 600
<i>Phragmites australis</i> (stems and leaves)	8	1	20	1	410
<i>Phragmites australis</i> (rootstock)	10	1	55	2	1 100
<i>Typha latifolia</i>	3	1	15.3	0.8	310
Mussels					
<i>Anodonta</i> sp. (soft parts)	7	1	117	5	2 400
<i>Anodonta</i> sp. (shells)	16	1	19	1	390
Fish					
<i>Rutilus rutilus</i> (flesh and bones)	110	4	631	19	12 900
<i>Rutilus rutilus</i> (heads and guts)	36	2	166	5	3 400
<i>Perca fluviatilis</i> (flesh and bones)	130	7	845	42	17 200
<i>Perca fluviatilis</i> (heads and guts)	85	4	274	11	5 600
Surface water	0.049	0.003			
Surface sediment layers (0-8 cm)			1 100 - 4 500		
Total amount in sediments (Bq m ⁻²)			31 800 - 36 700		

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Behavior of Sr-90 and transuranic elements in three areas in Finland

Tarja K. Ikäheimonen, Vesa-Pekka Vartti and Erkki Ilus

STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Introduction

This study was included in the INDOFERN Project with the aim of evaluating accumulation of radionuclides by different organisms. The study was carried out in three areas (both terrestrial and aquatic): in the Mänttä area in Central Finland and in the environs of the Loviisa and Olkiluoto Nuclear Power Plants (Fig. 1). Detailed descriptions of the areas, as well as of sampling and samples are given in the papers of Ilus et al. (2005, these Proceedings).

Methods used in the strontium and transuranic analyses

All the samples were dried before the gamma spectrometric measurements. For dried samples, three different pretreatment methods were used: ashing (only for Sr analysis), wet ashing with strong nitric and hydrochloric acids by Tecator wet-ashing equipment or acid leaching with microwave burning in the oven.

Separation of Sr-90 was performed by an ion chromatographic extraction method with Eichrom Sr-resin. Activity of Sr-90 was measured with its daughter nuclide Y-90 by a low background proportional counter. A detailed description of the method is given elsewhere (Ikäheimonen and Vartti, 1997).

Plutonium was separated with an anion exchange method, the alpha activity of Pu-238 and Pu-239,240 was measured by alphaspectrometry and the beta activity (Pu-241) with a liquid scintillation spectrometry (Ikäheimonen, 2003). The separation of americium was done after the plutonium separation with two different methods: with several ion exchanges and liquid extractions or with a new method using Eichroms different resins (Ikäheimonen 2003). Activity of Am-241 was measured alphaspectrometrically.

Results and discussion

Vertical distributions of Sr-90, Pu-239,240 and Am-241 (only in Mänttä) in soil profiles in the three areas are presented in Figs 2 - 4. The total amounts (Bq/m^2) of the nuclides in the whole soil cores to the depth of 15 cm (Sr-90 and Pu-239,240) or 10 cm (Am-241) are presented in Fig. 5. The largest amounts of the nuclides were found in Loviisa and the smallest in Olkiluoto. The differences were due to the different amounts of Chernobyl fallout in the areas, but also to different soil types: a part of old global fallout was probably trickled away horizontally or vertically in Olkiluoto.

The concentrations of Sr-90, Pu-238,240 and Am-241 in various species of mushrooms, vascular plants or parts of the plants and berries are presented in Tables 1-3. The highest Sr-90 concentrations were found in *Ebilobium angustifolium*, being 70 - 90 Bq/kg d.w., and *Empetrum nigrum*, 15 - 60 Bq/kg d.w. Concentrations of more than 10 Bq/kg d.w. were also detected in leaves of birch (*Betula pendula*), in berries of *Empetrum nigrum* and in ferns (*Dryopteris carthusiana*, *Dryopteris expansa*, *Polypodium vulgare*). The Sr-90 concentrations in mushrooms were less than 10 Bq/kg d.w. and varied considerably from one species to another.

The concentrations of Pu-239,240 were below the detection limits in mushrooms and berries. Detectable amounts of Pu-239,240 were found in ferns. In certain twigs and leaves, and in *Scirpus palustris*, the concentrations were low, less than 0.1 Bq/kg d.w. Am-241 was detected in ferns, but also in a *Cantharellus tubaeformis* sample and in *Calluna vulgaris*, in which the Pu-239,240 concentrations were below the detection limits.

Aggregated transfer factors (T_{agg}) of Sr-90 for mushrooms and other plants and those of Pu-239,240 for plants are presented in Figs 6 - 8. Vascular plants are better indicators of Sr-90 than mushrooms. *Empetrum nigrum* and *Ebilobium angustifolium* accumulate best of Sr-90, the T_{agg} being 0.16 m²/kg. Aggregated transfer factors of Pu-239,240 were about two decades lower, being about 0.001 m²/kg for *Scirpus palustris* (maximum).

The mean concentrations of Sr-90 in fresh and sea water were 7.3 and 12 Bq/m³, respectively, and those of Pu-239,240 and Am-241 were 0.03 and 0.003 Bq/m³, and 0.008 and 0.002 (or less) Bq/m³, respectively. The concentrations of Sr-90, Pu-239,240 and Am-241 in biota samples in fresh water (Mänttä) and marine environments (Loviisa and Olkiluoto) are presented in Tables 4 - 5. The highest concentrations of Sr-90 in fresh water environment were detected in shells and flesh of freshwater clam, *Anodonta sp.*, and in marine environment in *Saduria entomon* and *Macoma balthica*. In *Anodonta sp.* (both shells and flesh), also Pu-239,240 and Am-241 were detected. Pu-239,240 was detectable in almost all the marine samples.

Concentration factors (CF) of Sr-90 and Pu-239,240 for biota in fresh water and marine environments are presented in Figs 9 -10. The CFs of Pu-239,240 were roughly at the same level or greater than those of Sr-90, especially in the marine environment. Best indicator organism for Sr in the fresh water environment was *Anodonta sp.*, and then *Nuphar lutea* (CFs 10³ - 10⁴); and *Macoma balthica* and *Fucus vesiculosus* in the marine environment. Roots of *Nymphaea candida* and flesh of *Anodonta sp.* accumulated best Pu-239,240 in fresh water environment; the CFs were 10³ - 10⁴. The CFs of Pu-239,240 were greater in the marine environment compared to those in fresh water environment. *Phytoplankton* and *periphyton* accumulate most efficiently Pu-239,240 in the marine environment, the CFs being about 10⁵. The behavior of plutonium and americium were different; americium was generally more bioavailable.

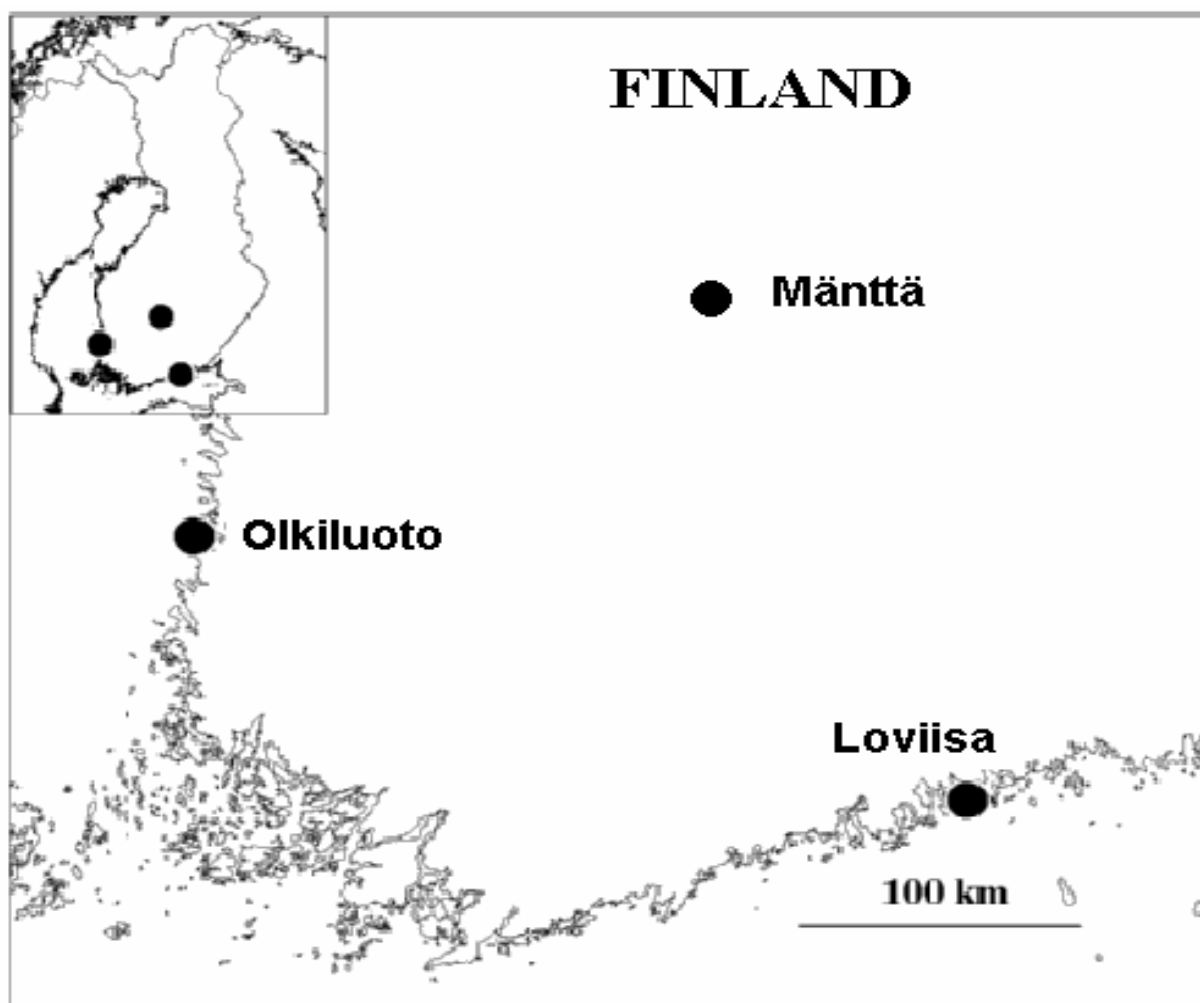


Figure 1. Location of the sampling areas.

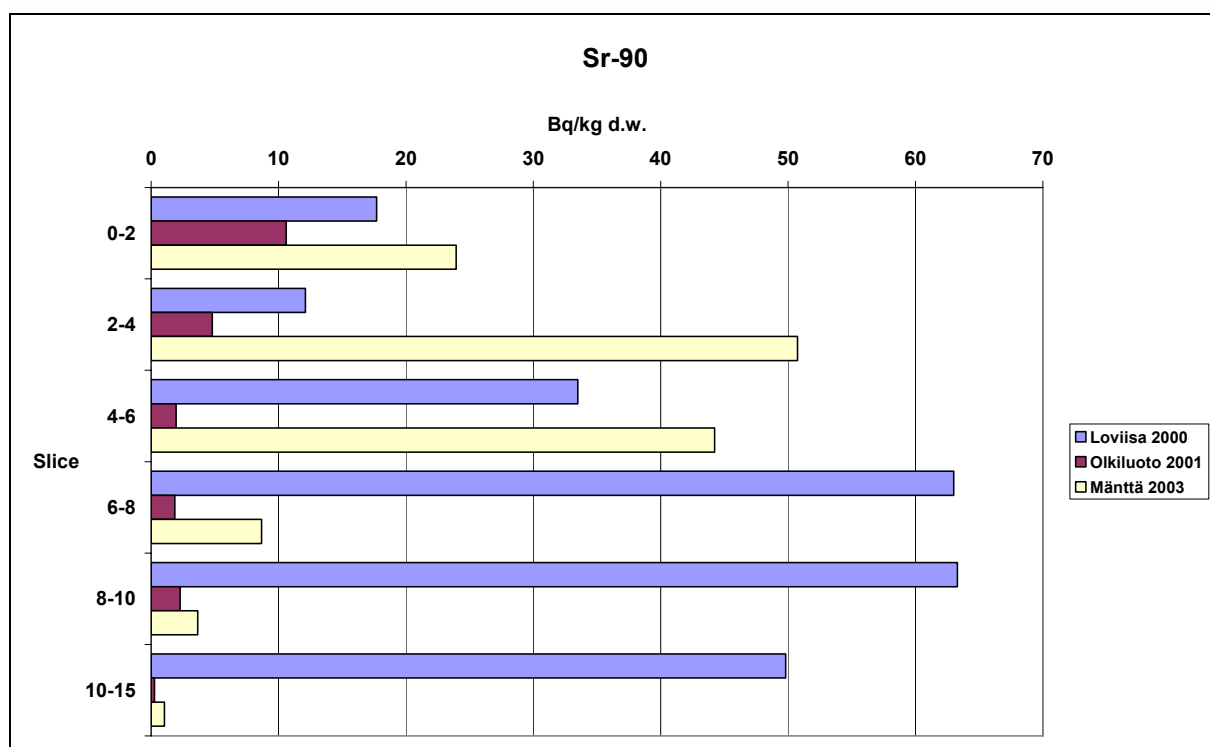


Figure 2. Soil profiles of Sr-90 (Bq/kg d.w.) in Loviisa, Olkiluoto and Mänttä.

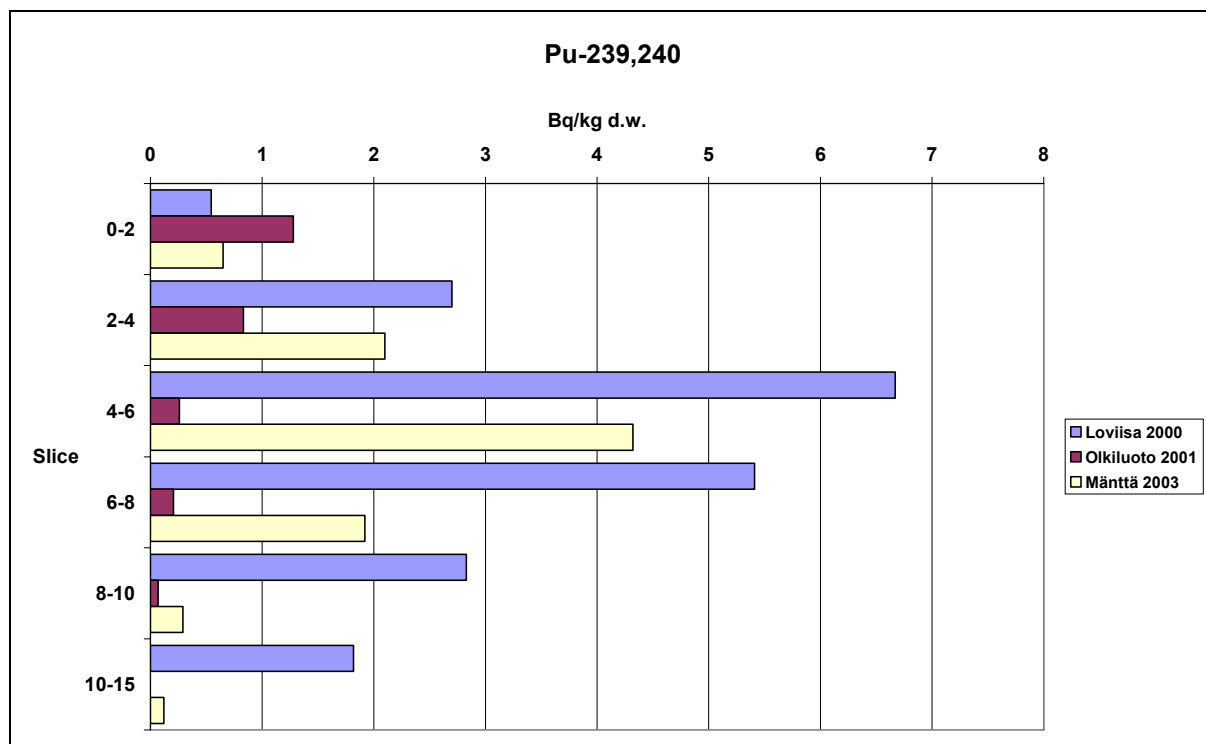


Figure 3. Soil profiles of Pu-239,240 (Bq/kg d.w.) in Loviisa, Olkiluoto and Mänttä.

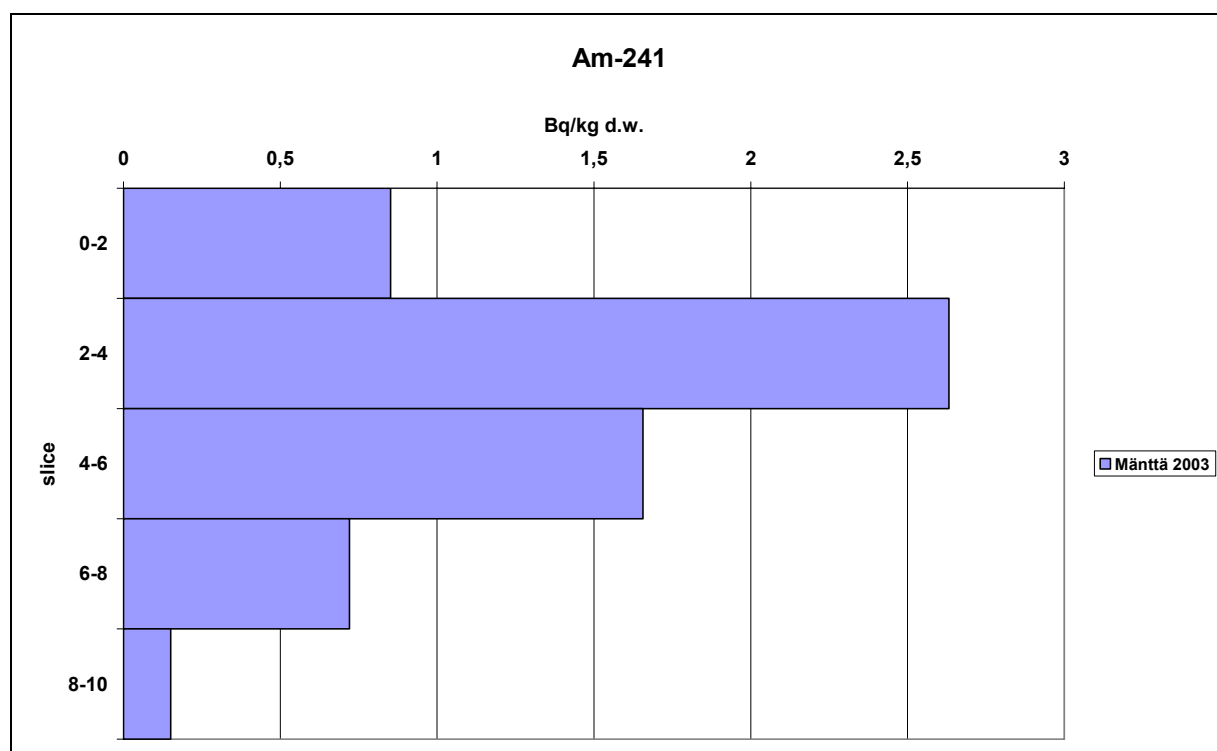


Figure 4. Soil profile of Am-241 (Bq/kg d.w.) in Mänttä

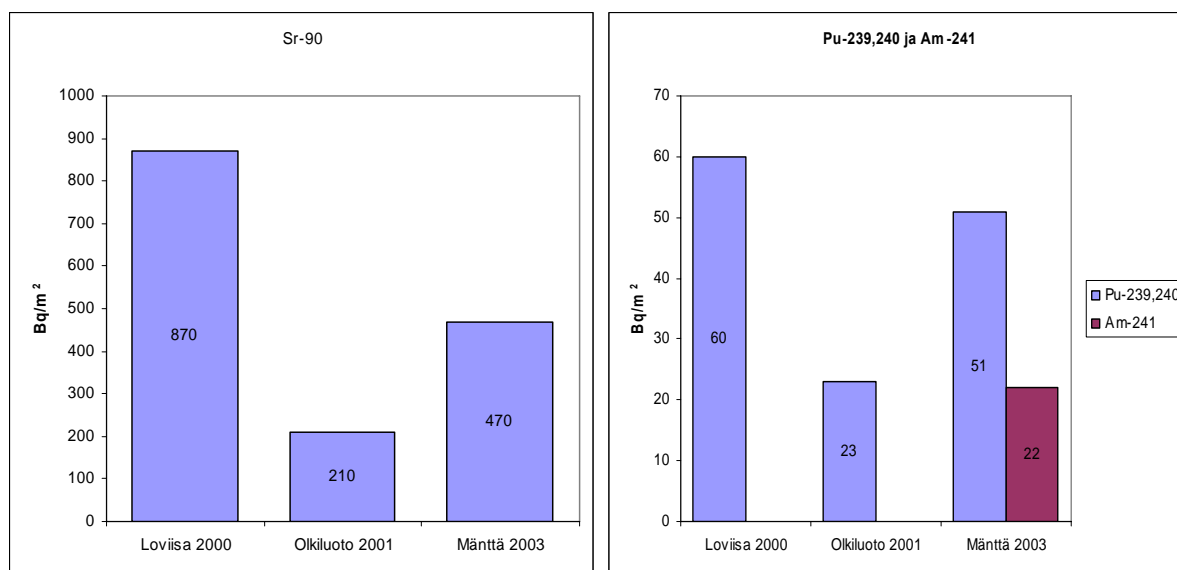


Figure 5. Total amounts of Sr-90, Pu-239,240 and Am-241 in soil cores in Loviisa, Olkiluoto and Mänttä.

Table 1. Activity concentrations of Sr-90, Pu-239,240 and Am-241 (Bq/kg d.w.) in mushrooms.

Species	Area	Bq/kg d.w.		
		Sr-90	Pu-239,240	Am-241
<i>Lactarius sp.</i>	Loviisa	0.2	<0.016	
	Olkiluoto	0.8		
<i>Leccinum versipelle</i>	Loviisa	4.9	<0.02	
<i>Hydnum rufescens</i>	Loviisa	0.8		
<i>Rozites caperata</i>	Loviisa	0.3	<0.05	
<i>Cantharellus tubaeformis</i>	Olkiluoto	1.3	<0.021	0.11 (46)
<i>Boletus edulis</i>	Mänttä	0.3		
<i>Hygrophorus camarophyllus</i>	Mänttä	5.4		
<i>Russula sp.</i>	Mänttä	0.5-1.4		
<i>Cortinarius traganus</i>	Mänttä	4.7		

Table 2. Activity concentrations of Sr-90, Pu-239,240 and Am-241 (Bq/kg d.w.) in other terrestrial plant samples.

Species	Area	Sr-90	Pu-239,240	Am-241
Lichen				
<i>Cladina spp.</i>	Loviisa*	1.0 (6)	0.017 (32)	-
	Olkiluoto	2.5 (6)	<0.02	-
Moss				
<i>Polytrichum sp.</i>	Loviisa	1.7 (6)	-	-
	Olkiluoto	3.9 (6)	-	-
Leaves				
<i>Betula pendula</i>	Loviisa	-	-	<0.04
	Olkiluoto	-	0.016 (39)	<0.01
	Mänttä	46 (5)	-	-
Needles				
<i>Pinus sylvestris</i>	Mänttä	5.2 (5)	-	-
<i>Picea abies</i>	Mänttä	31 (5)	-	-
Twigs and leaves				
<i>Vaccinium myrtillus</i>	Loviisa	11 (5)	-	-
	Olkiluoto	22 (5)	0.017 (36)	-
	Mänttä	27 (5)	<0.008	<0.02
<i>Empetrum nigrum</i>	Olkiluoto	61 (5)	0.01 (40)	<0.03
	Mänttä	16 (5)	-	-
Ferns				
<i>Dryopteris expansa</i>	Olkiluoto	26 (5)	-	0.05 (22)
<i>Dryopteris carthusiana</i>	Mänttä	30 - 47 (5)	0.01 (42)	0.10 (14)
<i>Polypodium vulgare</i>	Loviisa	35 (4)	0.01 (50)	-
	Olkiluoto	32 (5)	0.007 (45)	-
<i>Ebilobium angustifolium</i>	Mänttä	68 - 89 (5)	<0.009	<0.02
<i>Calluna vulgaris</i>	Mänttä	14(5)	<0.008	0.03 (31)
<i>Scirpus palustris</i>	Mänttä	4.9 (5)	0.05	-
<i>Ledum palustre</i>	Mänttä	7.1 (5)	<0.002	-

Table 3. Activity concentrations of Sr-90, Pu-239,240 and Am-241 (Bq/kg d.w.) in berries.

Species	Area	Sr-90	Pu-239,240	Am-241
<i>Vaccinium myrtillus</i>	Loviisa	2.2 (10)	-	-
	Olkiluoto	1.5 (6)	<0.01	-
<i>Empetrum nigrum</i>	Olkiluoto	13.6 (6)	<0.001	<0.02
<i>Vaccinium vitis-idaea</i>	Loviisa	-	-	-
	Olkiluoto	0.86 (6)	-	-

Table 4. Activity concentrations of Sr-90, Pu-239,240 and Am-241 (Bq/kg d.w.) in certain fresh water organisms in Mänttä.

Species	Sr-90	Pu-239,240	Am-241
<i>Equisetum fluviatile</i>	9.9 (5)	0.018 (40)	-
<i>Phragmites australis</i>	4.71 (6)	< 0.004	-
<i>Phragmites australis</i> , roots	2.85 (7)	< 0.05	-
<i>Nymphaea candida</i>	7.7 (5)	0.017 (36)	-
<i>Nymphaea candida</i> , roots	6.6 (5)	0.17 (12)	-
<i>Nuphar lutea</i>	17.6 (5)	< 0.016	-
<i>Anodonta sp.</i> , flesh	56.1 (5)	0.06 (15)	0.06 (17)
<i>Anodonta sp.</i> , shell	189 (5)	0.016 (33)	0.02 (28)
<i>Perca fluviatilis</i> , flesh and bones		<0.008	< 0.05

Table 5. Activity concentrations of Sr-90 and Pu-239,240 (Bq/kg d.w.) in certain brackish-water organisms in Loviisa and Olkiluoto.

Species	Sr-90	Pu-239,240
<i>Phytoplankton</i>		
Loviisa	-	0.04 (18)
Olkiluoto	-	0.3 (7)
<i>Myriophyllum spicatum</i>		
Loviisa	-	0.05 (15)
Olkiluoto	-	0.04 (16)
<i>Periphyton</i>		
Loviisa	-	0.20 (9)
Olkiluoto	-	0.21 (9)
<i>Potamogeton pectinatus</i>		
Loviisa	-	< 0.03
Olkiluoto	-	0.10 (12)
<i>Saduria entomon</i>		
Loviisa	13 - 19 (6)	0.02 (24)
<i>Fucus vesiculosus</i>		
Loviisa	8.5 - 9.6 (6)	0.06 - 0.08
Olkiluoto	7.9 - 16.5 (6)	0.06 - 0.09

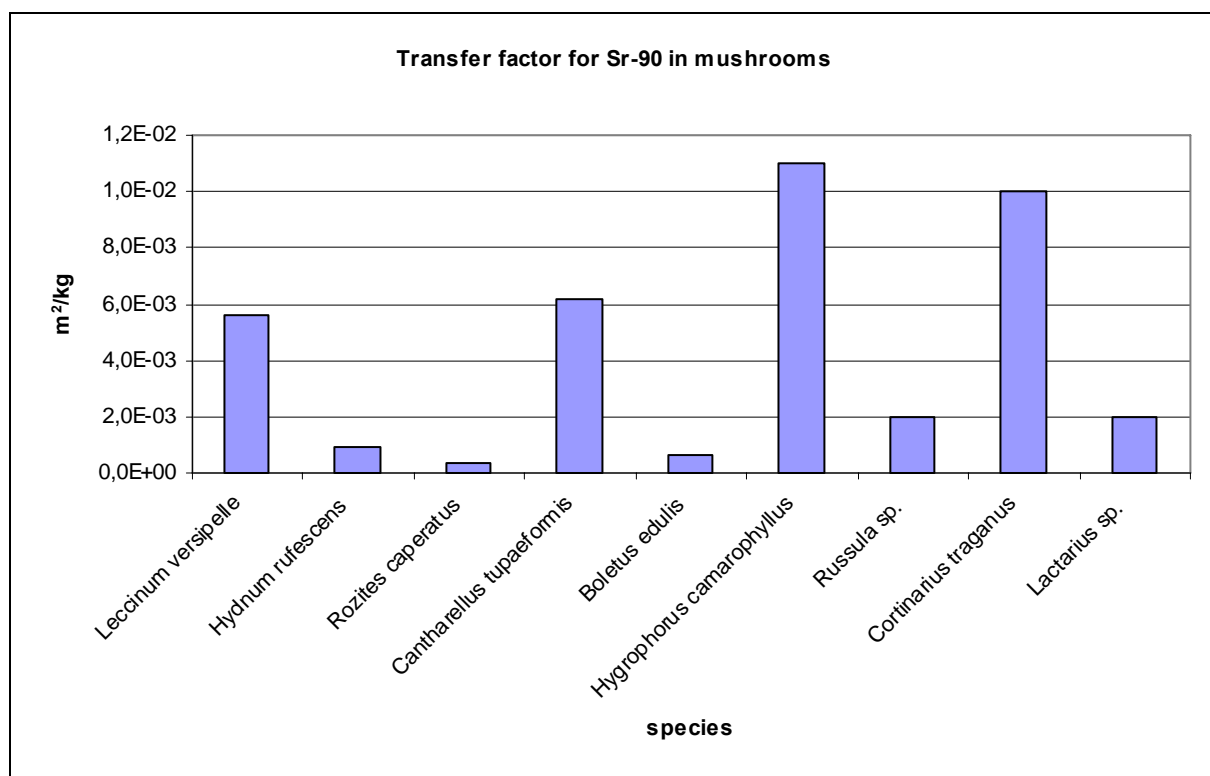


Figure 6. Aggregated transfer factors of Sr-90 for certain mushrooms.

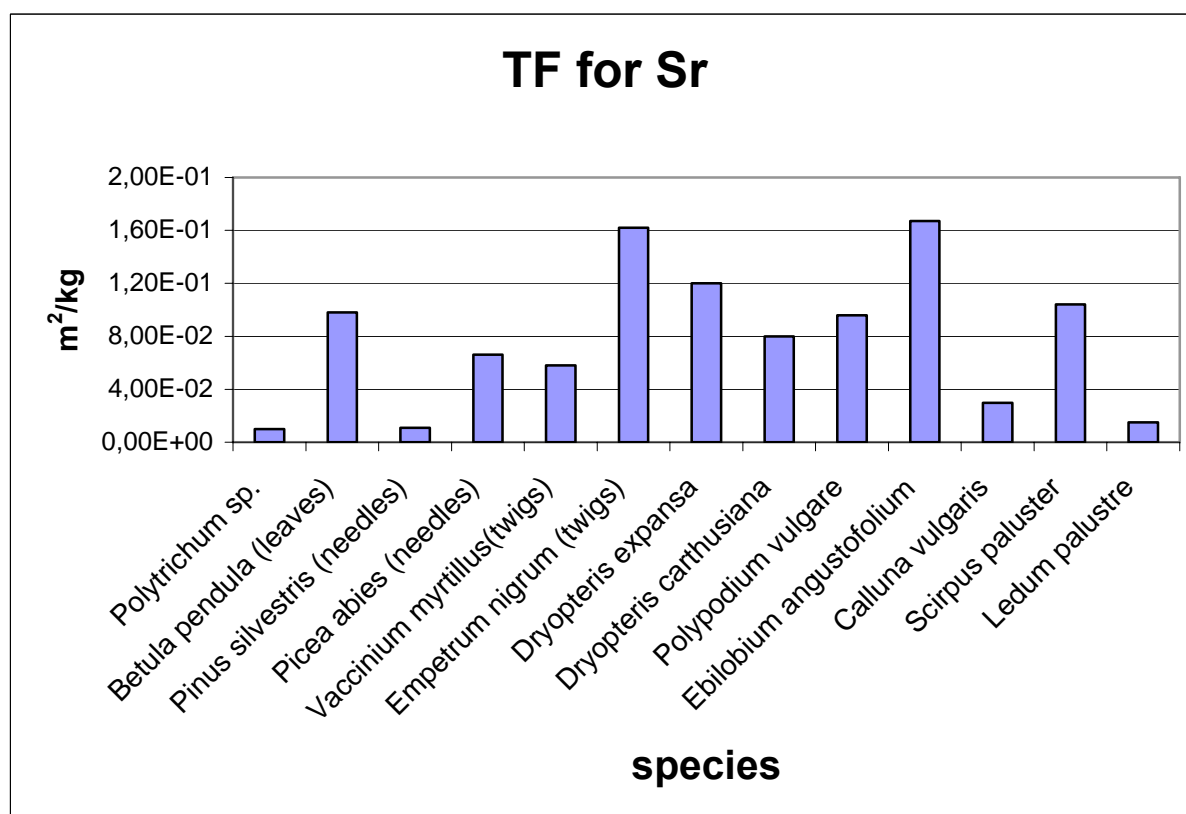


Figure 7. Aggregated transfer factors of Sr-90 for certain terrestrial plants except mushrooms.

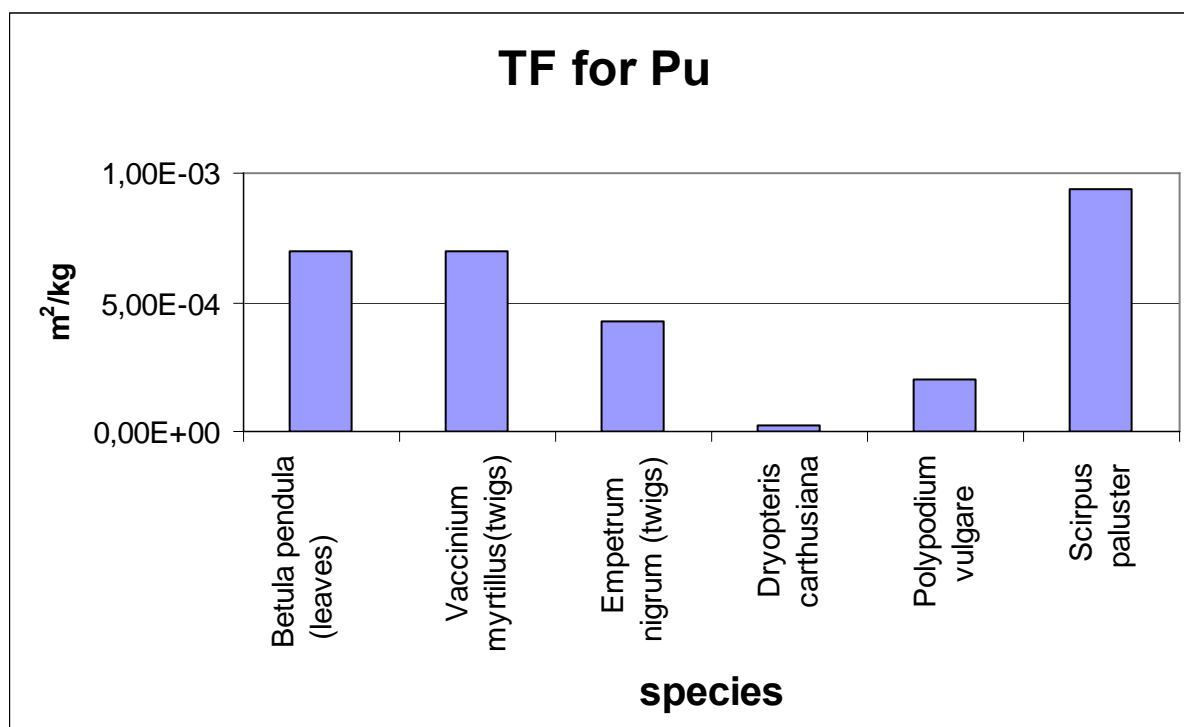


Figure 8. Aggregated transfer factors of Pu-239,240 for certain plant species.

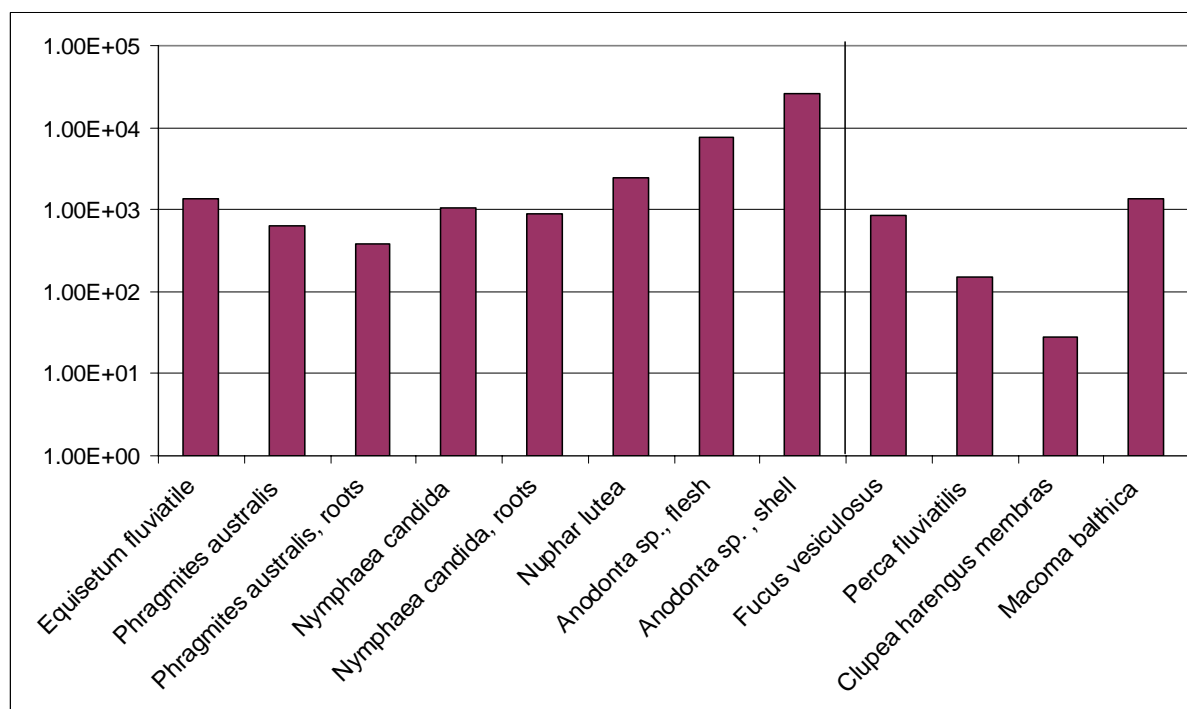


Figure 9. Concentration factors of Sr-90 for certain fresh water and marine organisms.

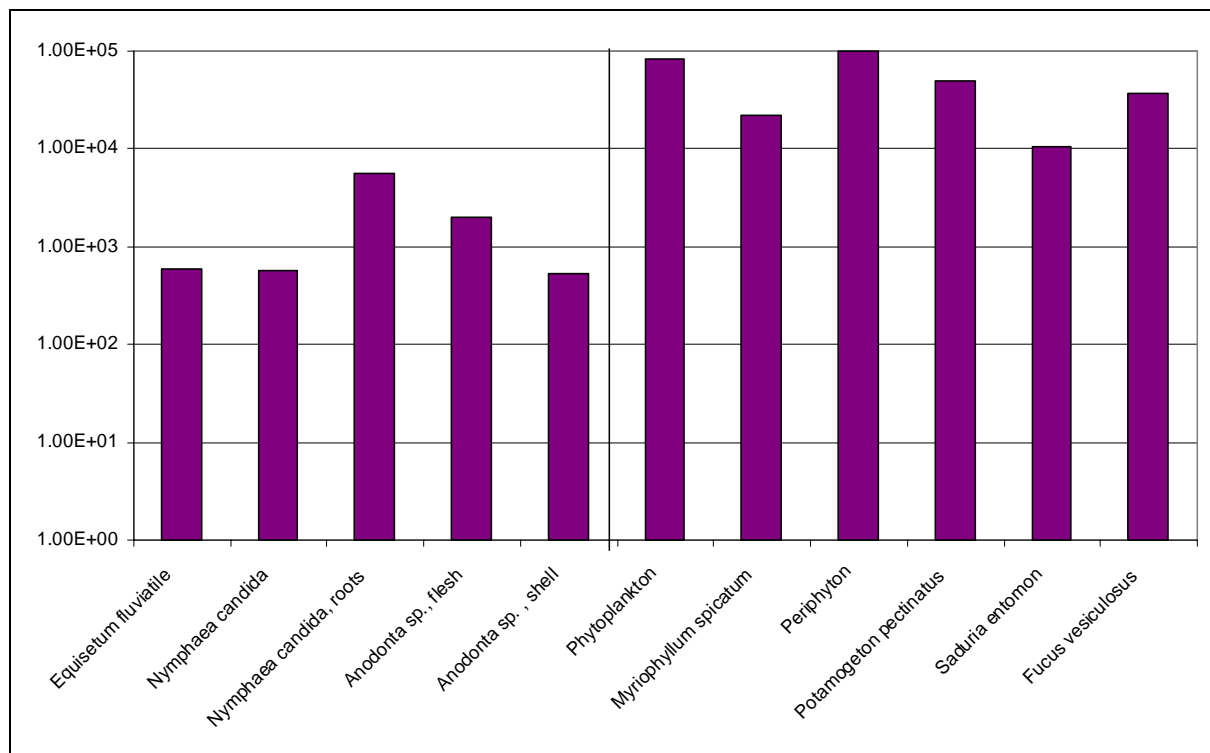


Figure 10. Concentration factors of Pu-239,240 for certain fresh water and marine organisms.

Radionuclides in Seal Flesh and Liver

Sven P. Nielsen

Risø National Laboratory, DK-4000, Roskilde, Denmark

Background

Concentrations of naturally occurring ^{210}Po in seals are relatively high due to their high consumption of fish, which contain ^{210}Po of typically 0.5-1 Bq/kg fw. This investigation was carried out in order to

- Determine concentrations of radionuclides including ^{210}Po in seal flesh and liver, and
- Determine the variability within the organs of same animal
- Estimate transfer factors.

Only few data on naturally occurring radionuclides in biota are available. Such data are of general interest in connection with radioactivity in the environment and uptake in biota and transfer to man, and of special interest in connection with determining radiation doses to biota from environmental radioactivity.

Methods

Flesh and liver samples were collected from a single harp seal (*Phoca groenlandica*) killed in Bylot Sound at Thule, Greenland, in August 2003. Five sub samples (50-100 g) were taken from flesh and liver and homogenised in a blender. The sub samples were analysed in duplicate (10 g). Polonium-210 was determined by alpha spectrometry after chemical separation and spontaneous deposition onto silver disks. Lead-210 was determined by repeated analysis of ^{210}Po in each sample after allowing time for ingrowth.

Caesium-137 was determined by gamma spectrometry of ashed samples, and plutonium was determined by radiochemical analysis followed by alpha spectrometry.

Results

The results for polonium are shown in the Tables 1 and 2, and in Figure 1. Results for lead are shown in Tables 3 and 4 and Figure 2 while results for radiocaesium and plutonium are shown in Table 5.

Table 1. Polonium-210 in seal flesh (Bq/kg fw).

Sample	Replicate 1		Replicate 2	
	Bq/kg	1 sd	Bq/kg	1 sd
1	10.4	0.3	10.3	0.3
2	6.6	0.3	6.7	0.2
3	8.9	0.2	8.7	0.3
4	6.3	0.2	6.5	0.3
5	8.4	0.3	8.6	0.3

Table 2. Polonium-210 in seal liver (Bq/kg fw).

Sample	Replicate 1		Replicate 2	
	Bq/kg	1 sd	Bq/kg	1 sd
1	44.7	1.2	42.3	1.2
2	48.6	1.3	46.2	1.2
3	39.7	1.1	37.5	1.0
4	40.9	1.1	44.1	1.1
5	39.6	0.9	44.0	1.1

Table 3. Lead-210 in seal flesh (Bq/kg fw).

Sample	Replicate 1		Replicate 2	
	Bq/kg	1 sd	Bq/kg	1 sd
1	0.31	0.02	0.21	0.01
2	0.28	0.03	0.28	0.02
3	0.42	0.03	0.20	0.02
4	0.56	0.03	0.23	0.02
5	0.36	0.03	0.26	0.02

Table 4. Lead-210 in seal liver (Bq/kg fw).

Sample	Replicate 1		Replicate 2	
	Bq/kg	1 sd	Bq/kg	1 sd
1	0.33	0.03	0.14	0.02
2	0.20	0.02	0.17	0.02
3	0.45	0.05	0.18	0.02
4	0.49	0.05	0.16	0.02
5	0.22	0.02	0.17	0.02

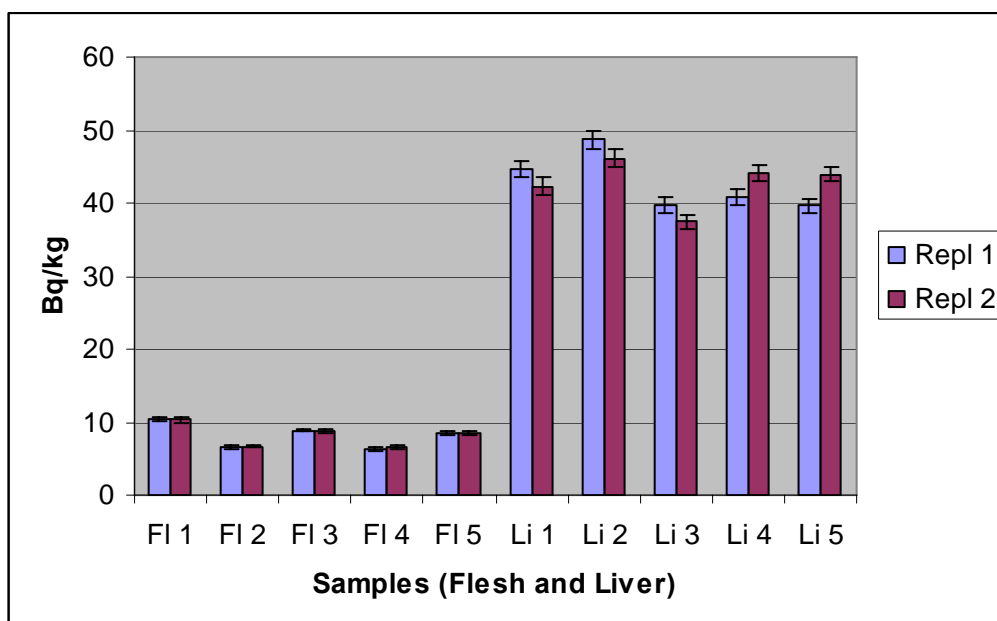


Figure 1. Concentrations (Bq/kg fw) of ²¹⁰Po in samples of seal flesh and liver with error bars showing uncertainties at one standard deviation. Each sample is analysed in duplicate.

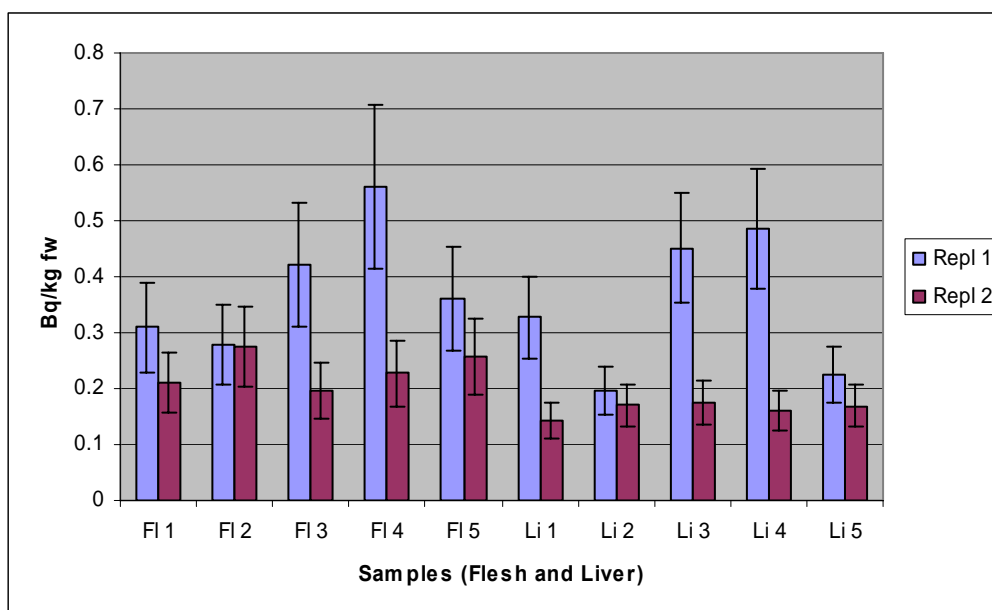


Figure 2. Concentrations (Bq/kg fw) of ²¹⁰Pb in samples of seal flesh and liver with error bars showing uncertainties at one standard deviation. Each sample is analysed in duplicate.

Table 5. Caesium-137 and Pu-239,240 in seal flesh and liver (Bq/kg fw).

	Cs-137		Pu-239,240	
	Bq/kg	1 sd	Bq/kg	1 sd
Flesh	0.20	0.02	<0.0002	
Liver	0.26	0.03	0.002	0.0003

The average concentration of ²¹⁰Po in seal flesh is 8.1 Bq/kg fw (28 Bq/kg dw) with a variation across the five sub samples of 20% (1 sd). The statistical uncertainties for ²¹⁰Po in the flesh samples were 3-4% (1 sd) in agreement with the observed variability of the duplicate samples.

The average concentration of ²¹⁰Pb in seal flesh is 0.31 Bq/kg fw (1.0 Bq/kg dw) with a variation across the five sub samples of 17% (1 sd). The statistical uncertainties for ²¹⁰Pb in the flesh samples were 6-10% (1 sd).

The average concentration of ²¹⁰Po in seal liver is 43 Bq/kg fw (151 Bq/kg dw) with a variation across the five sub samples of 7% (1 sd). The statistical uncertainties for ²¹⁰Po in the liver samples were 3% (1 sd) to which an additional variability of about 4% (1 sd) had to be combined to obtain agreement with the observed variability of the duplicate samples. The latter variability is believed to be due to incomplete homogenisation of sub samples.

The average concentration of ²¹⁰Pb in seal liver is 0.25 Bq/kg fw (1.0 Bq/kg dw) with a variation across the five sub samples of 26% (1 sd). The statistical uncertainties for ²¹⁰Pb in the liver samples were about 10% (1 sd).

Concentrations of ¹³⁷Cs were found at 0.20 Bq/kg fw in flesh and 0.26 Bq/kg fw in liver. Concentrations of plutonium were low, 0.002 Bq/kg fw in liver and below the detection limit of 0.0002 Bq/kg fw in flesh.

Conclusion

The concentrations of ^{210}Po in a seal were found to be 8 Bq/kg fw in flesh (28 Bq/kg dw) and 43 Bq/kg in liver (150 Bq/kg dw). The ^{210}Po is inhomogeneously distributed in flesh corresponding to a standard deviation of 1.6 Bq/kg fw and in liver corresponding to a standard deviation of 2.4 Bq/kg fw. The concentrations of ^{210}Pb in seal flesh are found at 0.31 Bq/kg.

Concentrations of ^{210}Po in marine fish are typically around 1 Bq/kg and in seawater typically around 1 Bq/m³. For the animal studied this gives estimated concentration ratios between ^{210}Po in seal and fish of 8 and 40 for flesh and liver, respectively (Bq/kg fw seal per Bq/kg fw fish). The aggregated transfer of ^{210}Po from water gives concentration ratios of 8 and 40 Bq/kg fw per Bq/m³ seawater for seal flesh and liver, respectively (or 28 and 151 Bq/kg dw per Bq/m³ seawater for seal flesh and liver).

Concentrations of ^{210}Pb in marine fish are typically around 0.2 Bq/kg and around 1 Bq/m³ in seawater. This gives a concentration ratio between ^{210}Pb in seal flesh and fish of 1.5 Bq/kg fw seal per Bq/kg fw fish. The aggregated transfer of ^{210}Pb from water gives a concentration ratio of 0.2 Bq/kg fw per Bq/m³ seawater for seal flesh (or 0.7 Bq/kg dw per Bq/m³).

Concentrations of ^{137}Cs and $^{239,240}\text{Pu}$ in seawater at Thule were found at 2 and 0.01-0.04 Bq/m³, respectively. The aggregated transfer of ^{137}Cs from water gives a concentration ratio of 0.1 Bq/kg fw per Bq/m³ for seal flesh and liver, while the aggregated transfer of $^{239,240}\text{Pu}$ from water gives concentration ratios in the range 0.05-0.2 Bq kg⁻¹ fw per Bq/m³ for liver and <0.02 Bq kg⁻¹ fw per Bq/m³ for flesh. The observed concentration ratios for ^{137}Cs agree with the range of values (0.03-1 Bq kg⁻¹ fw per Bq/m³) recommended by the IAEA (2004). However, the observed concentration ratio for Pu in seal liver is considerably higher than the range recommended by the IAEA (0.003-0.02 Bq kg⁻¹ fw per Bq/m³). The reason for this could be due to the fact, that the harp seal in West Greenland has a high intake of pelagic crustaceans, which in Bylot Sound near Thule contain elevated levels of plutonium. The values recommended by the IAEA are based on data derived from the Irish Sea where seal feeding habits may differ from those in North West Greenland.

Reference

IAEA, 2004. Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. Technical Report Series No. 422. International Atomic Energy Agency, Vienna.

Seaweed, fish and Crustaceans as bioindicators for ^{99}Tc released to marine environment.

Anicke Jerpetjøn, Deborah Oughton and Lindis Skipperud

Norwegian University of Life Sciences,
Department of Plant and Environmental Sciences
P.O. Box 5003 N-1432 Ås

Background

Technetium-99 (^{99}Tc), produced in the fission of ^{235}U , ^{238}U and ^{239}Pu , is a beta-emitter ($E_{\beta\text{max}} = 293\text{keV}$) with a half-life ($t_{1/2}$) of 2×10^5 years. The main sources of ^{99}Tc are global fallout from nuclear weapons testing and discharges from reprocessing plants for spent nuclear fuel in North-Western Europe [1]. Global fallout resulted in a ^{99}Tc concentration in the North Atlantic of $\sim 5 \text{ mBq m}^{-3}$ [2]. Radioactive wastes have been discharged from the reprocessing plants Sellafield (Cumbria, UK) (Fig.1), into the Irish Sea and La Hague (France) into the English Channel since 1952 and 1966 respectively [3].

In seawater, ^{99}Tc exists at the soluble pertechnetate ion, TcO_4^- , and is transported by ocean currents from Cumbria via the North Channel, across the North Sea to the coast of Norway, where it is carried northwards by the Norwegian Coastal Current.

Between 1981 and 1993, discharges of ^{99}Tc from Sellafield were below 7 TBq/a (BNFL, 1981-1996). In the first quarter of 1994 the discharges significantly increased with the opening of the new waste treatment plant Enhanced Actinide Removal Plant (EARP), and subsequent treatment of a backlog of radioactive wastes stored at the site [4;5]. According to BNFL, 72 TBq of ^{99}Tc were discharged in 1994, 190 TBq in 1995, and 150 TBq in 1996.



Figure 1. BNFL Sellafield nuclear facility in Cumbria, the main source of ^{99}Tc .

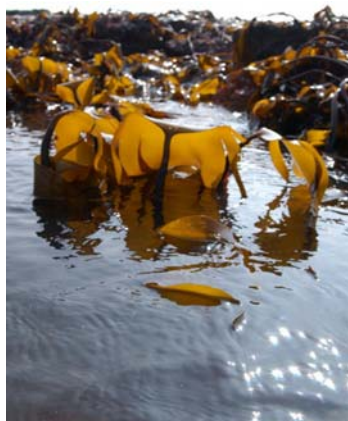


Figure 2. *Laminaria hyperborea*.

In Norway, seaweed has been used as a bioindicator for ^{99}Tc , in order to follow changes in ^{99}Tc activity along the Norwegian coastline over an extended time period. The measurement has been carried out on the large brown algae *Laminaria hyperborea* (Fig.2), which grows on bedrock or other stable substrata from extreme low water to depths depending on light penetration and sea urchin grazing (typically from 8 m depth in coastal waters to 30 m in open coastal waters). The brown algae consist of a stiff, tapering stem up to 2 meters, and a broad, deeply divided (5-20 fingers) leave [6]. The lifetime is up till 15 years. The main growing season is during winter. In spring the old leave is shed and replaced [7]. In *Laminaria hyperborea*, the proportion of growth allocated to various regions of the plant is reported to vary with the age of the plant and its habitat [8]. The proportion of growth allocated to the stem and hapteron, for instance, increases

with exposure, the latter probably helping the plant to remain attached and help it to survive in exposed localities [8]. In one-year-old plants however, growth mainly occurs in the lamina in order to maximize the area for photosynthesis in the light limited understory. The algae are of interest as a bioindicator, furthermore it is harvested commercially for alginate extraction, which in turn is used in various foodstuffs.

Experimental work

Since 1996 *Laminaria hyperborea* has been collected each month from two different sites along the Norwegian coastline (Kvitsøy and Steinshamn), and transported over night to the Isotope laboratory (Norwegian University of Life Sciences). Here the algae was separated into leave and stem, dried and grounded. ^{99}Tc was extracted using miCFowave acid digestion followed by chemical separation on TEVA ion-exchange columns [9]. $^{99\text{m}}\text{Tc}$ was used as a yield monitor for the chemical separation. The concentrations of ^{99}Tc in the extracts were measured by ICP-MS [10]. Together with the collection of seaweed, water samples of 5 litres were collected at the same sites. In addition, activity concentrations in different marine organisms such as fish and Crustaceans have been measured and concentration factors (CF) calculated, using literature values for the concentration in the water.



Figure 3. Map showing algae sampling sites on the Norwegian coastline and the BNFL Sellafield.

Results and discussion

There may be several factors influencing the accumulation of ^{99}Tc in *Laminaria hyperborea*, including growth patterns, seasonal changes in ocean circulation and short-term variations in actual discharge of ^{99}Tc from Sellafield.

The results show that seasonal fluctuations are apparent, with higher activity concentrations in the winter/spring than in the summer/autumn. Overall the activity concentrations of ^{99}Tc in stems are higher than the activity concentration in leaves. Results from Kvitsøy indicate an increase in ^{99}Tc activity in both stems and leave from 1996 until early 2000, then a decrease between 2000 and 2004. In general, activity concentrations decreased faster in leaves than in stems. Results from Steinshamn, further downstream from the discharge, show a similar pattern, but with a continuing increase in stems for ^{99}Tc activity concentration up to July 2002.

The observations could reflect two processes. First, the leaves are in rapid equilibrium with the water, whereas the stems have a slower exchange rate with the water, thus give an indication of concentrations in water over a longer period of time. Second, the leaves are shed

annually; hence the retention-time is limited for this part of the plant. The activity concentrations in the leaves reach a “peak” before shed, but the activity in the stems, which constitutes approximately 60 % of the algal biomass, remained at elevated levels (Fig.4).

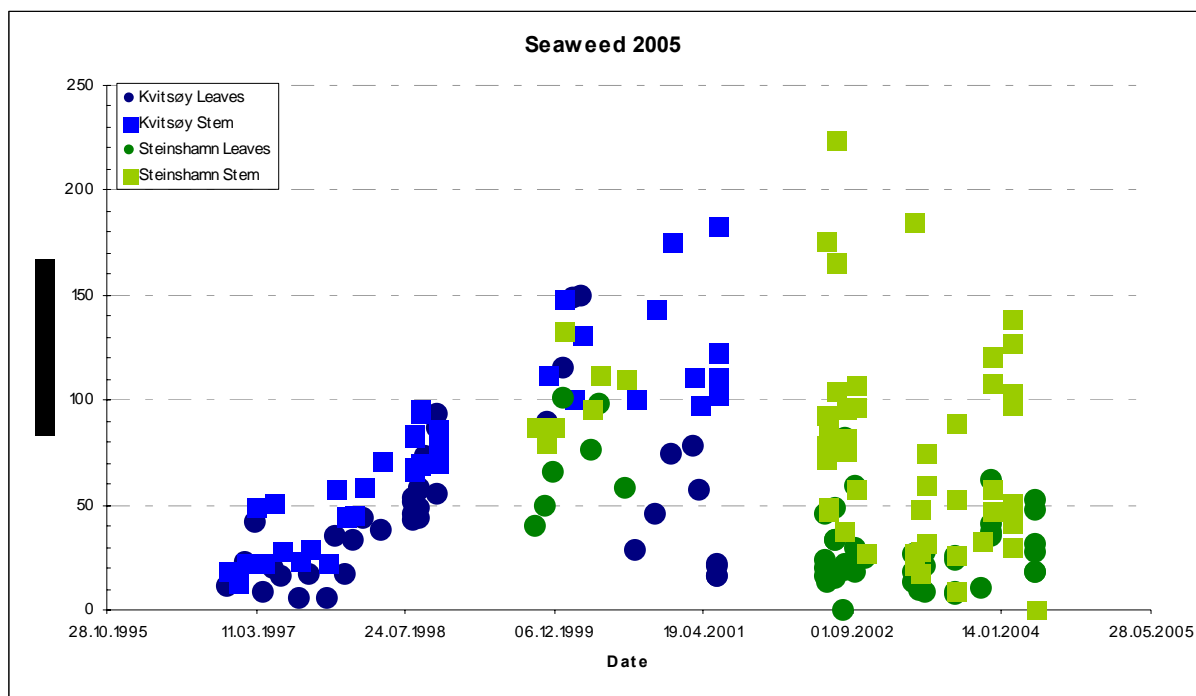


Figure 4. Activity of ^{99}Tc in the leaves and stems of *Laminaria hyperboreana* collected from Kvitsøy and Steinshamn since 1996.

Activity in the leaves suggests that the peak of the plume from Sellafield has reached these sites.

The activity concentration of ^{99}Tc in seaweed collected between 1996 and 2005 showed a range from 6-150 Bq/Kg in leaves and 10-185 Bq/Kg in stems. Whereas ^{99}Tc activity concentration in the water collected along the Norwegian coast between 2003 and 2005 showed a range from 0.1-0.4 Bq/m³. Using seawater and seaweed collected between 2003-2005, concentration factors (CF) were calculated. The CF is calculated by dividing the activity concentration in the organism on the average activity concentration in the water between 2003 and 2005. The CF ranged from 13 to 235 m³/Kg for stems and from 9 to 67 m³/Kg for leaves.

In addition, activity concentrations of ^{99}Tc in different marine organisms (both from the Irish Sea and from along the Norwegian coast) have been measured. Using literature values for ^{99}Tc concentration in water, the Irish Sea 22 Bq/m³ [11] and the Norwegian coastline 0.5 Bq/m³ [Pers.com Risøe/NRPA], CF's was calculated. Activity concentrations in lobster from the Irish Sea was high, as expected, showing activity concentrations of 842 ± 11 Bq/kg and 751 ± 7 Bq/kg dry weight, giving concentration factors of 4210 and 3755 m³/Kg respectively. These data support the CF data for lobster found by Smith et al. [11]. ^{99}Tc activity concentrations in Crustaceans from the Norwegian coast ranged from 0.12 to 0.61 Bq/kg for Crabs and from 34 to 79 Bq/kg for Lobster. This gave CF's in Crab ranging from 0.24 to 1.22 m³/Kg, and for lobster from 68 to 158 m³/Kg.

Activity levels of salmon filet collected from Norwegian fish farms showed 0.11-0.80 Bq/Kg giving CF levels ranging from 0.21 to 1.60 m³/Kg. Herring showed activity concentrations from 0.039 to 0.080 Bq/Kg, giving CF values of 0.08 and 0.16 m³/Kg. Only one sample of wrinkle was measured (activity concentration of 1.3 Bq/Kg), and the CF calculated was 2.6 m³/Kg, which makes it interesting for further investigations. Overall the data varies a lot, which is a problem when measuring natural organisms. Habitat, growth, and eating habits are among many different factors influencing the uptake of ⁹⁹Tc. According to C. Verthe *et al.* fish does obtain ⁹⁹Tc mainly through food, and not directly from the water [12].

Conclusions

Along the Norwegian coast, ⁹⁹Tc discharged from nuclear installations is found in seaweed, Crustaceans and fish. The activity concentrations of ⁹⁹Tc in seaweed peaked around July 2002, giving a concentration ratio (CF) for the stems up to 121 m³/Kg. The CF in different marine organisms differs between species, and within species; however, the CF is found to be highest for lobster, 4210 and 3755 m³/Kg in the Irish Sea and 68 to 158 m³/Kg along the Norwegian coast. ⁹⁹Tc activity concentrations in Crabs from the Norwegian coast ranged from 0.12 to 0.61 Bq/kg giving CF's in Crab ranging from 0.24 to 1.22 m³/Kg. Salmon filet collected from Norwegian fish farms showed CF levels ranging from 0.21 to 1.39 m³/Kg, whereas Herring showed CF values of 0.08 and 0.16 m³/Kg. Overall the data varies a lot, which is a problem when measuring natural organisms. Habitat, growth, and eating habits are among many different factors influencing the uptake of ⁹⁹Tc. Further investigation on CF and how ⁹⁹Tc are obtained by different species in the marine environment, and also different organs of the organisms, is needed.

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Cs-137 in the Hungarian environment, especially in the meat of wild animals

Sándor Tarjám, Beáta Varga

National Food Investigation Institute, P.O. Box 1740, Budapest 94, H-1456, Hungary

The Radiological Monitoring Network of Ministry of Agriculture and Regional Development (RMN) consist of 19 county-laboratories and a central laboratory in Budapest. Sampling, sample preparation is carried out in every county laboratory and screening methods are also used there. Half of the laboratories are able to use nuclide selective methods with high sensitivity. The monitoring program of the RMN focus on the major agricultural products and foodstuffs produced in the country. Above them the natural environment is also kept under control by regular in-situ gamma-spectrometry measurements and through the sampling of bioindicators. In this way RMN is able to provide data about the specific activity of different radionuclides in different medium, time dependency and spatial distribution of radionuclides, in order to give base to assess ingestion doses and to give support to the decision makers in the case of emergency.

In the cause of producing consistent and comparable dataset year by year, above the usual accreditation requirements the central laboratory lays emphasis upon the quality assurance activity with regular meetings to keep the colleges well trained and with organising intercomparison runs. For this purpose well characterized, natural samples were chosen, from which large amount was possible to collect and the activity level of interested nuclides were about or slightly above the typical natural levels.

The concentration of natural radionuclides, like daughters of U(Ra)- and Th-series and ^{40}K are determined by the geological parameters of living territory of wild animals. Radionuclides with long half-life from the nuclear weapons-tests and accidental releases of nuclear power plants give the main part of the anthropogenic contamination of the environment, ^{137}Cs and ^{90}Sr isotopes are of higher importance. The spatial distribution of anthropogenic isotopes depends on the dry and wet fall-out, namely the meteorological conditions.

The specific concentration of ^{137}Cs only in some elements of food-chain is shown in the Figure 1 for giving the order of magnitude of the radiocaesium in Hungarian environment. In general, the highest contamination was found in natural and semi-natural environment and the produced products have lowest specific activity values as usual.

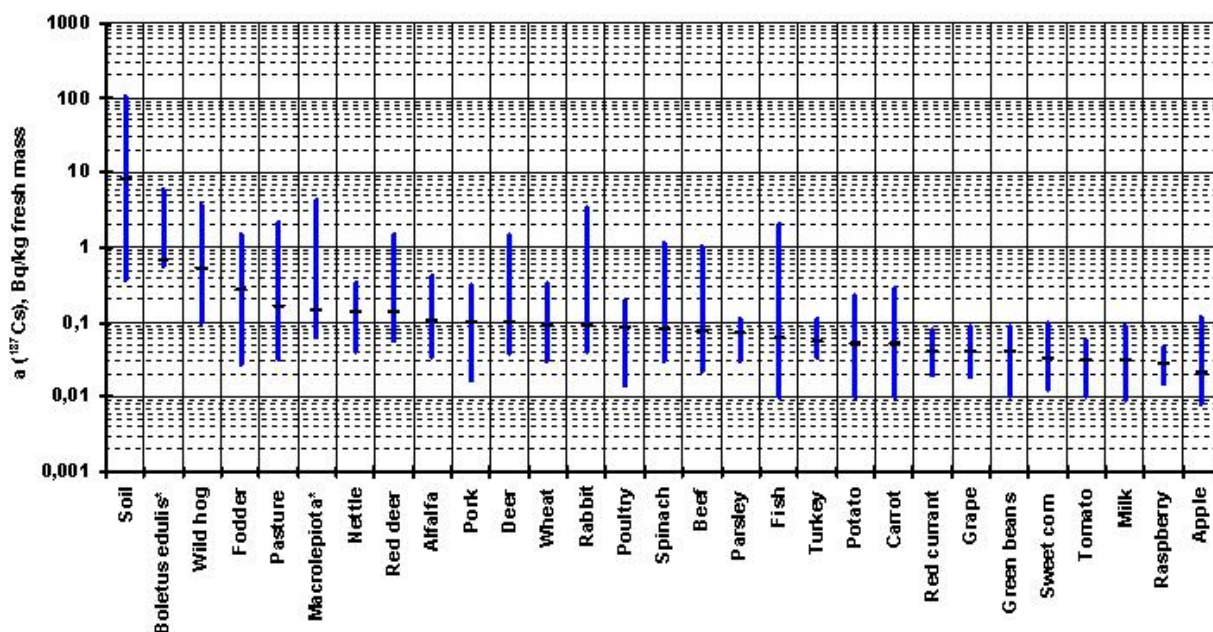


Figure 1. Specific activity of ^{137}Cs in different elements of food-chain (mushrooms are collected in forests).

As far as Hungary is concerned, the specific activity of anthropogenic radionuclides varies in a wide scale depending on the location. The concentration interval of the ^{137}Cs is 5-times wide and range of ^{90}Sr is 6-times wide. The typical average-concentrations of natural radionuclides in different counties differ 4-5-times from each other in the case of U(Ra) decay-serie, and 2-times in the case of Th-serie.

The specific activity of the most important isotopes in soil are the following:

U(Ra)	16-50 Bq/kg,
^{137}Cs	6-32 Bq/kg
^{90}Sr	0,6-4 Bq/kg

^{137}Cs -isotope is accumulated in the meat of animal, deriving from chemical property it can replace K in the tissues. Equilibrium is supposed between the ^{137}Cs content of the feed and its concentration in the meat, depending on the biological half-lives. ^{137}Cs -isotope is accumulated much more in some kind of mushrooms and mosses, they might be determined the ^{137}Cs -uptake, therefore it is very important to know well the eating-habit of different types of animals.

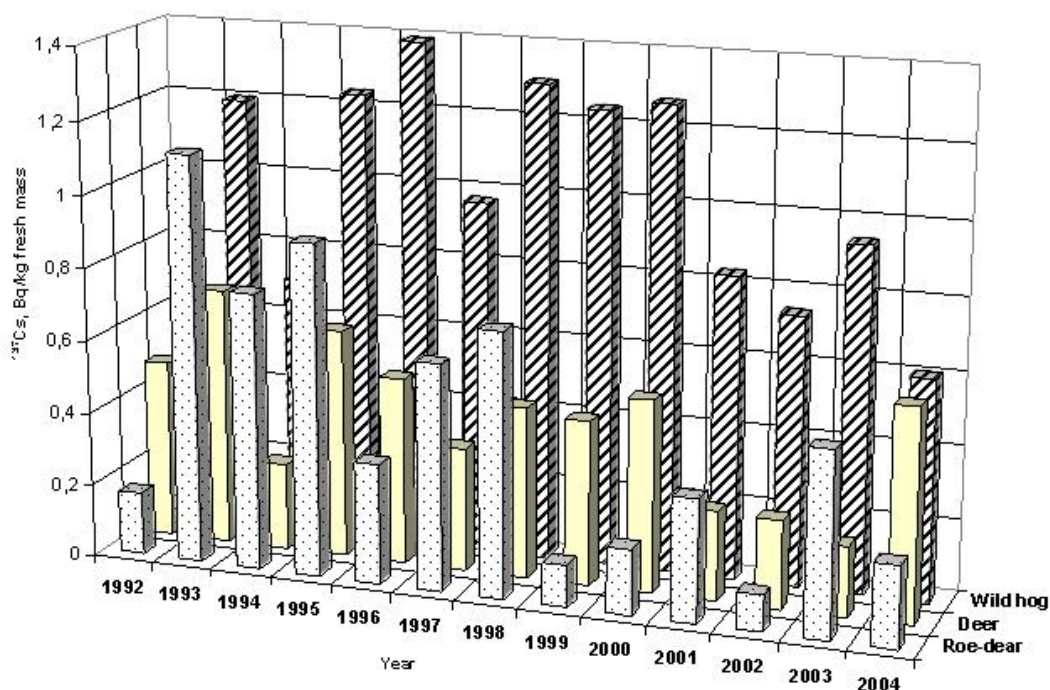
In the case of wild animals the elements of natural food-chain play the determinant role in the transport-processes of radionuclides. Intervention and assistance of human being may be supposed to have small or no effect on radionuclide-concentration, characteristic is given by the natural area. The consumption of animals living in natural circumstances is considered diversified regarding the seasons and spatial distribution also. Beside the pasture they consume lots of types of plants, some of them are considered herbs, sprout of different bushes, mushrooms. In winter they are fed with fodder produced mainly in the same area. From 1986 to 1991 the transfer of ^{137}Cs through the food-chain is considered rather high rate, data form 1992 may be handled as one dataset, because no further significant decrease in any type of the samples was observed. Meat samples were delivered by hunters with the information of the

place and date of the shot, typically 2-3 kg of meat samples were dried at 105°C, then ashed in 450°C. Ashes were measured by HPGe detectors, collection time was 80000 s. Table 1 shows the simply statistical evaluation of the result of the monitoring of 2000-2004 years.

Table 1. Specific activity of ^{137}Cs in some part of food-chain in the environment, 2000-2004.

	Soil Bq/kg dry mass	Pasture Bq/kg fresh mass	Roe-deer Bq/kg fresh mass	Deer Bq/kg fresh mass	Wild hog Bq/kg fresh mass
Number of samples	1078	703	53	121	140
Minimum	0,19	0,02	0,04	0,02	0,01
Maximum	114,2	5,80	2,70	5,65	11,04
Median	11,5	0,16	0,12	0,16	0,31
Average	15,3	0,24	0,29	0,37	0,87

The difference of the eating habit of the examined three types of animals determine the radionuclide content of the meat as it is shown in the table, the highest radiocaesium-contamination in the meat of wild hogs was observed. Mushrooms play an important role in the food chain regarding the radionuclides. They have higher specific activity mainly one order of magnitude comparing with the plant samples in the environment. However their consumption rate is much smaller than others they may determine the ingestion dose of animals. Regarding the spatial distribution the meat of wild animals shot in Zala and Somogy counties (south-west part of the country) contain more ^{137}Cs than the others. A correlation was observed, because these two counties have such a climate, which is favourable for mushrooms, so the possibility of the consumption by the animals is higher here. Generally the animals living in the parts of the forest from where they have access to the arable lands were less contaminated. The specific activity of the meat also depends on the amount and seasonal distribution of precipitation of their living area.



Taking into account the complexity of forest ecosystem, this topic needs further investigation, which has been started the last few months. Spatial variation could be determined in the case of these animals, because there are some investigations regarding the determination of living area of wild animals. Having these data and knowing the consumption habits of the animals further samples will be collected from the different types of plants and the soils also to quantify dependencies.

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Intercomparison of laboratory analyses of radionuclides in environmental samples, 2004-2005

Sven Nielsen

Risø National Laboratory, DK-4000 Roskilde, Denmark

Background

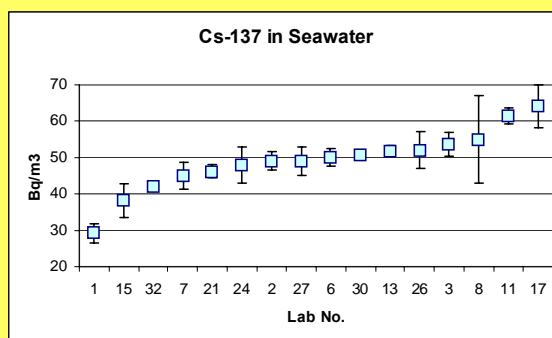
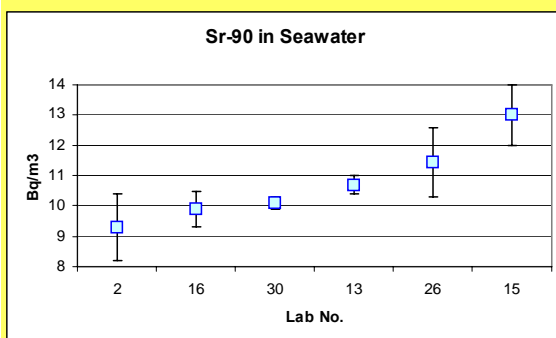
The LABINCO Project involves a comparison across laboratories of analytical results of radionuclides in environmental samples and foodstuffs. Participants were invited in spring 2004 and asked for interest in sample types. Samples were distributed in July 2004 to 40 participants in the Nordic and Baltic countries. Sample types and sample amounts including radionuclides recommended for analysis are shown in the tables below. Analytical results were received from 36 participants and with two participants submitting results from two local laboratories, 38 sets of results were available for the intercomparison. Graphical presentations of some of the results are shown in the following. Closed circles in the graphs represent reported detection limits.

Sample type, origin	Sample amount (kg)
Seawater - surface water from Danish waters, salinity 8 ‰	50
Lake water - from a Swedish lake	25
Tap water - spiked with tritium	2 x 1
Sediment - from Roskilde Fjord	0.2
Seaweed - from Danish waters	0.15
Fish meal - Danish origin	2
Soil - Swedish origin	0.2
Dry milk - Danish origin	1
Cereal - Swedish origin	0.5
Lucerne/alfalfa pellets – Danish origin	2
Soil, Certified Reference Material	0.1

Nuclide	Seawater	Lake water	Tap water	Sediment	Seaweed	Fish meal	Soil	Dry milk	Cereal	Lucerne	Soil CRM
Cs-137	X	X		X	X	X	X	X	X	X	X
Sr-90	X	X		X	X	X	X	X	X	X	X
Tc-99					X						
Pu-isotopes				X	X		X				X
Am-241				X	X		X				X
Ra-226				X	X		X				X
Th-232				X	X		X				X
K-40				X	X	X	X	X	X	X	X
Po-210				X	X		X				X
Tritium			X								
Total alpha		X									
Total beta		X									
Other gamma				X	X		X				X

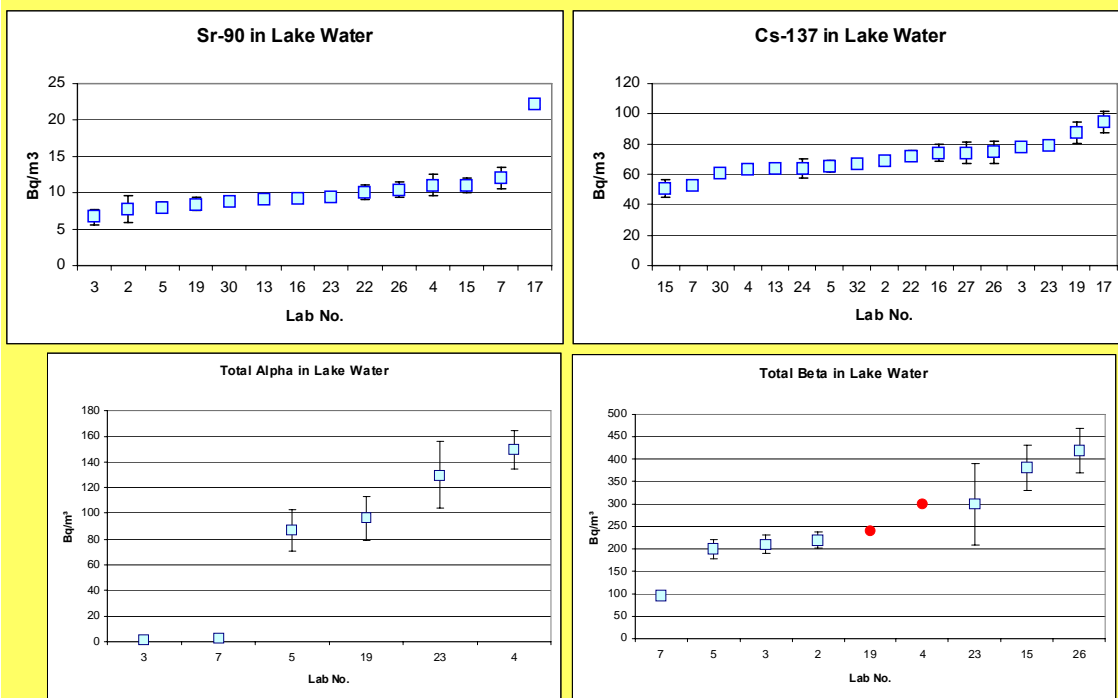
Seawater (Western Baltic)

RISO



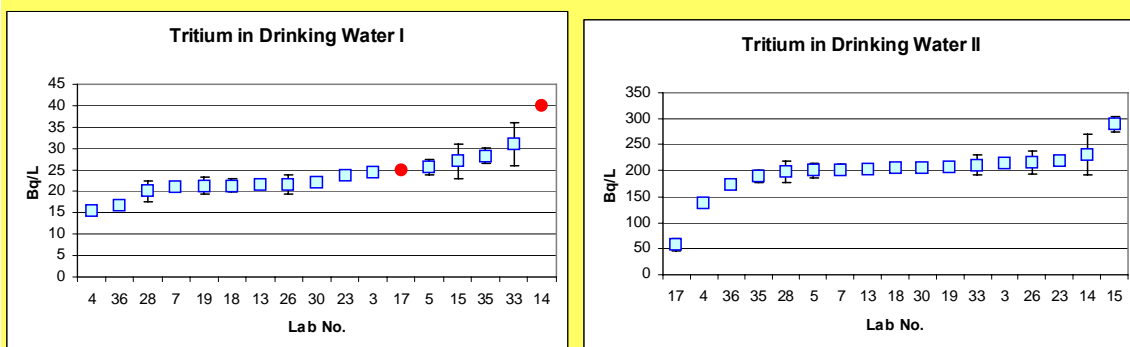
Lake Water (Hillesjön)

RISØ



Drinking Water (spiked with tritium)

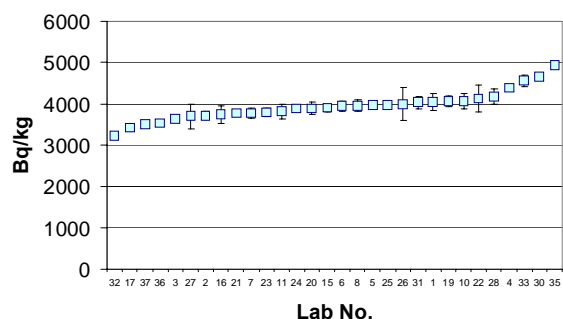
RISØ



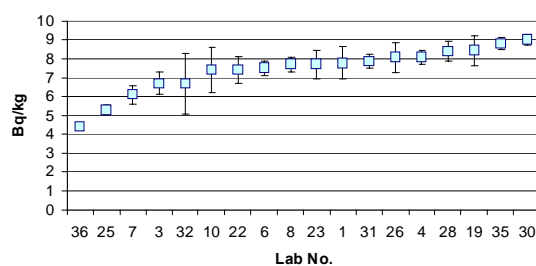
Soil (IAEA-375)

RISO

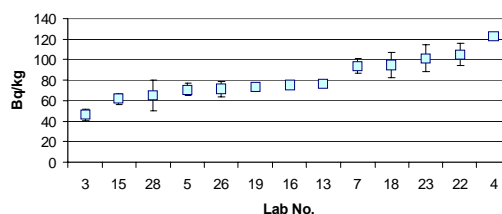
Cs-137 in Mineral Matrix III



Cs-134 in Mineral Matrix III



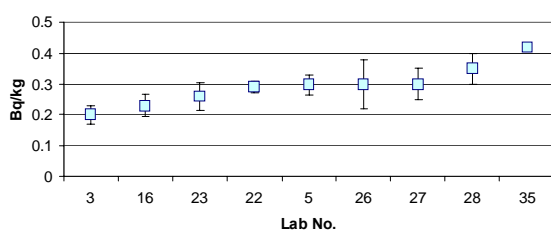
Sr-90 in Mineral Matrix III



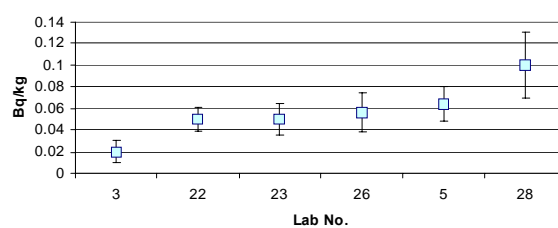
Soil (IAEA-375)

RISO

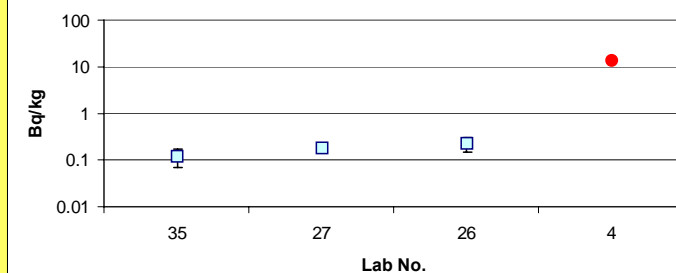
Pu-239 in Mineral Matrix III



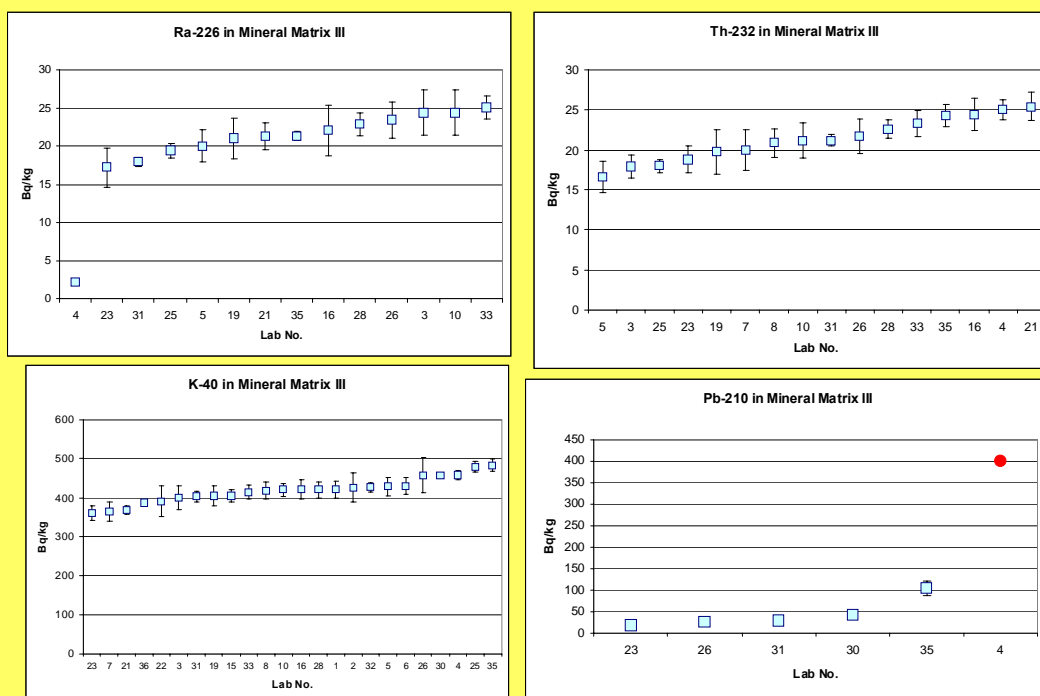
Pu-238 in Mineral Matrix III



Am-241 in Mineral Matrix III



Soil (IAEA-375)



Evaluation

The results were evaluated by comparing the laboratory results (x) to median values (x_m) for each sample type. The deviations from median values were calculated in terms of z-scores, $z = (x - x_m)/\sigma$, where σ is the target standard deviation of 10%. Z-scores were combined into rescaled sums of z-scores across sample types, $(\Sigma z)/n^{1/2}$, where n is the number of results, to test the bias and, furthermore, combined into sums of squares of z-scores, Σz^2 , to test the accuracy. Tests were carried out at the 99% level. The table below summarises the preliminary evaluation by radionuclide.

Nuclide	Laboratories submitting results	Laboratories passing evaluation criteria	
Sr-90	20	3	15 %
Tc-99	5	4	80 %
Cs-134	19	16	84 %
Cs-137	35	27	77 %
Pu-238	7	4	57 %
Pu-239	10	7	70 %
Am-241	6	1	17 %
Ra-226	20	14	70 %
Th-232	21	17	81 %
K-40	29	26	90 %
Pb-210	8	3	38 %
Po-210	3	2	67 %
Be-7	5	5	100 %
Tritium	17	12	71 %
Total alpha	6	2	33 %
Total beta	7	3	43 %

Conclusions

An intercomparison exercise of radionuclides in environmental samples and foodstuffs has been carried out. A total of 821 analytical results were received from 38 laboratories. Half of the laboratories are from the Nordic countries and the other half from Estonia, Latvia, Lithuania, Poland, Russia and Hungary.

The concentrations of radionuclides in the samples were unknown for what reason the evaluation was based on comparison with median values. However, one of the samples (soil) was a certified reference material (IAEA-375), and for this sample good agreement was found between the median values and the certified values.

The comparison shows generally good agreement between laboratories with some exceptions. Results for which the agreement is not good include ^{90}Sr , ^{241}Am , ^{210}Pb and total alpha and total beta. Less than half of the participants meet the evaluation criteria concerning these results. For these analyses there seems to be a particular need for improved analytical quality and testing of the results in international intercomparison exercises.

Results of the intercomparison on natural radioactivity in groundwater arranged within NKS-RADCHEM project

Tarja K. Ikäheimonen

STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Introduction

This intercomparison was agreed to be arranged within the RADCHEM project to ensure the capability of Nordic laboratories for measurements of natural radioactivity. At the same time, analytical methods used in different laboratories were presented. STUK from Finland volunteered to arrange the intercomparison because it had a good possibility for collecting suitable groundwater samples for this purpose. Six laboratories participated in the intercomparison:

- Institute for Energy Technology, Norway
- Risoe National Laboratory, Denmark
- Lund University, Sweden
- Linköping University, Sweden
- Helsinki University, Finland
- STUK, Radiation and Nuclear Safety Authority, Finland

Sampling and pretreatment

In the Loviisa area, the bedrock is rapakivi-granite with a high uranium concentration. A drilled well in this area was chosen for the sampling site. The depth of the well is 57 m. Sampling was performed on June 14, 2005 (reference date). First, the water was run about 0.5 hour before the sampling. Then the water was sampled into two 30-liter cans. The samples for radon measurements were sampled directly into the liquid scintillation bottles. The samples were transported to the laboratory immediately and the samples of a large volume were acidified into 0.05 M with nitric acid by mixing well. The water from the cans was divided into one liter bottles to be transported to the other laboratories as soon as possible. The laboratories were asked to analyze as many natural nuclides as they were able to within the given time of about 4 months.

Analytical methods

A direct liquid scintillation method was used for measurements of Rn-222 in water.

For separation of uranium isotopes, two different methods were used: extraction with tributylphosphate (TBP) and anion exchange methods. Uranium was measured with alpha spectrometer by using U-232 as a yield tracer. U-234, U-235 (only one laboratory) and U-238 were measured.

For Pb-210 activity measurements, two different kinds of analysis methods were used. Direct gammaspectrometric measurement after evaporation and/or precipitation was used by two laboratories in the study. The other laboratories first separated Po from the samples and then measured Po-210 (Po-210 in the sample + Po-210 growth from Pb-210 during sampling and separation). After that the second Po separation and alpha measurement were performed after the in-growth. Both Pb-210 and Po-210 could be analyzed with these two measurements by the alpha spectrometer. Po-210 was also calculated using the Pb-210 result from the gammaspectrometric measurement without the in-growth time.

Ra-226 was measured with two different methods. A co-precipitation with BaSO₄ and alphaspectrometric measurement using Ba-133 as a yield tracer (measured with NaI counting) was used. Another method was the liquid scintillation method with evaporation and a four-week in-growth of all Ra-226 daughters. Ra-226 was measured via its daughter Po-214 in the alpha window of the liquid scintillation spectrum.

Results

The results for Rn-222, U-234, U-238, Pb-210, Po-210 and Ra-226 with two sigma total uncertainties are presented in the Figures 1 - 6. Only one laboratory measured U-238 and another measured Ra-228. Those results were 0.003 ± 0.001 Bq/l and 0.05 ± 0.02 Bq/l, respectively.

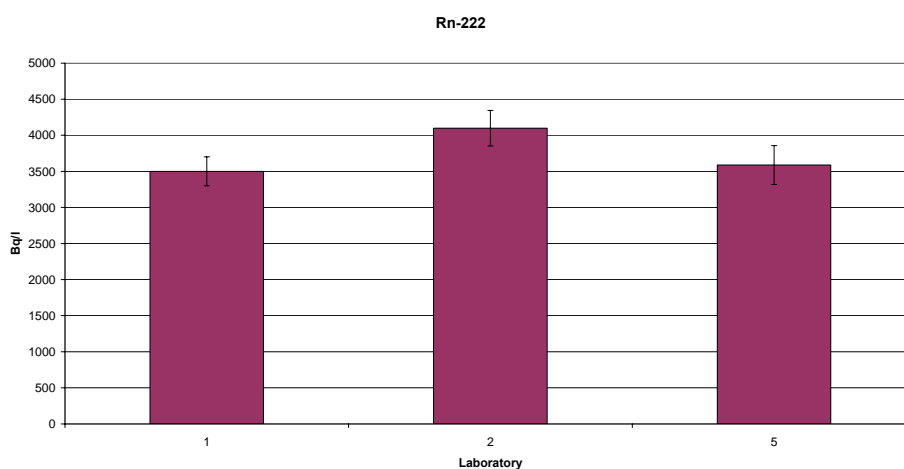


Figure 1. Results for Rn-222 with 2 sigma uncertainties.

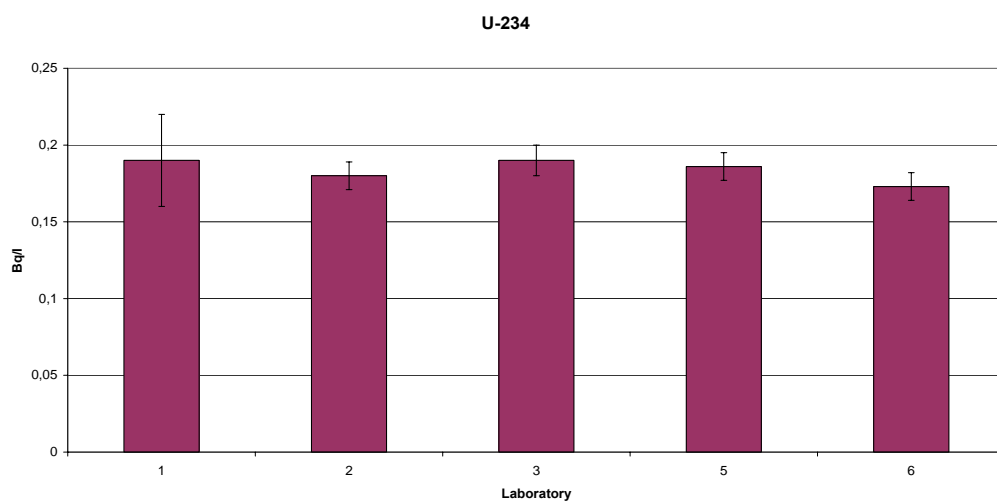


Figure 2. Results for U-234 with two sigma uncertainties.

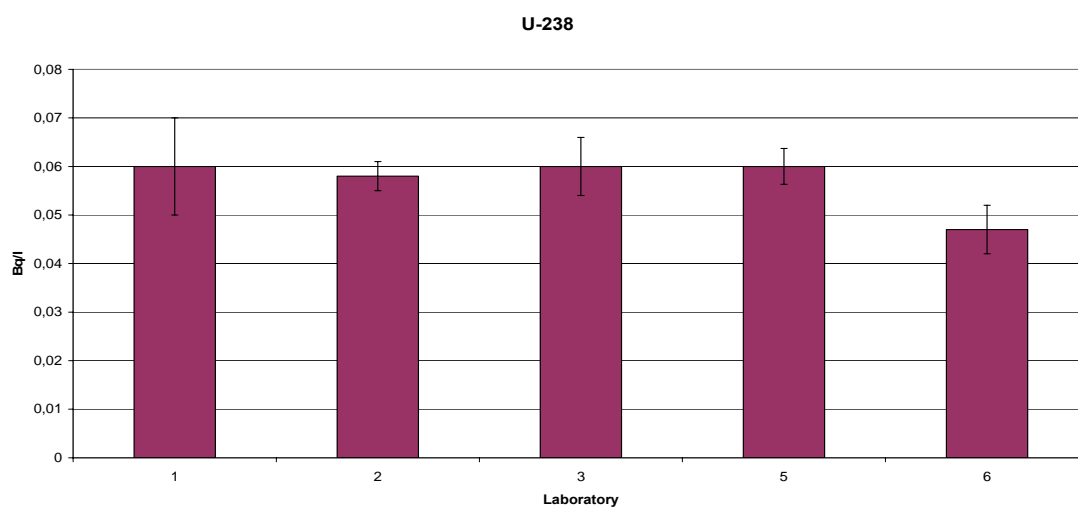


Figure 3. Results for U-238 with two sigma uncertainties.

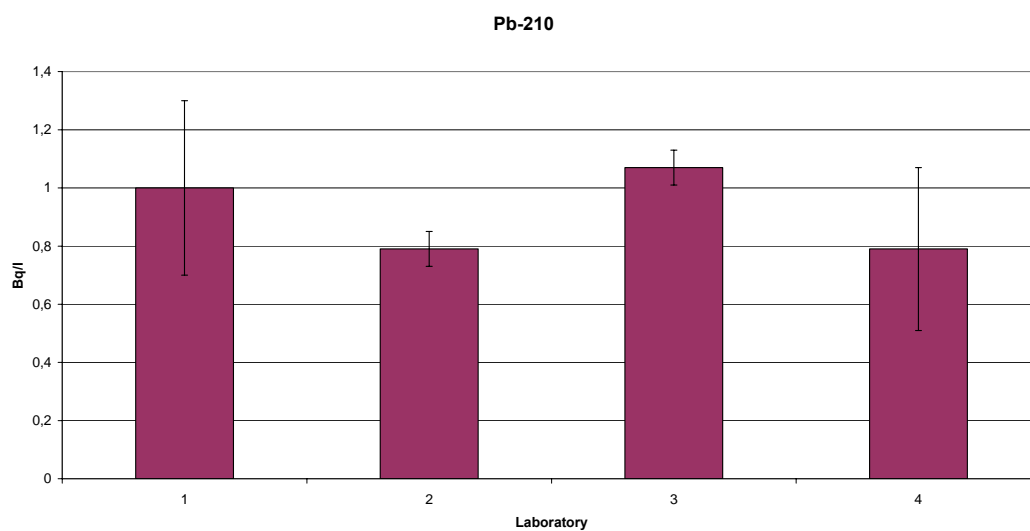


Figure 4. Results for Pb-210 with two sigma uncertainties.

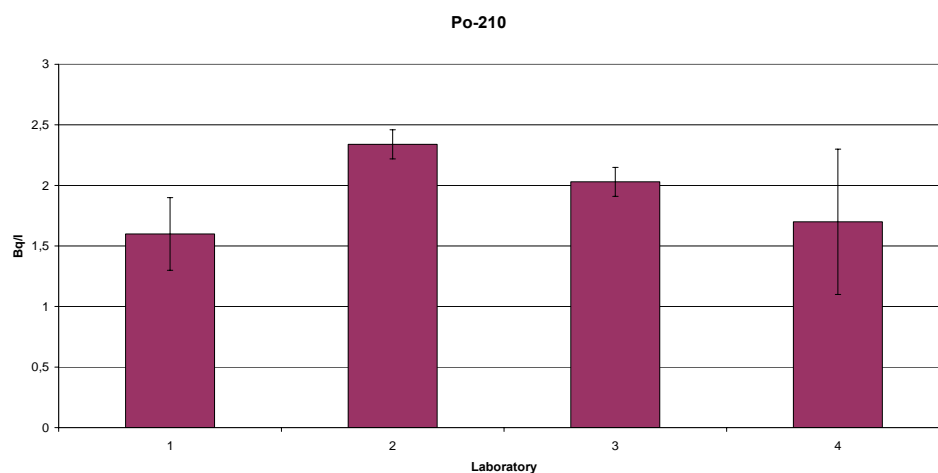


Figure 5. Results for Po-210 with two sigma uncertainties.

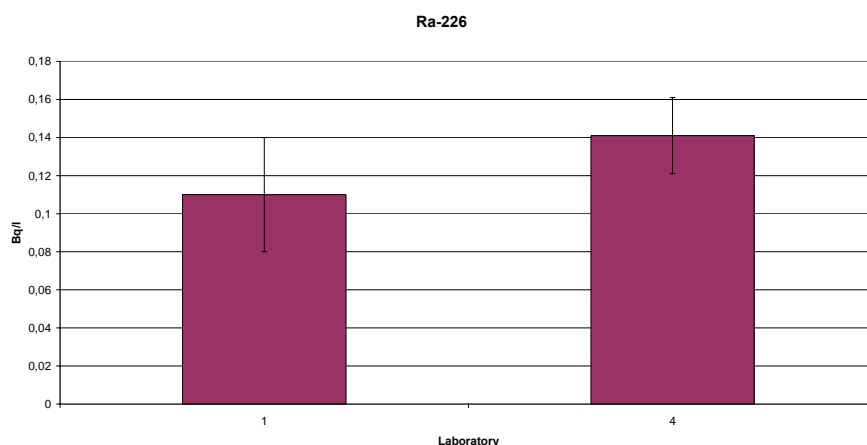


Figure 6. Results for Ra-226 with two sigma uncertainties.

The results for Rn-222, U-234 and Ra-226 were in good agreement with each other within the two sigma uncertainties. One result of U-238 was a little bit smaller than the others. The results for Pb-210 and Po-210 were not as good as the above mentioned. One reason for this could be the too small in-growth time of Po-210 after the first separation which was due to the tight timetable. Also, reported uncertainties varied greatly. In future it will be necessary to observe if the uncertainty estimations are consistent with each other in all laboratories. The accuracy of the presented results varied, too. This issue should also be discussed.

The results with the two sigma uncertainties with the mean value including 10% uncertainty limits are presented in the Table below. There all values fit within the limits of the mean.

Laboratory	Rn-222	U-234	U-238	Pb-210	Po-210	Ra-226
1	3300-3700	0.16-0.22	0.05-0.07	0.7-1.3	1.3-1.9	0.08-0.14
2	3850-4350	0.17-0.19	0.058-0.064	0.73-0.85	2.2-2.5	-
3	-	0.18-0.20	0.054-0.066	1.0-1.1	1.9-2.2	-
4	-	-	-	0.51-1.1	1.1-2.3	0.12-0.16
5	3320-3860	0.18-0.20	0.056-0.064	-	-	-
6	-	0.16-0.18	0.042-0.052	-	-	-
Mean ± 10 %	3360-4100	0.17-0.20	0.05-0.06	0.82-1.0	1.7-2.1	0.11-0.14

Conclusions

The intercomparison was considered very necessary and important. The results show that the general analytical capability for determination of natural radioactivity in drinking water in the Nordic laboratories is good. However, this kind of intercomparisons should be continued, as well as discussions of observations coming up during the intercomparison and reviewing of results.

Acknowledgements

I would like to acknowledge Rajdeep Sidhu from IFE, Per Roos from Risø National Laboratory, Elis Holm from Lund University, Jukka Lehto from Helsinki University, Håkan Petterson from Linköping University and Pia Vesterbacka from STUK for their personal contributions to the intercomparison.

Determination of cesium, americium and plutonium isotopes in the environmental samples

G. Lujanienė, J. Šapolaitė

Institute of Physics, Savanorių 239, LT-02300 Vilnius, Lithuania

The discovery of nuclear fission and the development of nuclear weapons and nuclear energy are related to the significant releases of radionuclides into the environment. The assessment of environmental impact of radioactive contamination requires determination of activity concentration of released radionuclides in various matrixes. The direct measurements are not always possible due to detection limit or interferences of radionuclides of different origin usually present in samples. Highly specific methods are necessary for pre-concentration and separation of radionuclides from samples of various matrixes.

Methods of ammonium molybdophosphate (AMF) and various transition metal Ferro cyanides both by precipitation and adsorption have been used for collecting radiocesium from natural waters. Ferro cyanides are valuable adsorbents for cesium. They are distinguished for high sorption efficiency and selective adsorption of cesium from sea water. Chemical properties and adsorption characteristics of Ferro cyanides have been thoroughly investigated [1]. According to the structures of the insoluble Ferro cyanides of bi and trivalent cations which may be represented as $M^{II}[M^{II}(NC)_6Fe]$ or $M^{III}_{1/3}[M^{III}(NC)_6Fe]$, the transition metal ions M standing outside the brackets are exchangeable for univalent cations.

For preparation of a Ferro cyanide exchanger, different techniques such as granulation by freezing, immobilization on different matrices, saturation of a cation exchanger and impregnation of different materials were used [2-4]. Cation exchangers saturated with Ferro cyanides as well as different fibers impregnated with mentioned substances are especially suitable for pre-concentration of cesium from sea water.

Fe Ferro cyanide method was used for determination of ^{137}Cs in sea and fresh water samples. This method allows to co-precipitate ^{137}Cs and ^{90}Sr from large volume samples with iron and other trace metals, naturally present in water, Ferro cyanides and calcium and strontium carbonates. This method was applied for cesium and strontium determination in water samples with different salinity (0 – 35 ‰).

Water samples were filtered through membrane filters 0.45 or 0.22 μm and were acidified (pH=2). Strontium carrier (as chloride, standardized) and cesium tracer (as ^{134}Cs , standard) for chemical yield determination were added to the water samples too. The samples were mixed at least for 2 hours. Then $FeCl_3$ and $CaCl_2$ (for samples of water salinity <6 ‰), $K_4[Fe(CN)_6]$ and Na_2CO_3 were added (pH=5). The sample was mixed and left to stand for 2 hours and then Na_2CO_3 was added. The samples were mixed until the sodium carbonate dissolved. Samples were left to stand over night (12 hours). The bulk of the supernatant was decanted; the residue was filtered, dried and placed into the standard plastic containers of 50 or 150 ml volume for the determination of ^{137}Cs with HPGe detector (resolution – 1.9 keV/1.33Mev, efficiency – 42%). Sr-90 (=Y-90) was determined by HDEHP-extraction of yttrium-90 and beta counting using the proportional Emberline FHT 770 T Multi-Low-Level-Counter.

The separation of the suspended matter was achieved using membrane filtration and sediment filter cartridges. Membrane filters of 0.22 µm, 0.45 µm and polypropylene Sediment Filter Cartridges (US Filter Plymouth Products) of 25 µm, 5 µm and 1 µm were applied. The activity of ^{137}Cs in some samples of suspended matter was close to detection limits for gamma-spectrometric measurements, therefore cesium was separated radiochemically. The samples were digested using HF/HNO_3 and HCl , and cesium after Ni Ferro cyanide pre-concentration was precipitated using saturated SbCl_3 and NH_4I solutions as $\text{Cs}_3\text{Sb}_2\text{I}_9$. Chemical yield was determined gravimetrically. The massic activity of cesium in suspended particles was measured using the proportional Emberline FHT 770 T Multi-Low-Level-Counter.

Precision of ^{137}Cs measurements by gamma-spectrometry was <10 %. Cesium activities derived from beta-counting had uncertainties from less than 10 to 20 %, depending on the activity of the measured source. Accuracy and precision of cesium determination in suspended particles were tested using NIST Standard Reference Material (SRM) No 4350B and No 4357 as well as samples originated from the Curonian Lagoon and from the Baltic Sea. The results of the comparison are presented in Table 1. Some data on cesium activities in the Baltic sea water and the Curonian Lagoon are presented in Table 2.

Table 1. Cesium activities in suspended particle and bottom sediment samples determined by gamma (I) and beta (II) measurements.

	^{137}Cs , Bq/kg (I)	$^{137+134}\text{Cs}$, Bq/kg (II)
Open sea water	91 ± 27	84 ± 8
Near shore water	210 ± 24	205 ± 7
Curonian Lagoon	223 ± 25	227 ± 10
Klaipeda port	197 ± 12	200 ± 20
SRM No 4350B	29 ± 3	27 ± 3
SRM No 4357	12.7 ± 1.0	12.9 ± 1.0

Table 2. Ranges of ^{137}Cs activities in water (Bq/m^3) and suspended particles (Bq/kg) in the Curonian Lagoon and the Baltic Sea (1999-2004).

	^{137}Cs , Bq/m^3	Sites, n	^{137}Cs , Bq/kg	Sites, n
Open sea water	41 - 80	21	80 - 970	20
Near shore water	75 - 98	24	15 - 372	24
Klaipeda port	45 - 52	4	144 - 197	4
Curonian Lagoon	1 - 54	12	16 - 223	12

Ion exchange, liquid-liquid extraction and precipitation techniques have been widely used for separation and pre-concentration of actinides. However, these traditional methods are time-consuming and generate large amounts of acidic and organic waste.

At present the separation methods based on extraction chromatography have become popular because Eichrom resins are highly selective, and allow an efficient analytical procedure to be performed. In addition, they produce significantly less acidic waste and hazardous organic solvents than the most traditional procedures. The extraction chromatography methods provide high selectivity actinide separation and can be useful in cleaning plutonium isotopic samples for mass spectrometry. However, the application of the extraction chromatographic methods published is not always suitable for environmental samples. Moreover, there are

some problems in complete separation of the alpha-emitting natural radionuclides, the traces of which interfere significantly the determination of low concentrations of Pu and Am and their isotopic activity ratios.

The radiochemical analyses of monthly samples of aerosol ashes (about 30 g) were performed. For separation of Pu isotopes the TOPO/cyclohexane extraction and radiochemical purification using UTEVA resin were performed, Am was separated after TOPO/cyclohexane extraction using TRU and TEVA resins (100–150 μm). ^{242}Pu and ^{243}Am were used as tracers in the separation procedure. The alpha spectrometry measurements of Pu and Am isotopes deposited on a stainless steel disc were carried out with the Alphaquattro (Silena) spectrometer. Accuracy and precision of analysis were tested using NIST SRM No 4350B and 4357. Precision of measurements by alpha spectrometry was $\text{Pu} \leq 8\%$, $\text{Am} \leq 10\%$. After alpha spectrometry samples were selected for mass spectrometric measurements. Plutonium isotopes were washed from stainless steel discs and after purification $^{240}\text{Pu}/^{239}\text{Pu}$ ratio was determined by ICP–MS.

The measurements carried out on aerosol samples collected in Vilnius in 1997–1999 indicated the presence of alpha emitting radionuclides. The activity concentrations of $^{239,240}\text{Pu}$ and ^{241}Am ranged from 0.4 to 18 and from 0.3 to 9.7 nBq/m^3 , respectively.

The origin of contamination source was identified using meteorological data of backward trajectories and atom ratio of radionuclides. For comparison measurements were performed on aerosol samples collected during the Chernobyl accident and the contaminated Chernobyl soil.

In some samples the high activity ratio of $^{238}\text{Pu}/^{239,240}\text{Pu}$ up to 0.5 was observed. The presence of the Chernobyl derived plutonium was confirmed by ICP-MS (atom ratio $^{240}\text{Pu}/^{239}\text{Pu}$ up to 0.40 was found).

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Practicable Monte Carlo Calibration of HPGe Detectors for Environmental Measurements.

Mark Dowdall

Norwegian Radiation Protection Authority
Polar Environmental Centre, 9296 Tromsø, Norway

Introduction

The advent of high purity germanium (HPGe) and silicon (Si) detectors during the course of the 1960's revolutionized the field of gamma ray spectrometry and such detectors remain the mainstay of the majority of radioanalytical laboratories around the world due to the non-destructive nature of the technique and the generally high resolving power of such detectors. The use of HPGe detectors to produce accurate and precise results is primarily reliant on the establishment of the instruments ability to detect gamma radiation incident on the detector at various energies. The traditional approach to the efficiency calibration of semi-conductor gamma-ray detectors is to directly determine the detectors response to gamma radiation of various energies by exposing the detector to known activities of selected specific isotopes usually in the form of single or multi-isotope standard solutions. The efficiencies calculated for the chosen isotopes/energies are then extrapolated across the desired energy range of interest to produce an efficiency curve subsequently used to calculate the activities of isotopes present in samples whose gamma-ray emissions may or may not have been included in the suite of gamma-rays used to produce the original curve.

Although this approach is almost ubiquitous in practice and initially appears quite straightforward, the process includes a number of pitfalls and considerations that must be made to ensure accurate and precise results. Of some concern is the time and expense required to conduct such empirical calibrations, limitations imposed by the procedure regarding sample sizes and matrices, the generation of waste radioactive materials, availability of isotope solutions, uncertainties introduced as part of the curve fitting routines and the necessity to perform a variety of corrections to establish the link between the original calibration and the spectral information from the sample. Because of these factors, mathematical calibration of detectors has received attention in the past and the practicable implementation of such techniques has been the focus of much research in recent years as increases in computing power and other factors have ameliorated some of the problems associated with the techniques.

Mathematical Efficiency Calibration of HPGe Detectors

Perhaps the first attempt at mathematical procedures for the calibration of HPGe detectors was the semi-empirical procedure proposed by Moens et al (1981) and elaborated upon by Moens and Hoste (1983). A three stage procedure, the method involved determining the efficiency of axially positioned reference point sources and then determining the solid angle of the desired source-detector configuration relative to the solid angle as defined for the reference source and the detector. Establishment of this ratio allowed correction of the reference peak efficiency to provide a value for the desired source-detector configuration. Although the

method took into account various attenuation factors, the integrative process was extremely constrictive in relation to the range of geometries that could be calibrated for.

For low energy gamma-ray photons where the photoelectric effect is dominant the variation of efficiency with energy may be relatively well estimated using the product of the probability that the photon reaches the detector and the probability of its full absorption in the active volume of the crystal. Debertin and Helmer (1988) used this product to approximate the intrinsic efficiency of a Ge detector up to energies of approximately 70 keV, their model assuming either no interaction on passing through the detector or that all interactions contribute to the full energy photopeak. For higher energies where the two other interaction processes (Compton Scattering and Pair Production) become relevant, the situation is complicated and no fully satisfactory relationship has yet been devised. The semi-empirical suggestions of Freeman and Jenkin (1966) and Harvey (1970), which incorporate all three interaction processes, were unable to eliminate the need for empirical appraisal of the efficiency. This area has been revisited however with some success being reported for large volume HPGe detectors by Sudarshan and Singh (1991). A suite of methods that may offer some promise however are those most often generally described as Monte Carlo methods.

The general Monte Carlo method is a computational technique for the solution of problems that depend on probability in some manner and are typically used where exact descriptions of a process may be impracticable to solve by direct methods for some reason. The Monte Carlo process uses a stochastic model that describes the process in question and a set of high quality random numbers is then used to sample the probability distribution functions as described by the model being used. For situations of the type being considered here (i.e. radiation transport) the probability distribution functions are defined by the interaction cross sections of interest. Monte Carlo methods as applied to detector calibration are based on the simulation of individual photon histories and the tracking of photons from the source on their path through the detector and associated materials. As the photon passes through the detector material it undergoes a variety of interactions as described previously and produces electrons, positrons, and a full range of secondary photons such as Bremsstrahlung, fluorescence radiation and annihilation particles. The Monte Carlo method then tracks each of these as they pass through the detector. At each and every point of interaction the probability of each interaction occurring is calculated and the potential scattering angles are incorporated to ascertain the final result of the interactions. As all events are followed through to their final condition, the entire spectral distribution is recreated.

Although previously used in relation to NaI detectors, as detectors based on semi-conductors began to achieve predominance in the early 1970's the focus of attention had shifted towards the application of Monte Carlo methods to the calculation of efficiencies for such detector types. Wainio and Knoll (1966), De Castro Faria and Levesque (1967) and Peterman et al. (1972) reported on the application of Monte Carlo methods to the determination of efficiencies for semi-conductor detectors and as methods were developed, the methods were applied to non-point sources and detectors of various size and shape. Kushelevski and Alfassi (1975), Vano (1975), Somorjai (1975), Haase et al. (1993) and Winn (1993) report on various extensions and modifications of Monte Carlo methods towards problems relating to volumetric sources and different geometrical shapes such as Marinelli beakers.

Three factors limited the adoption of Monte Carlo based techniques between the 1970's and the mid 1990's. The first of these was related to the computing power necessary to produce acceptable results. Early attempts to calculate efficiencies using Monte Carlo methods could only produce values within approximately 10% of empirically derived values with relatively

high uncertainties that could not compete with the 4-5% uncertainty of empirical data. This was due to a number of factors but possibly of prime importance was the amount of time required to track the number of histories necessary to reduce the uncertainty in the calculated value to an acceptable level. The second factor relates to the detector itself. Despite modern manufacturing methods there remains significant uncertainty with respect to the active volumes of individual HPGe detectors. Although manufacturers do provide information regarding the physical size of individual detectors, uncertainties remain regarding insensitive regions of the detectors, in particular with respect to the dead layers at the contacts. These uncertainties can be reduced to some extent by the use of radiography to allow direct measurement of crystal dimensions and using a large number of simulations, which would have been unfeasible 30 years ago, to achieve the best agreement between the calculated and empirically derived efficiency values. The third factor relates to the nuclear data used in the calculations. Mass attenuation coefficients used in determining the interaction of photons with matter typically had uncertainties of the order of 5% up to the 1980's and the uncertainties could be considerably worse for factors related to photoelectric and Compton interactions. The quality of such data has improved in recent years as a result of concerted effort regarding the quality and evaluation of nuclear data.

While the above factors have limited the application of Monte Carlo techniques to HPGe calibration, a fourth less tangible but nonetheless valid problem exists in relation to the gap between the designers of codes for Monte Carlo computations and the end users or gamma spectrometrists. The practicability of implementing such codes has been limited due to the nature of the codes themselves, their dissemination and the manner in which information relating to geometries and detectors is entered. These inputs typically consist of lengthy, complex text files in which small mistakes may go unnoticed until the final phases of the simulation. In combination with the way the codes have been disseminated and the common requirement that the codes must be compiled by the end users themselves, it is noticeable that the implementation of such routines has most often been reported by academic/research institutions with access to the necessary skill base and the penetration of Monte Carlo methods into institutions dealing with, for example, monitoring, has been rather limited.

Implementation of a Monte Carlo Method for HPGe Calibration

The purpose of the work reported upon here was the practicable calibration of a HPGe detector for routine environmental measurements. Although a number of codes exist and recent years have seen the production of a variety of user interfaces for such codes, the software selected for this work was the Virtual Gamma-ray Spectrometry Laboratory (VGSL) developed under the auspices of the CTBTO (Planteda, 2002). This package is a Windows based interface to the well known MCNP5 code from the Los Alamos National Laboratory designed to allow the operation of the code for the purposes most gamma ray spectrometrists find most useful, namely the production of detection efficiency values for HPGe detectors. The detector used throughout this study was a standard p-type HPGe detector, the nominal specifications being FWHM of 1.9 keV at 1332 keV, relative efficiency of 40.7% and a peak to Compton ratio of 67:1. The detector was cooled using an electronic cryogenic unit as opposed to more conventional liquid nitrogen.

The primary inputs for implementing Monte Carlo routines are the detector and geometries descriptions. As the latter can be controlled by the end user, the description of the detector must by necessity be derived from manufacturer's specifications or by other means. Such specifications have often proved to be less accurate than required and to this end some work

was performed in relation to characterizing the detector used. This involved taking CAT scans (Fig 1.) of the detector and its housing and using measurements derived from these scans to check and modify the manufacturer's specifications.



Figure 1. CAT scanning of the chosen detector for further characterizing and one view of the resulting scan.

While X-ray analysis or CAT scans of the detector are useful for the checking of gross dimensions of the detector, important facets such as the front dead layer thickness cannot be determined by this method. To establish an appropriate measure of this factor primarily, certified point sources of with low uncertainties as to activity were used to empirically determine detector efficiency at a number of distances normal and coaxial to the detector face. Then the optimized dimensions of the detector were used, with the nominal dead layer value, to simulate the detectors response using the software for the detector-source configurations employed. The dead layer thickness of the detector model was then modified incrementally to produce good agreement between the empirical efficiencies and the simulated values. Once all relevant dimensions had been decided, the efficiency of the detector was simulated for a variety of point sources and then a series of volumetric sources to ascertain the applicability of the simulated efficiency values.

Results and Discussion

Measurement of the detector indicated some deviations between specification and measured values for some dimensions of relevance, primarily the detector-endcap distance which was modified to 8 mm from a nominal value of 4.5 mm and slight deviations with respect to crystal length and radius although these remained within the manufacturers tolerances. To achieve good agreement between the simulated and empirical data it was necessary to extend the front dead layer by 0.3 mm to 0.8 mm although it should be noted that this value probably constitutes an average value over the face of the crystal as it is unlikely that the layer is of constant (or even well defined) thickness. The differences between simulated efficiency and empirical data using nominal and optimized dimensions are displayed in Fig. 2. As can be seen, major discrepancies are observed for low energies using nominal values, the agreement being much better using the optimized values. Simulating the peak efficiency of a variety of point sources at various distances produced relatively good agreement (Fig. 3), in most cases

the deviation between simulated and empirical being within tolerances determined using the uncertainty of the source activity and uncertainties in peak area etc. The deviations for the photopeaks at 122 and 136 keV remain the worst at all distances although it should be noted that in all cases they were less than 5% and comparable with the deviations that could be expected from fitting any of the more common functions to empirical data in order to interpolate efficiency curves. Analysis of the situation seems to indicate that much of the discrepancy seems to occur as a result of differences in how a real system and a simulation view events. In a real life system it is possible that some counts, due to vagaries in pulse processing for example, may not be included in the peak area determination yet these counts may very well be included in the area in the simulated system or, conversely, a peak area determination may underestimate a peak area for whatever reason relative to the simulation. The simulation produced good agreement over a wide range of energies, Fig. 4 displaying the agreement between simulated and empirical efficiency values for a range of isotopes that include isotopes normally displaying quite strong coincidence summation events but positioned far enough away to negate the necessity for correction.

Simulations were also run for a series of certified volumetric sources of various densities (0.5 to 2.0), constructed primarily of water equivalent plastic with inclusion of other materials to affect the density. Good agreement was once again obtained for the available energies (Fig. 5.) the vast majority of the simulated efficiency values being within the tolerance allowed.

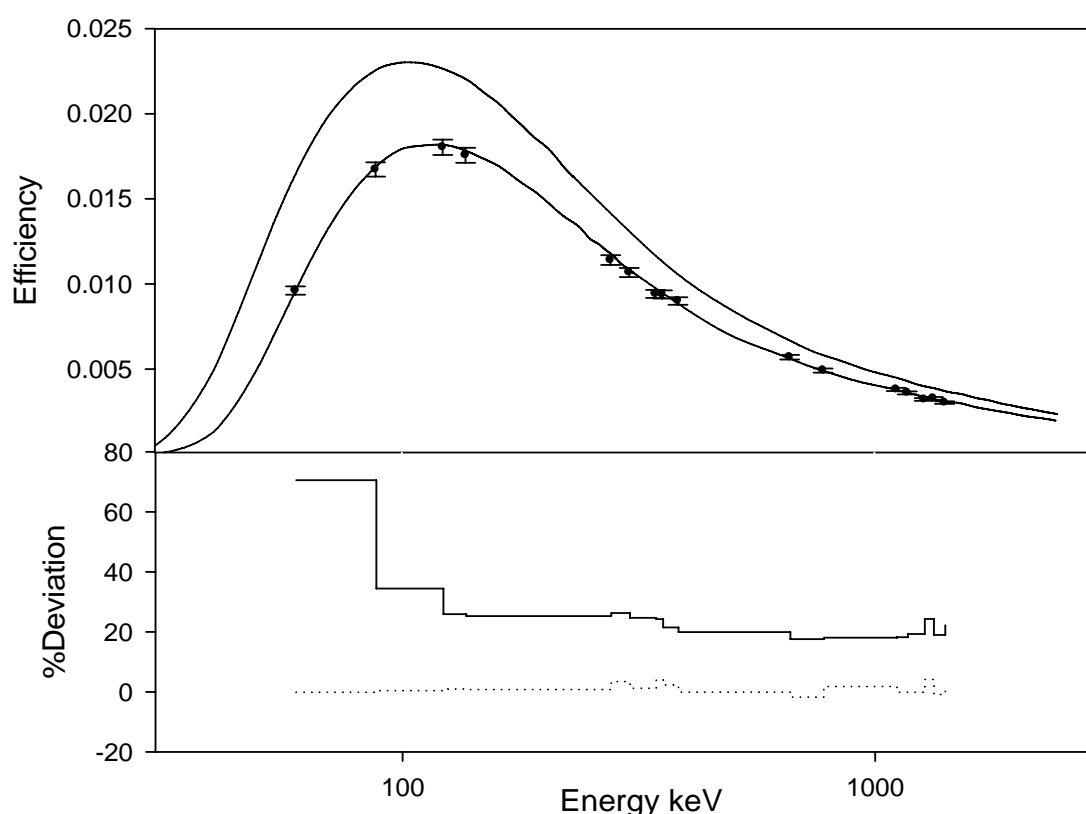


Figure 2. Plot of two Monte Carlo derived peak efficiency curves compared to empirically derived efficiencies (points) for the nominal detector dimensions (top curve) and optimised dimensions (bottom curve). The bottom graph represents the deviation (computed – empirical as a percentage of the empirical) for both nominal and optimised detectors as a function of incident photon energy.

Aside from the advantages of being able to calculate efficiency values with uncertainties comparable to those produced by empirical methods, deployment of Monte Carlo methods produce a series of secondary advantages. Determination of factors to correct for matrix or density effects are simplified without tedious calculation or the limitations implied in the application of transmission measurements and true coincidence summation corrections are simplified to a large extent. *A priori* determinations of detector efficiency for proposed geometries or matrices are possible for assessing optimum configurations of detector and geometry. As well as producing efficiency data, once the detector has been modeled it is possible to simulate spectra of any combination of radionuclides in any matrix once the nuclear data has been obtained in appropriate form, calculate coincidence correction factors for any appropriate isotope or calculate density/matrix effects for any sample type.

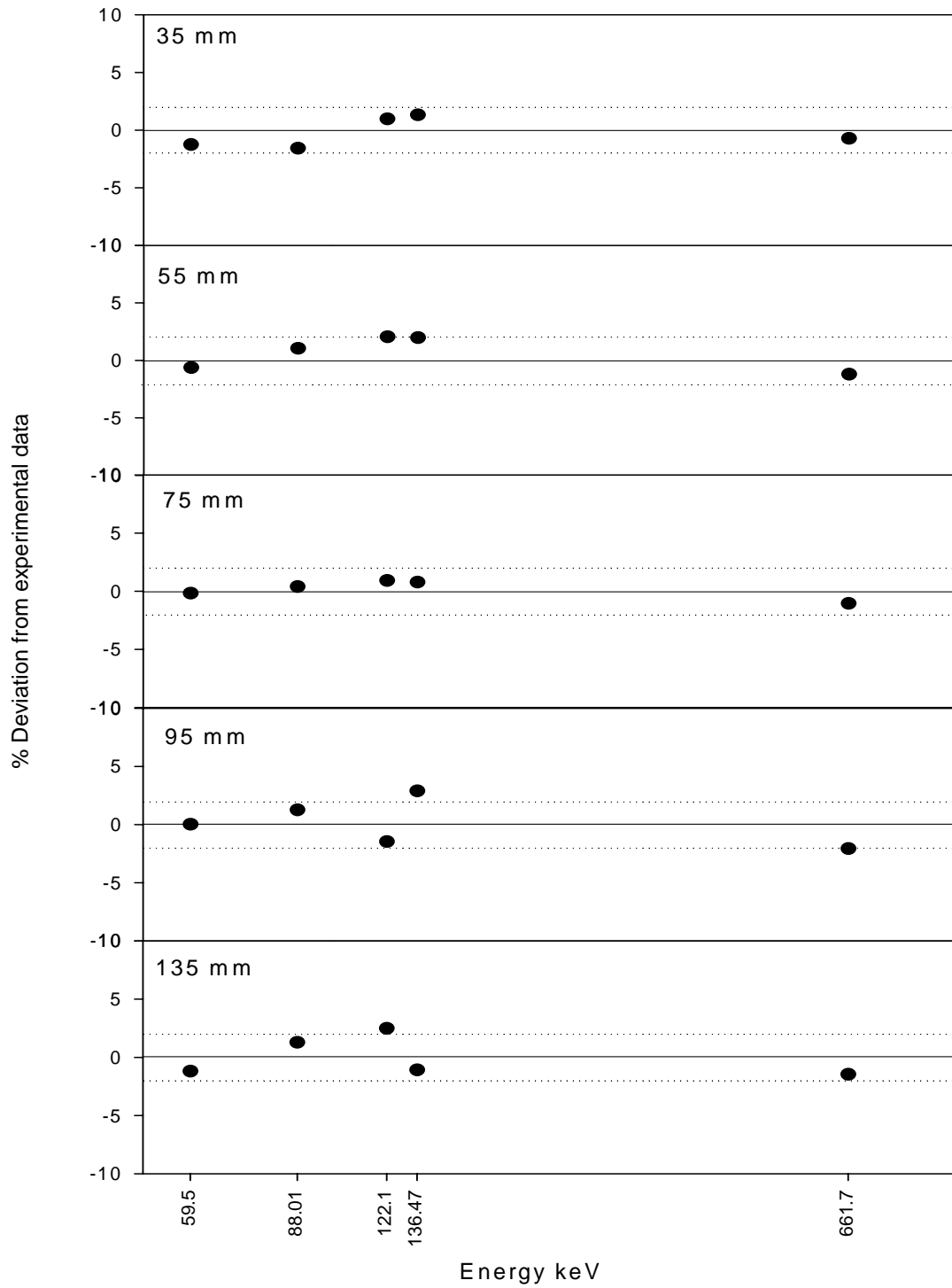


Figure 3. Deviations of calculated peak efficiency values from empirical data (calculated – empirical as percentage of empirical) for point sources at increasing distance from the detector endcap for various photon energies. Dashed lines are 1σ uncertainties in empirical values.

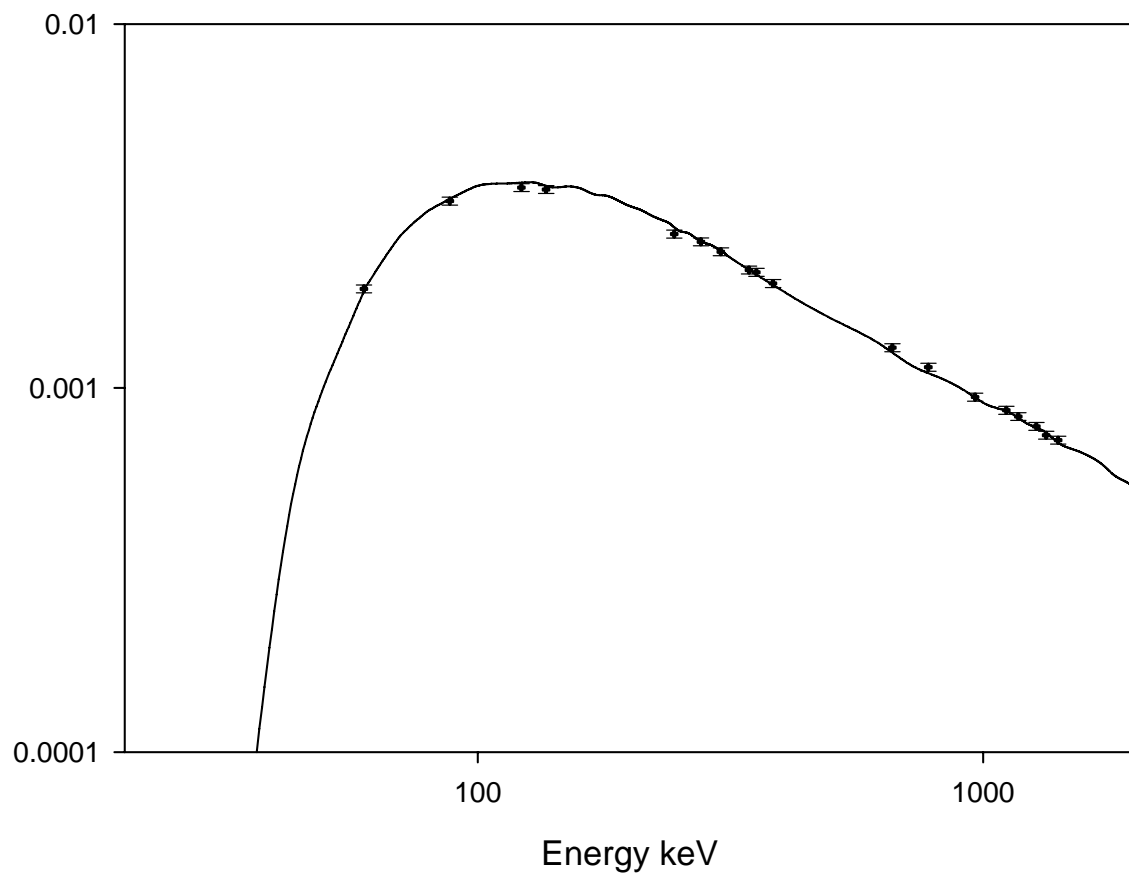


Figure 4. Calculated peak efficiency curve for point sources at a distance of 195 mm from the detector endcap and empirical data including emissions from ^{133}Ba , ^{60}Co , ^{22}Na and ^{152}Eu normally vulnerable to true coincidence summation.

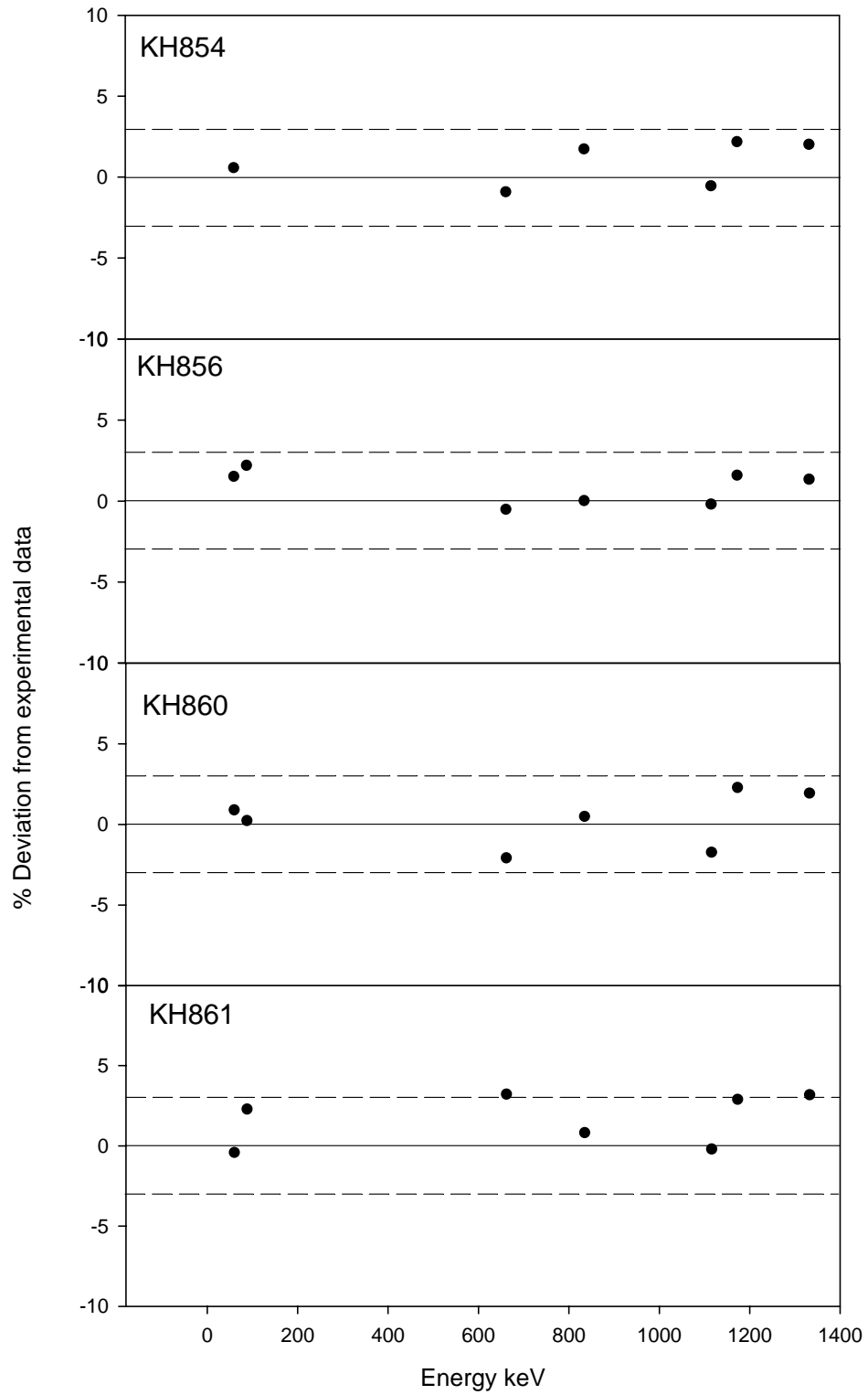


Figure 5. Deviations of calculated peak efficiency values from empirical data (calculated – empirical as percentage of empirical) for close geometry volumetric sources for various photon energies. Dashed lines are 1σ uncertainties in empirical values.

Conclusions

Recent advances in available computing power have allowed for the development of Monte Carlo codes as practicable alternatives to empirical calibration of HPGe detectors. Various interfaces have appeared in recent years facilitating the use of Monte Carlo codes by practitioners or institutions where previously the deployment of such routines would have required specialist skills. Testing of Monte Carlo codes indicates that such routines may offer a practicable alternative to traditional calibration routines with none of the drawbacks of such procedures and a concomitant set of advantages that appear to offer significant benefits while producing efficiency data that is comparable to that produced by empirical methods.

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Radioecology in Estonia - developments

Merle Lust¹, Enn Realo²

¹Estonian Radiation Protection Centre, Kopli 76, Tallinn, Estonia

²Institute of Physics, University of Tartu, Riia 142, Tartu, Estonia

Background

The history of radioecology in Estonia is relatively short, as its establishment as a discipline occurred only after the declaration of independence in 1991. There were several factors, which played important role in the developments - one of them was certainly the need to get to know better the situation with the environmental radiation and to get for example a better picture (true information) of the deposition of radionuclides in Estonia after the Chernobyl accident. NKS and co-operation with the Nordic countries has played an important role in the establishment of analysis facilities, as well as in multiple other developments. This paper will address the developments in two main institutions: Estonian Radiation Protection Centre (ERPC) and Institute of Physics, University of Tartu (UT). At the University of Tartu radioecology problems are studied in the nuclear spectroscopy laboratory of the Institute of Physics. Over the years it has become a training base for the environmental physicists in the field. The ERPC is the advisory body responsible for providing expertise and advice to the regulatory authorities in their work. Both the ERPC and the University of Tartu have laboratories where the following radioactivity measurement methods are available: low-background HPGe gamma spectrometry, low-energy HPGe gamma spectrometry, liquid scintillation spectrometry. The majority of measurement testing is performed on environmental samples.

Activities

Due to the low number of institutions participating in radioecology studies, the specialised knowledge base available in Estonia is quite scarce. However, the number of topics addressed in Estonia is comparatively long - beginning with the development of nuclear spectrometric methods and progressing to safety assessments for radioactive waste storage. Also of note are studies of artificial and natural radioactivity (including the migration of some radionuclides) in the soil, monitoring of artificial radionuclides in varying samples such as air, water, local fuels and ashes. All measurement information has been used in radiological modelling and in the improvement of safety assessment methods. During the last few years there has been a greater emphasis on quality assurance methods in sampling and analysis. Both laboratories have actively participated in international intercomparison exercises. An important section of radioecology activities within Estonia is the lecture series for students of the University of Tartu which provide information on environmental radioactivity and radiation protection through the UT Department of Environmental Physics and UT Türi College. The ERPC has additionally organized several introductory courses for other interested parties. The ERPC has also conducted several radon mapping programmes in co-operation with the Estonian Geological Survey. The following section will provide an overview of these activities.

Chernobyl deposition and modelling of the ^{137}C migration in soil

The first nationwide study carried out in 1991-1993 determined both geographic and depth distributions of deposited ^{134}Cs and ^{137}Cs in the Estonian soil [1,2]. According to the results of this study and of an independent air-borne scanning, radiocaesium originating from the 1986 Chernobyl accident with the mean deposition of 2 kBq m^{-2} was distributed in an extremely irregular fashion across the country. Estimates showed that approximately 2/3 of the total ^{137}C inventory in Estonia was to be found in Ida-Virumaa County, north-east Estonia. Deposits in this area reached 40 kBq m^{-2} .

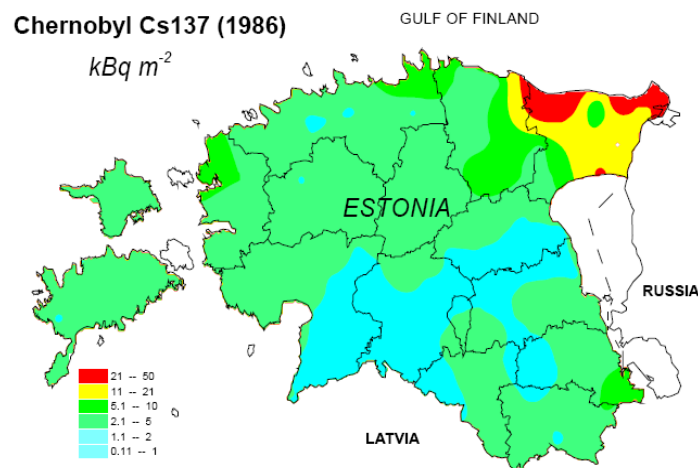


Figure 1. Chernobyl radiocaesium ^{137}Cs in soil. Recalculated from the ^{134}Cs inventory, the reference date: 26.04.1986.

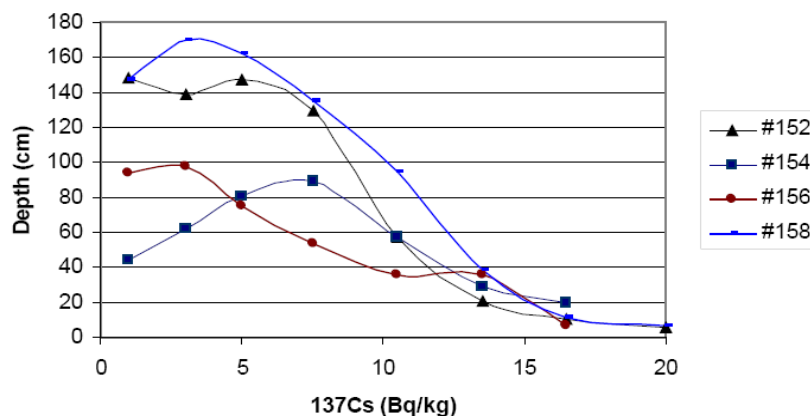


Figure 2. Depth distribution of radiocaesium in soil: 4 locations in NE Estonia (2001).

The region was revisited for soil sampling in 1998-2001 [3,4]. Undisturbed soil profiles to a depth of approximately 20 cm were collected, and 2-3 cm sample core slices analysed using a low-background HPGe gamma spectrometer. As in the previous study, both total depositions and depth distributions of the deposited ^{134}Cs (where possible) and ^{137}Cs activity concentrations were determined. Depth distributions of deposited activity showed considerable site-specific variations. In addition, it appeared that in comparison with the previous study, clear features of time-dependent migration to deeper soil layers were evident.

The preliminary modelling has demonstrated [4] a lognormal distribution with varying parameters that fits satisfactorily the determined depth-distributions of the Chernobyl radiocaesium activity concentration. An attempt was made to apply a single model to describe the observed time-dependent depth-distribution pattern over the time period of 1986-2001: in general, a slow migration rate, and consequently a strong retention of radiocaesium in the upper soil layers follow from the found weak time-dependence of the model fitting parameters.

Releases of radionuclides and assessment of doses from power production

Power production in Estonia is based mainly on the use of oil-shale in two large power plants in the north-east of Estonia. Oil-shale contains natural radionuclides, thus changes in the concentration of flyash during the power production process were measured. As a result, the estimates for radioactivity that is released with ashes to the atmosphere were derived [5-8]. A MATLAB-based software package was utilised and, in parallel, the CAP88PC software package was used for modelling of the long-term atmospheric dispersion and of geographical distribution of the deposited oil-shale ash radionuclides. Annual individual doses via external exposure, inhalation and ingestion pathways from oil-shale-based energy production were calculated, as well as the deposition amounts of flyash radionuclides in the surroundings of power plants. Total annual effective doses reach values up to 20 $\mu\text{Sv/a}$. The modelled ^{226}Ra deposition fluxes were used for a comparison with the measured ones. In the vicinity of power plants, depth-dependent activity concentrations of natural radionuclides in the soil have been determined by gamma spectrometry. As a result of the long-term operation of these power plants and ash deposition, an increased concentration of natural radionuclides has been found in the surrounding air and surface soil.

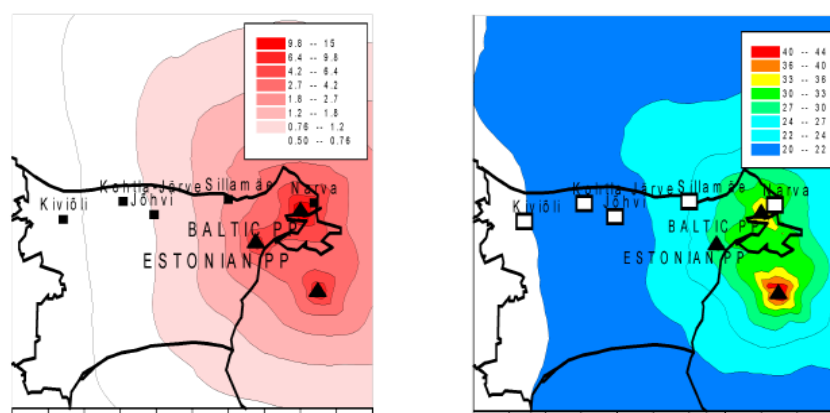


Figure 3. ^{210}Pb deposition flux ($\text{Bq m}^{-2} \text{a}^{-1}$) and annual doses ($\mu\text{Sv/a}$). The CAP88PC atmospheric transfer modelling software was utilised including local ash data (1995), radioactive ash contents and long-term local meteorology.

²¹⁰Pb in air and in soils

In north-east Estonia, ²¹⁰Pb activity concentrations in air and in natural surface soils have been determined. The region is characterized by varying Ra (U) activity concentrations [9] and is affected by a significant ash radionuclide deposition from two large oil-shale power plants. The activity concentrations of ²¹⁰Pb in surface air and depth distributions in natural surface soil profiles have been determined using HPGe gamma spectrometry [10,11]. In addition, weekly aerosol filter samples collected by a high-volume air sampler in Narva-Jõesuu have been used (Fig.4). The observed air concentrations of ²¹⁰Pb vary in the range of 100 - 1900 $\mu\text{Bq m}^{-3}$ with the average of 450 $\mu\text{Bq m}^{-3}$. The latter is comparable to 540 $\mu\text{Bq m}^{-3}$ determined for the Estonian inland air. The observed inverse correlation between the ²¹⁰Pb activity concentration in air and the precipitation rate confirms the role of wet deposition in removing lead from the air.

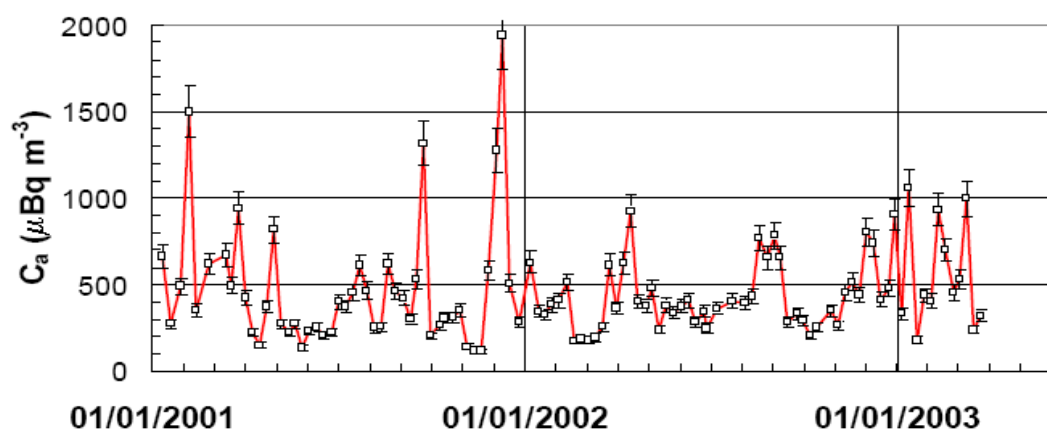


Figure 4. Weekly ²¹⁰Pb activity concentration ($\mu\text{Bq m}^{-3}$) in near-surface air, Narva-Jõesuu, 2001-2003.

In the study [12] two groups of low-Ra soil profiles from sites have been used for analysis: (a) far from the power plants and (b) near the power plants. In addition, the ²²⁶Ra activity concentrations in the range 14 – 35 Bq kg^{-1} and radon emanation coefficients in the range of 0.1 - 0.5 have been determined.

Depth-dependent concentrations of unsupported ²¹⁰Pb have been fitted using a 1D diffusion model and a compartmental transfer model to determine the transfer parameters in soil. About 2/3 of the unsupported ²¹⁰Pb inventory is bound within the upper 0-10 cm soil horizon. The average annual atmospheric deposition fluxes of ²¹⁰Pb with a mean value of 164 $\text{Bq m}^{-2} \text{a}^{-1}$ and an average deposition velocity of 12 mm s^{-1} have been found for natural locations, group (a). The maximum fluxes with a mean value of 321 $\text{Bq m}^{-2} \text{a}^{-1}$ have been found near the power plants (Fig.5). The results support an assumption about the technologically enhanced ²¹⁰Pb concentration in air near the power plants in north-east Estonia.

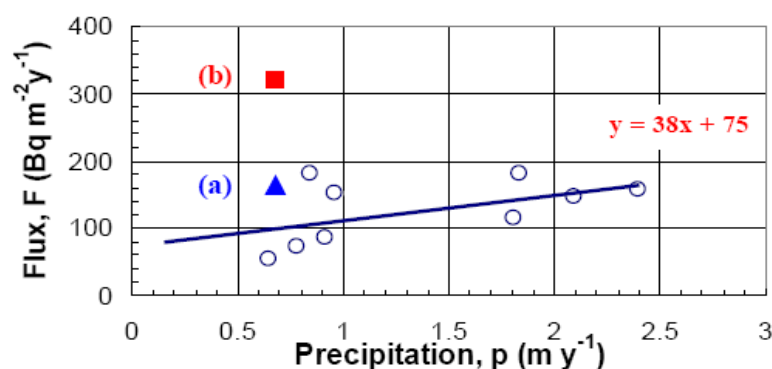


Figure 5. Comparison of ^{210}Pb deposition fluxes from atmosphere to the ground, F , vs precipitation rates, p : (a) natural sites, (b) sites near power plants. O – fluxes determined for other countries in Europe (references in [8, 10-12]).

Environmental radiation monitoring

In the framework of the monitoring program for environmental ionizing radiation in Estonia, the total external gamma dose rate, radioactivity of airborne particles and aerosols, content of artificial radioisotopes in river and drinking water, as well as in milk produced in Estonia and in the total diet, are monitored. 10 automated stations monitor the total external gamma dose rate. Data measured by this network is published on the webpage of the Estonian Radiation Protection Centre [13]. According to the monitored gamma spectra used for the evaluation, the gamma-dose is primarily due to natural radioisotopes. The concentration of Cs-137 in the atmosphere caused by the global fallout is negligible. However, in the north eastern part of Estonia, peak activity values have been caused by the re-suspension of the Chernobyl deposition to the atmosphere. Fig. 6 presents an example of the time-dependent Cs-137 concentration in air measured by the automatic station in Narva-Jõesuu (NE Estonia) in 1997-2003. In Estonia, Narva-Jõesuu is the monitoring station closest to the Sosnovõi Bor Nuclear Power Plant. However, in the measured spectra, other artificial radioisotopes indicating a substantial release of radioactive material from the nuclear power plant have not been detected.

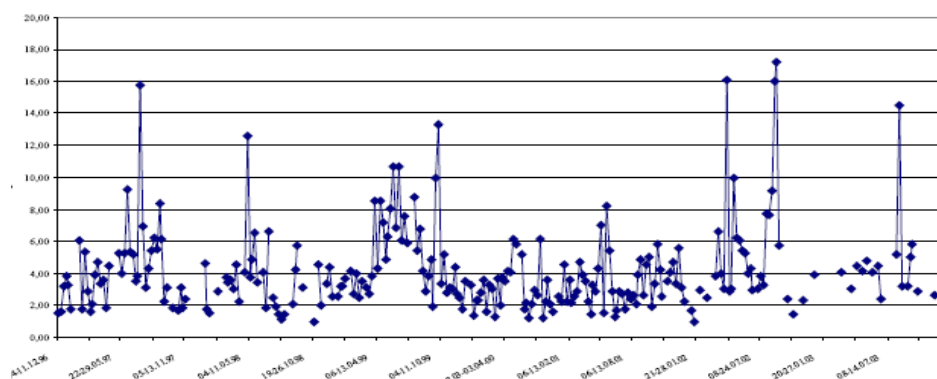


Figure 6. Radiocaesium concentrations ($\mu\text{Bq}/\text{m}^3$) in air in Narva-Jõesuu, 1997-2003.

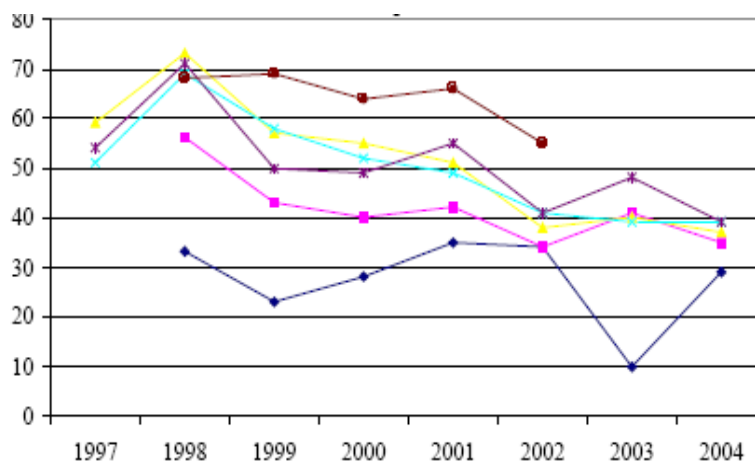


Figure 7. Annual radiocaesium concentrations (Bq/m³) in water (6 locations), Gulf of Finland.

Samples from the marine environment have been collected by five monitoring stations situated in Estonia under the framework of the HELCOM MORS program. There is a generally decreasing trend of the ¹³⁷Cs activity in the surface water of Gulf of Finland, which can also be seen in Fig. 7.

Conclusions

Taking into account that radioecology in Estonia is only slightly over a decade in age, our achievements have been quite remarkable. However, there are several difficulties similar to those typical of neighbouring states, e.g. lack of interest and insufficient challenges for students wishing to specialize in the field. Nonetheless, problems remain to be solved. In the near future, for example, the following issues should be addressed: additional studies connected to the drinking water directive, continuation of the public dose estimation, future safety assessment studies, etc.

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Nordic Collaboration on the use of Mass-Spectrometers for the Analysis of Radioisotopes (NorcMass)

Per Roos¹ Ulrika Nygren² Christina Greis³ Lindis Skipperud⁴ Håkan Pettersson⁵

- 1) Risø National Laboratory, Dk-4000 Roskilde, Denmark
- 2) Swedish Defence Research Agency, FOI, Umeå, Sweden
- 3) Department of Natural Sciences, Örebro University, Sweden.
- 4) Department of soil- and water sciences. Agricultural University of Norway, Norway.
- 5) Department of Radiation Physics, Linköping University, Sweden

Introduction

The ICP-MS technique has during the last 20 years become increasingly important in the analysis of long-lived radioisotopes. Although useful both with respect to rapid measurements as well as performing highly accurate isotope ratio determinations and analysis of extremely low levels the technique requires careful attention. The need for routine use in producing data often overshadows the need to look into the many parameters governing the quality of data. This is particularly important in the analysis of long lived radioisotopes due to the often extremely low concentrations and/or the need to perform isotopic ratio determinations with high accuracy of extreme ratios (U&Th isotopes). The purpose of the NorcMass project was to bring together Nordic scientists with a common interest in analysing Pu and U isotopes using ICP-MS and to stimulate ideas of how to better perform such measurements. More specifically the purpose was to identify and suggest solutions to existing problems within high precision isotope ratio measurements (of U and Pu) and analysis of ultra low levels of long lived radioisotopes performed on single collector instruments. Due to the lack of existing low-level reference materials with certified concentrations a production of a Nordic isotope ratio reference material (for $^{240}\text{Pu}/^{239}\text{Pu}$, ^{237}Np and $^{234}\text{U}/^{235}\text{U}/^{238}\text{U}$) was performed. This presentation deals with the production and measurements of Pu-isotopes on the NorcMass reference material.

Why a low-level reference material?

Although there are several reference materials available (eg IAEA) few have $^{239}\text{Pu}/^{240}\text{Pu}$ data and almost none have $^{237}\text{Np}/^{239}\text{Pu}$ -data. Those who have (eg IAEA-384) have very high concentrations and are not useful for testing analytical methods designed for low-level measurements. They are also often of different composition (eg IAEA-384 being a coral sample) than ordinary soils and are usually distributed in small amounts preventing testing at the 10-50g analytical scale. The aim of the reference material in the NorcMass-project was to produce a low-level ($<<1\text{mBq/g}$ of ^{239}Pu) sample of sufficient amount to allow individual laboratories to perform several tests without risk of using up the material

Even though the major use of ICP-MS instruments is to directly analyse elements in a variety of matrixes without any chemical purification this approach is not valid for actinides at environmental concentrations. Actually, the radiochemical methods normally used when separating Pu and Np for radiometric determination is usually not sufficiently good for ICP-

MS. The reason for this is the combination of a relatively low mass resolution and the presence of uranium in all samples. Normally the mass ratio between U and Pu is in the order of 10^6 - 10^8 in soil/sediments which is far too high to be neglected in the ICP-MS measurements. An example of how uranium disturbs the Pu and Np analysis in an ordinary ICP-MS analysis is shown in figure 1. Even though radiochemical methods were applied to remove uranium in this sample (for the original purpose of analysing Pu with alpha spectrometry) the removal was not sufficient for ICP-MS.

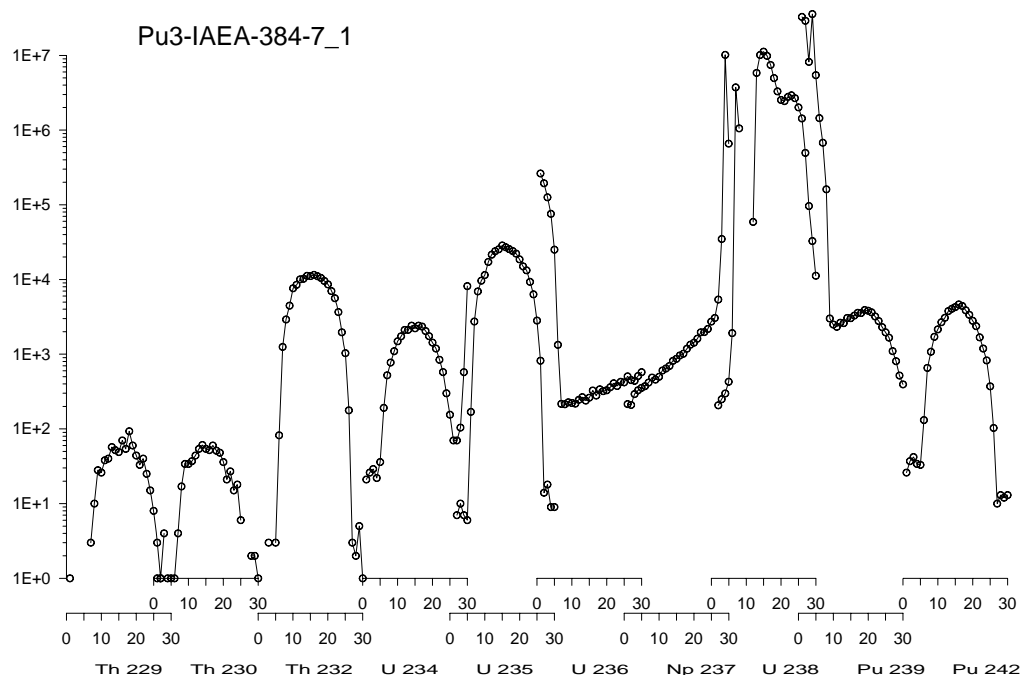


Figure 1. Mass-spectrum of a radiochemically purified sample of the reference material IAEA-384 (coral from Mururoa). The purified sample was analysed by alpha spectrometry, then stripped from the disc and measured by ICP-MS. Pu-242 was used as a yield determinant.

The use of appropriate radiochemistry is thus vital in order to be able to perform accurate Pu or Np analysis with ICP-MS. The use of a low-level soil/sediment with known concentrations is therefore of great help in testing the procedure

Origin and preparation of the reference material

The top 10cm of 2mm sieved soil from 12 different Danish locations collected during 2003 were pooled at Risoe Research Centre, Denmark. The Soil was further sieved through 0.6 and finally through a 0.4 mm sieve and coarsely mixed by hand. Following a single homogenization of all soil for 30 minutes in a large volume mixer a total of 17 kg soil remained. A second batch of 8.5 kg soil with the same origin was prepared simultaneously but was mixed separately and therefore not added to the larger soilsample. It is however expected that this smaller soilsample have an almost identical composition as the larger one.

Analysis

A total of 512.9g of the sieved and mixed reference soil was ashed in a large glass tray at 550 °C for 12h. The weight of ashed soil was recorded and a total of 47.3g ash subsampled for Pu-analysis by alpha spectrometry. The remaining sample was leached in 500 ml *aqua regia* over night, filtered through GF/A glass fiber filters and the leached ash thoroughly washed repeatedly with 1M HCl until a total volume of 1.5 litre. After precipitation using ammonia, amphoteric elements were removed by 6M NaOH and the final Fe(OH)₃ precipitate washed two times with weak ammonia. Plutonium was separated from the soil by a two-column ion exchange method

The so far purified sample was diluted to a total of 32.98g in 1M HNO₃. From this 0.488g was taken (SampleR1) for U-analysis and two 10g samples (10.030g and 10.008g) was transferred to plastic bottles and sent to two of the other participating laboratories together with 1.2kg of the original reference soil. The remaining 12.45g of solution was finally purified by a TTA-extraction procedure and the final solution diluted to 5ml with 1 M HNO₃. From this 0.5ml (Sample R2) was taken for U-Pu analysis. The remaining 4.5ml was diluted to 10ml (Sample R3) by deionized water (ELGA-water) and used for the Pu-isotopic measurement. This final sample thus represents a total of 72.77g ashed soil or 78.46g dry soil.

At Risoe a PlasmaTrace2 (PT2) HR-ICP-MS with an ultrasonic nebuliser was used to provide a first check on concentrations and homogeneity of the material. Analysis was performed for uranium and plutonium in the three samples R1-R3. Tuning of the instrument before analysis was performed using a 0.5 ppb uranium solution. The reference soil sample (R3) in a 10 ml volume was continuously pumped into the USN and 15 separate measurements of ²³⁸U, ²³⁹Pu and ²⁴⁰Pu was performed in the peak jumping mode.

Results

The total peak counts for each isotope in sample R3 is given in Table 1 below.

Table 1. Raw data counts for each isotope measured.

Run No.	²³⁸ U	²³⁹ Pu	²⁴⁰ Pu	Atom ²⁴⁰ Pu/ ²³⁹ Pu
1	1223	5000	911	0.18220
2	1180	5310	990	0.18644
3	1143	5368	923	0.17194
4	1082	5196	955	0.18380
5	1135	5111	964	0.18861
6	1058	5185	957	0.18457
7	1069	5140	979	0.19047
8	1117	5174	938	0.18129
9	980	4937	975	0.19749
10	1081	5004	967	0.19325
11	1031	5013	959	0.19130
12	991	5080	968	0.19055
13	1012	4985	894	0.17934
14	958	5179	966	0.18652
15	1020	4899	948	0.19351
Atom ratio:	0.1868±0.0065 (1 std.dev)			

The analysis of sample R1 showed that the levels of uranium already was sufficiently low to make the plutonium analysis without having any large corrections to the mass 239 due to tailing and UH^+ . Sample R2 showed that after the TTA-extraction the uranium was removed to the extent where the dominating source of uranium is the chemicals themselves. This was concluded from nearly identical uranium amounts found in the R2-sample and the 8M HNO_3 used for backextraction from TTA. The fraction of counts in mass 239 due to the presence of uranium was measured to be $3.2 \cdot 10^{-5}$ which means that the correction to mass 239 in this case was less than one count.

Due to the low count rates no dead-time corrections was applied to any of the isotopes. A standard U-solution (NBL-U112a) was used to monitor mass-fractionation. This was found to be insignificant and no corrections have been applied to the raw data.

Alpha-spectrometric measurements

Four subsamples of ashed material used for alpha spectrometry were separated for plutonium on a single ion exchange column. The samples were counted for 15 days on PIPS-detectors. Results are presented in table 2.

Table 2. Results from alpha spectrometric analysis of Pu on 10g samples of the Noremass reference material.

Sample	Mass of dry soil	$^{239+240}\text{Pu}$ [mBq/g]	$^{238}\text{Pu}/^{239+240}\text{Pu}$
K1	12.38	0.237±0.009	0.032±0.007
K2	9.60	0.249±0.012	0.037±0.009
K3	18.67	0.220±0.007	0.031±0.005
K4	10.41	0.247±0.011	0.032±0.007
Average		0.238±0.0135	0.033±0.0027

Unfortunately there is an inverse correlation between Pu-activity and sample mass, which indicates incomplete isotopic exchange between tracer and analyte.

The samples were measured before equilibrium had been attained between ^{228}Th and ^{224}Ra so even if the very small ^{238}Pu peak was visibly separated from the tiny ^{228}Th peak the $^{238}\text{Pu}/^{239+240}\text{Pu}$ values should be treated with some care.

Measurements of ^{237}Np

The reference material was also subject to ^{237}Np analysis. Eight subsamples of 10g each were analysed for ^{237}Np and Pu-isotopes using ^{242}Pu as tracer for both elements. The results are given in table 3 below.

Table 3: Summary of determinations of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in the Normass reference soil by participants.

Sample	Atom $^{240}\text{Pu}/^{239}\text{Pu}$	Atom $^{237}\text{Np}/^{239}\text{Pu}$	$^{239+240}\text{Pu}$ via ICP-MS	$^{239+240}\text{Pu}$ via alpha spectrometry
Soil-1	0.1908	0.283	0.240	0.251
Soil-2	0.1818	0.362	0.246	0.239
Soil-3	0.1969	0.329	0.239	0.238
Soil-4	0.1965	0.318	0.238	0.235
Soil-5	0.1895	0.294	0.227	0.231
Soil-6	0.1887	0.304	0.239	0.238
Soil-7	0.1928	0.316	0.240	0.246
Soil-8	0.1790	0.371	0.240	0.233
Atom ratio:	0.1895 ± 0.023	0.322 ± 0.01	0.238 ± 0.018	0.238 ± 0.087

In table 4 results are presented for measurement on the Pu-solution prepared from the reference material. Three laboratories performed the analysis.

Table 4. Results of $^{240}\text{Pu}/^{239}\text{Pu}$ on leached soil solution from the Normass reference material prepared and pre-separated at Risoe. A comparison with published $^{240}\text{Pu}/^{239}\text{Pu}$ data for a Danish soil is also given.

Participant	$^{240}\text{Pu}/^{239}\text{Pu}$
Lab #1	0.187 ± 0.004
Lab #2	0.193 ± 0.004
Lab #3	0.187 ± 0.006
Kelley et.al. 'Roskilde soil' (Sci.Tot. Env. 237/238, 1999, 483-500)	0.1904 ± 0.0012

Determinations of $^{240}\text{Pu}/^{239}\text{Pu}$ on the Normass soil.

In table 5 data for $^{240}\text{Pu}/^{239}\text{Pu}$ ratios on the reference soil prepared in the individual laboratories are presented. In this case sample mass typically ranged between 5-20g and was thus an order of magnitude less than what was used in the pre-separated soil solution supplied along with the reference soil.

Table 5. Participating laboratories results on $^{240}\text{Pu}/^{239}\text{Pu}$ ratios when analysing the Normass reference soil.

	$^{240}\text{Pu}/^{239}\text{Pu}$
Lab #1	0.215 ± 0.021
Lab #2	0.11 ± 0.02
Lab #3	0.189 ± 0.0062

Comments to the results

The pre-separated soil solution prepared at Risoe before sending out to the two other laboratories contained Pu from approximately 100g soil. This is more than the individual laboratories used in their own leaching/dissolution of the soil and therefore the Pu-signal

intensity in this sample was higher. Furthermore, the solution was pre-purified at Risø and the final sample might therefore be expected to be cleaner than the individual soil leach/dissolution samples performed. The difference in results between the solution and raw soil samples is therefore due to both sample size (signal intensity) and sample purity. Only one of the laboratories performed the ^{237}Np analysis so no comparison can be done on this subject.

Even if the reference material was expected to be homogenous it should be kept in mind that it was pooled from several Danish locations and there may thus be a risk, even though small, that $^{240}\text{Pu}/^{239}\text{Pu}$ ratio as well as the $^{237}\text{Np}/^{239}\text{Pu}$ ratio varies in the sample due to either incomplete mixing or due to the presence of individual fall-out particles.

Variation in the fall-out ratios are well known and for the Nordic countries published values are given in the table below.

Table 6. Expected variation in fallout ratio $^{239}\text{Pu}/^{240}\text{Pu}$ & $^{237}\text{Np}/^{239}\text{Pu}$ in the Scandinavian countries. From Kelley et.al. (Sci.Tot. Env. 237/238, 1999, 483-500).

	$^{240}\text{Pu}/^{239}\text{Pu}$	$^{237}\text{Np}/^{239}\text{Pu}$
Reykjavik	0.1801 ± 0.0006	0.474 ± 0.01
Bergen	0.1738 ± 0.0006	0.391 ± 0.009
Oslo	0.1790 ± 0.0007	0.447 ± 0.009
Roskilde	0.1904 ± 0.0012	0.531 ± 0.013

Age determination of plutonium using inductively coupled plasma mass spectrometry

Ulrika Nygren,^{a,b,#} Henrik Ramebäck^a and Calle Nilsson^a

^aSwedish Defence Research Agency (FOI), Division of NBC Analysis, SE-901 82 Umeå, Sweden

^bDivision of Chemistry, Luleå University of Technology, SE-971 87 Luleå, Sweden

[#]Corresponding author, e-mail ulrika.nygren@foi.se

Introduction

In the analysis of the origin and history of nuclear material (uranium and plutonium), parameters such as isotopic composition and time since the last purification (the age) of the material are of vital importance. Areas where such determinations are, or can be applied include, e.g., nuclear forensics (illicit trafficking) and as a tool in a verification regime for a future fissile material cut-off treaty (FMCT). The age of plutonium material can be determined by analysing the relationship between the isotopes of Pu and their daughter nuclides.

The ratio $^{241}\text{Pu}/^{241}\text{Am}$ has been utilised in the age determination of plutonium using γ -spectrometry.^{1,2} The benefit of using γ -spectrometry is the possibility to perform non-destructive analysis, but the technique suffers from low sensitivity. For the determination of plutonium isotopes where the γ -emission probabilities are low, measurable quantities are in the mg region and more.³ Wallenius et al. have in a series of publications, exploited different types of mass spectrometers for age determination of Pu. They applied secondary ion mass spectrometry (SIMS) for the dating of single plutonium particles,⁴ and thermal ionisation MS (TIMS) and quadrupole-based inductively coupled plasma MS (ICP-QMS) for the age determination of plutonium reference materials and plutonium pellet samples.^{5, 6, 7} The age determinations were based on the $^{238}\text{Pu}/^{234}\text{U}$, $^{239}\text{Pu}/^{235}\text{U}$ and $^{240}\text{Pu}/^{236}\text{U}$ ratios, and the results were generally in good agreement with the reported ages. However, some discrepancies were identified, e.g., too low determined ages from the $^{238}\text{Pu}/^{234}\text{U}$ ratio and too high from the $^{239}\text{Pu}/^{235}\text{U}$ ratio when analysing PuO_2 and MOX particles using SIMS.^{4, 7} This was due to interference from ^{238}U in the determination of ^{238}Pu causing a too high determined $^{238}\text{Pu}/^{234}\text{U}$ ratio, and contamination from ^{235}U causing an underestimated $^{239}\text{Pu}/^{235}\text{U}$ ratio. It should also be mentioned that the ratio $^{242}\text{Pu}/^{238}\text{U}$ was not utilised in these studies due to the high sensitivity to contamination from ^{238}U on age determination based on this ratio.

A possibility to avoid the problem of uranium contamination is to utilise the ratio $^{241}\text{Pu}/^{241}\text{Am}$. Determination of this ratio using mass spectrometry would result in more sensitive analysis compared to γ -spectrometry. However, since these nuclides are isobars, a thorough chemical separation prior to analysis will be required. The aim of this study was to develop a method for age determination of plutonium material based on chemical separation and the analysis of ^{241}Pu and ^{241}Am using inductively coupled plasma mass spectrometry.

Experimental

The age determinations were performed on two plutonium reference material solutions, IRMM081 (^{239}Pu) and IRMM083 (^{240}Pu) (both Institute of Reference Materials and Measurements, Geel, Belgium), and on one sediment sample from the Enewetak Atoll, Marshall islands (reference material IAEA367, International Atomic Energy Agency, Vienna, Austria).

Sample pre-treatment

Lithium borate fusion: The sediment reference material was dissolved using lithium borate fusion. Approximately 0.5 g sample was weighed and mixed with 1.5 g Spectroflux100B in a graphite crucible, and 0.2 ml of a spike solution containing ^{243}Am ($55 \text{ pg}\cdot\text{g}^{-1}$) and ^{242}Pu ($240 \text{ pg}\cdot\text{g}^{-1}$) was added to the sample (this amount was also weighed to the accuracy of 0.1 mg, which was the accuracy used for all sample and spike weighing in this work.). Samples were then fused at 1050°C in a furnace for 10-15 minutes and transferred (after cooling) to a glass beaker containing 50 ml 1.4 M HNO_3 . The melt was dissolved under heating ($\geq 100^\circ\text{C}$) and stirring and the solution was allowed to evaporate to half of the initial volume. In order to prevent precipitation of silicic acids during the following separation procedure, 0.2 g PEG2000 was added to flocculate the dissolved Si. The samples were left at room temperature overnight and then the precipitate was removed by filtering. This resulted in a HNO_3 concentration of approximately 2.8 M which was suitable for the following solid phase extraction procedure.

Solid phase extraction separation: The separations of Am and Pu were performed using solid-phase extraction. Material not pre-treated by fusion, i.e., IRMM083, was added to a load solution of 5 ml 3 M HNO_3 together with 0.2 ml spike solution as described above. Aliquots of 0.1 ml IRMM083 diluted to $11.8 \text{ ng } ^{240}\text{Pu g}^{-1}$ were used for the analysis. A red-ox procedure, first described by Moore and Hudgens⁸, was used to adjust Pu to Pu(IV) while keeping Am as trivalent. First, Pu was reduced to the trivalent state by adding $\text{NH}_2\text{OH}\cdot\text{HCl}$ to a concentration of 15 mg ml^{-1} and heating at 90°C for 5 minutes. After cooling to approximately 40°C , NaNO_2 was added at a concentration of 18 mg ml^{-1} to oxidise Pu(III) to Pu(IV).

1 ml TEVA resin and 2 ml TRU resin were added to 10 ml plastic columns. The columns were mounted in series with TEVA upstream of TRU and the resins were conditioned with 5 ml 3 M HNO_3 . The sample was loaded on the resins and the beaker was rinsed with 3 ml 3 M HNO_3 , which was added to the columns. Under these conditions, Pu(IV) is retained on the TEVA resin while Am(III) is retained on TRU.⁹ The columns were then separated from each other and the TEVA column was rinsed with 20 ml 3 M HNO_3 to remove uranium. Plutonium was then eluted using 5 ml 0.1% oxalic acid or 0.1% HEDPA¹⁰ (the elution using 0.1% oxalic acid was found to be incomplete, probably due to precipitation of Pu-oxalate on the resin, why the eluent was changed to 0.1% HEDPA). The TRU-column was mounted on top of a column containing 0.5 ml Ln-resin that had been pre-conditioned with 1 ml 0.01 M HNO_3 . 20 ml 0.01 M HNO_3 was then added to the columns to transfer americium from TRU to the Ln-resin. Americium was eluted from the Ln-resin using 3 ml 0.5 M HNO_3 .

Instrumentation

Measurements were performed using an Element2 ICP-SFMS instrument (Thermo-Finnigan, Bremen, Germany). The instrument was equipped with a semi-demountable Fassel quartz torch and a CD-2 Guard Electrode. Sample introduction was performed using a conical nebuliser (GlassExpansion, Melbourne, Australia) and a Scott-type double-pass spray chamber. Self-aspiration, resulting in a sample flow rate of approximately 0.2 ml min^{-1} , was used throughout the investigation. The guard electrode was grounded and the automatic dead time correction inactivated. Torch position, lens parameters and nebuliser gas flow rates were optimised daily to obtain a maximum sensitivity for ^{238}U . All measurements were performed using low resolution ($m/\Delta m = 300$).

Calculation of the age of Pu-material

The determination of the age of Pu-material was based on the mother/daughter relationship between ^{241}Pu and ^{241}Am . This relationship is described by the following equation

$$t = \frac{1}{\lambda_1 - \lambda_2} \cdot \ln \left[1 - \left(\frac{\lambda_2}{\lambda_1} - 1 \right) \frac{N_2}{N_1} \right]. \quad (1)$$

In these equations, t is the age of the material, λ_1 and λ_2 are the decay constants of ^{241}Pu and ^{241}Am , respectively, and N_1 and N_2 are the amounts of ^{241}Pu and ^{241}Am at the time of analysis.

Results and discussion

The determined isotope amount ratio of $^{241}\text{Am}/^{241}\text{Pu}$ and the corresponding ages are shown in Table 1. Determinations were performed on 3 to 5 sub-samples of each material, plus one γ -spectrometric analysis of IRMM081 and IRMM083. No γ -spectrometric dating could be performed on IAEA367 due to the low concentrations of the analytes in this material. The table also specifies the reference ages of the materials. These are based on the date of purification for the IRMM materials¹¹, and the time period when the US conducted nuclear weapons testing at the Enewetak Atoll was used as reference date for the Pacific Ocean sediment.¹² As can be seen from the table, the ages determined by ICP-SFMS correspond well (as indicated by the expanded standard uncertainties, $k=2$) with the reference ages as well as the γ -spectrometric determinations.

Table 1. Results of the age determinations. The results from the γ -spectrometric measurements are included as well (numbers in italics). The determined age of IRMM081 has been corrected for an off-set age of 2.4 y due to residual of ^{241}Am after purification of the plutonium material.

Sample	Analysis date	$^{241}\text{Am}/^{241}\text{Pu}$	Determined age / y	Corrected age / y	Reference age / y	Reference date
IRMM081:1	2005-04-04	3.69 ± 0.20	32.5 ± 0.9	30.1	29.9	May 1975
IRMM081:2	2005-04-12	3.88 ± 0.34	33.4 ± 1.5	30.9		
IRMM081:3	2005-04-12	4.04 ± 0.40	34.1 ± 1.8	31.7		
IRMM081:4	2005-04-12	3.83 ± 0.38	33.1 ± 1.7	30.7		
IRMM081:5	2005-04-12	4.10 ± 0.45	34.3 ± 1.8	31.9		
<i>IRMM081:γ</i>	<i>2005-04-20</i>	<i>3.36 ± 0.94</i>	<i>31 ± 4.6</i>	28.6		
IRMM083:1	2005-03-14	0.741 ± 0.018	11.6 ± 0.2		11.7	1993-06-15
IRMM083:2	2005-03-21	0.753 ± 0.018	11.7 ± 0.2			
IRMM083:3	2005-04-12	0.756 ± 0.018	11.8 ± 0.2			
<i>IRMM083:γ</i>	<i>2005-04-30</i>	<i>0.79 ± 0.23</i>	<i>12.1 ± 2.7</i>			
IAEA367:1	2005-02-23	10.4 ± 2.5	51.5 ± 4.7			1948-1958
IAEA367:2	2005-02-23	10.0 ± 3.4	50.6 ± 6.6			
IAEA367:3	2005-02-23	10.3 ± 3.8	51.3 ± 7.1			

A condition for age determination based on this kind of mother/daughter nuclide ratio is that there is no amount of daughter nuclide present at $t=0$. However, it was found that IRMM081 contained ^{241}Am at a concentration of $250 \mu\text{g g}^{-1}$ directly after purification. Calculations based on this concentration and the concentration of ^{241}Pu at the time of separation using Eq. 1 results in an off-set in the age of 2.4 years for which the determined age in Table 1 has been corrected. It should be noticed that the purification process used for IRMM081 at the reference time $t=0$ was electrolysis, while anion-exchange of Pu(IV) was used for IRMM083. Hence, it seems as if Am at least to some extent has not been separated from Pu in the electrolysis process while the use of anion exchange has resulted in a more thorough separation. The possibility of incomplete separation and/or contamination of, e.g., natural uranium demonstrate the value of utilising more than one mother/daughter nuclide ratio for a more accurate age determination.

Unexpectedly, the analysis of IRMM081 required a lithium-borate fusion in order to achieve similar chemical behaviour of the Pu in IRMM081 and the added ^{242}Pu spike. If a fusion was not conducted, the Pu from IRMM081 was lost to a higher degree than the ^{242}Pu spike in the solid phase extraction separation. The reason for this is not known at the present, but one explanation might be that some of the Pu in IRMM081 is in polymeric form. However, further examination needs to be conducted in order to explain this behaviour.

A critical parameter in developing this method was a thorough separation of ^{241}Pu and ^{241}Am since these nuclides would otherwise mutually interfere during the measurements. The performance of the separation was evaluated by monitoring the amounts of ^{243}Am in the Pu fraction and ^{242}Pu in the Am fraction. No significant levels could be found and hence the detection limits, defined as 3 times the standard uncertainty of the blank, were used to estimate separation factors for Am and Pu. The resulting separation factors were ≥ 13000 for Pu and ≥ 3900 for Am. Considering that the expected ratios of $^{241}\text{Pu}/^{241}\text{Am}$ will vary from 20 to 0.06 for ages from 1 to 60 years, these separation factors are more than adequate.

Initial experiments showed that about 1% of the plutonium ended up in the americium fraction when only the TEVA- and TRU-resins were used and hence the separation factor was

improved by the addition of Ln-resin. The extractant in Ln-resin is HDEHP, which has been employed for the separation of americium from the other actinides in liquid-liquid extraction.¹³ The retention of americium on Ln-resin is strong at low acid strength, while the retention on TRU requires high acid strengths. Plutonium is retained on TEVA as a tetravalent nitrate complex. If any of the plutonium passes through TEVA it would be as tri-, penta- or hexavalent plutonium or due to overload of tetravalent plutonium on the resin. All of these oxidation states are retained on TRU at 3 M HNO₃, with the possible exception of Pu(V). When americium is eluted from TRU using 10 mM HNO₃, Pu(III), Pu(IV) and possibly Pu(V) would co-elute, while Pu(VI) would still be retained. Of the former three species, Pu(III) and Pu(IV) will be extracted by HDEHP. When americium is eluted from Ln-resin using 0.5 M HNO₃, Pu(III) would co-elute but not Pu(IV), which is highly extractable by HDEHP at this acid concentration.¹⁴ Thus, transformation of Pu(III) to higher oxidation states is critical in order to achieve a good separation from Am/Cm and the oxidation procedure used in this work has proven to be sufficient for this purpose. The use of Ln-resin provides not only a clean up of the americium fraction from plutonium, but also a factor of six more concentrated fraction compared to the 20 ml 0.01M HNO₃ required to elute Am from the TRU resin.¹⁵

The results of a more thorough analysis of IAEA367 are displayed in Table 2. It can be seen that the concentrations of Pu and Am in IAEA367 varied by factor of about two between the different sub-samples, suggesting that the material is not homogenous in terms of concentration in such small aliquots as 0.5 g. The isotope amount ratios of the plutonium isotopes, as well as the ratios of ²⁴¹Pu and ²⁴¹Am, were, however, homogenous in each sub-sample, in spite of the varying concentrations (see Tables 2 and 3). This means that no significant fractionation between Pu and Am had occurred before 1982 when this material was collected (or afterwards).

It should also be noticed (Table 2) that the concentration of ²⁴¹Pu in this material is low, varying between 21 and 39 fg g⁻¹. The amount of sample used for the analysis was about 0.5 g which, in combination with a chemical yield of about 60% and a fraction volume of 5 ml, results in concentrations of only 1.3 to 2.3 fg g⁻¹ in the analysed solution.

Table 2. Results of Pu from the analyses of IAEA367.

Sample	Conc / pg·g ⁻¹		Isotope amount ratio			
	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	²⁴⁰ Pu/ ²³⁹ Pu	²⁴¹ Pu/ ²³⁹ Pu
IAEA367:1	15.5 ± 0.14	4.41 ± 0.071	0.0392 ± 0.0084	0.405 ± 0.014	0.283 ± 0.005	0.00250 ± 0.0006
IAEA367:2	8.62 ± 0.10	2.57 ± 0.056	0.0248 ± 0.0078	0.245 ± 0.010	0.297 ± 0.007	0.00285 ± 0.0009
IAEA367:3	8.11 ± 0.10	2.45 ± 0.052	0.0215 ± 0.0071	0.220 ± 0.010	0.301 ± 0.007	0.00262 ± 0.0009

The observed amount ratios of the plutonium isotopes are also shown in Table 2. The average ratio of ²⁴⁰Pu / ²³⁹Pu is 0.29 (± 0.01) which is higher than that ratio of global fall-out of 0.20.¹⁶ Different constructions of nuclear weapons were tested at Enewetak, both fission and thermonuclear (fusion) weapons. The first fusion weapon ever tested, “Mike”, detonated at the Enewetak Atoll on October 31, 1952 with a yield of 10.4 Mton. The ²⁴⁰Pu / ²³⁹Pu ratio in the debris from this test has been reported to be 0.363 ± 0.004 (the uncertainty representing one “standard error”).¹⁷ A relatively high ²⁴⁰Pu / ²³⁹Pu ratio is characteristic of thermonuclear weapons due to high neutron fluencies, thus creating ²³⁹Pu and ²⁴⁰Pu from neutron capture by

^{238}U present in the uranium blanket.¹⁸ Hence, the ratio of 0.29 found in this study indicates significant contribution of debris from thermonuclear devices, which would limit the actual reference date for the age determination to the range 1952 to 1958. Further “successful” thermonuclear tests yielding more than 1 Mton were conducted at Enewetak in 1954 (“Nectar” 1.69 Mton), 1956 (“Apache” 1.85 Mton) and 1958 (“Koa” 1.37 Mton, “Oak” 8.9 Mton and “Pine” 2 Mton).^{19, 20} The results displayed in Table 2 are centred around 1954, but the uncertainties in the age determinations are too large make a more specific conclusion on what test the Pu originates from.

Conclusions

The method presented is well suited for age determination of plutonium material based on the $^{241}\text{Am}/^{241}\text{Pu}$ isotope amount ratio. A thorough separation of Pu and Am was achieved from the solid phase extraction procedure used, and the determined ages of the Pu materials corresponded well with the age reported by the reference material suppliers. Also the γ -spectrometric analyses of the two samples with higher concentrations showed a good agreement with the mass spectrometric determinations. The use of ICP-SFMS is advantageous due to its high sensitivity and relatively simple sample preparation. The utilisation of the $^{241}\text{Am}/^{241}\text{Pu}$ pair for age determination is valuable compared to using a Pu/U pair since contamination or mixing with U from other sources are of higher probability in most case compared to contamination from ^{241}Am . However, a condition for this kind of age determination is that the daughter nuclide was not present in the material at $t=0$ or that this concentration is known. Thus, if this information is not available a more reliable dating can then be based on both the $^{241}\text{Am}/^{241}\text{Pu}$ and a Pu/U ratio.

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RadChem – Radiochemical analysis in emergency and routine situations

Rajdeep Sidhu¹, Elis Holm², Per Roos³, Jukka Lehto⁴, Tarja K. Ikäheimonen⁵, Håkan Pettersson⁶, Christina Greis⁷ and Ulrika Nygren⁸

¹ Institute for Energy Technology, IFE, P.O.Box 40, NO-2027 Kjeller, Norway

² Lund University, SE-22 185 Lund, Sweden

³ Risø National Laboratory, DK-4000 Roskilde, Denmark

⁴ Laboratory of Radiochemistry, P.O.Box 55, FIN-00014 University of Helsinki

⁵ STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

⁶ Department of Radiation Physics, Linköping University, Sweden

⁷ Department of Natural Sciences, Örebro University, Sweden

⁸ Swedish Defence Research Agency, FOI, Umeå, Sweden

Background and introduction

An accurate determination of radionuclides from various sources in the environment is essential for assessment of the potential hazards and suitable countermeasures both in case of accidents, authorised release and routine surveillance. Reliable radiochemical separation and detection techniques are needed for accurate determination of alpha and beta emitters. Rapid analytical methods are needed in the case of an accident for early decision-making.

The objective of the first part of this project (RadChem-2004) has been to compare and evaluate radiochemical procedures used at Nordic laboratories for the determination of strontium, uranium, plutonium, americium and curium.

A similar task was undertaken in 1985 in the framework of NKS (NKA): “The Sampling and Analysing Methods of Radionuclides used in The Nordic Countries for Environmental Samples”, Edited by Tarja K. Taipale [1]. Since then new separation methods and instrumentation has been introduced and is in use in most laboratories.

To gather detailed information on the procedures in use, a questionnaire regarding various aspects of radionuclide determination was developed and distributed to all (sixteen) relevant laboratories in the Nordic countries. The response and the procedures used by each laboratory were then discussed between those who answered the questionnaire. A report summarising the findings and providing recommendation on suitable practice has been submitted to NKS.

The second part of the project (RadChem-2005) is more practically orientated. Each participant has worked with improvement of existing procedures or development of new procedures. One participant has also arranged an intercomparison exercise on the determination of natural radionuclides in ground water.

Radiochemical procedures

Strontium

Separation:	<i>Fuming nitric acid precipitation combined with carbonate, hydroxide and chromate precipitations, solvent extraction (TBP), or extraction chromatography (Sr-Resin)</i>
Source preparation:	SrCO ₃ precipitation for direct determination or ingrowth of ⁹⁰ Y and Y-precipitation (hydroxide or oxalate) for ⁹⁰ Sr determination
Activity determination:	Beta counter (proportional or GM) or LSC

Uranium

Separation:	Anion exchange chromatography (from HCl or HNO ₃), solvent extraction (TBP), or extraction chromatography (UTEVA)
Source prep:	Electrodeposition or co-precipitation with rare earth fluorides
Activity determination:	Alpha spectrometry, ICP-MS or fluorescence

Plutonium

Separation:	Anion exchange chromatography (HNO ₃), solvent extraction (TBP, TIOA, TTA), or extraction chromatography (TRU)
Source prep:	Electrodeposition or co-precipitation with rare earth fluorides
Activity determination:	Alpha spectrometry, ICP-MS, or LSC (²⁴¹ Pu)

Americium and curium

Separation:	Anion exchange chromatography combined with TIOA, HDEHP-TBP extraction and cation exchange chromatography
Source preparation:	Electrodeposition or co-precipitation with rare earth fluorides
Activity determination:	Alpha spectrometry

Main conclusions on the determination of Sr, U, Pu, Am and Cm

The detailed information provided by the labs on their practise regarding the specified analysis, is very valuable. The benefits are obvious as radiochemical analysis now can be studied and compared in detail. It is now 20 years ago such a study was last undertaken in the Nordic countries. Although, most of the techniques in use are still the same, some deviations can be seen: Besides Pu separation using anion exchange chromatography, there is not a single procedure that is used in all labs. More labs are doing americium determination. Due to the commercial availability of extraction chromatographic resins, more labs are now using this technique. A comparison of the results provided by different labs in the NKS-B LABINCO excersise, will also provide a direct comparison of the different procedures in use. Most labs are satisfied with the procedures they use.

- Developing individual matrix-dependant separation procedures can speed up the analytical methods, but this requires a more stringent supervision.
- Smaller ion exchange columns and reduced “washing and elution volumes” for the separation of actinides can be used to speed up the analysis without negative effects.
- Do not throw away Pu-sources as they can be used to get valuable information on ²⁴¹Pu content.

- Problems with low Am-yields and poor Am-separation when using TRU- and TEVA-Resin.
- HCl-TiCl₃ elution of Pu from TRU-Resin eliminates Po-interference caused when Pu is eluted with ammonium oxalate.
- Not always necessary to separate trivalent actinides from lanthanides.
- One lab has occasional problem with poor separation of Pu from uranium when using TIOA extraction and anion exchange (2x) for Pu-separation (drainage water).
- Low Pu-recoveries when using TTA extraction.
- Several labs are working with the development of sequential procedures.
- Occasional high background when Sr-Resin is used for Sr separation (effluent water).
- Observe ²²⁸Ac in the ⁹⁰Y fraction.
- Low yields when using the “fuming nitric acid” procedure for the determination of ⁹⁰Sr in seawater and seaweed.
- Indirect determination of ⁹⁰Sr in seawater using ⁹⁰Y separation and determination should be tested.
- Avoid the use of fuming nitric acid.

RadChem-2005

Intercomparison on the determination of natural radionuclides in ground water

An intercomparison exercise on the determination of natural radionuclides in ground water has been arranged. Six labs participated in the determination of ²¹⁰Pb, ²¹⁰Po, U-isotopes, ²²²Rn, ²²⁶Ra and ²²⁸Ra and the results show a very good consistency between the labs.

Sequential determination of Sr, U, Pu, Am and Cm in urine

A procedure for the sequential determination of ⁹⁰Sr, U-isotopes, ^{238,239,240}Pu, ²⁴¹Am and Cm-isotopes in urine samples has been developed. The analytes are first pre-concentrated using calcium phosphate co-precipitation and after destruction of the phosphates, they are separated from interferences and each other using three extraction chromatography columns stacked on each other (Sr-, UTEVA- and TRU-Resin). The procedure has been tested in PROCORAD urine intercomparison exercise with good results, and is used at one of the labs as a routine procedure.

Determination of Am and Cm in environmental samples

Americium and curium determinations in environmental samples are often troublesome due to their low activities and interference from other elements and radionuclides. One lab has worked on a more secure procedure for their separation using extraction chromatographic resins (Diphonix-, TRU- and TEVA-Resin). The results so far show that the yields and spectra are satisfactory when analysing water, soil and sediment samples, but the procedure requires modification for biota samples.

Age determination of Pu

Age of a plutonium source containing ^{241}Pu can be determined by the determination of ingrown ^{241}Am . When using mass spectrometric techniques for activity determination this requires a good chemical separation between ^{241}Pu and ^{241}Am . One lab has developed a method using extraction chromatographic resins TEVA- TRU- and Ln-Resin for this purpose. Analysis of reference solutions and environmental samples show good agreement with reported values.

Rapid determination of Pu using ICP-MS

One participant is studying different aspects of plutonium determination with the aim of developing a rapid procedure for plutonium determination with ICP-MS:

- Leaching of plutonium from different matrices with different acids using traditional hot plate leaching and microwave assisted leaching
- Preconcentration using rare earth fluorides or $\text{Fe}(\text{OH})_3$
- Use of different ion exchange resins
- Use of different extractants (TIOA, TBP, TTA)
- Use of extraction chromatography
- Use of ICP-MS vs alpha spectrometry

Determination of U, Pu and Am in emergency situations

Two participants have worked with simplification of U, Pu and Am determination in emergency situations. Proposed methods:

Americium and plutonium

- Ashing, acid leaching and addition of tracers
- Hydroxide precipitation using NH_3
- Oxalate precipitation followed by gamma spectrometry of ^{241}Am and ^{243}Am or
- Fluoride precipitation followed by gamma spectrometry of ^{241}Am and ^{243}Am
- Destruction of the oxalates or fluorides and separation of Pu(IV) using TTA followed by activity determination using combinations of LSC, alpha spectrometry or ICP-MS.

Uranium

Extraction with TBP from 8 M HNO_3 , rinse with 1.2 M HCl and elution with water followed by alpha spectrometry. Increased levels of uranium can also be confirmed by direct determination of ^{235}U using gamma spectrometry.

Various other work

- Ultra low level measurements using ICP-MS
- ^{90}Sr in milk using extraction chromatography
- ^{90}Sr via ^{90}Y in seawater
- Usefulness of stable lead as tracer for ^{210}Pb

Radioactive contamination of milk from the Nordic countries

Håvard Thørring,

Norwegian Radiation Protection Authority, P.O.Box 55, NO-1332 Østerås, Norway

Introduction

For the NKS-B EcoDoses project – “Improving radiological assessment of doses to man from terrestrial ecosystems”, data concerning ^{137}Cs and ^{90}Sr in cow’s milk were collated from all Nordic countries for both the Nuclear Weapons Fallout and the post-Chernobyl periods. An overview of the collated data for ^{137}Cs is shown in Figure 1.

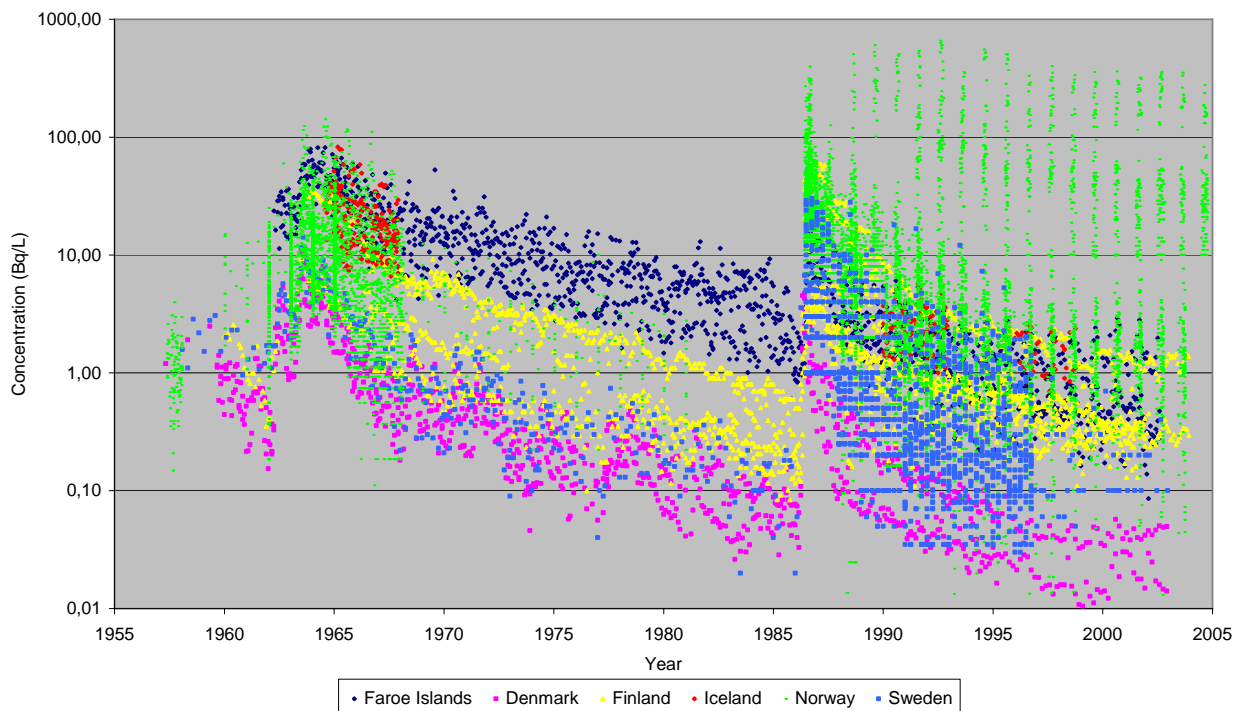


Figure 1. Caesium-137 in cow’s milk from the Nordic countries.

Summary of data

Caesium-137

For the NWF-period, the contamination levels of caesium-137 in milk vary considerably - dependent on factors such as deposition level and soil properties in different regions. Generally, precipitation rich areas such as the Faroe Islands, Iceland and western parts of Norway show the highest activity concentrations of ^{137}Cs . The importance of soil properties for milk concentrations is demonstrated by comparing the data from Lapland (North-Finland) and south-western Finland. During the NWF-period the deposition levels were similar in these two regions, yet the contamination level in milk from Lapland is several times as high as the corresponding levels from the south west. In northern Finland there are predominantly nutrient deficient organic soils which give a higher transfer of ^{137}Cs compared to the clayish soils in the south west. Denmark shows the generally lowest levels of caesium-137 for the NWF-period, most likely due to the clayish soils in these areas.

Looking at Figure 1, the post-Chernobyl data are clearly much higher in certain parts of Norway compared to other Nordic countries. This is mainly due to high deposition in some Norwegian areas. Many farms joined the summer monitoring program in 1988-89 which explains why the peak values are found around these years instead of in 1986-87. There are regional differences in the reported data for Sweden, Finland and Denmark as well - the higher level samples are from the mid-eastern part of Sweden, western Finland and Jutland in Denmark.

Strontium-90

The Faroese data show the highest Nordic contamination levels for the NWF-period, while the Norwegian data generally are highest for the post-Chernobyl period. There is a gap in contamination levels for the NWF data for Norway. The higher levels are found in Bergen and Florø, situated on the west coast of Norway. Due to the high precipitation rate in this coastal area the estimated deposition was higher here than in other parts of the country. No large regional differences within other countries are apparent for ^{90}Sr . This indicates that soil types are less important in determining the transfer factors to milk for ^{90}Sr than for ^{137}Cs .

A more thorough description of the Nordic milk data is given in the EcoDoses annual reports 2003 and 2004 (Bergan *et al.* 2004, 2005).

Time trends and effective ecological half lives

From the collated milk data, long and detailed data series with good consistency were selected for a more thorough study of time trends and effective ecological half lives – emphasising regional variations. These series are briefly described in Table 1. Unfortunately, the selected data series comprises only three of the Nordic countries, namely Denmark, Faroe Islands and Finland.

Table 1. Selected time series from Nordic countries.

DATA SERIES	MILK TYPE	SAMPLING	TIME PERIOD
West Denmark (Jutland)	Dry milk	~Monthly	1959-02
East Denmark (Islands)	Dry milk	~Monthly	1959-02
Faroe Islands (Thorshavn)	Dairy milk	~Monthly	1962-02
Faroe Islands (Klaksvig)	Locally produced milk	~Monthly	1962-02
Faroe Islands (Tvoroyri)	Locally produced milk	~Monthly	1962-02
North Finland	Dairy milk (Kursu)	~Monthly	1963-88
	Dairy milk (Rovaniemi)		1986-02
South-west Finland	Dry milk	~Monthly	1960-89
	Dairy milk		1986-02
West Finland	Dairy milk	~Monthly	1966-73 1978-04
East Finland	Dairy milk	~Monthly	1978-04

Dual exponential regressions were performed on each data-series. Since the milk data show considerable seasonal variation, the monthly values were combined to an annual average before the regression analysis was performed. The model expression was as follows:

$$Y = \ln \left[A1 \frac{\ln 2 \cdot t}{T1} + A2 \frac{\ln 2 \cdot t}{T2} \right]$$

where,

A1: Start activity, component 1

T1: Effective ecological half-life for component 1 (“fast component”)

A2: Start activity, component 2

T2: Effective ecological half-life for component 2 (“slow component”)

t : Time elapsed since reference date

For ^{137}Cs , the starting point of the NWF-period regressions was ~1964, and the end point was 1985 (December). For the post-Chernobyl period the starting point was 1986 (summer), whereas the end points were according to the length of the available time-series (see Table 1).

Strontium-90 regressions were performed for the whole period (1964-present). In addition, regressions were run for the NWF-period to allow direct comparison with caesium-137.

Caesium-137

Figure 2 shows the results for the time-series from Western Finland, Jutland in Denmark, and Thorshavn (Faroe Islands). The regression lines seem to represent the time development of ^{137}Cs -contamination in milk fairly well. Effective ecological half lives for all of the investigated time-series (including the three above) are summarised in Table 2.

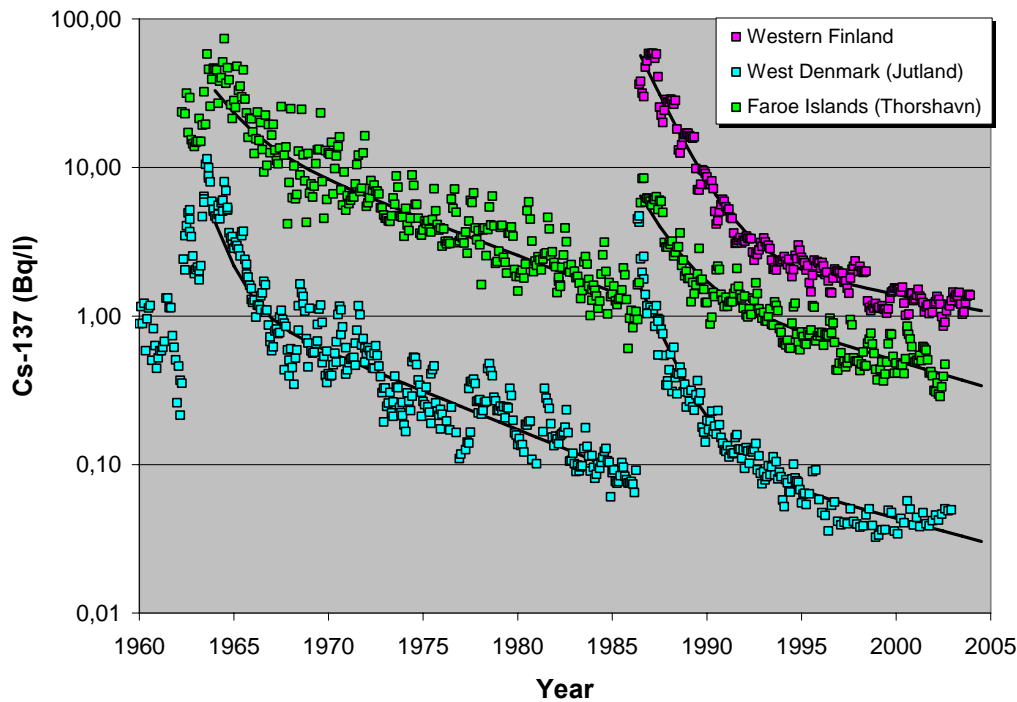


Figure 2. Caesium-137 dual exponential regressions for selected time-series before and after the Chernobyl accident.

Table 2. Effective ecological half lives for ^{137}Cs in milk from various regions in three Nordic countries.

Caesium-137	$T1 \pm SE^1$ (years)	$T2 \pm SE^1$ (years)	R^2
NWF-period			
West Denmark (Jutland)	0.7 ± 0.1	5.8 ± 0.2	0.91
East Denmark (Islands)	0.8 ± 0.1	6.0 ± 0.3	0.88
Faroe Islands (Thorshavn)	1.1 ± 0.2	6.2 ± 0.4	0.88
Faroe Island (Klaksvig) ²	(1.9 ± 0.3)	-	(0.81)
Faroe Island (Tvøroyri)	1.0 ± 0.3	6.5 ± 0.3	0.85
North Finland (Kursu)	1.0 ± 0.1	5.4 ± 0.1	0.97
South-western Finland	1.0 ± 0.1	7.2 ± 0.4	0.93
Post-Chernobyl			
West Denmark (Jutland)	1.0 ± 0.1	8.8 ± 1.3	0.94
East Denmark (Islands)	0.5 ± 0.0	6.6 ± 0.6	0.90
Faroe Islands (Thorshavn)	1.1 ± 0.2	8.0 ± 1.0	0.89
Faroe Islands (Klaksvig) ²	(1.4 ± 0.3)	-	(0.77)
Faroe Islands (Tvøroyri) ²	(1.6 ± 0.2)	-	(0.82)
North Finland (Rovaniemi)	1.3 ± 0.2	9.0 ± 1.0	0.94
West Finland	1.1 ± 0.1	12.6 ± 2.2	0.95
East Finland	0.8 ± 0.1	8.3 ± 0.7	0.83
South-West Finland ²	(1.1 ± 0.1)	-	(0.94)

¹Asymptotic standard error

²T2 showed anomalous results with very large standard error and has not been presented. T1 is therefore given in brackets.

Looking at the estimated effective ecological half lives, we see that T1 is around 1 year for all sites for both periods. For the NWF-period, T2 is around 6 years for all localities except for south-western Finland that shows a slightly longer T2 half-life (about 7 years). T2 half-lives for the post-Chernobyl period are generally longer. In addition there seem to be more pronounced geographical variations: The shortest T2 half-life is found for the Danish Islands (about 7 years), while the data from western Finland gives a T2 half-life of about 13 years.

Strontium-90

Effective ecological half lives for all of the investigated time-series are presented in Table 3. Generally, the regression lines represent the time development of ^{90}Sr -contamination in milk quite well (as evident from R^2 values).

Table 3. Effective ecological half lives for ^{90}Sr in milk from various regions in three Nordic countries.

Strontium-90	T1±SE¹ (years)	T2±SE¹ (years)	R²
NWF-period			
West-Denmark (Jutland)	0.9±0.1	7.0±0.2	0.96
East-Denmark (Islands)	1.1±0.1	7.6±0.3	0.94
Faroe Islands (Thorshavn)	0.8±0.4	3.8±0.1	0.96
Faroe Islands (Klaksvig)	0.8±0.2	4.2±0.1	0.95
Faroe Islands (Tvøroyri)	0.7±0.2	4.4±0.1	0.94
North Finland (Kursu)	1.6±0.1	11.9±1.2	0.94
South-western Finland	1.2±0.1	12.0±0.6	0.95
Whole period			
West-Denmark (Jutland)	1.4±0.1	8.5±0.2	0.97
East-Denmark (Islands)	1.8±0.1	10.6±0.4	0.96
Faroe Islands (Thorshavn) ²	(3.4±0.1)	-	(0.98)
Faroe Islands (Klaksvig) ²	(3.5±0.1)	-	(0.97)
Faroe Islands (Tvøroyri) ²	(3.8±0.1)	-	(0.96)
North Finland (Kursu)	1.6±0.1	12.0±0.3	0.96
South-western Finland	1.2±0.1	11.6±0.2	0.93

¹Asymptotic standard error

²T2 showed anomalous results with very large standard error and has not been presented. T1 is therefore given in brackets.

T1 for ^{90}Sr for the NWF-period is about 1 year for all sites. In contrast, there is a considerable geographical variation in T2 half-lives for this period: The shortest T2 half-lives were found for the Faroe Islands (about 4 years), whereas T2s for the Finnish data are about 12 years.

The Danish sites show slightly longer half-lives re both components (T1 and T2) using 2003 as the end point of regression, whereas the half lives for the Finnish sites do not seem to differ much considering either the whole period or the NWF-period. Note that the Faroese series do not give satisfactory effective ecological half-lives for T2. The reason for this is that there has been no decrease in concentrations of ^{90}Sr in milk after ~1990 (Figure 4). Accordingly, T2 half lives for these series will be very high (with a large appurtenant uncertainty). T2s for the Faroese series have therefore not been reported in Table 3. A similar trend is also observed

for some of the ^{137}Cs time series. This, for instance, applies to the post-Chernobyl ^{137}Cs -series from South-western Finland and Tvøroyri (Faroe Islands) (Table 2).

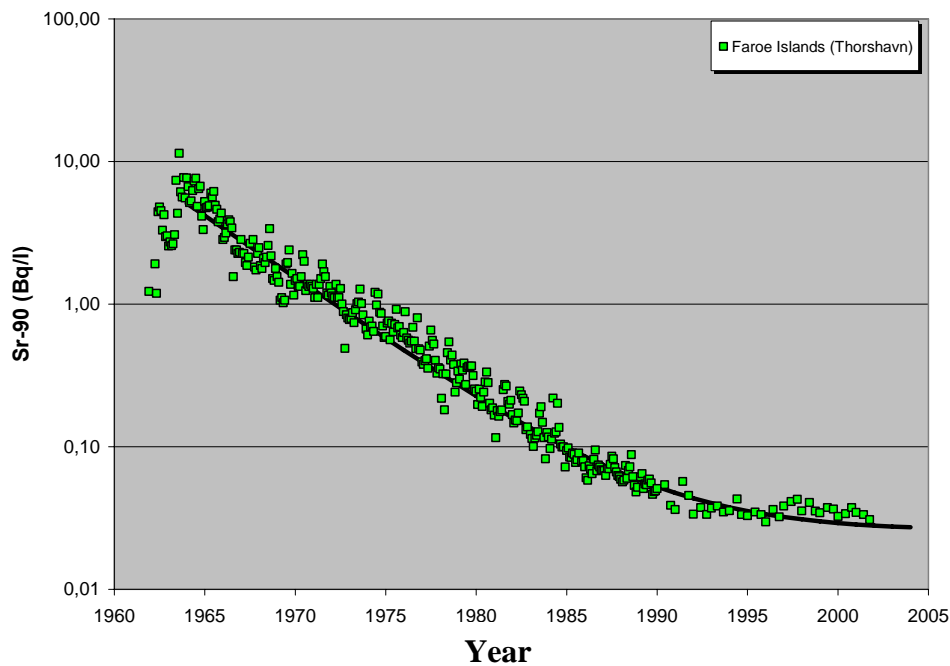


Figure 3. Strontium-90 in milk from Thorshavn (Faroe Islands) where anomalous effective half lives for the “slow component” were observed. Estimated dual exponential regression lines are indicated.

Conclusions

Considerable variations in activity concentrations of ^{137}Cs and ^{90}Sr were observed between countries or regions due to factors such as different precipitation patterns, soil types and Chernobyl deposition. The observed time trends indicate that the factors influencing the ecological half-life for ^{90}Sr are not entirely the same as for ^{137}Cs in the pasture – milk system. For ^{137}Cs the effective ecological half-lives seem to be fairly equal for the different investigated regions. Slightly longer T2s were observed for the post-Chernobyl period for this radionuclide. For ^{90}Sr the long component varies between 4 and 12 years. Finally, for some series (both ^{137}Cs and ^{90}Sr) a “no decrease” in activity concentrations has been observed for recent years.

Acknowledgements

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Adaptation of ECOSYS for Nordic food chain modelling

Kasper G. Andersson & Sven P. Nielsen

Risø National Laboratory, P.O. Box 49, DK-4000, Roskilde, Denmark

Abstract

The ECOSYS model is the basic foodchain model in the current European state-of-the-art systems for decision-aiding in connection with major nuclear accidents. To apply this model optimally in the decision process in a Nordic country contaminated due to an accident, it is necessary to change a number of the default parameters in ECOSYS to reflect Nordic, national or regional conditions. A number of potentially problematic parameters are identified and examined in this context. The importance of obtaining adequate parameters for, e.g., human consumption habits, animal feeding regimes, crop development dynamics and soil-plant transfer processes are discussed. Also the generic importance of considering particle size in connection with deposition modelling is demonstrated.

Introduction

In the European state-of-the-art decision support systems, ARGOS and RODOS, the modules for simulating the transfer of radioactive material in foodchains, respectively FDM and FDMT, are in essence identical with the ECOSYS model. The ECOSYS model default parameters were derived from experience obtained in central Europe, or more specifically, southern Germany. Many of the parameters describing the processes determining the foodchain transfer of radionuclides may however be subject to considerable variation according to local conditions. Such parameters include soil type and condition, sowing times, harvesting times, feeding regimes for animals, human consumption habits, and dependence of plant development on season. This paper aims to pinpoint main features of the ECOSYS model that would need to be addressed/alterd for the model to be applicable in a Nordic area. A number of alternative input parameters are described and their implications for contaminant contents in food products, and thereby for ingestion dose, are discussed. Also some generic comments are given to the ECOSYS model. The work was carried out as part of the NKS-B/ECODOSES project.

Methods and results

Food products and consumption habits

Agricultural traditions will differ between different countries. For instance, the agricultural products of a land area in a Nordic country will not on average be the same as the products of a corresponding land area in southern Germany. These are dictated by factors such as local consumption habits, export traditions, climate and geology. The ECOSYS model works with local consumption figures rather than production figures. It is in ECOSYS possible to allow

for that only a fraction of the food consumed in the area of interest is produced locally (the rest is assumed to have been produced in an uncontaminated area). It is very important to apply reliable and representative age-specific survey data for consumption rates and import fractions. The defaults in ECOSYS are for Bavarian conditions, and reflect local traditions specific to that area. The applied data should ideally have been acquired recently, as consumption habits may change significantly over a period of few years. For instance, in Denmark, the intake of morning cereals by children (ages 4-14) has been halved over the period 1995-2002, whereas it is unchanged for adults (Fagt et al., 2002). The same pattern is seen for the consumption of rye bread. Also, the butter consumption has gone down by about one-third, but the consumption of green salad and fruit has gone up by more than 50%. The survey also revealed that there can be some (rather limited, but significant) geographical variation within the country. For instance, inhabitants of Copenhagen consume about one-third less potatoes than do villagers (Groth & Fagt, 2002). ECOSYS works with discrete values for consumption rates, and not with statistical distributions. Therefore, 'critical group' individuals consuming higher than average rates of particularly strongly contaminated food products might be missed out. In ECOSYS, the user can enter the seasonality of consumption of the various crops, but what is perhaps even more crucial, particularly in a Nordic country, would be the seasonality of import of the crops, which can not be entered in the model.

Crop development stage and contaminant deposition

The stage of development of the canopy of crops at the time of contaminant deposition is highly important to consider in the estimation of both wet and dry deposition velocities. The deposition process thus has a pronounced seasonality. This seasonality in turn is location specific, as for instance growth seasons in Nordic countries are significantly different from those in Southern Germany, for which ECOSYS default parameters apply. In the ECOSYS model, this is generally accommodated by the incorporation of the leaf area index (LAI), which is defined as the one-sided leaf area in the crop canopy per unit ground area. The Chernobyl accident showed that for a surface such as grass, the deposition velocity of particulate contaminants as well as of elemental iodine gas can vary considerably (Roed, 1990). However, when the length (roughness) of the grass is taken into account by dividing the deposition velocity by the mass per unit area relationship, the figures are generally seen to exhibit only little variation.

The influence of LAI on deposition velocity is in ECOSYS incorporated by: $v_d = v_{d, \max} \text{LAI}_d / \text{LAI}_{\max}$, where $v_{d, \max}$ is the maximum deposition velocity for the given plant type, assuming fully developed foliage, LAI_{\max} is the corresponding LAI for the fully developed plant, and LAI_d is the LAI at the actual time of deposition. If there are differences in the stage of plant development at the time of harvest in different countries, this should thus be addressed by adjusting these parameters. A potential problem in using the ECOSYS model is that it by default only distinguishes between aerosols, elemental iodine and organic iodine. This means that all aerosols are treated the same way, regardless of size, and judging from the default deposition velocities, the 'standard' aerosol has an aerodynamic diameter of some few microns. Table 1 shows some typical measured values of v_d of aerosols of different sizes to fully developed vegetation (wheat and lettuce) (Watterson & Nicholson, 1996). The figures speak for themselves: if the default values of ECOSYS are not changed to adequately represent the aerosol size with which the radionuclide in question is associated, the deposition estimates can be wrong by several orders of magnitude. Even if only sparse information is

available in the early phase on the nature of the contaminating incident, this will give some idea of likely particle sizes of the key contaminants.

Table 1. Representative measured values of $v_{d, \max}$ for wheat and lettuce.

Particle diameter [μm]	0.5	4	10	20
$v_{d, \max}$ [10^{-4} m s^{-1}]	4	15	50	300

Values of LAI_d have been investigated experimentally by the Danish Institute of Agricultural Sciences for a number of crops, including winter wheat, spring wheat, spring barley, winter barley, grass, clover grass, peas, spring rape, carrots, winter rye, triticale, lupin, beets, maize, oats and potatoes. Based on the findings, a model was developed to describe LAI from sowing times, harvesting times, fertilising status and soil temperature. These parameters would be expected to govern most of the geographical variation in LAI, so that the Danish model could be used for any area, by inserting location-specific parameters. In general, most

of the plant growth will occur exponentially with time: $\text{LAI} = \text{LAI}_i * \{[\exp(2.4 \frac{S_t - S_E}{S_m - S_E}) - 1] /$

$10\}$ (Plauborg & Olesen, 1991). Here, LAI_i is the initial LAI prior to the main plant development, S_t is the temperature sum (sum of the products of days by average diurnal temperatures) from the start of growth to the time in question, S_E is the temperature sum from growth start, at which the exponential growth starts, and S_m is the temperature sum from growth start required to fully develop the plant. Needless to say, all these temperature sums are plant specific. Normally, soil temperature variation with depth will not be very significant over depths that would be relevant to the rooting zone of most crops.

The sensitivity of dry deposition to variations in LAI is demonstrated by the values in Table 2, which shows results of runs of the ECOSYS model with its default values, assuming a dry deposition on respectively the 15th of May and the 15th of June. A corresponding variation could be observed from deposition on a single day in different countries, due to differences in climate. The figures clearly show that the contents of radiocaesium in the crops would in the first year after deposition be very sensitive to the time of deposition, but not in the following years, where root uptake is the dominant mechanism of contamination.

Table 2. ECOSYS run examples showing crop contaminant concentrations after a dry deposition of caesium on respectively the 15th of May and the 15th of June.

		Activity concentration [Bq kg^{-1}]					
Time of deposition	Time following deposition	Spring wheat whole	Spring wheat flour	Spring wheat bran	Winter wheat whole	Winter wheat flour	Winter wheat bran
15 May	1 month	0	0	0	0	0	0
	6 months	1.6E-01	7.9E-02	4.7E-01	2.0E+00	9.8E-01	5.9E+00
	1 year	1.6E-01	7.8E-02	4.7E-01	1.9E+00	9.7E-01	5.8E+00
	2 years	2.1E-03	1.0E-03	6.2E-03	2.1E-03	1.0E-03	6.3E-03
	50 years	1.0E-05	5.0E-06	3.0E-05	1.0E-05	5.0E-06	3.0E-05
15 June	1 month	0	0	0	0	0	0
	6 months	5.9E+00	2.9E+00	1.8E+01	9.0E+00	4.5E+00	2.7E+01
	1 year	5.8E+00	2.9E+00	1.7E+01	8.9E+00	4.4E+00	2.7E+01
	2 years	2.1E-03	1.1E-03	6.4E-03	2.1E-03	1.1E-03	6.4E-03
	50 years	1.0E-05	5.0E-06	3.0E-05	1.0E-05	5.0E-06	3.0E-05

As mentioned above, the LAI will also affect the wet deposition interception factors of plants. This is in the ECOSYS model taken into account by the formula (Müller & Pröhl, 1993):

$$f_{w,i} = \frac{LAI_i \cdot S_i}{R} \cdot \left(1 - \exp\left(\frac{-\ln 2}{3 \cdot S_i} \cdot R\right) \right)$$

where $f_{w,i}$ is the interception factor for plant type i , S_i is the retention coefficient of plant type i , LAI_i is the leaf area index of plant i at the time of the event, and R is the amount of rainfall in the event.

Animal specific feeding rations and their seasonal variation

Feeding regimes for the various farm animals will vary between countries, or even smaller areas, according to both tradition and climate. In Table 3, the ECOSYS default feeding regime (Southern Germany) is compared with a typical Danish feeding regime, based on information acquired from the internet site of the Danish Agricultural Advisory Service and from communication with experienced Danish farmers.

Table 3. Comparison of typical feeding regimes for Southern Germany and Denmark.

	Southern Germany	Denmark
Lactating cattle	Hay until 10 th May, grass until 9 th Nov., hay until 31 st Dec.	Maize silage (70 %), grass silage (30 %) all year.
Beef cattle	96 % maize silage, 2 % winter barley, 2 % winter wheat all year	Grass silage until 15 th May, grass until 15 th Oct, grass silage until 31 st Dec.
Goats/sheep	Hay until 10 th May, grass until 9 th Nov., hay until 31 st Dec.	Hay/straw until 15 th May, grass until 15 th Sept., hay/straw until 31 st Dec.
Pigs	50 % winter barley, 50 % winter wheat all year	90 % winter barley, 10 % soy flour all year.
Chickens/hens	Winter wheat all year	Winter wheat all year

As can be seen, there are significant differences, particularly in what cattle are fed. One of the feedstuffs is not by default considered in ECOSYS (soy flour), and the model implications of using silage products are not clear. There are also significant differences in the grazing periods, due to differences in climate.

In relation to the influence of feedstuffs on dose, it has previously been demonstrated that a decisive parameter is the time of the year at which the contamination occurs (Riesen et al., 1996). Table 4 shows an example of the impact that the feeding regime could have on the concentrations of contaminants in food products (following dry deposition), unless countermeasures are actively taken. The difference between results of German and Danish feeding regimes is here rather extreme, as it is assumed that the contamination occurs just before the German animal grazing season starts. In contrast, ECOSYS by default assumes that there is no direct dry deposition on maize.

Table 4. An example of ^{137}Cs contents in cream, butter and beef 6 months after an accidental dry contamination on the 1st of May. ECOSYS model results. The code 'D' designates that the animals were fed according to the Southern German feeding regime, whereas code 'DK' is for the Danish feeding regime.

Product	Bq/kg after 6 months (D)	Bq/kg after 6 months (DK)
Cream	$1.8 \cdot 10^0$	$5.4 \cdot 10^{-3}$
Butter	$5.2 \cdot 10^{-1}$	$1.5 \cdot 10^{-3}$
Beef (cow)	$8.6 \cdot 10^0$	$2.3 \cdot 10^{-2}$

Fixation and migration rates

Fixation rates are used in ECOSYS to describe the natural decrease in availability of radionuclides to plant uptake, due to strong fixation in soil substances. In ECOSYS, fixation rates are considered to practically exclusively have importance for radiocaesium (and to a much lesser extent for radiostrontium) (Müller & Pröhl, 1993). This is reasonable due to the very strong, selective fixation of the caesium cation on frayed edge surfaces of the lattice of certain clay minerals. The data applied in ECOSYS to describe the fixation/migration processes partially date back to pre-Chernobyl experiments conducted to mimic the behaviour of radionuclides released after an accident (Bachhuber et al., 1984). However, the fixation process will be critically dependent on the physicochemical form of the radionuclides at deposition. The most recent reference applied for the parameterisation seems to be from 1987 (Frissel and Koster, 1987). The caesium fixation half-life applied by default in ECOSYS is 8.7 years. Since the Chernobyl accident, other workers have investigated the dynamics of the process of natural fixation of caesium in soils, using broadly consistent methods based on the protocol described by Tessier et al (1979). Even in loamy sand with comparatively little clay, Krouglov et al (1998) found that the process of fixation of caesium, to an extent where it could only be removed by strong acid dissolving minerals, had a half-life of only 2.6 years. This suggests that the amount of caesium available for root uptake might be significantly overestimated by using the ECOSYS default parameter. Also for instance investigations made by Andersson & Roed (1994) in Russian and Swedish soils suggest that by far the majority of the radiocaesium deposited from the Chernobyl accident (80 % +) was virtually irreversibly fixed after four years. In very sandy areas where the clay content is extremely low, as well as in very organic-rich soils, the fixation process can be slower. This will be reflected by an increased downward contaminant migration. The illustrations below (Fig. 1) show an example of the influences of clay content on fixation/migration. Both illustrations show the vertical radiocaesium distribution in an undisturbed soil in the Bryansk Region (Russia), ten years after the Chernobyl accident (Roed et al., 1998). The conditions of contaminant deposition are reported to be identical. However, the profile shown to the left is for a very shallow, sandy soil, whereas the one to the right had a clay content comparable to that typically found in Scandinavian soils.

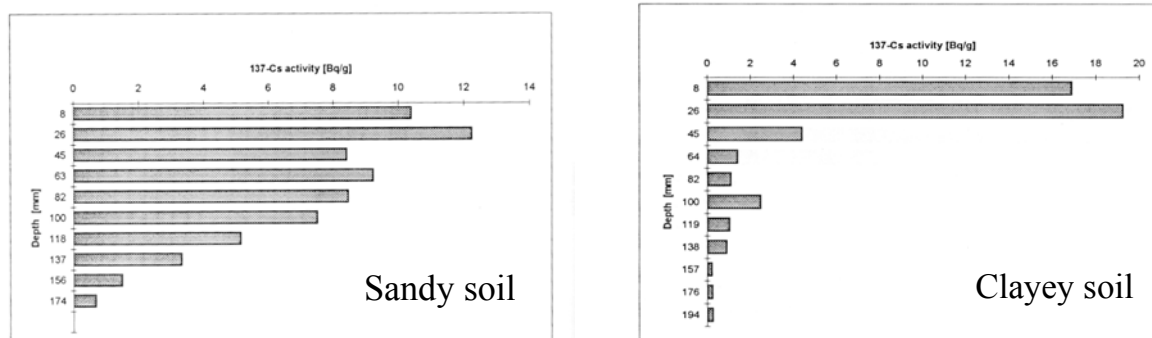


Figure 1. Vertical radiocaesium profiles in two soils in the Bryansk region. Samples taken 10 years after the Chernobyl accident contaminated the area.

The difference in ingestion dose estimates between assuming a caesium fixation half-life of 8.7 years and 2.6 years is easily found by running the ECOSYS model. Naturally, the influence is negligible the first year, but could become important in calculating long term dose prognoses if it were assumed that no doses are received from the first year's harvest (a consumption ban may be introduced here if the contamination level is high). The difference in dose received from 3rd year's harvest will be some 30 %, whereas it will be about a factor of 6 for the dose from the 10th year's harvest.

'Leaching rates' or 'migration rates' are applied in ECOSYS to describe the process of decrease in availability of contaminants for uptake by migration out of the root zone. This is a parameter that might possibly be problematic to give a general estimate of, as has been done in ECOSYS. These rates will depend on the depth of the root zone in the soil (by default in ECOSYS assumed to be 25 cm). This will in turn depend on the nutrient/fertilising status and soil type, and will be crop type specific, as root zone depths will differ between crops. It is difficult to assess how much this may matter, but in most cases, leaching rates, particularly of caesium, would be expected to be exceedingly slow. This seems to be adequately reflected by the ECOSYS default parameters. However, as demonstrated above, much higher migration rates must be expected for very sandy areas.

Transfer factors soil-plant

Due to the uptake competition between contaminant ions and their equivalent natural macro-nutrients, as well as variation in ion exchange potentials offered by different types of soil, both with respect to total cation exchange capacity and selective strong fixation of particularly caesium in minerals, contaminant transfer factors from soil to plant will in general be strongly dependent on soil type / fertilising status. Indeed, this is recognised in the ECOSYS model, where it is stressed next to the data table of default transfer factors that these are representative only of intensive farm management in Germany. A comprehensive literature study of transfer factors of relevance also to Nordic conditions was published a few years back (Kostiainen et al., 2002). Transfer factors are here given for different soil characteristics. The influence of soil conditions on crop contaminant concentration can be demonstrated through sample runs of ECOSYS, assuming that a dry deposition of caesium took place on the 1st of May. The transfer factors were here changed from the ECOSYS defaults to recommended values for mineral soil, taken from the above literature study. This made the Cs

concentrations in second year's leafy vegetable harvest rise by a factor of about 6, whereas the corresponding concentration in root vegetables rose by a factor of about 4.

Conclusions

A number of parameters were identified, which need careful considerations if the ECOSYS model is to be applied to describe radiological consequences in Nordic areas of a nuclear accident. The importance of using reliable, recent and location-specific information on consumption habits as well as on the seasonality of import of the different food products was stressed. Crucial contaminant deposition parameters depend on the crop development stage at the time of deposition. In turn, crop development dynamics depends on climate as well as on sowing times and fertilising status. A relatively simple model that can be used to estimate the seasonal crop development as a function of such parameters was briefly described. Currently, ECOSYS does not take into account the implications of particle size on deposition velocity. It was demonstrated that this could under some circumstances lead to estimates that are wrong by orders of magnitude. In an example it was demonstrated that feeding seasons and regimes may have a highly important bearing on contaminant concentrations in food products. Fixation of radiocaesium in soil will affect uptake by crops. The default fixation half-life applied in ECOSYS seems long compared with recently reported measured values. The difference was found to have significant effect on dose contributions received after the first few years. A short calculation example demonstrates the importance of applying soil-plant transfer parameters that are relevant for the particular area under study.

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Human metabolism of caesium

C. L. Rääf¹, R. Falk², T. Rahola³, B. Lauridsen⁴, S. Soogard-Hansen⁴, L. Skuterud⁵

¹ Department of Radiation physics, Lund University, Malmö university Hospital, SE-205 02 Malmö, Sweden

² Swedish Radiation Protection Authority, SE-171 16 Stockholm, Sweden

³ STUK – Radiation and Nuclear Safety Authority, P.O. 14, FI-00881 Helsinki, Finland

⁴ Risø National Laboratory, P.O. 49, DK-4000 Roskilde, Denmark

⁵ NRPA – Norwegian Radiation Protection Authority, P.O. 55, NO-1332 Østerås, Norway

Abstract

A study of the human biokinetics of caesium in two forms, *i.*) incorporated in foodstuff (^{137}Cs in perch and mushrooms) and *ii.*) in ionic state (^{134}Cs in aqueous solution) has been carried out at the department of Radiation Physics in Malmö, starting in 2001. The results of the pilot study was published in 2004, and a continuation of that study is now undertaken by means of NKS funding (NKS-B Cskinetik). The aim is to, *i.*) investigate whether Scandinavian populations exhibit shorter biological half-time of radiocaesium than other populations; *ii.*) extend the biokinetic study to additional human subjects from the other Nordic countries.

Results from the continued study further indicate a near complete absorption of radiocaesium in the gastro-intestinal tract, be it in ion state or contained in food matrix. So far, the literature survey of Nordic studies on biokinetics of Cs does not suggest that the biological half-time is shorter among Scandinavians, although there is a considerable variation between individuals.

Introduction

Huge amounts of data on Cs-kinetics in human adults exist today (e.g.; Leggett, 2003⁽¹⁾; Schwartz and Dunning, 1982⁽²⁾), but there are still uncertainties of the variation in uptake fractions and biological half-times with regard to the chemical state of Cs when ingested. There is also suggestion of regional variances in the biokinetics (ICRP, 1989⁽³⁾). In order to improve the accuracy of predictions of long-term transfer of Cs deposited in the Nordic ecosystems to humans, it is important to study more thoroughly the variance of the biokinetics of Cs. A shorter biological half-time in humans than the 110 days presented by the ICRP, may in part explain the discrepancies observed between the measured ^{137}Cs body burdens and estimations based on detailed food surveys in some Nordic human populations (Referens).

In April 2004 the project CsKinetic was launched within the NKS-B programme with the aim of investigating the human biokinetics of radiocaesium, specifically targeting the results from studies carried out in the Nordic countries. The project is a continuation of a study on conducted in Sweden where human adults have ingested known amounts of ^{134}Cs and ^{137}Cs and subsequently have been subject to excretion analysis as well as whole body countings (Rääf et al., 2004⁽⁴⁾).

Materials and methods

Biokinetic model

It is verified from many studies that the Cs-retention in human adults, $R(t)$ as a function of time in days, follow a two-component exponential model (Eq. 1). ICRP (ICRP, 1989) has suggested generic values to be applied conservatively to humans, where it is assumed that full absorption of Cs occurs when entering the gastro-intestinal tract, that is $f_0=1$ in Eq.1. The caesium in plasma is then assumed to be distributed to viscera and muscle tissues, and then released from the body with a short-term biological half-time of $T_1=2$ days ($a_1=10\%$ of initial retention), and a major component ($a_2=90\%$ of initial uptake), with a biological half-time of $T_2=110$ days.

$$R(t) = f_0 \left(a_1 \cdot e^{-\frac{\ln 2}{T_1} t} + a_2 \cdot e^{-\frac{\ln 2}{T_2} t} \right) \quad \text{Eq. 1}$$

For continuous intakes of Cs spanning over several years a semi-equilibrium is achieved in the intake and excretion. The ratio between the whole-body content and equilibrium excretion rate is defined by ICRP (ICRP, 1989) as the equivalent biological half-time, T_e , and is approximately related to the retention parameters as in Eq. 2.

$$T_e = \ln(2) \frac{\text{observed body-content}}{\text{observed 24h excretion}} \approx a_1 \cdot T_1 + a_2 \cdot T_2 \quad \text{Eq. 2}$$

Present-day levels of ^{137}Cs in humans in urban Nordic populations are very low, typically between 50 and 100 Bq (Räaf et al., 2005⁽⁵⁾), and are not assumed to have interfered with the studies conducted on the subjects at Risö. The retention function can thus be applied to the experimental data, which represents a single intake of radiocaesium.

Collection of samples

The supply of food portions containing concentrations of ^{137}Cs in sufficient amount to accommodate biokinetic studies had run out in 2003 during the experiments conducted at the Dept of Radiation physics in Malmö. The portions of fishmeal originated from samples taken from an area south of the city of Gävle in Sweden (Figure 1), which was heavily affected by the Chernobyl fallout (average ground deposition of ^{137}Cs in municipality exceeds 50 kBq m^{-2}). The species collected was perch and mushrooms, which contained up to 10 kBq kg^{-1} dry matter. At the beginning of this NKS-B study there was thus need for additional collection of environmental samples that could be used for human biokinetic experiments. Concentrations of 5 kBq kg^{-1} ^{137}Cs and higher in the fish or mushrooms are required in order to enable detectable levels (within reasonable acquisition times) by gamma spectrometry in excreta the first few days upon ingestion.



Figure 1. Map over the location of the lake Hedsjön.

A brief survey of possible food samples to be used for the kinetic study was done in late spring 2004. It was concluded that reindeer meat in Norway and Sweden contains too little (less than 5 kBq kg^{-1}) to be of use. Instead it was decided to return to the area outside the city of Gävle in Sweden, where the food samples of the previous study were collected. On August 11, 2004, eight number of perch, of about 1 kg size each, were caught in the lake Hedsjön (Figure 1), south of Gävle. The individual concentrations of ^{137}Cs were determined to be, on average, 7 kBq kg^{-1} (Figure 2), at the SSI in Stockholm before being sent to Malmö for preparation to edible food portions. This means that it may require up to 5 hectogrammes of fish meat in order to have more than $2 \text{ kBq } ^{137}\text{Cs}$ in edible form (taking into account cooking losses of up to 30%).

Subjects

Three adults (one females, two males) volunteered to participate in the kinetic study. The subjects reside in Denmark, and have readily access to gamma spectrometry facilities at the Risø National laboratory. More subjects are desired, but tedious sampling and access to whole-body countings at narrow time intervals after ingestion of food portions, make the recruitment of subjects a challenge. The subjects are preferably selected from personnel and their relatives, working at laboratories equipped with whole-body counters.

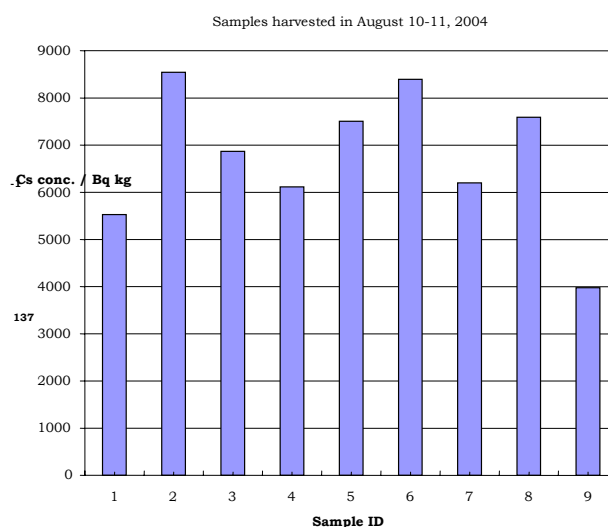


Figure 2. ¹³⁷Cs contents in perch (Samples 1-8) and pike (Sample 9) from lake Hedsjön, Gävle, Sweden.

Experiment

Food preparation of the fish caught in Hedsjön into food portions was carried out in Malmö in September 2004. The fishmeal was portioned into 110ml plastic flasks that were used as a reference geometry in both Malmö and Risö. In connection with the food processing three aliquots of ¹³⁴CsCl were prepared for the subjects in the study. The food portions and aliquots were brought to Risö in the beginning of October 2004, where the three volunteers ingested the amount on October 6 and 7. Daily urine samples (from all three subjects) and fractional faecal samples (from two of the subjects) were collected during five days after the ingestion. The urine was analysed by gamma spectrometry at Risö, whereas the faecal samples were sent for further preparation before being analysed by gamma spectrometry at the Dept of Radiation physics to Malmö. The intention has been to use the same sample geometry; a 110 ml plastic flask, when determining the ^{134,137}Cs content in the food portions, the ¹³⁴CsCl-aliquots, the faecal and the urine samples, in order to enhance the accuracy of the determined values.

Results and discussion

Excretion of radiocaesium

The urinary excretion in fractions of ingested amount of the three subjects varied from 0.09 to 0.17, due to individual variation (Figure 3). No systematic difference between ¹³⁴Cs and ¹³⁷Cs incorporated in perch has been observed. The dispersion in the fractional excretion of faeces was even less than for urine (Figure 3). The results are in accordance with the findings of the Swedish pilot study (Räaf et al., 2004⁽⁴⁾), which indicate no difference in the gastro-intestinal uptake of Cs incorporated in certain food matrices and Cs in ionic state (aqueous solution). In Figure 4 are plotted both the new data from Denmark and the results of the previous study in Sweden. The plot illustrates that the difference between the accumulated fractional excretion

of ^{137}Cs and ^{134}Cs as a function of time elapsed after ingestion, averaged over all participants, is close to zero %.

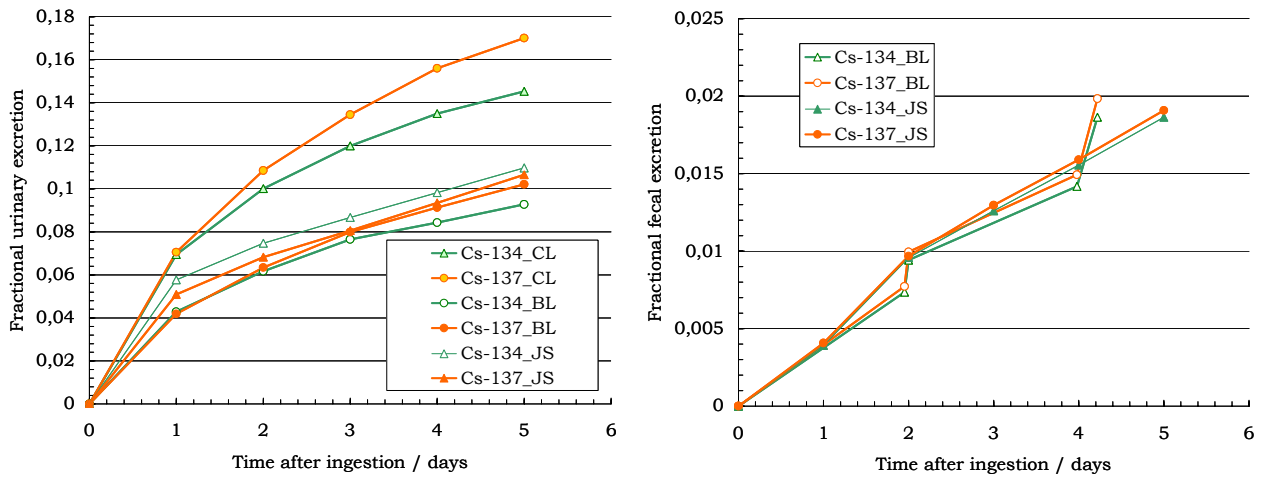


Figure 3. Fractional excretion of urine and faeces in three adult subjects upon ingestion of ^{137}Cs in perch and of $^{134}\text{CsCl}$ in aqueous solution.

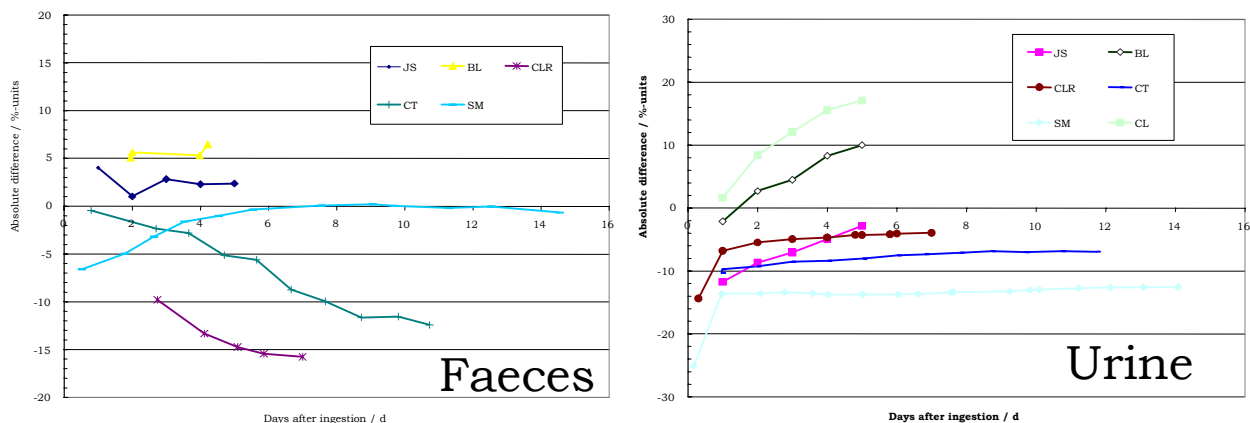


Figure 4. Difference in accumulated fractional excretion of ^{134}Cs and ^{137}C in five Nordic subjects. The absolute difference refers to the fraction excreted ^{134}Cs – fraction ^{137}Cs expressed in %-units.

In subjects BL and JSH, the accumulated faecal excretion over 4 days was less than 2% of the ingested amount for both forms of Cs. This suggests an uptake fraction from the gastrointestinal tract, f_0 , of about 98%, irrespectively of the two forms of Cs ingested. The result is in accordance with the previous study (Rääf et al., 2004⁽⁴⁾), as well as with other international studies, which all point to a near 100% uptake to plasma immediately after intake. Discrepancies between estimated average ^{137}Cs concentration in human population using the ICRP-model (NKS-studie I Norge) do therefore not appear to originate from erroneous assumption of complete plasma uptake.

Whole-body retention of radiocaesium

The observed biological half-times, using the retention model given in Eq. 1, showed no difference between the two forms of radiocaesium in subject BL (Table 1). Curve fits to ^{134}Cs data for the subject did not exhibit realistic values unless data points shortly and long time after the ingestion were considered separately. Data from participants JSH and CL are still pending.

Table 1. Short-term and long-term biological half-time of ^{134}Cs and ^{137}Cs observed for subject BL.

Subject	T_1	T_2	a_1	a_2
^{134}Cs	6.5±N/A	79±2.5	N/A	N/A
^{137}Cs	2.4±0.4	84±4	0.35	0.65

Conclusions

The following can be concluded from the study;

- There appears to be no difference in the initial uptake and excretion of radiocaesium between caesium incorporated into food tissue and in ionic form. This is further evidenced by the additional results of this study.
- Faecal excretion during the first few days appears to account for a minor fraction, <2-3%, of the initial gastrointestinal content of caesium, indicating a near 100% uptake in the gut to plasma. This means also that the ICRP-model from 1989 is realistic with respect to the uptake fraction.

Future activities

- The participants of the project have compiled a literature study containing more than 50 references on biokinetic studies of radiocaesium, with special focus on studies including some form of excretion sampling. There is more work to be done regarding the literature study of Nordic biokinetic studies on human radiocaesium metabolism, but a draft has been submitted to the NKS-B co-ordinator.
- There is still foodstuff containing contamination of ^{137}Cs , such as perch and pike, which is readily available to human biokinetic experiments. The time schedule of further human experiments depends on various factors, availability of sampling and access to whole-body counters, etc. It is, however, expected that the other Nordic countries can contribute to additional human volunteers within a 1-year period.

Acknowledgement

The project co-ordinator is very much grateful of the funding provided by the NKS, under the NKS-B programme.

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Guidance for sampling in forests for radionuclide analysis and update of the Nordic forest radioecology network

E. Strålberg¹, L. Aro², A. H. Plamboeck³, A. Rantavaara⁴, and V. Vetikko⁴

¹ Institute for Energy Technology, IFE, P.O.Box 40, NO-2027 Kjeller, Norway

² Finnish Forest Research Institute, METLA, Parkano Research Station, FI-39700 Parkano, Finland

³ The Swedish Defence Research Agency, FOI, SE-90182 Umeå, Sweden

⁴ STUK - Radiation and Nuclear Safety Authority, P.O.Box 14, FIN-00881 Helsinki, Finland

Abstract

The aims of the NKS project FOREST are to compile a sampling guide for radionuclide analysis of northern forests and to strengthen the Nordic collaboration in the field of forest radioecology by creating a network for scientists. Sampling procedures for soil, trees and understorey vegetation are prepared in collaboration with radioecologists and forest researchers from Finland, Norway and Sweden. Besides the practical instructions for collecting samples in the forest ecosystems, the guide will also contain examples on sampling strategies, guidelines for site description, and quality assurance. At the end of the project a workshop will be organised where the sampling methodology will be discussed. Future challenges in the field of Nordic forest radioecology will also be discussed at this workshop, as will the functions of the network for forest radioecologists and forest scientists.

Introduction

The project was motivated by a need for intensified Nordic collaboration in forest radioecology. No projects focusing on sampling methodology in forest ecosystems have been carried out internationally as a Nordic Nuclear Safety Research (NKS) or an EU-project earlier.

Qualified sampling is a necessity in analysis of forest products and tree stands potentially or actually contaminated by radionuclides. The complex structure of vegetation and soil in forests and time-dependent changes in radionuclide contents in various forest compartments require special skills and experience to carry out suitable sampling of vegetation and soil. Vegetation needs to be sectioned in a logical way. First, it is important to obtain comparable radionuclide contents and second, certain compartment structures are needed for radionuclide analysis in order to complete and improve the knowledge for radioecological modelling. For calibration of external dose rates from the ground, representative samples showing the depth distribution of radionuclides and sufficient information on the site characteristics are necessary. Besides the need for representative data, there is a requirement for a representative

analysis of forests of different vegetation types and structures. Furthermore, this has to be balanced against the very laborious sampling and analysis methods.

Samples of forest soil and vegetation have often been collected with various techniques by multidisciplinary research groups, but without documentation that would ensure the comparability of radioactivity data. With an intensified Nordic collaboration a better comparability of new data on radioactivity in forests can be obtained. To achieve this, the project will prepare a sampling guide that will provide information on the significance of sampling to the accuracy, comparability and reliability of data. The use of these data will then further improve the reliability of model parameters derived from the data, and the assessment of radiation doses after radiological accidents.

Concerning the new generation of scientists entering this field, the challenge is to collect relevant new information on time-dependent changes in forest ecosystems. To achieve this, knowing the basics of radioecology and nutrient cycling in forests are necessary. The sampling methodology has to be connected with the processes related to the transfer of radionuclides in forests.

Sampling guide

A guide for sampling in forest ecosystems will be prepared. The guide is planned to cover examples on sampling strategies, guidelines for site description, quality assurance, and practical instructions for collecting samples in the forest ecosystems. The aim of the sampling guide is to improve the accuracy, comparability and representativeness of new data on forest radioactivity. Thereby the reliability of model parameters derived from the data, and the assessment of radiation doses after radiological accidents will also be improved.

Preferred procedures for sampling will vary depending on the purpose of the study. The purpose therefore needs to be considered in the design of the sampling strategy. Different purposes will be, for example, national and international forest ecosystem monitoring based on recommended methods, surveillance of human exposure to radiation, potential needs for exposure assessment of biota, studies of processes and mechanisms of distribution of radionuclides within the forest ecosystem. Site description (e.g. fertility, vegetation structure, tree stand characteristics, topography, hydrology) is essential for further utilisation of analysis results.

Sampling strategies for soil and vegetation will be discussed in the sampling guide. Random, grid or stratified sampling designs are common for field studies. Other important factors to be considered are the size of samples (e.g. due to detection limits), the number of replicates of samples, composite samples versus single samples, resources for sample preparation and analysis, transport and storage of samples, availability of sample objects, timing and frequencies of sampling. Sampling equipment will be described giving a reasonable basis for making choices between them.

The guide will devote one chapter to each of the compartments

- Soil
- Fungi
- Understorey vegetation
- Trees

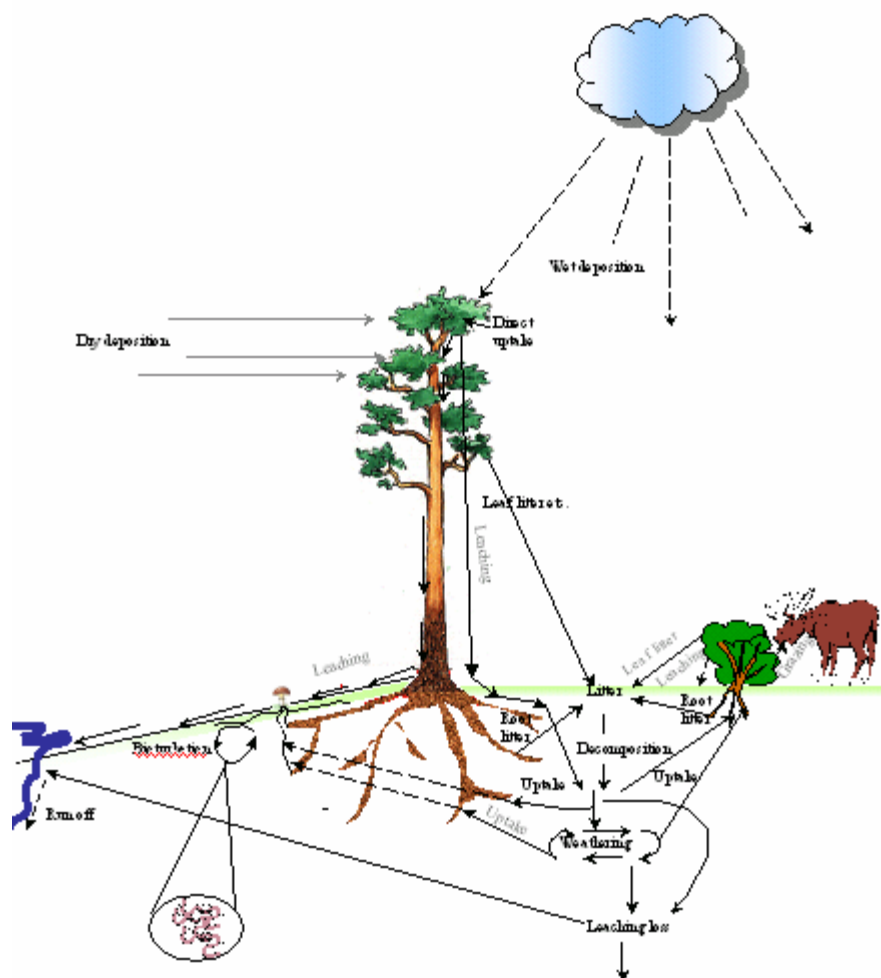


Figure 1. Cycling of radionuclides in forests, showing some of the pathways for radionuclides through the atmosphere-plant-soil-animal system.

Quality assurance of all stages in sampling is one of the main aims of the study. Avoiding cross contamination in field conditions and in the laboratory is essential in radioanalytical work. Also the number of samples and their statistical implications are crucial. However, costs of sample preparation and analysis often have an influence on the number of analyses.

A draft of the guide will be submitted to the NKS in December 2005. The finalised draft of the guide will be completed by the participants in 2006. The finalised draft will be refereed by external experts on field studies of forest ecosystems and discussed at a workshop in 2007. The final version of the sampling guide will be submitted for publication within two months after the workshop. Activities planned for 2006 and 2007 will be carried out only if further funding is granted from the NKS.

Network for forest radioecologists and workshop in 2007

A nordic network will be formed during the work with the sampling guide and a workshop is planned for 2007. The FOREST project can provide a new starting point for intensified Nordic collaboration in forest radioecology. The working methods of the network could be exchange of information, and organisation of topical workshops as a forum for presentation of new research and latest results. Forestry experts will also be invited to take part in the network, as they need information for building their own preparedness for solving problems caused by radionuclide contamination.

The first venue for the Nordic network for forest radioecologists will be at the planned workshop in 2007. The workshop will cover presentation of the results of the project, and provide a forum for scientific communication. The workshop will further cover:

- discussion and critical comments on the sampling guide
- recent results in the field of forest radioecology and viewpoints for research in the future
- the practices to maintain the network as an active discussion forum, and the forms and resources needed for scientific collaboration.

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Date	August 2006
Project/Sub Project	NKS-B
No. of pages	184
No. of tables	72
No. of illustrations	89
No. of references	0
Abstract	<p>This report contains the proceedings of the NKS-B Summary Seminar held on 24-25 October 2005 in Tartu, Estonia. The aim of the seminar was to provide a forum for presenting and discussing the results obtained in the NKS-B programme during the project period 2002 - 2005. The main attention was focused on radioecology and measurement techniques including presentations on the work done in the Projects INDOFERN (New Indicator Organisms for Environmental Radioactivity), LABINCO (Intercomparison of Laboratory Analyses of Radionuclides in Environmental Samples) and ECODOSES (Improving Radiological Assessments of Doses to Humans from Terrestrial Ecosystems). The total number of presentations in the seminar was 27.</p> <p>The seminar was also the final seminar of the four-year INDOFERN Project. The objective of the project was to identify new indicator organisms and biomarkers for assessment of environmental radioactivity in normal and emergency situations. The goal was to search new useful organisms accumulating effectively and specifically certain radionuclides in various Nordic ecosystems (forest, fresh water, marine), and to compare their indicator value to those of the earlier known indicators. The project yielded new data on the occurrence and transport of radionuclides in a wide scale of Nordic ecosystems. A summary of the whole project, and summaries of the work done in all the participating laboratories were presented in 13 presentations in the seminar.</p>
Key words	Radioecology, terrestrial radioecology, aquatic radioecology, indicator organisms, laboratory analyses of radionuclides, modelling, sampling, doses to man