



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

NKS-70
ISBN 87-7893-126-6

**Proceedings of the 8th Nordic Seminar
on Radioecology**
25-28 February 2001, Rovaniemi, Finland

Erkki Ilus (editor)
STUK, Finland

April 2002

Abstract

This report contains proceedings of the 8th Nordic Seminar on Radioecology held on February 25-28, 2001 in Rovaniemi, Finland. The Seminar was arranged by STUK - Radiation and Nuclear Safety Authority of Finland and supported by the NKS. The Seminar was intended to be a "final forum" of the four-year NKS radioecology project BOK-2, *Radioecological and Environmental Consequences*, which was focused on the consequences of releases of man-made radionuclides into the environment. The programme of the Seminar consisted of 3 invited lectures, 31 oral presentations and 22 poster presentations dealing with marine, terrestrial and freshwater radioecology, methods, foodstuffs, models, whole-body counting and doses to man.

Key words

Radioecology; marine radioecology, terrestrial radioecology, freshwater radioecology, methods, foodstuffs, modelling, doses to man

NKS-70
ISBN 87-7893-126-6

Pitney Bowes Management Services Denmark A/S, 2002

The report can be obtained from
NKS Secretariat
P.O. Box 30
DK – 4000 Roskilde
Denmark

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

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Preface

This report contains proceedings of the 8th Nordic Seminar on Radioecology held on February 25-28, 2001 in Rovaniemi, Finland. The Seminar was arranged by STUK - Radiation and Nuclear Safety Authority of Finland and supported by the NKS (Nordic Nuclear Safety Research). The previous Nordic Seminars on Radioecology were held in Norrköping, Sweden (1976), Helsingør, Denmark (1979), Hyvinkää, Finland (1982), Gol, Norway (1985), Rättvik, Sweden (1988), Torshavn, Faroe Islands (1991) and Reykjavik, Iceland (1996). The programme of the Seminar consisted of 3 invited lectures, 31 oral presentations and 22 poster presentations. A total of 72 participants from 8 countries (including Estonia and Lithuania) attended the Seminar.

The Seminar was also intended to be a 'final forum' of the four-year NKS radioecology project, BOK-2, in which results of the project would be presented. The BOK-2 project, *Radioecological and Environmental Consequences* (1998-2001) was focused on the consequences of releases of man-made radionuclides into the environment. The project included 2 subprojects: *Important Nordic Food Chains* (BOK-2.1) and *Radioactive Tracers in Nordic Sea Areas* (BOK-2). The BOK-2.1 subproject was further divided into 2 main sections: *Radioecological Vulnerability* (BOK-2.1.1) and *Internal Doses* (BOK-2.1.2), and the components of the BOK-2.2 sub-project were *Seawater Transport* (BOK-2.2.1) and *Biological and Biogeochemical Processes* (BOK-2.2.2).

Twelve of the presentations in the Rovaniemi Seminar dealt with marine radioecology and 11 with terrestrial radioecology (including posters). Eight of these dealt with freshwater radioecology, 8 with methods, 6 with foodstuffs, 4 with models, 4 with whole-body counting and 2 with doses. Four presentations are published as refereed papers in *Boreal Environment Research*. Only abstracts of these papers are published in the present report. The Seminar was finished with a panel discussion on the future of radioecology in the Nordic countries. The Seminar itself demonstrated the vitality and wide range of Nordic radioecology.

Finally, I would express my gratitude to Mrs. Raisa Tiililä for her valuable help in arranging the Seminar and editing these proceedings.

Erkki Ilus
editor

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Experiences in radioecology during the period 1964 - 2000

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Introduction

My presentation deals with the experiences gained in the field of environmental behaviour of radionuclides from global fallout while working at the Department of Radiochemistry, University of Helsinki since 1964.

Testing nuclear weapons in the open atmosphere began to arouse world-wide concern in the middle of the 1950s. Worrying news concerning high radioactivities in precipitation and surface air came especially from Japan and the United States. Nuclear testing period was strongly in progress and hydrogen bomb detonations were carried out in the Soviet Union and by the USA in the Nevada and Pacific Ocean test sites. As early as in 1956 W.F. Libby from University of Chicago published in *Science* a study of the occurrence of radioactive strontium in global fallout as a result of nuclear detonations (1). The ^{90}Sr isotope has later been widely used to describe the distribution of radioactive fallout from the atmospheric nuclear weapons testing as a function of the latitude. The ^{90}Sr analysis from soil revealed that the maximum fallout was found at the latitude 40° - 50°N . In Finland (60° – 70°N) the level of radioactive deposition was about 60 percent of maximum level.

The nuclear weapons tests have been the major source of global radioactive fallout. During the period of 1945-1998 altogether about 2000 tests have been made in the atmosphere, underwater and underground. The nuclear weapons test-ban agreement signed by the United States, United Kingdom and the Soviet Union early in 1963 eliminated further testing in the open atmosphere by the three signatory powers. Only France and China have conducted some tens of small-scale nuclear tests in the atmosphere after the year 1962. The total number of the underground tests performed is about 1500 and the frequency since 1963 up to late 1980s has been about 50 underground tests per year. However, if we are considering the global radioactive fallout the most significant atmospheric nuclear weapons tests were conducted during the period of 1945 – 1962 and especially during the years 1961 and 1962 (Table 1).

Table 1. Estimated yields of atmospheric nuclear weapons tests^a.

	Period	No. of tests	Estimated yield (MT)	
			Fission	Total
United States	1945-1962	193	72	139
USSR	1949-1962	142	111	358
United Kingdom	1952-1953	21	11	17
France	1960-1974	45	11	12
China	1964-1980	22	13	21
Total		423	218	547

^aFrom UNSCEAR (1982) (2)

The estimated total explosion yield from the atmospheric weapons tests during the period 1945 – 1980 has been some 550 MT out of which about 520 MT originates from the tests carried out before the year 1963 (2).

Other significant sources of global radioactive fallout have been the re-entry of the navigational satellites carrying to atmosphere a radioisotope power generator (SNAP 9-A) and the reactor accident of the Chernobyl nuclear power station.

Radioecological studies in Finnish Lapland

After the first observations of global radioactive contamination from nuclear weapons testing numerous projects were started to elucidate the behaviour of radionuclides in environment and especially their transfer to man. The volume of radioecological studies was the highest in the United States where the first national symposium on radioecology was organised at Fort Collins, Colorado, in 1961 (3).

At the end of the 1950s several preliminary investigations were carried out on the ^{90}Sr and ^{137}Cs contents of foods and feeds in the Finnish Lapland. The results showed that the concentration of these radionuclides were quite high, especially in lichens, which grow slowly, and in reindeer, whose principle feed is lichen during the main part of the year. A relatively high ^{137}Cs -content was observed in cow's milk and lake fish in Lapland, too. It was evident that in Finnish Lapland and also in other similar arctic regions exceptional food chains of long-lived radionuclides exist. Therefore, detailed investigations of such food chains were started.

At the first part of these studies a dietary survey of the Finnish Lapps was performed at the beginning of 1960s. These studies showed that the contribution of reindeer meat and liver in the diet of the Sami population is 80 – 90 % and that of the fish about 10 % (4). In 1960, a regular collection of plant and animal samples for analysis of ^{90}Sr and ^{137}Cs was also started.

Determination of body burden of ^{137}Cs

K. Liden in Sweden performed the first measurements of the body burden of ^{137}Cs in reindeer herders. The three Lapps measured contained 30 to 40 times more ^{137}Cs than the Swedes on an average. Prof. Liden understood the importance of this result and developed in the summer of 1961 a semiportable whole-body counter which was used in the determination of ^{137}Cs body burden in about 150 Swedish Lapps at Jokkmokk in Northern Sweden in September 1961. One month later (in October) 180 persons were counted at Inari, Finland, with the same equipment as a Finnish-Swedish co-operation.

Due to the new period of nuclear weapons tests that started in 1961 a mobile whole-body counter was installed at the Department of Radiochemistry by a grant obtained from the Finnish Atomic Energy Commission. This mobile counter consisted of a 512-channel analyser, which was one of the first analysers manufactured by the Finnish Cable Works, NaI (Tl) crystal and massive lead shielding. In this counter a chair geometry was used (5). With this mobile wholebody counter 250 persons of the Sami population were measured in the Finnish Lapland in May 1962. The study was carried out in three Lapland counties, Inari, Utsjoki and Enontekiö. The group investigated were selected from church registers by a random sampling method. The ^{137}Cs body burden of this group has been determined up to the year 1997. The results for the male reindeer herders of Inari are given

in the Figure 1. The maximum average body burden of ^{137}Cs 55 kBq was observed in 1965 two years after the maximum of the ^{137}Cs fallout. After the Chernobyl accident the corresponding average value increased from 5 kBq just before the accident to 14 kBq in 1988.

I started as a researcher and teaching assistant at the Department of Radiochemistry in fall 1964. My task in the field of radioecology was to develop radiochemical separation methods for determination of extremely low concentrations of radionuclides in environmental samples. As my research topic for the PhD degree I started the studies on behaviour of the ^{55}Fe , produced as an activation product in the atmospheric nuclear weapons tests, in the Finnish Lapland. This research was financed by the grant from the US Atomic Energy Commission to Professor Jorma K. Miettinen. In addition, I got acquainted with the atomic absorption spectrophotometry that was quite a new method to determine the trace concentrations of elements in different materials.

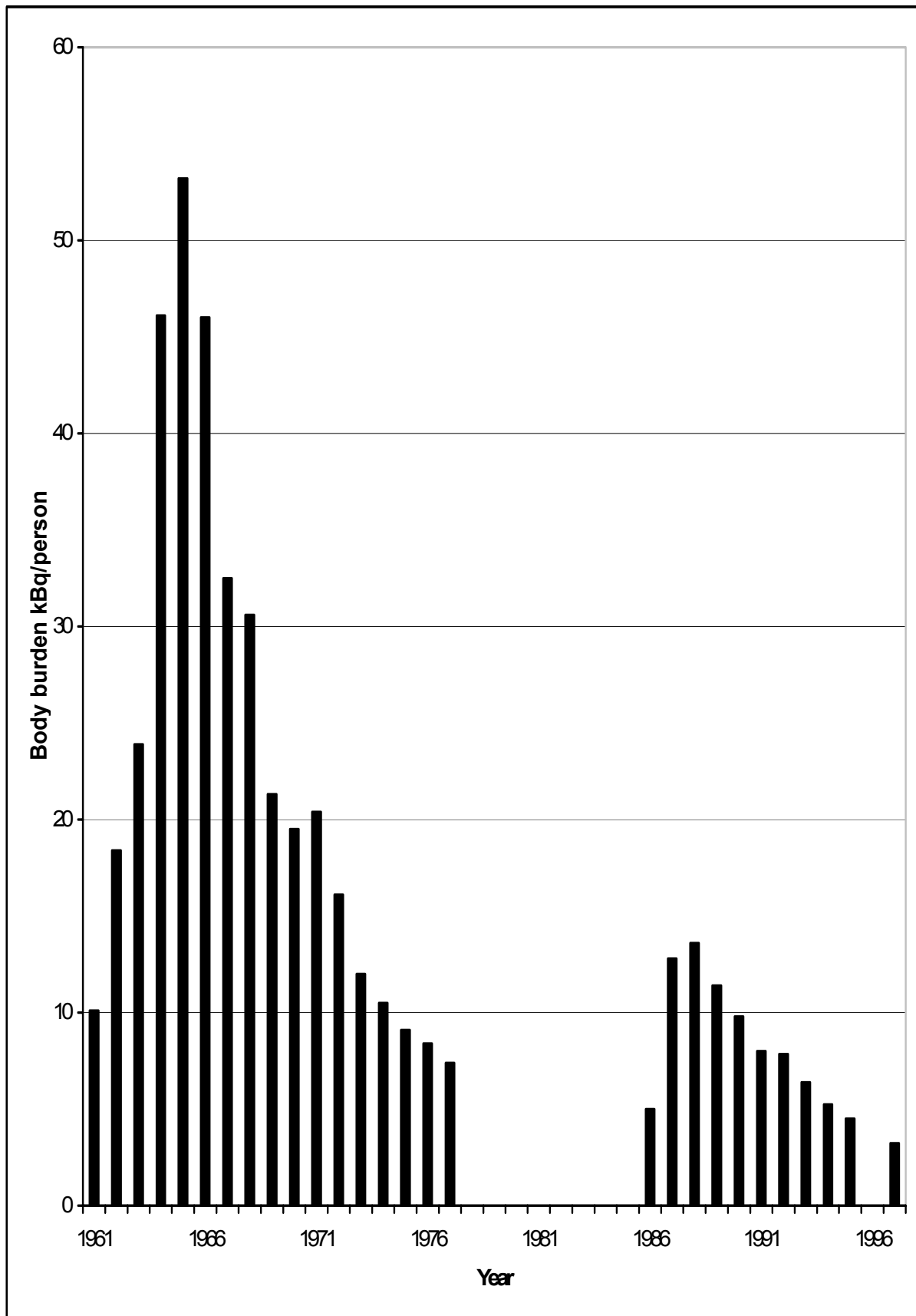


Figure 1. The mean body burden of ^{137}Cs in male reindeer herders of Inari.

Other radionuclides studied in Finnish Lapland

The radionuclides and food chains studies are indicated in the table 2.

Table 2. The transfer of radionuclides along foodchains to man in Finnish Lapland. The radionuclides investigated are given in parenthesis.

1. Lichen - Reindeer - Man (Cs-137, Sr-90, Fe-55, Pu-238, Pu-239,240, Am-241, Cm-242, Pb-210, Po-210)
2. Lake water - Fish - Man (Cs-137, Fe-55, Pb-210, Po-210)
3. Lake water - Sedge and horsetail - Cow's milk - Man (Cs-137, Sr-90)
4. Seawater - Plankton - Fish - Man (Fe-55, Cs-137)

⁹⁰Sr

⁹⁰Sr in reindeer is nearly solely in the bones that are not eaten by man and therefor there is a break in the chain. However, in the bone of the Lapps two times higher ⁹⁰Sr concentrations have been found than in the bone of southern Finns.

⁵⁵Fe

At the first phase of the ⁵⁵Fe studies a method for determination of low concentrations of ⁵⁵Fe from environmental samples was developed. In this method iron was separated from dissolved samples by liquid extraction and cation exchange and electrodeposited onto a copper plate from which the ⁵⁵Fe concentration was determined by means of a thin-windowed proportional counter (10). The maximum amount of iron to be electroplated onto copper plate was about 10 mg. The low background liquid scintillation counters equipped with a pulse shape analyser that allows discrimination between alpha and beta pulses offer at present the best method for the radioassay of ⁵⁵Fe. By liquid scintillation counting the samples containing up to 100 mg stable iron can be counted.

The body burden of ⁵⁵Fe in the male Lapps was 3.3 - 6.3 kBq and in females 3.7 - 5.9 kBq in 1966. Over 98 % of ⁵⁵Fe in the diet of reindeer herding Inari Lapps originated from reindeer meat, liver and blood, but 6 -15 % of ⁵⁵Fe in the diet of the fishing Utsjoki Lapps was derived from salmon of the river Teno and from cod caught in the Arctic Ocean. In the Sevettijärvi Skolt Lapps the specific activity of ⁵⁵Fe (Bq/mg Fe) in blood was in women of fertile age about twofold compared to that of men in the same age group (Figure 2). This is a consequence of the increase in the rate of iron absorption from the diet caused by the iron loss during menstruation. The results of ⁵⁵Fe studies are presented in the first doctoral theses made at the Laboratory of Radiochemistry in 1969 (10).

²¹⁰Po and ²¹⁰Pb

Studies on natural radionuclides of the uranium decay series by Kauranen and Miettinen indicated that the arctic food chain lichen–reindeer–man maintains higher ^{210}Po concentration in soft tissues of reindeer herding Lapps than in those of the southern Finns. The ^{210}Po concentration in reindeer meat varied between 3.2 - 12.4 Bq per kg wet weight and ratio of ^{210}Po to ^{210}Pb from 11 to 42. In reindeer liver the corresponding values were 37.7 - 174 Bq/kg wet wt. and 3.1 - 5.8 for ^{210}Po concentration and the $^{210}\text{Po}/^{210}\text{Pb}$ ratio, respectively (11). Rahola and Miettinen estimated in 1969 the total radiation dose received by the present generation of the Lapps between 1956 and 1985 (12). They found that the two nuclides, ^{137}Cs and ^{210}Po , completely dominated the dose to the whole body and genitals. The dose commitment for the natural radionuclide ^{210}Po was estimated to be in Lapps' gonads 2.5 rem/30 yrs and in their liver 5 rem/30yrs (12).

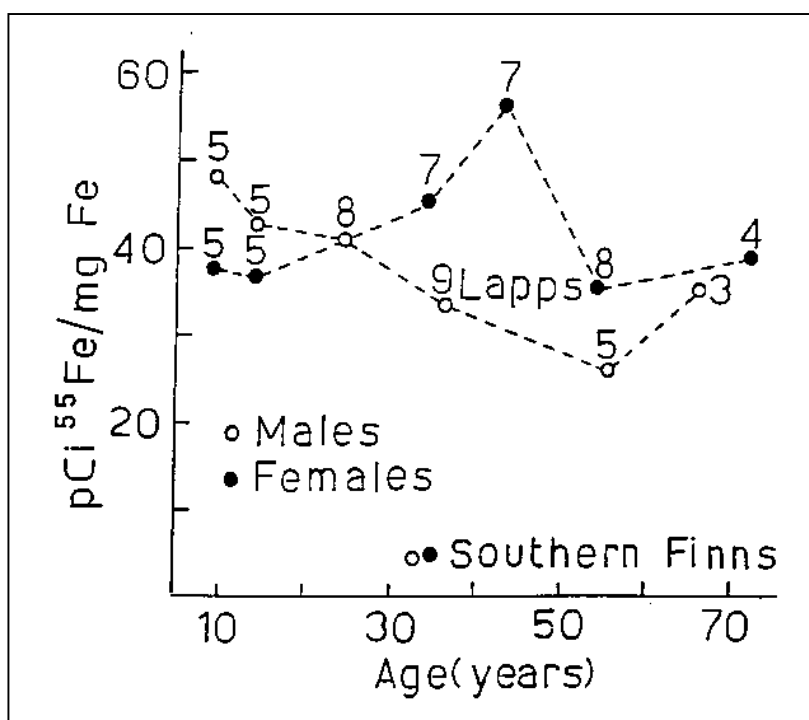


Figure 2. Concentration of ^{55}Fe in the Severtijärvi Skolt Lapps as a function of age in March 1966 (10).

Radioecological Symposium in Stockholm, 1966

This large international symposium on “Radioecological Concentration Processes” was important for the radioactivity studies carried out in Lapland. A big delegation from the Soviet Union in this conference was the first one ever attended an international radioecological conference. Thus, it was possible for the first time to compare the behaviour of ^{137}Cs along the food chain lichen–reindeer–man in the whole arctic zone; in the arctic regions of the USSR, in Alaska and in Lapland. The time of the meeting was very suitable because there was plenty of data available from the maximum period of the global fallout, 1962–1964. The Swedish and Finnish results of the ^{137}Cs body burden in the Lapps decreased in spring 1966 for the first time after the heavy nuclear testing period 1961 - 1962. The decrease from the year 1965 to 1966 varied in different studies from 15% to 25%. About 100 papers were presented and almost 60 % of the papers dealt with ^{90}Sr and ^{137}Cs . At that time no studies on plutonium and americium in the environment were presented. Instead, there was one paper dealing with “Biochemical food chains of uranium in aquatic and terraneous organisms” (6).

In three papers the transfer of ^{55}Fe along the food chains to man was studied. B. Persson and T. Jaakkola had investigated the ^{55}Fe in the food chain lichen-reindeer-man in Swedish and Finnish Lapland (7,8). The body burdens of the ^{55}Fe in Sweden and Finland were 3.7 and 5.9 kBq for males and 7.4 and 9.6 kBq for females, respectively. For natives of Bethel, Alaska who eat large quantities of salmon H.E. Palmer and T.M. Beasley reported the average body burden of ^{55}Fe 25 kBq. In one person the body burden of 54 kBq was found (9).

As a result of the discussions with colleagues from the Soviet Union in Stockholm scientific co-operation between the Institute of Radiation Hygiene in Leningrad and Department of Radiochemistry, University of Helsinki, started. The proceedings of the Stockholm symposium can be regarded still today as one of the basic books in radioecology.

The studies of the transuranic elements in 1973 - 1986

The studies on the transfer of transuranium nuclides along the food chain lichen – reindeer – man started at the Department of Radiochemistry in 1973. From the very beginning plenty of work has been devoted to developing radioanalytical method for determination of plutonium and americium in different biological materials. Due to the high number of samples to be analysed a fast and reliable method had to be developed. For plutonium a method based on one anion exchange step and electroplating onto platinum or stainless steel disc was developed (13). The determination of extremely low concentrations of ^{241}Am from environmental samples was very laborious due to difficulties to separate americium from three valency lanthanides and other cations. Separation method of americium used includes several ion exchange steps, liquid extraction and one or two coprecipitation steps.

Pu in air and plants

The maximum concentration of $^{239,240}\text{Pu}$ in ground air at Helsinki, about 900 aCi/m³ (33 μBq/m³) was found in the spring of 1963. The level in Finnish Lapland was about 20 % lower. In the 1960s and 1970s the radioactivity ratio of $^{239,240}\text{Pu}/^{137}\text{Cs}$ in the ground level was on the average 0.013 (Figure 3).

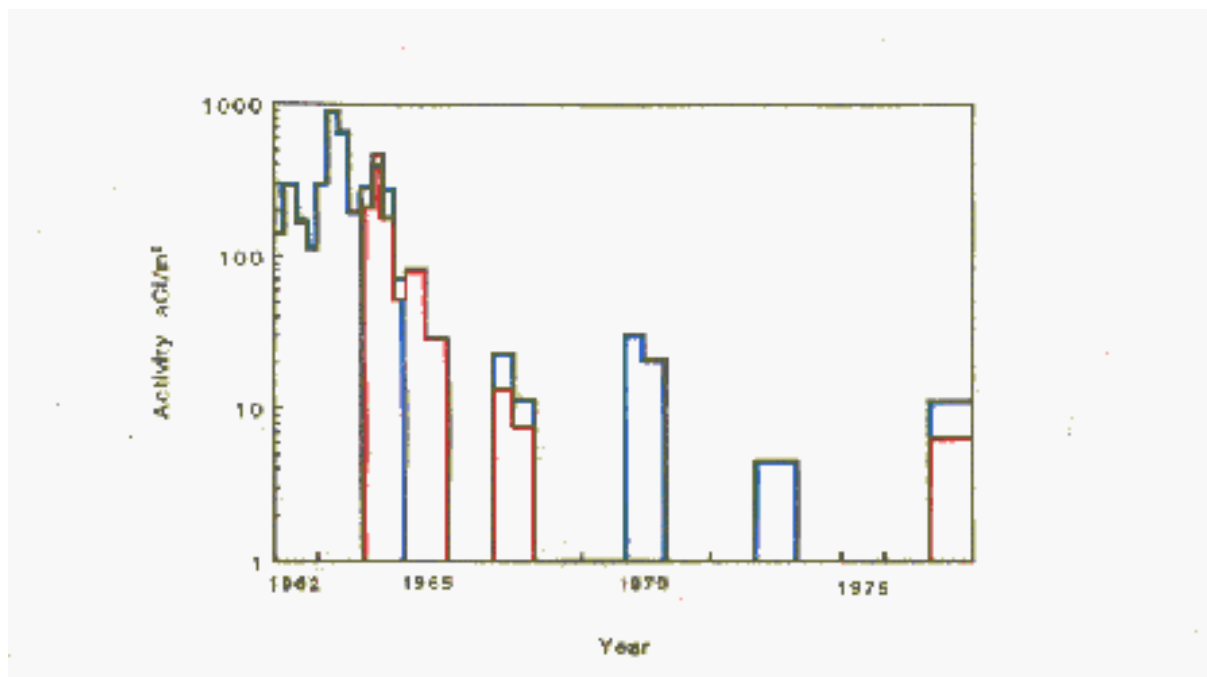


Figure 3. $^{239,240}\text{Pu}$ concentration in ground level air at Helsinki, 60°N (blue) and at Utsjoki, 60°N (red) during 1962-1977.

Plutonium in plants and reindeer

Plutonium concentrations of plants in 1960 – 1977 are presented in figure 4 (15).

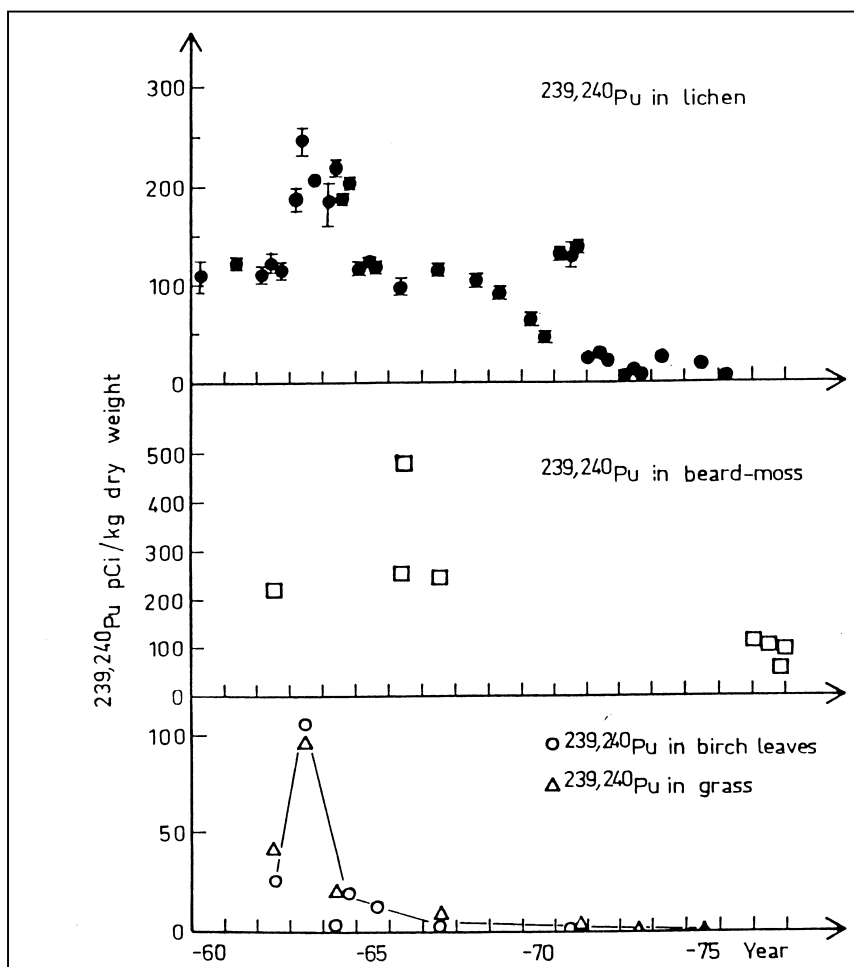


Figure 4. $^{239,240}\text{Pu}$ in lichen (*Cladonia alpestris*), beard moss (*Alectoria species*), in grass and in birch leaves in Finland during 1960-1977 (15).

In reindeer, about 60 % of the plutonium content was in liver, from 10 % to 40 % in the skeleton and about 10 % in muscle tissue. The average concentration $^{239,140}\text{Pu}$ in reindeer liver was 0.56 Bq/kg dry wt. in 1967-1968. The value of 4.0×10^{-5} was calculated for the gastrointestinal tracts absorption of plutonium in reindeer.

Plutonium in man

The plutonium content of the Lapps and southern Finns has been determined by analysing the autopsy tissue samples (Table 3, Figure 5) (14). The ^{210}Po results in the table 3 confirm the accumulation of plutonium in soft tissues of Lapps observed by Kauranen and Miettinen in placenta samples in 1960s (11).

Table 3. $^{239,240}\text{Pu}$, ^{137}Cs , ^{210}Pb and ^{210}Po in liver of male Lapps and southern Finns. The results are given as pCi/kg wet wt. (14)

Case NO Lapps	$^{239,240}\text{Pu}$	^{137}Cs	^{210}Pb	^{210}Po
1E/77 1)	0.27	500	3.5	44.2
2V/77 1)	0.15	3800	8.0	62.2
3A/77	0.29	100	3.0	10.8
4P/78	1.00	3100	-	-
5S/79	0.39	3300	-	-
Mean	0.42	2160	-	-
Southern 2) Finns	0.32 (48)	<50	7.37 (7)	15.4 (7)

- 1) The liver and this case was fatty degenerated
- 2) The number of persons is given in parenthesis

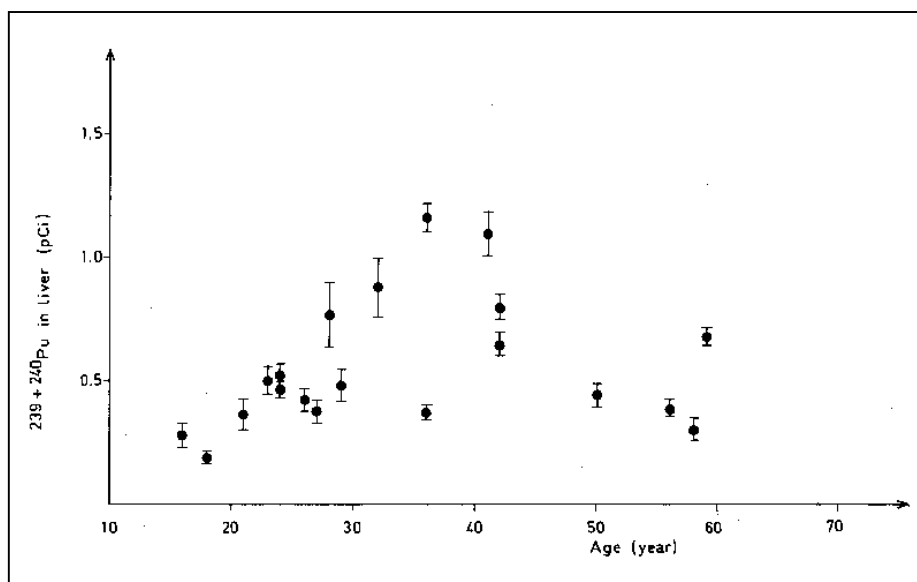


Figure 5. $^{239,240}\text{Pu}$ in liver of southern Finns in 1976 as a function of the person's age (14).

Using the concentration of plutonium in air and tissues of reindeer the ingestion and inhalation intake of $^{239,240}\text{Pu}$ for the period 1945 - 1978 has been estimated. The inhalation intake of the males Lapps has been about 0.5 Bq and that of male southern Finns 0.7 Bq. The corresponding values for the ingestion intake was 20 Bq and 1.3 Bq, respectively. Using these values and a lung model, the calculated fractional absorption factor of plutonium in human GI-tract was the order 7×10^{-4} and in lungs 8×10^{-2} .

The studies after the Chernobyl accident in 1986

The nuclear power plant accident at Chernobyl on April 26, 1986 resulted in several new radioecological projects even in many institutions not earlier involved in this field. Just after the accident it was important to determine the concentration levels of radionuclides in deposition, soil, water, plant, animals and especially in foodstuffs important in human diet to estimate the possible countermeasures needed in view of the radiation protection point. The studies of the fallout from Chernobyl have produced new information on many radioecological aspects for which only modest knowledge has been available. Such aspects have been formation and behaviour of hot particles, occurrence of some radionuclides (e.g. Cm) in environment on which only limited data exist and the role of chemical form and oxidation state as well as the fixing mechanisms on the migration of radionuclides in the environment.

Based on the experience of 25 years in the field of radioecology the research of Department of Radiochemistry was focused after the Chernobyl accident on four topics: 1) Determination of distribution of Chernobyl-derived plutonium deposition in Finland. 2) Continuation of the studies on behaviour of radionuclides in the food chain lichen-reindeer-man in Finnish Lapland, 3) Occurrence of curium in terrestrial and aquatic environment, 4) Behaviour of radionuclides in the southern part of the lake Päijänne, at Asikkalanselkä.

- 1) The research topic one has been performed in co-operation with the Finnish Meteorological Institute and STUK Radiation and Nuclear Safety Authority. Based on plutonium results of lichen, peat, precipitation and surface soil samples a deposition map of plutonium was produced (16). The plutonium deposition in Finland was very unevenly distributed. Even in most contaminated areas the plutonium deposition was only some 10 percent of that in the global fallout from the weapons tests.
- 2) The studies on lichen–reindeer–man include the continuation of the determination of ^{137}Cs body burden of the same Sami population group selected in 1962. After the Chernobyl accident this study was carried out in co-operation with the Radiation and Nuclear Safety Authority. The results are given in figure 1. Plutonium, americium and curium have been analysed in the tissues of reindeer of the southernmost reindeer-herding district, Halla, where the highest fallout from the Chernobyl accident in the reindeer-herding area in Finland was found (Figure 6).
- 3) New information of behaviour of curium in environment has been received by analysing the ^{242}Cm isotope having the highest activity concentration of the alpha-emitting radionuclides from Chernobyl fallout. The ^{242}Cm isotope has been analysed in plant, sediment and reindeer tissue samples (17,18).
- 4) Since the year 1992 the studies on migration of ^{137}Cs and ^{90}Sr in the aquatic ecosystem at lake Päijänne have been performed in co-operation between the STUK Radiation and

Nuclear Safety Authority and Laboratory of Radiochemistry. Asikkalanselkä, the southern part of the lake Päijänne, is located in the area with the highest radioactive fallout in Finland from the Chernobyl accident. The investigation on behaviour of the fallout radionuclides in Asikkalanselkä is important because this basin is via about 100 km long Päijänne tunnel, the main source of raw water for about one million inhabitants in Helsinki area. The most important radionuclides studied have been ^{137}Cs , ^{134}Cs and ^{90}Sr . The results indicate fast removal of radio-caesium from water phase to sediment. In 1987 over 90 % of the fallout radio-caesium was found in sediment (Figure 7). The residence time of strontium in water is much longer than that of caesium.

In 1997 the aim of the study was to analyse some additional water, sediment and soil samples to yield complementary time depending transfer parameters for the model describing the transfer of ^{90}Sr and ^{137}Cs through the aquatic pathway (19).

The work as scientist in the field of environmental radioactivity and radioecology since the first half of the 1960s has been interesting and challenging all the time. The development of the counting techniques and radiochemical separations methods opened possibilities to increase the number of radionuclides studied in the environment and also lower detection limits for many radionuclides.

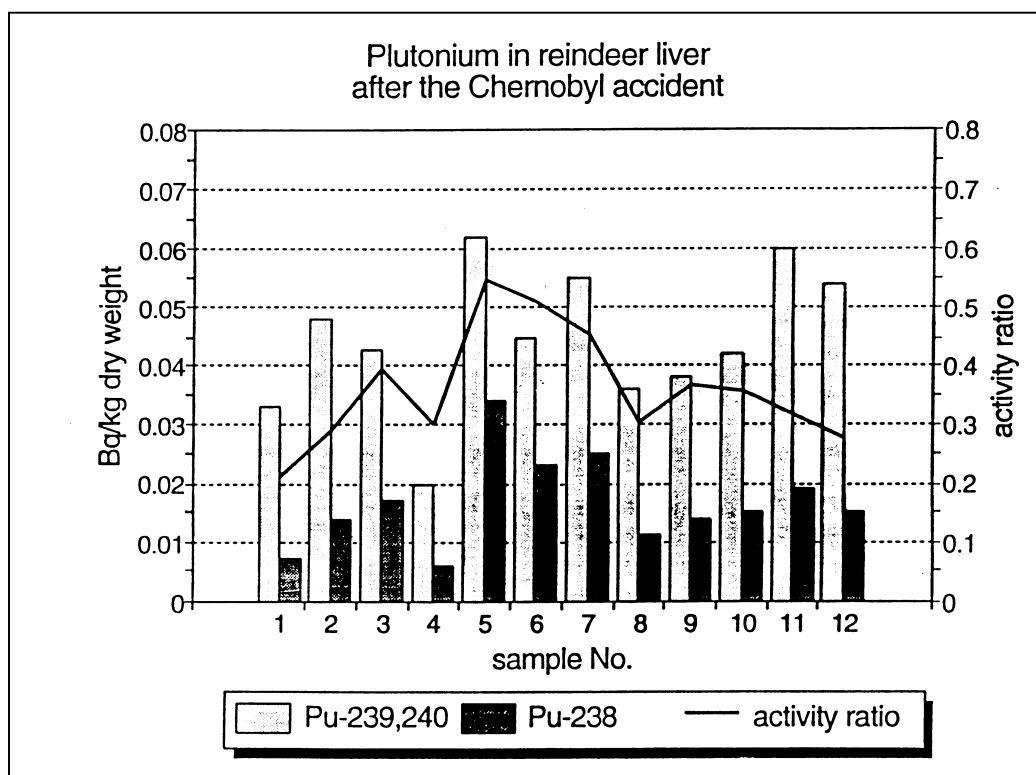


Figure 6. Plutonium in reindeer liver in Finland at southernmost reindeer herding district Halla, in 1986.

The volume of radioecological studies is significantly decreased compared to volume of 1960s and 1970s and the main topics are changed, too. The reasons for this was the cease of the nuclear weapons tests and also the wide information already obtained from the previous radioecological research programs which have improved our knowledge in this research field.

The Chernobyl accident proved the need of internationally sufficient research capability in the field of radioecology. The accident proved also usability of studies on radioactive food chains carried out in Finnish Lapland that helped to minimise the disturbances in the life of reindeer-herding population and in markets of reindeer meat.

The good international contacts between the researchers have always been characteristic for radioecology not least in the Nordic countries. In the period of 1959 – 1969 five symposia on “Radioactivity in Scandinavia” were organised and now we are participating “the Eight Nordic Seminar on Radioecology”.

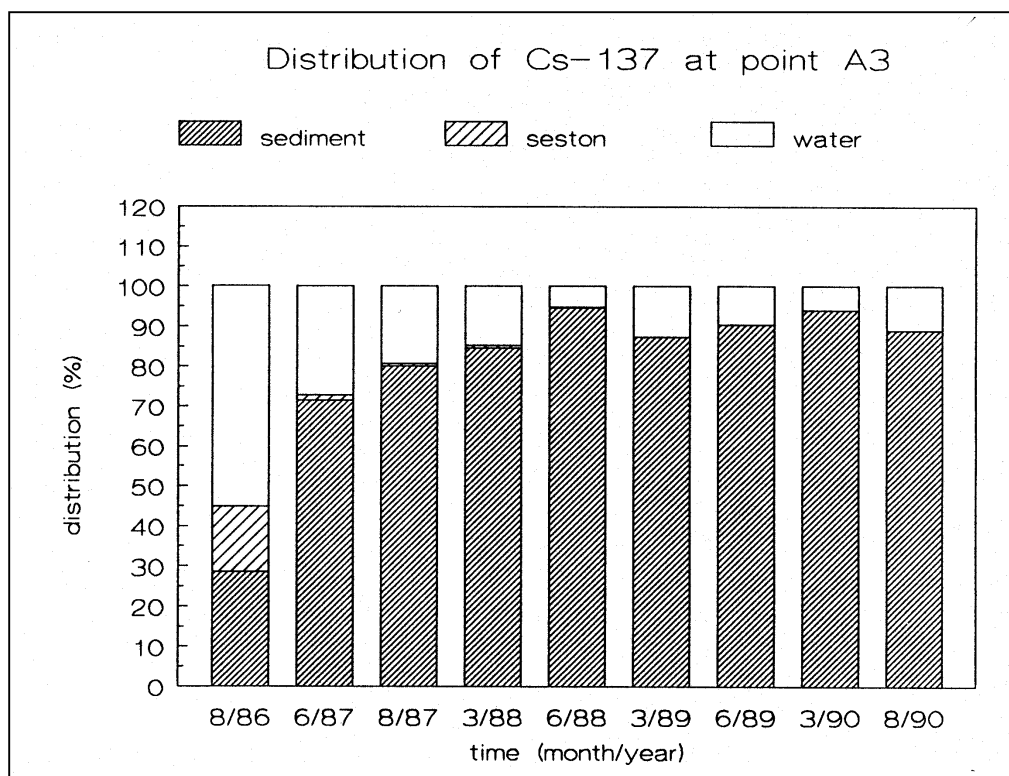


Figure 7. The distribution of ^{137}Cs between water, seston and underlying sediment at the Asikkalanselkä basin, Lake Päijänne in 1986-1990 (19).

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A Nordic view on perspectives for radioecology

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Abstract

At the turn of the millennium, several scientists have expressed their thoughts on the future of radioecology and related topics in the Journal of Environmental Radioactivity. These contributions are listed and commented upon. The role of radioecology in the 6th Euratom Framework Programme (2002-2006) is discussed. Previous Nordic radioecology seminars are summarised and trends identified, and suggestions are given for future Nordic activities and developments in the field of environmental radioactivity.

Introduction

Nearly twenty-five years have passed since the first Nordic radioecology seminar was organised in 1976. In many connections such a time span represents an occasion for looking back, evaluating the past and making guesses and wishes for the future. The fact that the Nordic radioecology seminars have continued for 25 years indicates a tradition with stamina.

Nuclear power plants were built in Scandinavia and elsewhere in the 1970's and this prompted the need for assessing the radiological consequences of possible releases. Following a NKS (Nordic Committee for Nuclear Safety Research) recommendation, the first Nordic seminar on radioecology took place in Nyköping, Sweden, and was followed by a number of joint projects. From the 1980's the use and development of nuclear power in Europe and the USA was generally not favoured by the media, the public and the politicians, partly influenced by major reactor accidents in the USA (TMI, 1979) and Ukraine (Chernobyl, 1986). The latter accident generated considerable interest in and funding to the field of radioecology in Europe, but today 15 years later the funding and public interest has changed and turned to other areas.

Editorials in Journal of Environmental Radioactivity

Many international journals are open to scientific papers dealing with radioecology but the Journal of Environmental Radioactivity (JER) is among the more prominent ones in this field. Inspired by the new millennium, the Editor Murdoch Baxter launched a series of short editorial articles in JER written by members of the Editorial Board (1). This millennium project has produced a number of editorials, which are of considerable interest to those who are contemplating the future of radioecology. The editorials are listed by occurrence in the journal in Table 1, which gives the authors and titles of the editorials.

The members of the Editorial Board have made significant scientific contributions in the field of environmental radioactivity and radioecology. The board members represent a wide range of scientific backgrounds and interests but the editorials indicate that the authors are all personally deeply involved in environmental radioactivity and that they care about the future development of

this field. The views presented in the editorials reflect diversity of opinion but with many common features.

Table 1. Millennial Editorial Series in Journal of Environmental Radioactivity.

Author	Title of Editorial
Murdoch Baxter and Per Strand (2)	JER/IUR: a new co-operation for the new millennium
Marian Scott (3)	Some millennium thoughts
Gabriele Voigt (4)	Trends in radioecology
Yoichiro Ohmomo (5)	A brief history of, and views on the future of, radioecology in Japan
Asker Aarkrog (6)	Trends in radioecology at the turn of the millennium
Erich Wirth (7)	Afterthoughts on Tokai-mura
Ward Whicker (8)	Radioecology: relevance to problems of the new millennium
Bernard Wilkins (9)	The development of countermeasure strategies in agricultural systems
Tom Hinton (10)	Strong Inference, Science Fairs, and Radioecology
Dennis Woodhead (11)	Whither marine radioactivity studies?
Elis Holm (12)	The Swedish nuclear dilemma
Gabriele Voigt (13)	Are radioecologists the dinosaurs of this century?
Gennady Polykarpov (14)	The future of radioecology: in partnership with chemo-ecology and eco-ethics
William Burnett (15)	So, what is environmental radioactivity anyway?
Brit Salbu (16)	Hot particles – a challenge within radioecology

One of the common points is that radioecologists and others have not been successful and effective to explain and communicate radiological aspects to the public and the media. This has been characteristic for the public debate since the 1970's following all major incidents and accidents involving radioactive material and nuclear facilities. The public debate concerning the military use of depleted uranium is a recent example of this situation.

Another common opinion is that radioecology may help to provide better tools for the remediation of environments contaminated with radioactive material. This illustrates the general development from basic science towards a more applied approach or, as the European Commission puts it: From science push to society pull.

A new scientific challenge in the field of environmental radioactivity is that of eco-dosimetry, i.e. the negative effect on the whole ecosystem. The precautionary and biodiversity principles are now applied in environmental protection against all pollution types and must also be applied for radioactivity. Radioecology will here find new land to cover in the development of primary dose rate criteria for the protection of flora and fauna and identification of reference and critical species.

A common concern is the problem of maintaining and developing radioecological expertise on the long term. It is becoming increasingly difficult to attract young scientists into this field due to reduced funding and the poor public image of nuclear energy.

However, new developments like the one mentioned on eco-dosimetry and new analytical techniques like mass spectrometry will help on this situation. Furthermore, radioecology holds a potential value to science and society in the use of radioactive tracers to understand and measure natural processes. One example is that disequilibrium within the natural U/Th decay series in water

and sediments can be used to give information on a range of environmental processes (e.g. sediment accumulation rates, particle fluxes, and ages of sediment layers).

European Trends

Preparations are under way to go beyond the current 5th Framework Programme of the European Union (1998-2002) and reflect on issues to be covered by the 6th Euratom Framework Programme (2002-2006). The Scientific and Technical Committee Euratom (STC) which is attached to and provides advice to the European Commission makes such reflections (17). The STC has presented its view on the general outline of a research strategy in a 20-50 year perspective, which is summarised in the following:

- EU depends on electricity at a reasonable price
- Euratom R&D strategy based on global sustainable development
- One third of electricity is based on nuclear in EU today
- Unrealistic that renewables can replace nuclear within 20-30 year
- R&D objective to ensure that future generations can choose between technologies
- Technical R&D therefore must include both fission and fusion
- Public financing needed for future scientific and technical infrastructure
- STC recommends an increased European nuclear R&D budget from 2002.

The STC mentions the following strategic items, which are of direct relevance to radiation protection and radioecology:

- Environmental impact assessments needed for nuclear waste disposal facilities
- Radiation protection criteria should be developed for the general protection of the environment
- Concern about elevated levels of natural radiation affect community planning
- Maintenance and development of competence in radiation protection is important to ensure high standards of healthy living and working conditions within the EU.

In spite of the lack of new nuclear build in Europe there seems to be room for nuclear energy and related research beyond 2002 if the European Commission takes the advice of the STC.

Nordic Radioecology

Nordic collaboration on radioecology was active in the 1960's when meetings on Radioactivity in Scandinavia were held from 1962 to 1969. The first Nordic radioecology seminar in 1976 was held with the following headlines:

- Aquatic radioecology
- Terrestrial radioecology
- Methods, Environmental monitoring
- Transuranics
- Education and co-operation.

The subsequent Nordic radioecology seminars covered corresponding topics, and Table 2 shows a listing of the seminars and the number of presentations grouped by topics. Aquatic and terrestrial radioecology and methods seem to cover the majority of the presentations across the seminars with

less interest in models, doses, whole body measurements and education. The interest in education seems to have been particularly present in the 1970's, whereas the interest in whole body measurements seems to have intensified following the Chernobyl accident.

Table 2. Presentations at the Nordic radioecology seminars grouped by main topics.

Venue, year	Aquatic	Terrestrial	Models	Methods	Whole body	Doses	Education
Norrköping (SE), 1976	13	7		8			1
Helsingør (DK), 1979	17	16	1	2		1	3
Hyvinkää (FIN), 1982	14	12	3	9		2	
Gol (NO), 1985	14	11	4	9		5	
Rättvik (SE), 1988	14	20	8	13	8		
Torshavn (FO), 1992	12	22		8	2	3	
Reykjavik (IC), 1996	7	25	3	10	9	21	
Rovaniemi (FIN), 2001	20	19	2	9	2	2	

Generally, the seminars have attracted from 60 to 100 participants. The two exceptions are Rättvik in 1988 with 135 participants due to the Chernobyl accident two years earlier, and Reykjavik in 1996 with 182 participants where the seminar was organised back-to-back with the general meeting of the Nordic Society of Radiation Protection. Figure 1 shows the number of participants at the seminars by country.

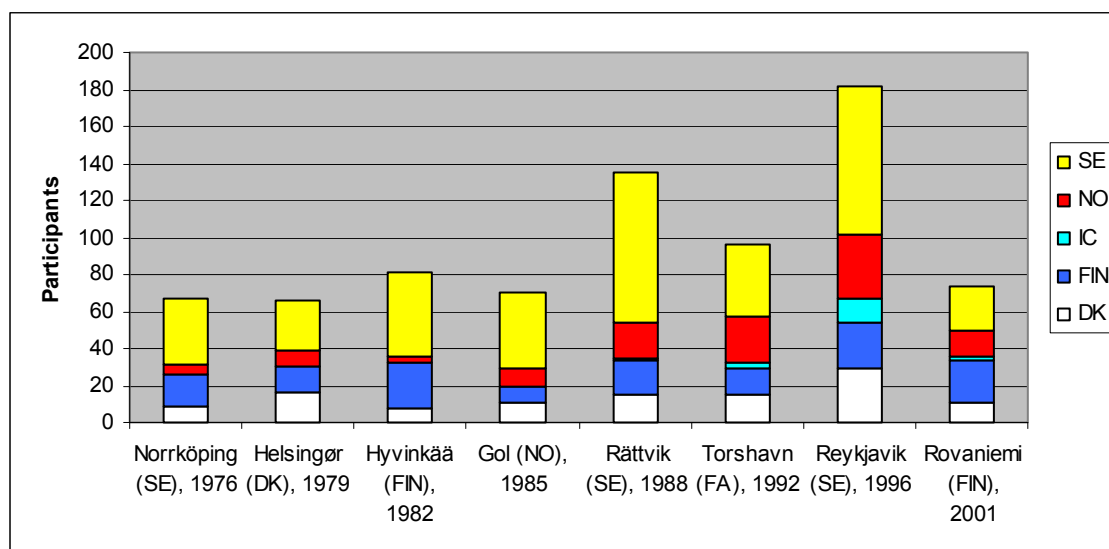


Figure 1. Number of participants at the Nordic radioecology seminars shown by country.

Perspectives for Nordic radioecology

Radioecology is a child of radiation protection and therefore linked to nuclear energy. Although nuclear energy presently is not the most popular means in Europe of producing electricity, there are no realistic alternatives for the next 10-20 years. Therefore there will continue to be a need to maintain and develop the competence in radiation protection including radioecology. The Nordic countries have an important tradition and background in radioecology as demonstrated from the radioecology seminars and the many internationally recognised Nordic scientists in this field.

Presently the Nordic co-operation on nuclear safety research including radioecology is facilitated through the NKS to a large extent. The NKS plays here an important role and should continue to do so. The many challenges for radioecology should be addressed and co-ordinated through the next NKS programmes. There are plenty of tasks to take up. Perhaps the issue of education and training is one that should have a higher priority in the future.

The time elapsed between the Nordic radioecology seminars during the 25-year period has increased rather steadily from about 2.5 to 4.5 years and if we make a linear extrapolation, the next seminar would be expected at Christmas Day 2005. Perhaps we can break the curve and meet again earlier than that.

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Radioactive contamination in Arctic - present situation and future challenges

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Introduction

There is currently a focus on radioactivity and the Arctic region. The reason for this is probably the high number of nuclear sources in parts of the Arctic and the vulnerability of Arctic systems to radioactive contamination. The Arctic environment is also perceived as a wilderness and the need for the protection of this wilderness against contamination is great. In the last decade information has also been released concerning the nuclear situation which has caused concern in many countries. Due to such concerns, the International Arctic Environmental Protection Strategy (IAEPS) was launched in 1991 and the Arctic Monitoring and Assessment Programme (AMAP) was established. AMAP is undertaking an assessment of the radioactive contamination of the Arctic and its radiological consequences. In 1996 IAEPS became part of the Arctic Council. AMAP presented one main report in 1997 [AMAP 1997] and another in 1998 [AMAP 1998]. There are also several other national, bilateral and international programmes in existence which deal with this issue. This paper summarises some of current knowledge about sources of radioactive contamination, vulnerability, exposure of man, and potential sources for radioactive contamination within Arctic and some views on the future needs for work concerning radioactivity in Arctic.

Past and present radioactive contamination

There are three main sources of radioactive contamination in the Arctic today; global fallout from nuclear tests, releases from nuclear reprocessing plants in Western Europe and Chernobyl fallout. A total of 518 atmospheric nuclear explosions took place up to 1980. The spatial distribution of ^{137}Cs of this fallout onto land is shown in Figure 1. [Strand et al. 1998].

The major source of radiocaesium to the Arctic from West European nuclear reprocessing plants is Sellafield in the UK. Releases started in 1952 and continue up to the present day with major releases having occurred between 1974 and 1982. The releases ^{99}Tc and ^{137}Cs from Sellafield and the subsequent contamination levels in the Barents Sea are shown in Figure 2. Releases of ^{99}Tc increased in 1994 from the reprocessing plant at Sellafield and have recently led to increased concentrations of ^{99}Tc in northern marine environments and further into the Arctic Seas.

Following the Chernobyl accident in 1986, a major release of radioactive material spread over large areas in Europe, with the radioactive cloud and fallout also reaching Arctic areas, mainly the North West Russia and Northern Fenno-Scandia, with deposition levels in the order of $1\text{--}5\text{ kBq/m}^2$ ^{137}Cs . Deposition levels in the order of $10\text{--}200\text{ kBq/m}^2$ were, however, observed just south of the Polar Circle. Radioactive contamination of the North and Baltic Seas provides continuously indirect contamination to the Arctic marine environment via transport pathways along the Norwegian coast.

There do also exist several other sources which have given small or only localised contamination such as the Thule accident in Greenland, where an American plane carrying nuclear bombs was

involved, from the accident with the Komsomolets submarine in the Norwegian Sea, waste storage sites and from dumping of radioactive waste in the sea. Former releases from the nuclear reprocessing plant at Mayak in the Urals, even if it has not contaminated Arctic areas to a substantial degree with earlier discharges, is today a considerable source contained in the environment and, through the potential for transport of radionuclides via the Ob River, is a source to be considered as a threat to the Arctic environment. The releases of radioactive caesium and strontium from Mayak were nearly 100 times higher than the releases of these radionuclides from Chernobyl. Today the radionuclide inventory is stored mainly in lakes and reservoirs, with potential risk for leakage to the rivers and into the Northern Seas.

Exposure of man from past release

The largest contribution to radiation doses from radioactive contamination to the population in northern areas has come from fallout from nuclear tests in the 1950's and '60's. In some areas also the Chernobyl fallout has contributed significantly to the total dose. The population in northern areas is exposed to a dose, from radioactive contamination, which is about 5 times higher than the dose for people in more temperate areas with the same deposition. This illustrates the higher transfer and vulnerability of the Arctic areas from ^{137}Cs contamination.

Indigenous people in the Arctic, mainly living off traditional food products from mountain, forest and lakes, e.g. reindeer-herding Saamis, receive the highest doses from radioactive contamination.

Radiation doses to both the average population and the special groups depend mainly on intake of locally-produced terrestrial products. In comparison, Arctic population groups with mainly marine products in their diet, receive relatively low doses. The most exposed population groups in the Arctic can on average receive up to 50 times higher individual doses than members of the average population.

The intake by the general Arctic population of radiocaesium from different foodstuffs during the period 1990-1994 in the 8 Arctic countries are shown in Figure 3 [Strand et al. 1998]. For the average Arctic populations, a range of different food products contribute to the total ^{137}Cs intake. Canada is an exception, with the domination of reindeer meat consumption in the intake of radiocaesium. The regional effect on dietary preferences is also clearly shown in Figure 3. For example, goat cheese is only an important source for radiocaesium intake in Norway. Mushrooms are important in Sweden, Finland and Russia. However, there is a considerable lack in knowledge concerning the importance of natural food products in the transfer of radionuclides to man. In contrast, people who consume only marine products, such as marine fish and mammals, receive doses that are at least an order of magnitude lower than people consuming terrestrial products such as reindeer/caribou meat, freshwater fish and mushrooms. The intake for the selected groups were considerably higher as shown in Figure 4. The major contributor to the intake for these groups was reindeer meat.

The major contribution ($\approx 15\,000$ manSv) to the collective dose to Arctic populations results from fallout from nuclear weapons testing, with a range of individual dose commitments between 1 and 150 mSv [Strand et al. 1998]. The second most important contribution (≈ 500 manSv) to collective dose within the Arctic derives from the Chernobyl accident, with individual dose commitments normally in the range 1 to 50 mSv. Individual annual doses to the most exposed residents of the Arctic from Chernobyl releases, however, could be approximately 10 to 20 mSv/y in the most affected areas. Countermeasures introduced by some countries following the Chernobyl accident

and, in some cases, maintained to the present day, have resulted in reduced individual doses and dose commitments. Releases from the Sellafield fuel reprocessing plant provide the third most important collective dose contribution (≈ 50 manSv) with a relatively small contribution to individual dose (*i.e.* in the range 0 to 0.05 mSv) [CEC 1990].

The total exposure of man from the dumping of nuclear waste is very small even taking into consideration the potential future dose. Smaller-scale releases from accidents in military operations, such as those in northern Russia, the plutonium spill at Thule and the loss of the *Komsomolets* submarine in the Norwegian Sea, have resulted in no significant increases in radiation exposures to the Arctic populations. For other releases to the environment, such as from the Mayak reprocessing plant, it has been difficult to assess the collective or individual doses to Arctic populations from the releases especially, before 1960.

Potential threats to Arctic and future need for risk and impact assessment

The concentration and number of nuclear installations and the potential for releases cause concern, especially since the vulnerability of Arctic populations is much greater than for populations in temperate areas due to the importance of terrestrial semi-natural exposure pathways. The largest threat to the environment and the population in the Arctic today is connected to potential accidents in nuclear power plants, during handling and storage of nuclear weapons, decommissioning and refuelling of nuclear powered vessels and during storage of radioactive waste.

The future work should concentrate on an assessment of the possible consequences of potential major radiation accidents or releases in the Arctic terrestrial or aquatic environments and their vicinity. Knowledge gaps relating to important pathways of human exposure and environmental contamination in the Arctic, such as long-term migration of radionuclides, changes with time in characteristics of the diet of different Arctic population groups, reasons for the high variability in levels determined in different foodstuffs need to be addressed. To fulfil some of these gaps, additional experimental, surveys and modelling work will be needed. High priority should be given to studying the site-specific vulnerability of particular Arctic regions and communities as an essential basis for the prediction of consequences of potential radioactive contamination. Radiation monitoring in the Arctic environment should be continued, in order to acquire both spatial and temporal information as input data for assessment. Finally, the need for developing a system for assessing the consequence of radiation exposure for Arctic flora and fauna has a high priority. There has historically been a focus on the consequences on peoples' health, not the effect on the environment itself, in assessing the impact of radioactive contamination. This work needs collaboration at an international level and, with this in mind, AMAP and IUR will work together on this topic.

Risk Management - need for closer links between Risk assessment and action program's

It is imperative that action is based on risk and impact assessments and furthermore that the results of such actions are reassessed. Currently, communication and interaction is poor between the existing Risk and Impact assessment Programs devised to assess and monitor contamination in the Arctic and the Action Programs tasked to devise strategies and respond to existing radioactive contamination sources by implementing short- or long-term solutions. It is vital to bridge this gap and foster an interdependence between the Risk Assessment and Practical Programs to improve monitoring, response strategies and the implementation of action plans.

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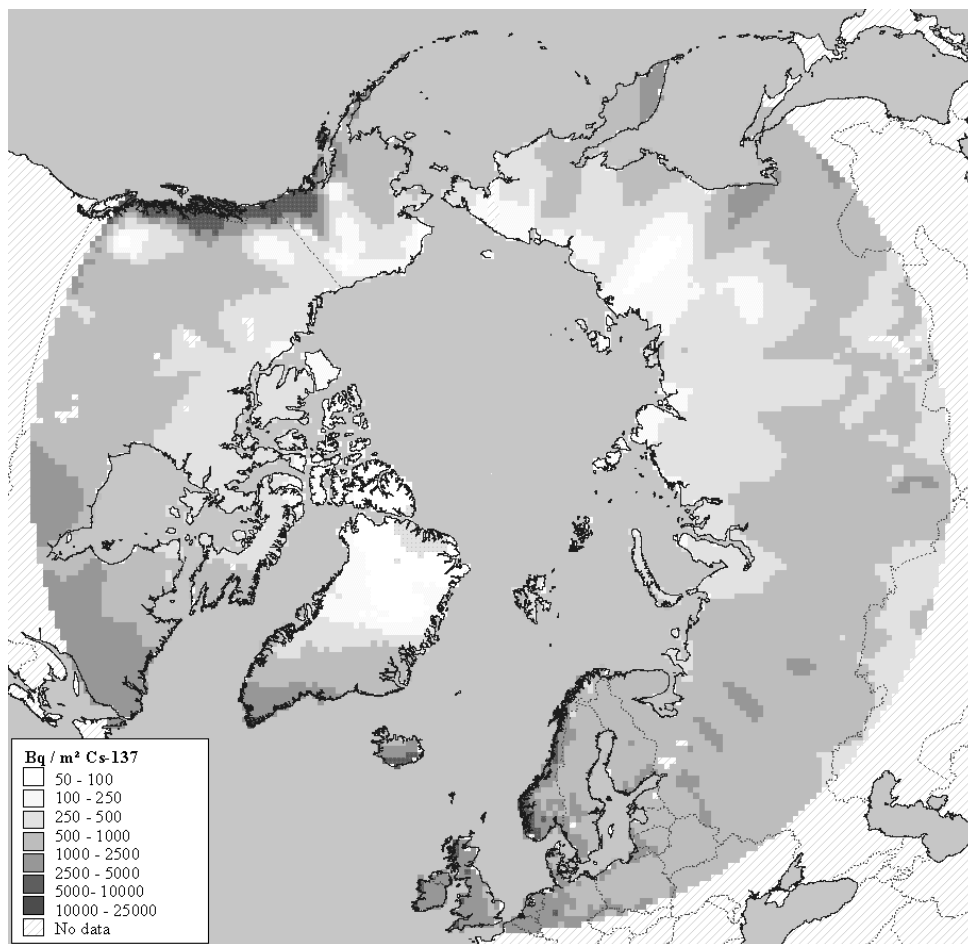


Figure 1. Estimated spatial distribution of ^{137}Cs fallout from nuclear weapons testing

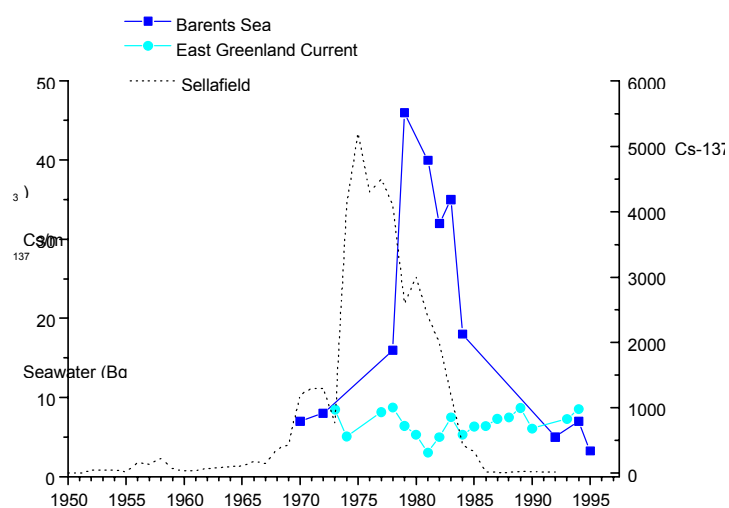


Figure 2. ^{137}Cs activity concentrations in the Barents Sea compared to the yearly releases from Sellafield.

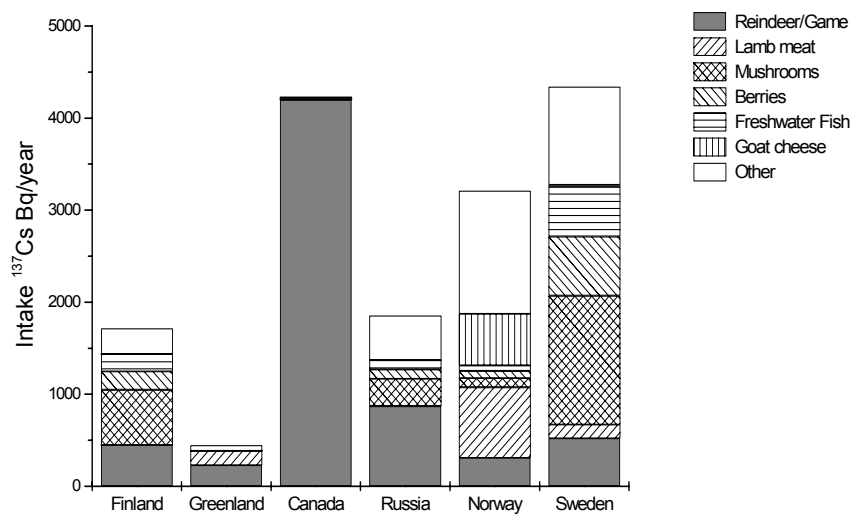


Figure 3. Yearly intakes by the average populations during 1990-1994

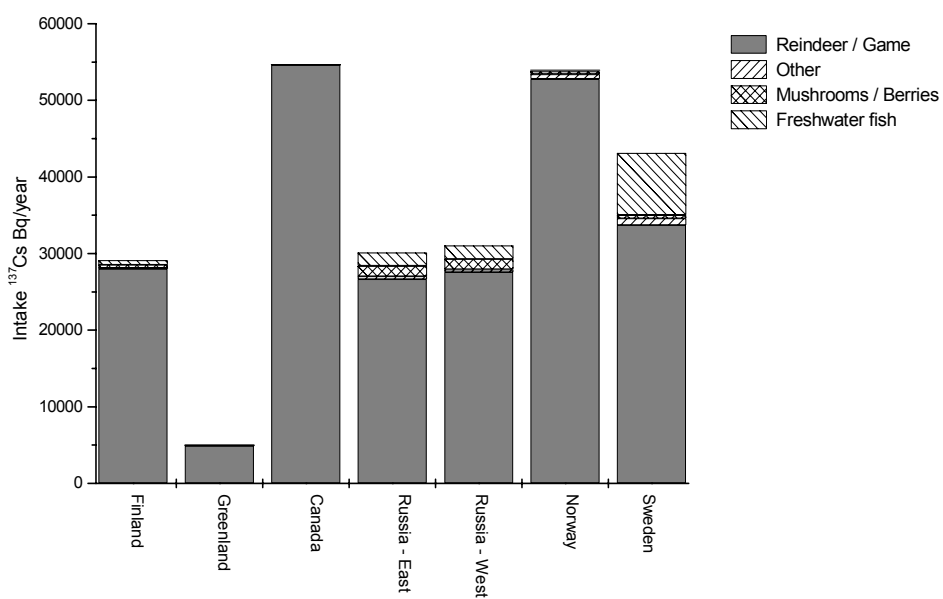


Figure 4. Yearly intakes by the selected indigenous groups, during 1990 -1994

Views on the effects of radioactive deposition in different environments

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Abstract

It is well known that after the atmospheric bomb tests in the 1950's and 1960's with rather even Cs-137 deposition all over Finland the concentrations in plants, animals and people were higher in Finnish Lapland than in other parts of the country. The enrichment of Cs-137 in different foodchains in subarctic and arctic environments were intensely followed. In 1986 after the Chernobyl accident the deposition was uneven varying from a few to about one hundred kilobecquerels per squaremeter. This resulted in big variations in activity concentrations in foodstuffs especially those of seminatural or wild origin.

In Finland different population groups where studied using whole-body counting as well as indirect methods to assess internal radiation doses. In this presentation results of measurements on three different groups of people will be given.

Two of them were studied already in the 60's, the Helsinki reference group and the group of reindeer herders from Lapland. The third was a group from Padasjoki an area with the highest Cs-137 deposition in Finland after 1986. The Helsinki group also represents the whole Finnish population as could be seen from measurements simultaneously done on a statistically chosen population group and on the Helsinki reference group.

In the Helsinki group as also for most of the people in the population group most foodstuffs were of agricultural origin as opposed to the special groups in Lapland and Padasjoki. In the latter groups the people consumed many foodstuffs of wild origin such as reindeer meat, freshwater fish, berries and mushrooms. The highest mean body content in the group of male reindeer herders was 55 kBq in 1965 but only 15 kBq in 1987 when the second maximum was noted. In the Helsinki and Padasjoki groups the highest maxima were noted in 1987, 3 and 12 kBq respectively. Our studies have shown the great influence of the consumption of foodstuffs of wild origin. To establish the correlation between deposition and body content and hence radiation doses it is necessary to introduce more parameters. As these parameters might change with time it is necessary to regularly re-evaluate the situation. It is important to do follow up studies on people to get better knowledge of the longterm behaviour of cesium in the foodchains and in man.

Cs-137 fallout in Iceland, model predictions and measurements

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Basically all the fallout Cs-137 in Iceland came from the atmospheric nuclear weapons tests in the late fifties and early sixties, the addition from the accident in the Chernobyl Nuclear Power Plant was relatively very small. Measurements of fallout from nuclear weapons tests started in Iceland over 40 years ago and samples of soil, vegetation and agricultural products have been collected from various places and measured during this period. Considerable variability has been seen in the results, even between places close to each other. This is understandable due to the mountainous terrain, changing strong winds and high levels of precipitation. This variability has been especially noticeable in the case of soil samples. The important role of uncultivated rangelands in Icelandic agriculture (e.g. for sheep farming) makes it necessary to estimate deposition for many remote areas. It has thus proven difficult to get a good overview of the distribution of the deposition and its subsequent transfer into agricultural products.

Over a year ago an attempt was made to assess the distribution of Cs-137 fallout in Iceland. The approach is based on a model predicting deposition using precipitation data, in a similar manner as that used previously within the Arctic Monitoring and Assessment Programme (AMAP) (Wright *et al.* 1999). One station close to Reykjavík (Rjúpnahæð, code in this paper: RJU) has a time series of Cs-137 deposition data and precipitation data from 1960 onwards. The AMAP deposition model was calibrated for Iceland by using deposition and precipitation data from this station.

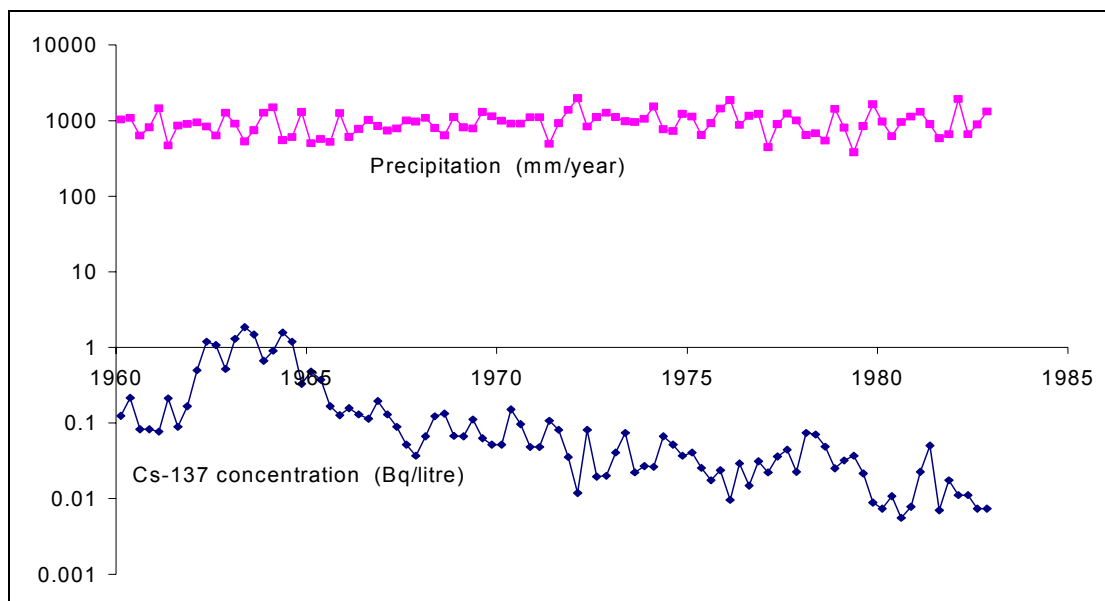


Figure 1. Precipitation rate and concentration of Cs-137 in precipitation at Rjúpnahæð (RJU) station. (Concentration levels refer to time of sampling).

Using these data and decay correcting the Cs-137 activity to last quarter of 1999 gave an estimate of 1.8 kBq/m^2 for each 1000 mm annual rainfall. A map showing predicted deposition was produced (Figure 2), what remained was to test the prediction against actual measurements.

A follow-up study was begun in the summer of 2000 aiming at obtaining a better overview of the Cs-137 deposition in Iceland and then to study systematically the behaviour of radiocaesium in different soil types, since the young and volcanic soils in Iceland are in many ways untypical compared to soils elsewhere. The preliminary results of the soil study are presented in a separate paper by Magnús Á Sigurgeirsson *et al*, "Radiocaesium (Cs-137) fallout in Iceland and its behaviour in Arctic volcanic soils".

Improvements were made to the soil sampling procedure. The choice of procedure is critical in an uneven terrain as found in Iceland. A new procedure was adapted, involving taking 20 cores for each sample. A description of the procedure adapted can be found in Isaksson (2000).

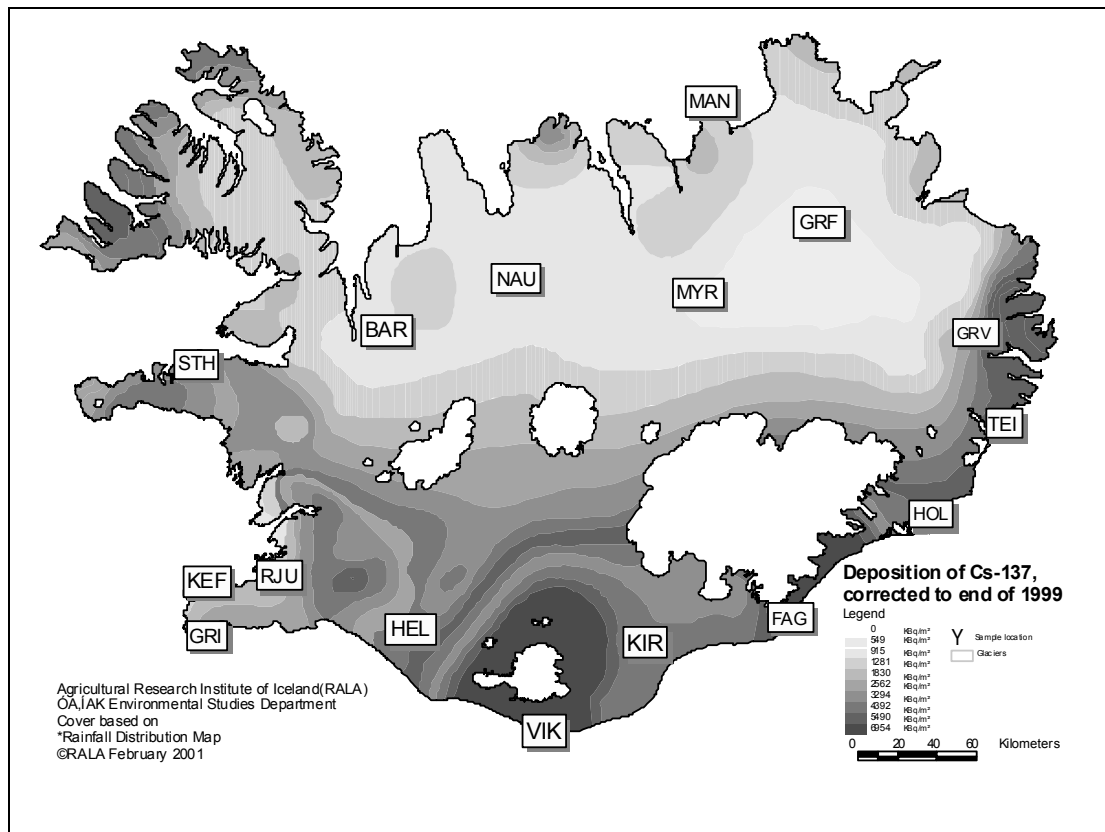


Figure 2. Map of predicted cumulative deposition of Cs-137 from atmospheric nuclear weapons testing, decay corrected to last quarter of 1999. Also shown are the 16 sites where soil samples were collected in autumn 2000.

Strong winds and uneven landscape help to create a very uneven precipitation pattern in Iceland. Accurate precipitation estimates were essential in order to be able to test the model through collecting soil samples and comparing predictions with measurements. In order to get as good precipitation data as possible; sampling was now undertaken close to meteorological stations where precipitation data were available from the sixties. Samples were collected from 16 such sites during the autumn of 2000. The sites were selected so that they would span a range of precipitation rates as well as representing different areas and soil types. The distribution of sites can be seen on the map of predicted deposition, in figure 2. The results of the comparison of model predictions and actual measurements are shown in Figure 3.

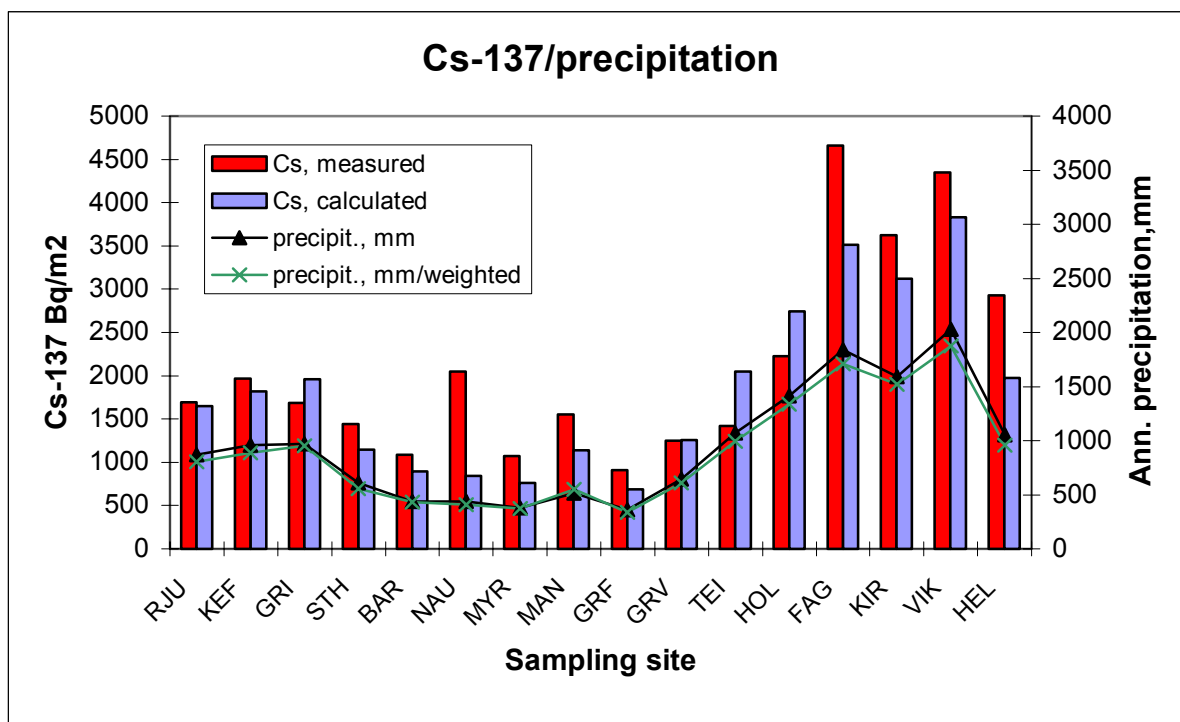


Figure 3. Cs-137 deposition at 16 sites close to meteorological stations, predicted values based on precipitation data and measured values. The annual precipitation is also shown, both as 8 year average (1960-1967) and as a weighted average with each month having weight corresponding to the concentration of Cs-137 observed in rainwater.

The correlation between measured and predicted values for Cs-137 deposition was very good for all places except one, Nautabú farm (NAU). The discrepancy may be due to difficulties with meteorological measurements at that site as well as some uncertainty about the previous use of the site, which is a hayfield close to the farmyard at Nautabú. The graph shows little difference whether a simple average is used for the years 1960-1967 or a weighted average (using the Cs-137 concentration in rainwater as a weighing factor). If the 30 year average was used, then a slight difference could be seen for some of the sites.

In the AMAP study data were analysed for 50 samples from Greenland, Norway and Russia from the period 1961-1985. A line through (0,0) was fitted to the data set using least squares regression. A r^2 value of 0.51 was obtained (Wright *et al.* 1999).

The study presented here gives a corresponding r^2 value of 0,95 and if the station NAU is removed the r^2 increases to 0,97. It should be borne in mind that it is easier to obtain good correlation for sites close to meteorological stations where precipitation has been measured in a consistent manner and soil samples taken by the same team over a very short period of time, which are subsequently processed and measured in the same manner.

The results show that very good estimates of Cs-137 deposition can be obtained, provided good precipitation data are available. The study is continuing. More soil samples will be collected. Work on a more detailed precipitation map of Iceland is in progress. When that will be ready it will be used for making a new deposition map.

Further analysis involves the use of the deposition map in a GIS system, combining it with a soil type map and the results of analysis of Cs-137 availability in different soil types. Those results are then being combined with a vegetation map in order to investigate the correlation of predicted concentration in vegetation in different areas to measured concentration in agricultural products there.

The study is carried out in co-operation between the Icelandic Radiation Protection Institute, the Agricultural Research Institute and the Icelandic Meteorological Office. This study is a part of the NKS/BOK-2 project and has received support from the NKS through the project.

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A regional survey of deposition of Cs-137, based on precipitation

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Abstract

A method to determine the amount of deposition of ^{137}Cs based on precipitation measurements has been tested in the province of Skåne (an area of about $100 \times 100 \text{ km}^2$) in the southern part of Sweden. By measuring the activity concentration of the precipitation at a single location the deposition over a larger region has been calculated by multiplying the activity concentration with the amount of precipitation obtained from a network of between 113 and 143 precipitation stations. The deposition of ^{137}Cs in the investigated area amounts to about $1\text{--}3 \text{ kBq/m}^2$ and is almost equally divided between fallout from nuclear weapons tests and from the Chernobyl nuclear accident. Both the pre-Chernobyl and post-Chernobyl depositions have been calculated from the precipitation measurements. The method was validated by measuring the activity concentration of ^{134}Cs and ^{137}Cs in soil samples, by comparisons with in situ measurements and by aerial surveys. The soil samples were collected at 16 sites distributed in a grid pattern over the whole of the investigated area and 16 sites distributed in a grid pattern over the city of Lund. Yearly samplings have also been performed between 1988 and 1998 at Blentarp. The agreement between calculated and measured deposition is good for the pre-Chernobyl, as well as, for the post-Chernobyl deposition. It may then be concluded that this method allows a good and detailed knowledge in retrospect of the deposition, as well as, a way to make a good first prognosis about fresh fallout.

Introduction

The nuclear weapons tests in the atmosphere which started in the 1940:ties and which caused a deposition of radioactive material all over Sweden, reached a peak between 1962 and 1966. This deposition amounts to 1 to 3 kBq/m^2 of ^{137}Cs . Due to the meteorological conditions the deposition is much more evenly spread both geographical and in time than the deposition caused by the Chernobyl accident in 1986. Because of the occurrence of ^{134}Cs together with ^{137}Cs in the Chernobyl deposition it is possible to distinguish the deposition from nuclear weapons tests from that of Chernobyl in soil samples (Isaksson & Erlandsson, 1995; 1998).

On 1st April 1962, regular measurements of the activity concentration in the air and in the precipitation were initiated by the National Defence Research Establishment at Ljungbyhed (situated almost in the middle of the investigated area) in Sweden (Lindblom, 1966; Bernström, 1969; 1974; De Geer *et al.*, 1978, FOA 41). The Elleson dataset (Elleson, 1993), based on a dense network of daily precipitation measurements, in combination with the Swedish Meteorological and Hydrological Institute (SMHI) rain-gauge network, also enables an extraordinarily detailed estimation of the spatial distribution of the precipitation in the area. Several investigations have

shown that the variations in the deposition are closely connected to the precipitation (Arnalds *et al.*, 1989; Mitchell *et al.*, 1990; Blagoeva & Zikovsky, 1995).

The aim of this investigation was to find a method to estimate the spatial distribution of the deposition of ^{137}Cs , not only from the Chernobyl accident in 1986, but also from the nuclear weapons tests from 1962–1977, over the province of Skåne in southern Sweden. The model was then validated by comparisons with activity measurements on soil samples, with *in situ* measurements (Arntsing *et al.*, 1991) and with aerial surveys (SGAB, 1986).

Materials and methods

For the determination of activity concentrations in ground-level air, a centrifugal pump with a capacity of 270 m³/h was used. The air was drawn, with a linear flow of 0.3 m/s, through 0.50 × 0.50 m² glass fibre filters mounted vertically about 1 m above the ground. The filters were then pressed and packed into 180 ml plastic containers. This pump construction and the filters used were the same as those used by the National Defence Research Establishment (Vintersved & DeGeer, 1982; Vintersved *et al.*, 1987).

A large collector, consisting of two 1 × 2 m² aluminium sheets, slightly angled with a length of guttering in between them, was used for the collection of precipitation at the Department of Physics in Lund. The collected rainwater was first filtered through 2–4 filter papers to remove coarse particles and the water was then passed through 30 ml Dowex 1-X8 anion and 30 ml Dowex 50 WX8 cation ion-exchange resin. The ion-exchange resin and the filter papers were dried and packed into 60 ml plastic containers. On some occasions, with a high activity concentration, it was possible to measure the rainwater directly in a 180 ml plastic container.

Soil samples were collected between 1988 and 1994 on horizontal ground, mainly with a metal tube, 8 cm in diameter, which was pushed into the ground to a depth of 10–20 cm. The soil core was then cut into slices, 2–3 cm in thickness. (Isaksson & Erlandsson, 1995; 1998). The activity concentrations of the various radionuclides were determined with a Ge(Li) detector (efficiency 18% and resolution 1.9 keV FWHM at 1332 keV) and efficiency calibration was carried out using samples of known activity with the same density as the collected samples, packed into containers of appropriate sizes (Bjurman, *et al.*, 1987).

The dense rain-gauge network was obtained by combining a regional network where daily measurements were made (Elleson, 1993), with precipitation measurements made by SMHI. For the period 1962–77, 65 stations in the Elleson network and 48 SMHI stations were used, giving a density of more than one station per 100 km². The quarterly accumulated precipitation was calculated for each station and missing data were estimated as a fraction of the precipitation values at nearby stations with a complete record for the corresponding time (Liljequist, 1970). For 1986, 104 stations from the Elleson network and 39 SMHI stations were used. The stations have complete records for May 1986, the most critical month concerning the deposition from the Chernobyl accident. For the rest of 1986, less than 10 % of the data was missing, and these data were interpolated as described above. Due to very low values of ^{137}Cs in the period 1987–1994, only data from 1986 have been included in the calculations.

To calculate the deposition 1962–77 we have utilised the activity concentrations of various radionuclides in the air which have been measured at Ljungbyhed, in the middle of the investigated area, since the second quarter of 1962 (DeGeer *et al.*, 1978). The deposition does not show any

dramatic variation with time, and measurements at other places in Sweden have also shown that the variation over an area such as Skåne ought to be small (Bernström, 1969). We have therefore assumed that the air activity concentration was the same over the whole area during the period in question. At Ljungbyhed, the deposition per unit precipitation in $\text{Bq/m}^2\cdot\text{mm}$, which is the same as the activity concentration in the precipitation in Bq/l , has also been measured quarterly (FOA 41). These activity concentrations were multiplied by the corresponding precipitation for the 113 stations and the deposition for each station was summed for the whole period. The calculated station values were then transformed into areal values (Isaksson et al., 2000).

The cloud from the Chernobyl accident did not reach Skåne until 7th May 1986. During the evening of May 7th, thundershowers occurred in the western part of the area with up to 10 mm of rain at some locations. The cold front passed over Skåne during the following day causing precipitation of up to 10 mm in the western part. During the following days, there were occasional showers and during 10th to 11th May evenly distributed showers all over the area. The first precipitation fell over Lund at 22.00 on 7th May. The deposition of ^{137}Cs per mm precipitation obtained from measurements made at Lund were multiplied by the corresponding amount of precipitation for 143 stations for May 8th, 9th–10th, 11th–31st, June–July and August–December, and the accumulated deposition for each station was calculated.

Results

Figure 1 shows the accumulated deposition for the years 1962–77, which is relatively evenly distributed over the area, with the topography having a slight effect. The deposition varies from 0.8 kBq/m^2 in the Southwest, to 1.5 kBq/m^2 in the north.

Figure 2 shows the deposition from 06.00 GMT on May 7th until 06.00 GMT on May 8th, 1986. The deposition is heterogeneously distributed in the south-western part of the area with a number of high point values. The spatial distribution of the deposition for the period 8th May 1986 to 31st December 1996 (Figure 3), is dominated by the deposition during this single 24-hour period. The calculated deposition, excluding May 8th, is quite evenly distributed over the whole area, with slightly higher values in the western part, which is mostly due to the distribution of precipitation on May 9th when the activity concentration in the precipitation was still quite high, and showing more events related deposition but with the same trend of higher values in the western part.

The total deposition for the years 1962–94, shown in Figure 4, was obtained as an overlay of the deposition maps for the deposition for 1962–77 and for 1986 (Figures 1 and 3). This figure shows the restricted local deposition caused by the rain showers, and also the agreement between the calculated accumulated deposition obtained from the precipitation measurements and the measured deposition obtained from the soil samples.

Comparisons between calculated and measured accumulated deposition at Lund and Blentarp

The accumulated ^{137}Cs deposition at Lund both before and after the Chernobyl accident was calculated and gave a mean deposition of 1.2 kBq/m^2 for the whole city. The activity concentrations of ^{137}Cs and ^{134}Cs in the soil have been measured in 1991 at 16 sites in Lund (Isaksson & Erlandsson, 1998). Because of the known ratio of the activity concentration of ^{134}Cs and ^{137}Cs in the deposition from the Chernobyl accident it has been possible to divide the deposition of ^{137}Cs into

pre- and post-Chernobyl deposition. Of the 16 measurements, 5 have been excluded. The mean value, given with a 95% confidence interval, of the total deposition is 3.05 ± 0.40 kBq/m², of the old deposition 0.86 ± 0.29 kBq/m² and of the new deposition 2.29 ± 0.16 kBq/m². As the pre-Chernobyl deposition is calculated from the total and post-Chernobyl depositions the uncertainty is large. The measured deposition values from the soil samples are in good agreement with the total calculated deposition of 3.23 ± 0.20 kBq/m² comprising the pre-Chernobyl deposition of 1.23 ± 0.12 kBq/m² and a post-Chernobyl deposition of 2.00 ± 0.15 kBq/m².

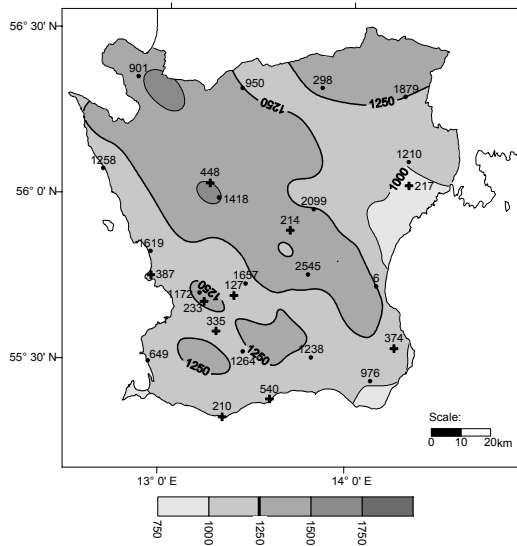


Figure 1. Deposition of ¹³⁷Cs (Bq/m²).
1962-77

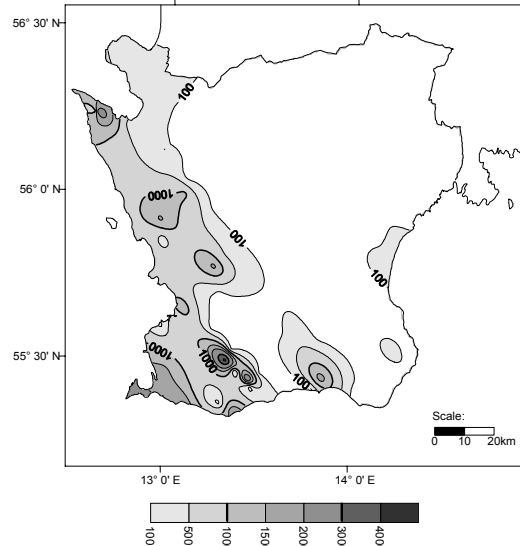


Figure 2. Deposition of ¹³⁷Cs (Bq/m²)
8th May 1986.

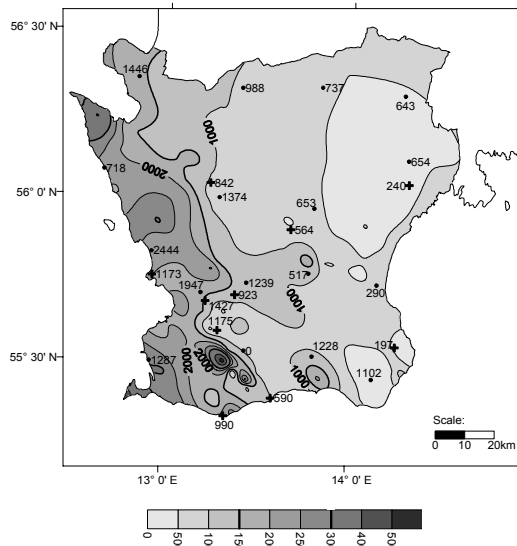


Figure 3. Deposition of ¹³⁷Cs (Bq/m²)
8th May 1986 to 31st December 1996.

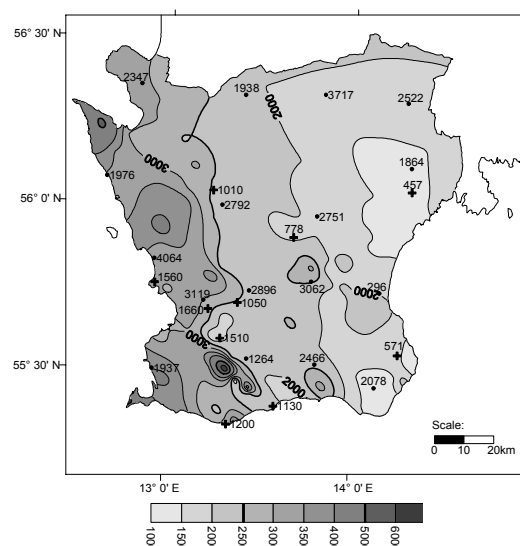


Figure 4. Deposition of ¹³⁷Cs (Bq/m²)
1962-77 and 1986.

The deposition at Blentarp (about 20 km from Lund) has been calculated as the mean for two precipitation stations, which are only about 5 km from Blentarp (Isaksson et al., 2001). For the period from 1962 to the first quarter of 1986 the calculated deposition is 1.41 ± 0.07 kBq m⁻², decay

corrected to 1986. This agrees well with the amount of bomb ^{137}Cs found in soil measurements, which was $1.60 \pm 0.38 \text{ kBq m}^{-2}$. For the period 8th May to the end of December 1986 the precipitation has been measured at three stations, all within 5 km from Blentarp. The mean value of the calculated deposition for Blentarp, corrected for decay to 1986, is $0.79 \pm 0.12 \text{ kBq m}^{-2}$, which is in good agreement with the $0.79 \pm 0.05 \text{ kBq m}^{-2}$ obtained from soil measurements in 1988. The total calculated deposition of ^{137}Cs at Blentarp is then $1.41 + 0.79 = 2.20 \pm 0.14 \text{ kBq m}^{-2}$, which is in good agreement with the mean value for the soil measurements, $2.28 \pm 0.34 \text{ kBq m}^{-2}$.

Comparisons between calculated and measured accumulated deposition in Skåne

The ratio of the calculated-to-measured pre-Chernobyl deposition for each soil sampling site is 0.96 ± 0.21 , when two of the 16 sites are excluded, and the ratio for the calculated post-Chernobyl deposition is 1.07 ± 0.38 , with the same sites excluded. The ratio of the calculated-to-measured total deposition is 0.96 ± 0.20 (excluding two sites). These ratios are close to one, but with uncertainties of about 20–40 %. Nevertheless they are rather good considering the fact that the post-Chernobyl values of ^{137}Cs are based on measurements of ^{134}Cs and that the pre-Chernobyl values are obtained by subtracting the post-Chernobyl deposition from the total deposition.

A comparison between the calculated pre-Chernobyl activity of ^{137}Cs from precipitation and from *in situ* measurements made by the National Defence Research Establishment (Arntsing, *et al.*, 1991) shows a ratio of about four. Because the results from the *in situ* measurements are given as equivalent surface deposition, *i.e.* assuming a plane source on the surface, the activity is underestimated due to the vertical distribution in the ground. Calculations have shown that this underestimation is reasonably well explained by the actual depth distribution (Isaksson *et al.*, 2000). The deposition of new ^{137}Cs shows a mean ratio between calculated and *in situ* values of about 1.5, with a rather large spread between the sites. These values probably reflect both migration of ^{137}Cs into the soil between the time of the fallout and the *in situ* measurements, as well as surface characteristics of the sites, and are in good agreement with measurements made by Arntsing *et al.* (1991).

Aerial measurements of ^{134}Cs (subsequently recalculated to ^{137}Cs) were performed by the Swedish Geological Company (SGAB, 1986) during the period May 9th to June 19th 1986 with complementary measurements during September and October. In Skåne, three regions can be seen (Figure 4) with different levels of deposition of ^{137}Cs : 0–2 kBq/m^2 in the eastern part of the province, 2–3 kBq/m^2 in the west and 3–5 kBq/m^2 in a smaller region close to the west coast. The border between the two regions of lowest deposition is, however, not distinct, as some of the values were close to the detection limit. The calculated post-Chernobyl deposition of ^{137}Cs (Figure 3) shows a similar pattern, with below 1.5 kBq/m^2 in the eastern parts and between 1.5 and 2 kBq/m^2 in the west.

Summary and conclusions

The distribution of ^{137}Cs deposition from nuclear weapons fallout and from the Chernobyl accident in the province of Skåne in southern Sweden have been calculated from deposition and precipitation measurements. Comparison with other types of measurements (soil sampling, *in situ* measurements and aerial measurements) shows good agreement, both for the nuclear weapons fallout and for the first year following the Chernobyl accident. The method described can therefore be considered as a valuable complement to other types of measurements when a larger region is surveyed for the

deposition of ^{137}Cs . Because precipitation measurements are made regularly at many meteorological stations in Sweden, the determination of deposition can be reduced to measurements of activity and precipitation at a few chosen sites, which enables the results to be produced shortly after a deposition event.

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Influence of airborne chemical substances on the behaviour of radionuclides in boreal forest ecosystems

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Abstract

Atmospheric deposition of chemical substances may influence the behaviour of radionuclides in soil and their uptake in plants. This is a fact that has so far received limited attention in radioecological studies. This paper presents briefly two cases where differences in atmospheric deposition of heavy metals from a strong pollution source and marine aerosols, respectively, influence the turnover of radionuclides in natural boreal ecosystems.

Introduction

The dominant natural sources of atmospheric deposition of chemical substances other than H₂O are marine aerosols (Na⁺, Cl⁻, SO₄²⁻, Mg⁺⁺, Ca⁺⁺, Sr⁺⁺, . . .), re-suspension of soil particles, volcanic activity, and biogenic emissions (e.g. CH₃I from marine organisms). In addition man has added significant amounts of chemical substances to the atmosphere in the form of air pollutants (acidic components, metal-containing aerosols, etc.). These substances are likely to interact with the surface to which they are supplied, and if the fluxes are significant, they may significantly modify the chemistry and even the microbiology of surface soils.

A very important type of chemical reactions influencing the turnover of radionuclides in soils is cation exchange. The most important classes of soil microparticles (clay, oxide, humus) all develop an excess of negative charges on their surfaces by different mechanisms. In the case of humus particles, which is the focus of the present paper, negative surface charges are produced by dissociating protons (H⁺) either from carboxylic (-COOH) or phenolic (-OH) groups. The excess negative surface charges attract cations to the particle surface and thus represent a certain cation exchange capacity.

The most important cations participating in these processes are the so-called “base” cations (Ca⁺⁺, Mg⁺⁺, K⁺, Na⁺) and those contributing to most of the soil acidity (H⁺, Al³⁺, Fe³⁺). But also cations of minor and trace elements (e.g. Rb⁺, Cs⁺, Co⁺⁺, Sr⁺⁺, Ba⁺⁺, Ra⁺⁺ . . .) participate in cation exchange processes, and radionuclides in cationic form are no exception! In the following two examples are presented to illustrate the significance of cation exchange processes in radioecology in general and in boreal ecosystems in particular.

The EPORA project

The main purpose of the EU-sponsored project EPORA (Suomela et al. 1999) was to examine potential effects of chemical pollutants on the soil-plant transfer of radionuclides previously supplied to the ecosystem. The study was carried out in five different stands of spruce forest located at distances ranging from 8 to 150 km from a copper-nickel smelter in Monchegorsk, north-western Russia. The main air pollutants emitted from the smelter are SO₂, Cu, and Ni, and it became clear that the heavy metals had the most obvious impact on the soil chemistry and thereby on plant uptake of major nutrients as well as of the radionuclides studied (¹³⁷Cs, ⁹⁰Sr, Pu).

Soil samples were collected from the humus layer (O_f, O_h), eluvial horizon (E), and illuvial horizon (B) in the podzolic soils, and samples of four dominant species of vascular plants, the grass *Deschampsia flexuosa* and the dwarf shrubs *Vaccinium myrtillus*, *Vaccinium vitis idaeae*, and *Empetrum nigrum*, were also collected. The concentrations of Ni and Cu in the O_f (raw humus) horizon, which has the highest density of fine plant roots, ranged from about 5000 ppm to less than 10 ppm. Of the total metal content about 10% of Ni and 2 % of Cu were in exchangeable form. Apparently the high content of exchangeable heavy metals strongly interfered with other cations in the O_f horizon at the sites with shortest distances from the smelter. For Mg e.g. the exchangeable concentration decreased from about 550 to about 150 ppm (Thørring et al. 1999), most probably because of leaching to lower soil horizons due to cation exchange with heavy metals. Similar loss from the O_f horizon was also observed for other macronutrients such as K, Ca, and Mn. Correspondingly low levels were observed in plants at the sites with shortest distance to the smelter.

Very similar results were observed for the radionuclides in question. The activity concentration of ¹³⁷Cs in the humus layer decreased from about 500 to 200 Bq m⁻² from the farthest to the nearest site, and the decline of ⁹⁰Sr was even greater. Similarly, whereas 80% of the total inventory of ¹³⁷Cs was found in the root zone (O + E horizon) at the farthest site, this figure was only 40% at the site nearest to the smelter. Corresponding values for ⁹⁰Sr were 74% and 18% respectively, and this also caused lower uptake of radionuclides on the sites situated most closely to the smelter (Suomela et al. 1999).

Radiocesium in surface soils in Norway

The fallout of ¹³⁷Cs in Norway following the Chernobyl accident was very inhomogeneous, ranging from almost nothing to over 100 kBq m⁻², as evident from analysis of samples of surface soils (Bache et al. 1986). In 1995 the collection of surface soil samples, originally intended to see the contribution of long range atmospheric transport to the heavy metal levels accumulated in the topsoil, was carried out all over the country. The upper 3 cm of the humus layer was sampled from podzolic soils, normally representing the O_f (raw humus) layer, with a defined surface area, and the ¹³⁷Cs activity was measured by gamma spectrometry using a germanium detector.

The sampling sites in the 1995 survey were not the same as in 1986, and the sampling procedures were also somewhat different. In order to relate the results from the 1995 survey to that in 1986, mean values were established on a municipality basis, and the 1995 value was expressed as a percentage of the corresponding number from 1986. The “relative retention factor” thus obtained appeared to vary among the 18 counties of Norway by a factor of four. Especially in the southern part great differences were evident between different counties. In order to explain these differences

it seems necessary to discuss some typical differences in precipitation chemistry between different parts of this territory:

- The eastern part of Southern Norway (Østlandet) is shielded from marine influence by high mountains: Langfjella to the west and Dovre to the north. This region therefore has a continental type climate with annual precipitation values of 300-700 mm. On the west side of the mountains (Vestlandet) the climate is oceanic with annual precipitation amounts typically within the range 1500-3000 mm. If we look at the fluxes of marine salt deposited the differences are even greater (Låg 1968). As an example the chloride concentration in small lakes, which is closely related to the input of marine ions with precipitation, varies by a factor of 20 between the coastal areas of Vestlandet and the upper valleys of Østlandet (Allen & Steinnes 1987).
- Along the coast of the southernmost part (Sørlandet) the deposition of acidic pollutants (sulphur and nitrogen compounds) in 1994, due to long range transport from other parts of Europe, was about 5 times higher than in the more northerly parts of Norway (Tørseth 1995). The annual precipitation in this region is 800 -2000 mm.

In order to emphasise the geographical differences observed in the retention of ^{137}Cs in the organic topsoil, “retention factor” values were calculated as follows:

Indre Østlandet (Hedmark, Oppland, Akershus, Buskerud):	72±6 %
Vestlandet (Hordaland, Sogn og Fjordane, Møre og Romsdal):	41±2 %
Sørlandet og Sør-vestlandet (Aust-Agder, Vest-Agder, Rogaland):	27±5%

These trends are most probably related to differences in atmospheric deposition between the regions discussed above. The most likely mechanism to cause the enhanced removal of ^{137}Cs from the topsoil in the coastal areas is cation exchange of Cs^+ ions with deposited marine cations, among which Mg^{++} appears to be the most likely candidate. In the south, where the retention of ^{137}Cs is the lowest, cations from acidic deposition (NH_4^+ , H^+) may have been active in addition to the marine ones in the replacement of ^{137}Cs from the surface soil layer.

Thus the retention of ^{137}Cs in organic topsoil appears to be considerably affected by more recent supply of cations from the atmosphere. From the results from the EPORA project it is clear that other radionuclides in cationic form may be similarly affected by atmospheric deposition of cations from natural or anthropogenic processes.

General conclusions

- Cation exchange processes seem to have been strongly underestimated in radioecological research dealing with soils. The results presented here indicate that such processes should be taken much more seriously in future research on environmental radioactivity.
- The possible influence of precipitation chemistry on the turnover of radionuclides in soil – plant systems has been ignored in radioecology so far, and deserves to be more systematically studied.

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Measurements of ^{137}Cs in the food-chain of lamb in the Faroe Islands in the period 1990-1999

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Abstract

Radiocaesium has been measured in the food-chain of lamb in nine uncultivated pastures in the Faroe Islands since 1990. The sampling programme includes soil from the uppermost 10 cm, mixed grass and lamb meat. Lamb faeces have been included since 1995. The results for the period 1990-99 show the following averages across the nine pastures: the ^{137}Cs deposition decreased from 5.8 kBq/m² in 1990 to 5.1 kBq/m² in 1999; the ^{137}Cs concentration in mixed grass decreased from 155 Bq/kg(dw) in 1990 to 51 Bq/kg(dw) in 1999; the ^{137}Cs concentration in lamb meat decreased from 26.0 Bq/kg(ww) in 1990 to 10.4 Bq/kg(ww) in 1999. Large variations are, however, observed between as well as within the pastures. Averaged across the pastures, the soil-to-grass transfer factor decreased from $30 \cdot 10^{-3} \text{ m}^2/\text{kg(dw)}$ in 1990 to $13 \cdot 10^{-3} \text{ m}^2/\text{kg(dw)}$ in 1999, and the aggregated soil-to-lamb (meat) transfer factor decreased from $5.5 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$ in 1990 to $2.0 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$ in 1999. The effective ecological half-life of ^{137}Cs in mixed grass and in lamb meat is estimated to be in the range 2.8-4.3 and 6.6-8.0 years, respectively.

Introduction

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

The paper presents results from measuring ^{137}Cs in soil, grass and lamb meat in nine uncultivated pastures in the Faroe Islands for the years 1990-99. Effective ecological half-lives and transfer factors are presented. Chemical characteristics of the soil have earlier been considered in connection with the observations (Joensen, 1999). Faroese results for 1990-93 have been presented in a Nordic context by Hove *et al.* (1994).

Material and methods

Soil, grass and lamb meat have been collected each year from nine uncultivated pastures across the country. Soil and grass were sampled in July-August from four randomly chosen 0.25m² microplots in each pasture. The grass was cut from each microplot before taking three soil cores with 5.7cm diameter and length 10cm. Samples from the microplots were measured separately. Neck muscle was collected from lamb at slaughter in October. Meat samples were typically taken from 5 lambs in each pasture (carcass weight around 12-13kg). Lamb faeces were collected for 1995-99 from fresh manure on the pasture ground, and measured as one sample per pasture each year. Soil samples were dried at room temperature, while grass and faeces were dried at 105 °C before measurement. Meat samples were kept frozen and thawed before individual measurement.

Results and discussion

The ¹³⁷Cs deposition in the uppermost 10cm soil layer is presented in Fig. 1, showing large temporal and spatial variation between and within the pastures. An exponential decay model was only acceptable for Hvalvík, where the effective ecological half-life (VAMP, 1992) was estimated to 11.6 years ($R^2 = 0.51$). R^2 was below 0.30 for other pastures. 50-80% of the deposition in the top 10 cm soil layer is found to be in the uppermost 5 cm. The ¹³⁷Cs deposition, averaged across the pastures, decreased from 5.8 kBq/m² in 1990 to 5.1 kBq/m² in 1999.

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

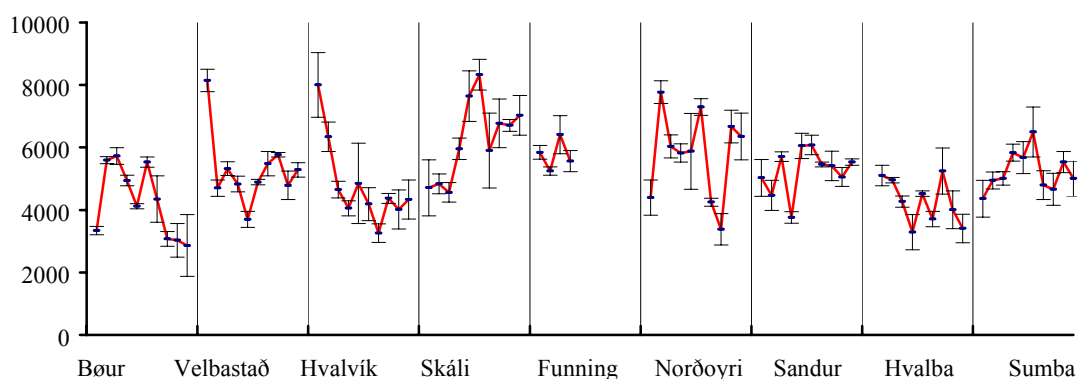


Figure 1. ¹³⁷Cs (Bq/m²) in 0-10cm soil layer 1990-99. Yearly averages \pm 1 standard error.

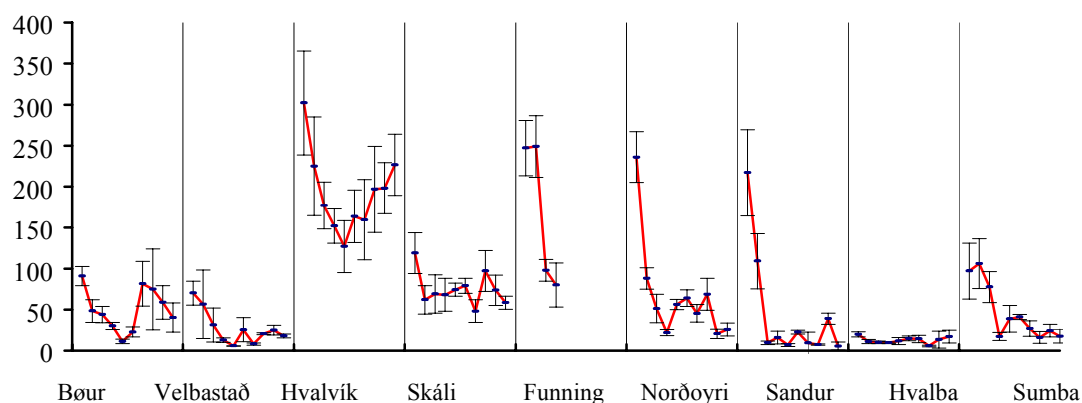


Figure 2. ^{137}Cs (Bq/kg dw) in mixed grass 1990-99. Yearly averages ± 1 standard error.

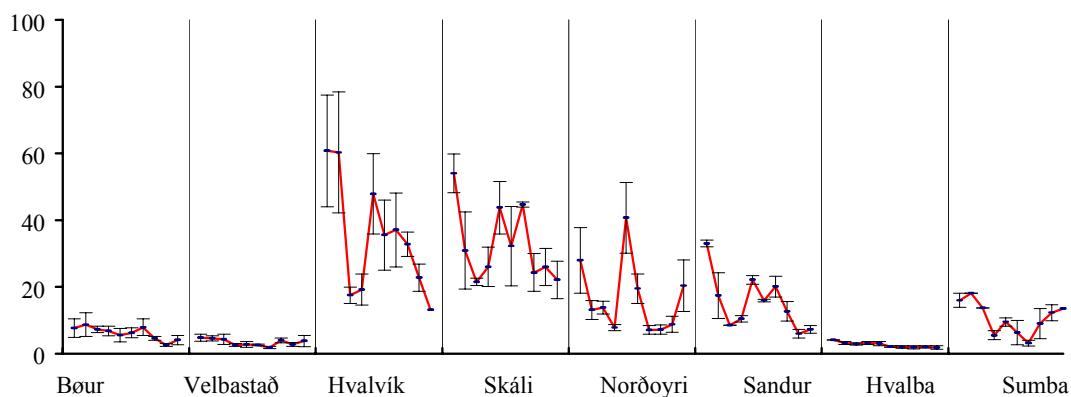


Figure 3. ^{137}Cs (Bq/kg ww) in lamb meat 1990-99. Yearly averages ± 1 standard error.

Table 1. Effective ecological half-life in years. Numbers in brackets represent R^2 from a linear regression between time and natural logarithm of ^{137}Cs concentration in the samples. No estimates if $R^2 < 0.300$.

	Bøur	Velbastað	Hvalvík	Skáli	Norðoyri	Sandur	Hvalba	Sumba
Grass	- (0.000)	- (0.221)	- (0.036)	- (0.104)	4.3 (0.438)	2.8 (0.379)	- (0.005)	3.6 (0.667)
Meat	7.2 (0.661)	- (0.162)	7.2 (0.300)	- (0.174)	- (0.089)	6.6 (0.493)	8.0 (0.873)	- (0.069)

The ^{137}Cs concentration in mixed grass (undetermined botanical composition) is observed to decrease in most pastures, with highest concentration in Hvalvík and lowest concentration in Hvalba (Fig. 2). Estimated effective ecological half-lives are presented in Table 1. The average ^{137}Cs concentration across the pastures was 155 Bq/kg(dw) in 1990 and 51 Bq/kg(dw) in 1999.

The ^{137}Cs concentration in lamb meat is presented in Fig. 3 (no meat samples from Funningur). Large standard errors express large variations between animals. An effective ecological half-life could be estimated in four pastures (Table 1). The average concentration across the pastures was 26.0 Bq/kg(ww) in 1990 and 10.4 Bq/kg(ww) in 1999.

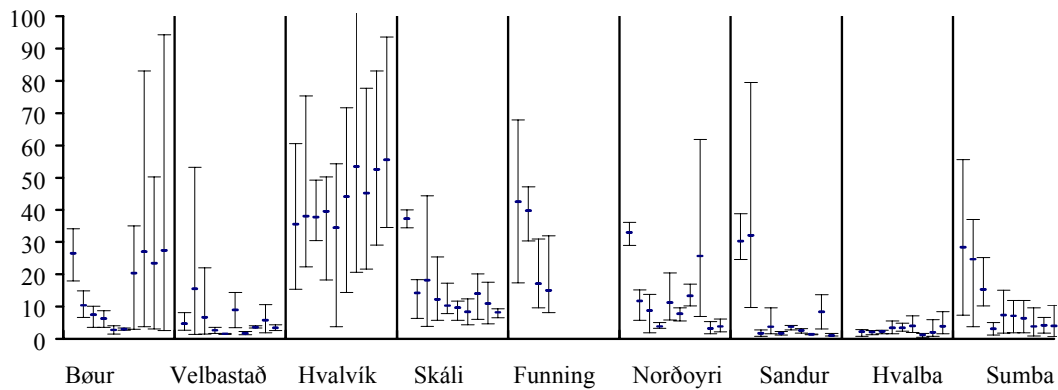


Figure 4. Soil-to-grass transfer factor for ^{137}Cs ($10^{-3} \text{ m}^2/\text{kg dw}$) 1990-99. Yearly averages and ranges.

The soil-to-grass transfer factor has been calculated for each 0.25 m^2 microplot used for the grass and soil sampling, as the ^{137}Cs concentration, Bq/kg(dw) , in grass divided by the ^{137}Cs deposition, kBq/m^2 , in the top 10 cm soil layer. The results in Fig. 4 show large geographical variations of the soil-to-grass transfer factors, and large differences between the microplots within the pastures. It is important to know the chemical characteristics of the soil in order to explain these observations. Knowledge to botanical composition of the mixed grass could also be a valuable explaining factor.

The soil-to-meat aggregated transfer factors have been calculated for a given pasture by averaging the ratio of the ^{137}Cs concentrations in individual meat samples to the averaged ^{137}Cs deposition in the pasture. The minimum and maximum mean values in a pasture for the period 1990-99 were $0.33 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$ and $11.5 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$, respectively.

The averages across the pastures showed the following results: the soil-to-grass transfer factor decreased from $30 \cdot 10^{-3} \text{ m}^2/\text{kg(dw)}$ in 1990 to $13 \cdot 10^{-3} \text{ m}^2/\text{kg(dw)}$ in 1999, and the soil-to-meat aggregated transfer factor decreased from $5.5 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$ in 1990 to $2.0 \cdot 10^{-3} \text{ m}^2/\text{kg(ww)}$ in 1999.

^{137}Cs has been measured in lamb faeces in the period 1995-99. The concentration was between 18 and 126 Bq/kg(dw) , and the yearly average across the pastures was between 53 and 72 Bq/kg(dw) . The average across the pastures for the meat/faeces concentration ratio and of the faeces/grass concentration ratio was 0.20 [Bq/kg(ww) per Bq/kg(dw)] and 2.1 [Bq/kg(dw) per Bq/kg(dw)], respectively.

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

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

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

Conclusion

Although the Faroe Islands has a total land area of only about 1400 km², large temporal and spatial variations are found for the measured ¹³⁷Cs activities and the calculated transfer factors. The effective ecological half-life has been estimated to be 2.8-4.3 years for grass and 6.6-8.0 years for lamb meat. Deposition of ¹³⁷Cs has not decreased significantly over the study period except for one pasture, where the effective ecological half-life was 11.6 years. These are of course rough estimates, because large variability is associated with this kind of measurements. Among the tree soil parameters potassium, pH and loss on ignition, loss on ignition is found to be most important for the transfer of ¹³⁷Cs in the food chain of lamb.

Acknowledgement

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

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¹³⁷Cs i samiska renskötare uppmätt med förenklad helkroppsmätning

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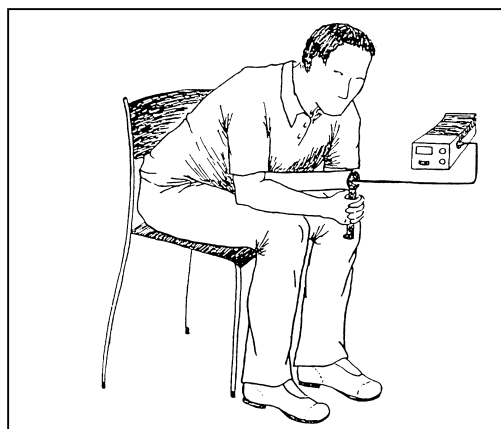
Sammandrag

Helkroppsaktiviteten hos en utvald grupp finska samer mättes 1962-77 och 1986-97 med mobil helkroppsmätare. I den mest utsatta gruppen, manliga renskötare steg medelvärde för kroppsaktiviteten ¹³⁷Cs till 53 kBq/person år 1965 och 14 kBq/person år 1997. Den effektiva halveringstiden för ¹³⁷Cs var 7 år på 1970-talet och 4,4 år på 1990-talet. Sommaren 2000 utfördes en kompletterande mätning med famngeometri. Resultatet bekräftade den trend, som observerats tidigare.

Inledning

När radionuklider i nedfallet från kärnvapenproven visade sig koncentreras i näringskedjor blev det under 1960-talet aktuellt att mäta den interna kontaminationen i de berörda befolkningsgrupperna. Konventionella helkroppsmätare med massiv avskärmning av stål eller bly kunde oftast inte användas i de avlägsna trakter det var fråga om. Palmer⁽¹⁾ beskrev först den så kallade famngeometrin, som innebär att personen som skall mätas sitter med en scintillationsdetektor i famnen och böjer sig över detektorn (figur 1). Metoden kom att användas i USA⁽²⁾, Kanada⁽³⁾, Norge⁽⁴⁾, Sverige⁽⁵⁾ och Sovjetunionen⁽⁶⁾. Efter Tjernobyk Katastrofen var problemet inte transport av tung utrustning till avlägsna orter, utan behovet att snabbt kunna mäta många personer; enligt Hill et al.⁽⁷⁾ utfördes 317 000 mätningar i Ryssland, Vitryssland och Ukraina. Famngeometrin användes av flera forskargrupper⁽⁸⁾, men också andra system togs i bruk.

Famngeometrin har två grava nackdelar. Om nukliden som skall mätas också förekommer i bakgrundsspektret, vilket var fallet efter Tjernoby, uppstår problem med att beakta bakgrundens andel, eftersom kroppen avskärmar en del av den externa strålningen samtidigt som den inre kontaminationen ökar pulsraten⁽⁹⁾. Det andra problemet är det stora beroendet av antropometriska faktorer (vikt, längd, midjemått) och av kroppsställningen under mätningen. I äldre källor redovisas kalibrationsfaktorer^(1, 2, 10, 11, 12), medan senare publikationer inte ger numeriska värden. Ryska forskare⁽¹³⁾ har kalibrerat efter midjemått eller vikt, skilt för positionen ”böjd över detektorn” och ”sittande upprätt”. Olika funktioner har använts för kalibrationsfaktorns beroende av kroppsvikten, men generellt kan beroendet beskrivas lineärt med formeln



Figur 1. Famngeometrin

$$A = R_n \cdot k \cdot m \quad (I)$$

där A är kroppsaktiviteten, R_n nettopulsraten, kroppsvikten och k en konstant, som för en 75 mm scintillationsdetektor får värden mellan 7,4 och 9,0 Bq·s·kg⁻¹. Westerlunds grupp⁽⁴⁾ valde emellertid att använda samma mäteffektivitet för alla personer, vilket kan försvaras med att viktkorrekturen är försumbar jämfört med andra felkällor.

Finska samers helkroppsaktivitet har mätts med blyavskärmade helkroppsräknare under 1961-1977 och 1986-1997. Sommaren 2000 utfördes en kompletterande mätning med famngeometri.

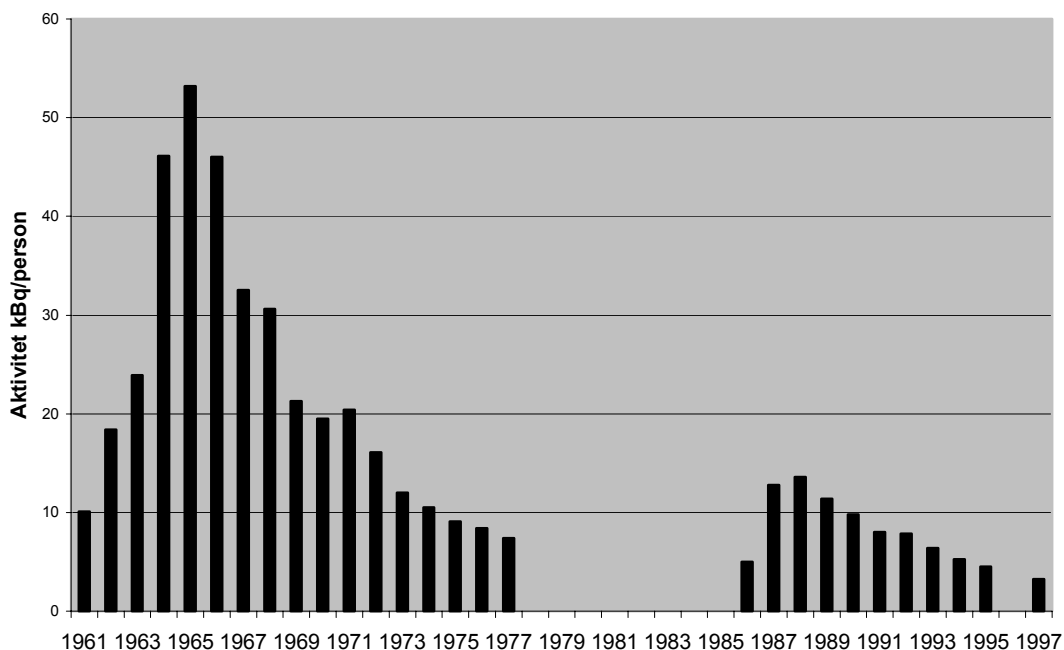
Material och metoder

Radiokemiska institutionen (sedan 1995 laboratoriet för radiokemi) påbörjade mätning av finska samer år 1961 med utrustning lånad från Sverige. Följande år konstruerades en mobil helkropps-mätare med stolgeometri, scintillationsdetektor och öppen blyavskärmning, s.k. shadow shielding⁽¹⁴⁾. Gruppen som mättes utvaldes slumpmässigt ur folkbokföringen och har senare kompletterats med yngre personer. Mätningarna gjordes på våren, vissa år också på hösten. Mätningarna pågick till 1977, återupptogs våren 1986 i samarbete med Strålsäkerhetscentralen (STUK) och avslutades 1997. STUKs utrustning använder halvledare, modifierad båggeometri och öppen avskärmning⁽¹⁵⁾. De två helkroppsmätarna har kalibrerats mot varandra genom att mäta samma personer med vardera utrustningen och genom att delta i interkalibrationer^(16,17).

Famngeometrin hade kalibrerats genom att mäta samer under 1970-talet både med famngeometri och med den avskärmade stolgeometrin^(11,12). Kalibreringen kompletterades genom att mäta NKS-fantomet i två geometrier, ”böjd över detektorn” och ”sittande upprätt” för olika kroppsvikter. Fantomet är konstruerat av polyetenblock, i vilka stavar med känd aktivitet sticks in, och det kan byggas upp för bädd-, stol- och famngeometri⁽¹⁷⁾. Sommaren 2000 mättes nio personer ur mätgruppen med famngeometrin.

Resultat och diskussion

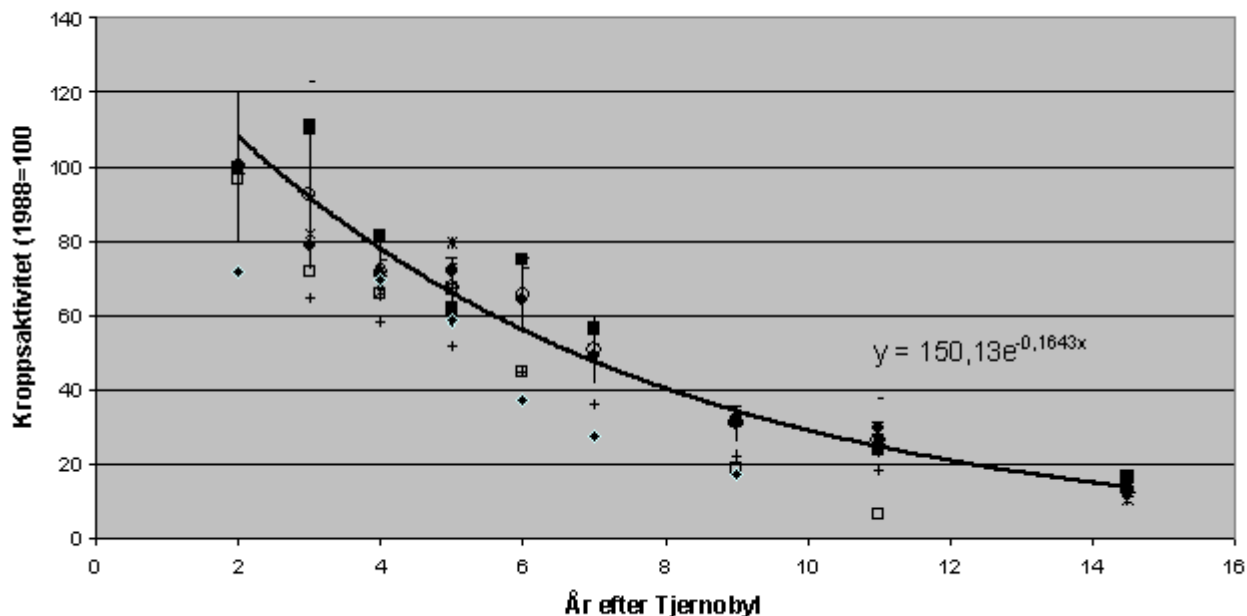
Mätresultaten för 1962-77 och 1986-97 visas i figur 2 för den mest exponerade gruppen, manliga renskötare från Enare kommun. Maximivärdena för ¹³⁷Cs uppmättes 1965, då medeltalet för hela gruppen var 53 kBq/person och den högsta individuella aktiviteten 100 kBq. Efter Tjernobyl var maximiåret 1988, då medeltalet var 14 kBq och den högsta aktiviteten hos en person 40 kBq. Den effektiva halveringstiden för samtliga mätta var under 1970-talet 7 år, samma som i Norge⁽⁴⁾.



Figur 2. ^{137}Cs i manliga renskötare från Enare

Efter Tjernobylolyckan förekom större variationer i mätresultaten, eftersom nedfallet var ojämnt fördelat. Det visade sig oändamålsenligt att beräkna den effektiva halveringstiden utgående från medeltalet av samtliga mätta. Istället utvaldes 97 personer, som deltagit i minst nio mätningar under perioden 1988-95. Alla personliga resultat normerades så, att kroppsaktiviteten våren 1988 sattes till 100. För varje år beräknades medeltalet av de normerade kroppsaktiviteterna, och ur medeltalen den effektiva halveringstiden. Resultatet blev 4,4 år. Kroppsaktiviteterna hos denna grupp är ungefär 10 % högre om våren än om hösten (utan korrektion för nuklidens sönderfall).

I augusti 2000 hade manliga renskötare kroppsaktiviteten 2000 – 5000 Bq ^{137}Cs /person, kvinnor och pensionärer omkring 1000 Bq/person. Den effektiva halveringstiden beräknades genom att normera kroppsaktiviteten 1988, som ovan beskrivits. Resultatet 1988-2000 blev 4,2 år (figur 3).



Figur 3. ^{137}Cs hos 9 personer 1988-2000.

Den effektiva halveringstiden efter Tjernobyl (4,4 år) är kortare än på 1970-talet (7 år). Orsaken är sannolikt den, att renskötare i Finland håller för många renar. Laven är i praktiken slutbetad, och renarna måste under vintern matas med hö och kraftfoder, som inte innehåller ^{137}Cs .

Kalibreringen av famngeometrin med NKS-fantomet gav för mätgeometrin "sittande böjd över detektorn" och en 75 mm scintillationskristall kalibrationsekvationen

$$A = R_n \cdot (-0,0612 \cdot m^2 + 12,787 \cdot m - 142,71) \quad (\text{II}),$$

där storheterna är samma som i ekvation (I), A uttrycks i Bq, R_n i s^{-1} och m i kg. Denna ekvation stämmer väl överens med den Travnikova⁽¹³⁾ använde.

Viktkalibreringens inverkan på mätresultaten kan demonstreras genom att anta att de 150 samer, som våren 1997 mättes med konventionell utrustning, hade mätts med famngeometrin. Deras kroppsvikt var mellan 45 och 101 kg, i medeltal 71 kg. Om kalibreringskoefficienten beräknas på medelvikten och tillämpas på alla mätta kommer visserligen kroppsaktiviteten för en 45-kilos person att överskattas med 48 %, och aktiviteten för en 100-kilos person att underskattas med 13 %. Medelvärdet för alla mättas kroppsaktivitet kommer däremot att vara endast 4 % för stort. Andra felkällor har försumrats i detta resonemang.

Fel kroppsställning innebär ett större systematiskt fel. Om kalibreringsekvation (II) används för en person, som inte kan inta rätt position utan sitter upprätt, blir resultatet 23 % i underkant.

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On neptunium-237 in Nordic waters

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Abstract

The actinide neptunium-237 ($T_{1/2}$: 2.14×10^6 yr.) was present in Northern Hemisphere fallout in atom ratios around 0.45 – 0.70 relative to the much shorter-lived plutonium-239 ($T_{1/2}$: 2.41×10^4 yr.), i.e. $^{237}\text{Np} / ^{239}\text{Pu}$ activity ratios in fallout was 0.005 – 0.008. Neptunium-237 levels in the Nordic waters are too low to be readily measurable by α -spectrometry – the predominant method for quantifying plutonium. With sensitive mass spectrometric methods ^{237}Np is however measurable in the Nordic waters – as probably in most surface seawater world-wide. Recent observations in Danish and Greenlandic sea-water has shown a much higher number of atoms of ^{237}Np than of ^{239}Pu – 20-50 times higher, i.e. as compared to the $^{237}\text{Np} / ^{239}\text{Pu}$ ratio in fallout, the present neptunium level in Nordic sea-water is up to 2 orders of magnitude higher.

The main reason for this observation is the different chemical behaviour of the two elements. Plutonium has a greater affinity for particle surfaces than neptunium, i.e. plutonium has a higher K_d than neptunium and it will therefore have a shorter environmental half-life in the water column. Therefore, we see much more fallout neptunium than fallout plutonium in surface seawater today. Of the same reason, neptunium discharged to coastal waters is much more likely to be transported over long distances than plutonium. Thus discharges from European reprocessing plants is contributing to the levels in Danish as well as in Arctic Ocean and Greenlandic seawater samples.

Time trends of anthropogenic radionuclides at Utsira

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Introduction

Institute for Energy Technology has annually collected the brown alga *Fucus vesiculosus* at several sampling locations along the Norwegian coast since 1980, mainly in August and September [1,2]. At the south-western location Utsira, monthly samples have been collected regularly since January 1986. Time series for ⁹⁹Tc and ¹³⁷Cs in *Fucus* samples from Utsira are presented in this study. The results will be discussed with regard to Sellafield discharges and outflow from the Baltic Sea.

Materials and methods

The *Fucus vesiculosus* samples were dried and homogenised before analysis. The pretreatment and the gamma spectrometric analyses have been performed at Kjeller and the analysis of ⁹⁹Tc at Lund. The *Fucus* samples were dried at 105 °C, homogenised and analysed for gamma emitters using high purity germanium gamma spectrometry and computerised spectrum analysis. Technetium was extracted by TBP from sulphuric acid-hydrogen fluoride media. Backextraction was done from sodium hydroxide solution from which media technetium was electrodeposited onto stainless steel discs. As radiochemical yield determinant ^{99m}Tc was used. After decay of the yield determinant technetium was measured by an anticoincidence shielded GM counter [3].

Results and discussion

Caesium-137

The transit time from Sellafield to the southern coast of Norway has been estimated to 3-4 years [4]. Knowledge of the annual discharge of ¹³⁷Cs from Sellafield year *i* [5,6] allow us to calculate *Fucus* transfer factors (FTFs) from Sellafield to the southernmost locations based on concentrations in *Fucus* in year *i*+4. The calculations were done for the years 1980-85 when other sources than Sellafield were negligible.

For transport of ¹³⁷Cs from the Baltic Sea, the transit time from the Kattegat is estimated to be 2 years to Utsira. The annual net water outflow from the Belt Sea to the Kattegat has been estimated to 1170 km³ [7]. By using published data for the ¹³⁷Cs concentrations (Bq m⁻³) in the Kattegat waters (box 71 of the Risø box model [7]) for year *i* [8] and measured concentrations of ¹³⁷Cs in *Fucus* (Bq kg⁻¹ d.w.) at Utsira for year *i*+2, "Baltic" FTFs have been calculated. These calculations were done for the years 1990-95 when other sources than the Chernobyl fallout were negligible. An average value of 0.035 (Bq kg⁻¹ d.w. per TBq year⁻¹) was found. The calculated FTFs do in fact

include the outflow of ^{137}Cs via Norwegian rivers to the Skagerrak, as this source also influences on the concentration of ^{137}Cs in the *Fucus*. Calculations show, however, that it probably contributes to much less than one third of the total input to these waters of Chernobyl related ^{137}Cs [9].

The *Fucus* transfer factors were then used to calculate the contribution from the different sources compared to the total amount of radiocaesium present in the *Fucus* at Utsira. The results are shown in Figure 1. Our calculations show that in the early 1980's, approximately all the radiocaesium in the seaweed at these sampling locations originated from Sellafield. After 1990 the contribution from Sellafield is nearly negligible. Since 1986, fallout from the Chernobyl accident makes the most important contribution to radiocaesium in *Fucus*.

Based on measured ^{137}Cs concentrations of 40-60 Bq m⁻³ in Kattegat sea water [8] and a net annual outflow of 1170 km³ [7], calculations show that 50-70 TBq of ^{137}Cs has been transported annually out of the Baltic Sea the last five years.

Technetium-99

The results for ^{99}Tc in the *Fucus* samples from Utsira from January 1995 to November 1999 are shown in Figure 2. The discharges of ^{99}Tc from the EARP plant at Sellafield started in April 1994. It is evident from the results that the discharge *front* has reached Utsira waters already in January 1997, a little less than 3 years later. This is somewhat faster than the *average* transport time of 3-4 years calculated for ^{137}Cs [4]. The annual discharges reached a peak in 1995 [5], as shown in Figure 3. The reduction in the ^{99}Tc concentration found in 1999 (see Fig. 2), indicates about the same *average* transport time for ^{99}Tc as for ^{137}Cs . Analyses of samples from 2000 may confirm this.

The variations of the concentration of ^{99}Tc shown in Figure 2 may be a combined effect of Sellafield discharge patterns and annual biophysical/-chemical mechanisms of uptake in the *Fucus*. The lower concentrations in the summer season indicate that ^{99}Tc is governed by some different mechanisms compared to ^{137}Cs which shows a concentration peak in *Fucus* in the summer season.

Annual mean concentrations of ^{99}Tc in monthly *Fucus* samples from Utsira have been calculated and is shown in Figure 3 together with annual Sellafield discharges. The *Fucus* values for 1995 and 1996 reflect the small discharges before 1994. Calculations give Sellafield FTFs of 4.2, 4.3 and 10.3 (Bq kg⁻¹ d.w. per TBq year⁻¹) for transit times of 3, 3.5 and 4 years, respectively.

Conclusions

The major source of ^{137}Cs in *Fucus vesiculosus* at the coast of southern Norway has since 1990 been the Chernobyl fallout, and most of this activity is due to the outflow from the Baltic Sea. The increased Sellafield discharges of ^{99}Tc as from 1994 reached Utsira early 1997. Preliminary calculations show *average* transit times of 3-4 years and *Fucus* transfer factors of 4-10 Bq kg⁻¹ d.w. per TBq year⁻¹ at Utsira.

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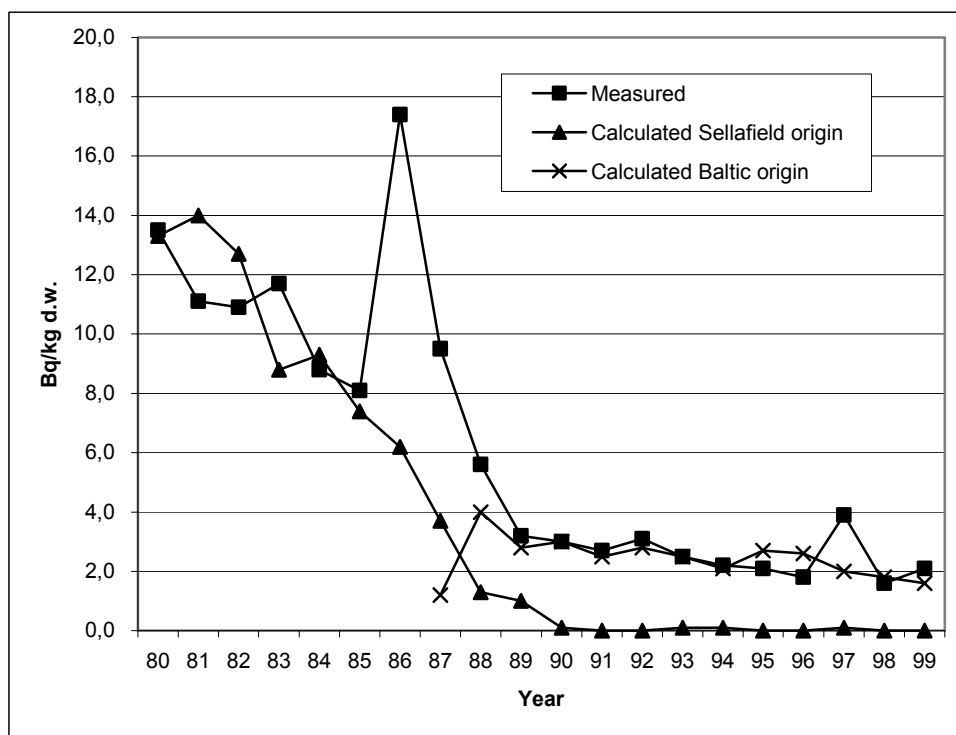


Figure 1. Calculated contributions from Sellafield and the Baltic Sea to the measured concentrations of ^{137}Cs in *Fucus vesiculosus* at Utsira.

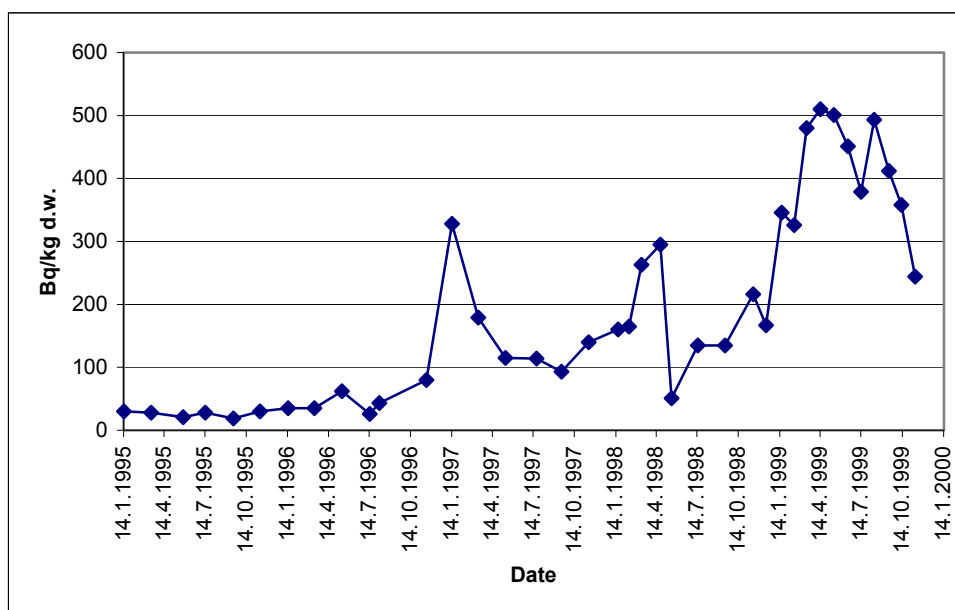


Figure 2. ^{99}Tc in *Fucus vesiculosus* at Utsira January 1995- November 1999.

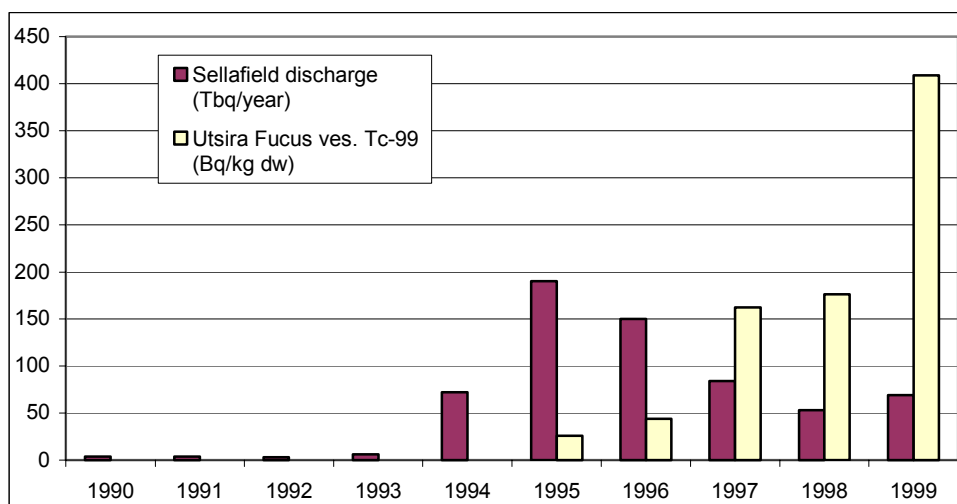


Figure 3. Annual means of ^{99}Tc discharges from Sellafield 1990-1999 and ^{99}Tc concentrations in *Fucus vesiculosus* at Utsira 1995-1999.

Technetium reduction and removal in a stratified fjord

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Introduction

The transport and behaviour of the long-lived ^{99}Tc isotope in the marine environment has become a matter of increased scientific and political interest over the past 10 years. In contrast to most radionuclides discharged from Sellafield, ^{99}Tc emissions increased approximately 20-fold in the 1990's as a result of the opening of the enhanced actinide removal process plant (EARP; BNFL 1978 - 1998). Technetium is discharged as the pertechnetate anion (TcO_4^-) which, having a negative charge, does not readily interact with particles and is therefore dispersed by tidal flow, away from the point of discharge. The conservative nature of pertechnetate makes it an ideal marine tracer, and has been used to map oceanographic transport from the Irish sea into Arctic waters.

By contrast to the high solubility of the pertechnetate anion, Tc(IV) is particle reactive, with a high affinity for iron oxyhydroxides and sulfides, and humic substances (Lieser & Bauscher, 1988). Chemical reduction occurs at Eh values below approximately +200mV (Lieser, 1995), and so reducing estuarine sediments are potential sinks for Tc. It has been demonstrated that significant amounts of Tc are retained in the anoxic sediments of the river Techa and salt marsh sediments of the Irish sea. (Aarkrog *et al.*, 1997; Morris *et al.*, 2000). However, it is also important to understand the fate of Tc in anoxic waters, as reduction in the water column and adhesion to settling particles may also be an important pathway for Tc immobilisation. In this study, the distribution of Tc in the water column of a stratified fjord has been measured to investigate the behaviour and fate of Tc on reaching reducing waters.

Sample Site

The sample site, Framvaren fjord, is an silled, stratified fjord of 180 m maximum depth, located at the southernmost tip of Norway. It has limited water exchange with the North sea via a network of fjords and a narrow, shallow channel (500 m long, 20 m wide, maximum depth 2.5 m) linking it with its neighbour, Helvik Fjord. Framvaren has a uniquely stable oxic/anoxic interface at approximately 18 m depth, with abundant microbial populations around this interface (Skei, 1988), providing an unique opportunity to examine the behaviour of trace metals and, in particular, the cycling and removal of trace metals.

The samples for this study were collected on two sampling expeditions. The first took place in November 1999, in order to establish whether Tc was measurable in the fjord. On this occasion, samples were also taken from Helvik fjord, and filtered (0.45 μm) and unfiltered samples were collected. On the second occasion, a much more detailed profile was sampled and samples from 5 depths were ultrafiltered. A sediment core was also taken from the deepest part of the fjord. Salinity, oxygen and temperature profiles were taken on both occasions using a CTD.

Methods

The methods were adapted from Chen *et al.* (1994), and Tagami and Uchida (1999), using ^{99m}Tc as the yield monitor. Briefly, the samples were acidified ($\text{pH} < 1.5$) with nitric acid, spiked and heated to destroy any organic matter. After heating, the pH was raised to pH 10 with NaOH, and the supernatant filtered through a paper filter. The supernatant was acidified to pH 6 and then pumped through a BioRad AG1-X4 column, preconditioned with 14.4 M HNO_3 (Chen *et al.*, 1994). The preconditioning stage is necessary to remove Ru from the resin, as it is the main interference when measuring Tc by ICP-MS. The columns were then washed with 5 column volumes of 0.5 M HNO_3 and then the Tc eluted with 5 column volumes of 10 M HNO_3 .

The 10 M HNO_3 elution was heated down to 0.25-0.5 ml, diluted with 25 ml deionised water (18.2 M Ω) and applied to a 2 ml TEVA.SpecTM column. The column was washed with 2 M HNO_3 (40 ml) to remove Ru and the Tc was eluted with 8 M HNO_3 (7 ml), as described in Tagami and Uchida (1999). When the initial volume of the samples was 25 or 50 l, the TEVA.SpecTM column was repeated. The yield was obtained by gamma counting in standard geometry. The 25 and 50 l samples required double spiking, first for the preconcentration stage and then for the TEVA.SpecTM columns. The samples were measured by ICP-MS.

Results

The total Tc concentration profile of the Framvaren water column is shown in Figure 1, including the data from both sampling expeditions. There was very little change in the Tc concentrations at depths of 4 and 21 m between the two expeditions, but there was a difference at 16 m. It is difficult to establish whether this difference is relevant, however, given the limited number of depths sampled on the first expedition.

The water in Helvik fjord at depths of 1 and 7 m was found to contain $0.74 \pm 0.02 \text{ mBq l}^{-1} \text{ }^{99}\text{Tc}$, which is within error of the maximum concentrations found at 6-8 m in Framvaren. This suggests that the main influx of water from Helvik fjord occurs at the depth of 6-8 m, and indeed, the salinity data give additional evidence of this. Seaweed collected from Utsira showed concentrations in the range 250 – 500 Bq kg^{-1} dry weight in 1999 (Christensen *et al.*, 2001), which corresponds to 2.5 – 5 mBq l^{-1} Tc in the seawater. The salinity of the seawater at Utsira is approximately 2.5 - 3 times higher than the water in the top 8 m of Framvaren, and so the Tc data suggest that North Sea seawater is transported into the fjord efficiently.

The distribution of Tc in the water column is shown in figure 2. Tc is taken up onto the particulate matter from a depth of 19 m, and the highest proportion of the Tc in the particulate is at a depth of 21 m, which was also the depth we observed the highest concentration of particulate matter. The fraction between 0.45 μm and 10 kDa only held measurable Tc between 21 and 23 m depth.

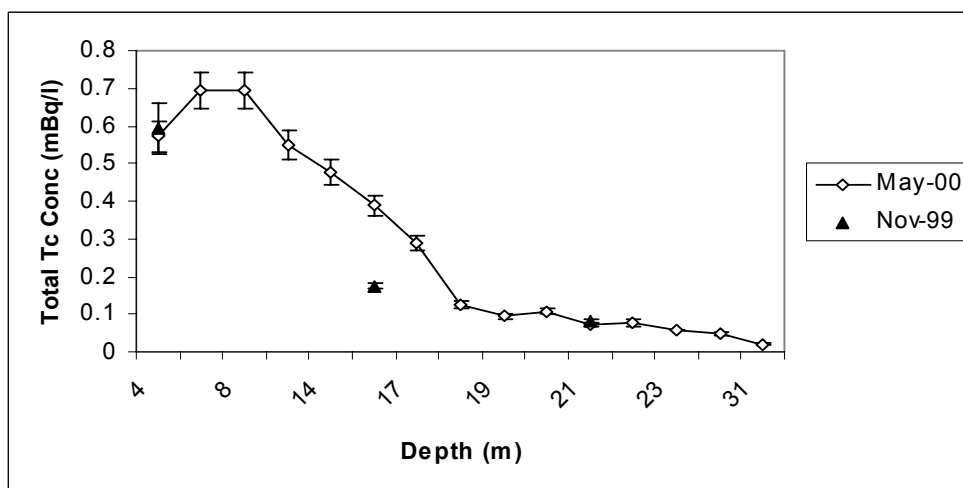


Figure 1. Vertical Profile of Total Tc in the Framvaren Water Column

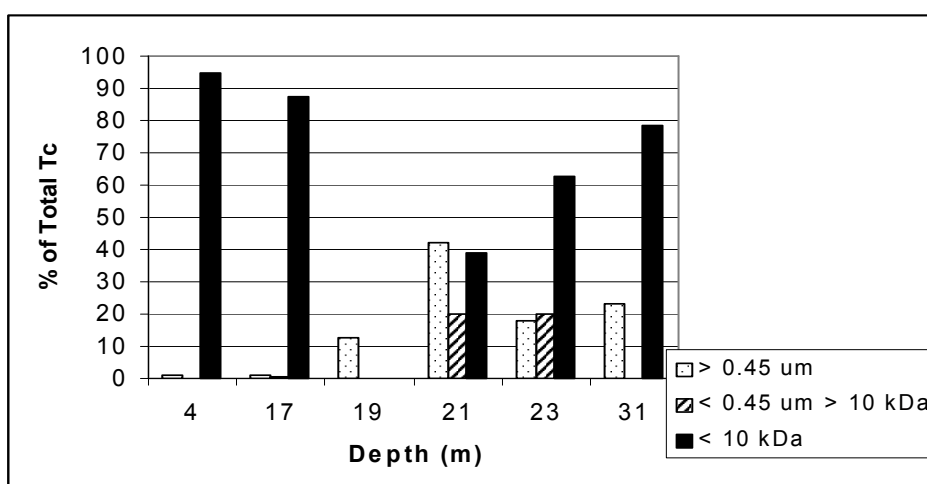


Figure 2. Distribution of Tc at Different Depths

The bulked sediment core analysed showed an overall Tc concentration of 3 Bq m^{-2} . This is a very rough estimate, given that it is based on one single core, but it shows that significant quantities of Tc have been deposited in the sediments, approximately 6 times greater than global fallout levels ($540 \pm 50 \text{ mBq m}^{-2}$; Holm, 1993)

Summary of Findings

- Technetium is being transported from Sellafield into the southern Norwegian fjords
- Slow mixing in the water column of Framvaren fjord results in vertical transport of the dissolved Tc to the oxic/anoxic interface
- Tc is reduced just below the interface and at 21 m 60% is sorbed to particulate and colloidal material

- Tc is carried to the sediments sorbed to the particulate material, where there is a current inventory of approximately 3 Bq m⁻².

Additional Work

Iron, manganese and DOC will be measured in our samples so that the data can be compared with other studies in the fjord to give a greater insight into the processes taking place. Sediment core profiles will also be analysed to assess the extent to which these sediments are a sink and the efficiency of Tc removal from the water column

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Inventory calculations in sediment samples with heterogeneous plutonium activity distribution

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Abstract

A method to determine the total inventory of a heterogeneously distributed contamination of marine sediments is described. The study site is the Bylot Sound off the Thule Airbase, NW Greenland, where marine sediments became contaminated with plutonium in 1968 after a nuclear weapons accident. The calculation is based on a gamma spectrometric screening of the ²⁴¹Am concentration in 450 one-gram aliquots from 6 sediment cores. A Monte Carlo programme then simulates a probable distribution of the activity, and based on that, a total inventory is estimated by integrating a double exponential function. The present data indicate a total inventory around 3.5 kg, which is 7 times higher than earlier estimates (0.5 kg). The difference is partly explained by the inclusion of hot particles in the present calculation. A large uncertainty is connected to this estimate, and it should be regarded as preliminary.

Introduction

Bylot Sound off the Thule Airbase, NW Greenland, was contaminated with weapons plutonium January 1968 when a US strategic bomber crashed on the sea ice with 4 nuclear weapons. Part of the weapons plutonium was released to the environment by conventional chemical explosions and further distributed in the fuel fire following the crash. The plutonium was present in an insoluble oxide form, and mainly associated to particles with an average size of 2 micrometers (USAF, 1970). During the months following the accident a clean up program was performed where most of the debris and contaminated ice was removed from the area. However, the impact caused a hole in the ice and it is possible that some of the weapons plutonium went through this crack initially and down to the sediment. In the following summer, during ice melting, the ice sheet drifted in a northerly direction in Bylot Sound causing a contamination of the sediments north of the impact point. More detailed descriptions of the accident are published elsewhere (i.e. Aarkrog 1971, 1977, 1984, 1997, Eriksson et al., 1999)

Earlier estimates of the ^{239,240}Pu inventory have been centred around 1- 1.6 TBq or approximately 0.5 kg (Aarkrog 1971, 1977, 1984, 1997, Eriksson et al., 1999). Although the inhomogeneous nature of the plutonium contamination was observed, no special attention was paid to the hot particles. The earlier works are solely based on radiochemistry and alpha spectrometry. Due to limited resolution of the alpha spectrometric equipment used, the ^{239,240}Pu peak will in some cases be growing into the ²⁴²Pu tracer peak for “hot” samples. These spectra were impossible to evaluate and was for that reason omitted in the inventory calculations.

The present work is a preliminary result based on a gamma screening experiment with sediment samples collected at the Thule-97 expedition. A method to determine the total inventory in sediments when the activity distribution is heterogeneous is described.

Methods and material

Aware of the problems with the hot particles that cause a heterogeneous activity distribution in the Thule area, we have tried to improve the inventory calculations for such sites.

A screening experiment has been performed which involves six sediment cores taken at the Thule-97 expedition. These cores were carefully selected with accordance to distance from the point of impact. From every 1-cm section of the well mixed freeze dried sections, five sub-samples were selected. Every sample was measured in a HPGe detector for 3600 seconds for determination of ^{241}Am . In total there were 450 samples. A constant $^{241}\text{Am}/^{239,240}\text{Pu}$ ratio was assumed. The ^{241}Am gamma measurements were “calibrated” to $^{239,240}\text{Pu}$ by dissolving and chemically separating the plutonium from 20 of the measured sub-samples, followed by alpha spectrometric determination of $^{239,240}\text{Pu}$. From this set of samples it is possible to combine 5^N sediment cores at every location, where N is the number of sections in the core from one location. A Monte Carlo program was written to simulate core selections of the set of sub-samples. The program was calculating the inventory (Bq m^{-2}) at each location and the inventory distribution was plotted. In this study 10 000 simulations were carried out. The total inventories were calculated from these results by fitting the data to a bi-exponential function and integrating over the area. It was assumed that the dissemination was equal in all directions and for that reason the integral could be simplified, as seen in equation 1, where $A(r, \theta)$ only depends on the distance from the impact point, r . The integration limit was determined in Eriksson et al, 1999.

$$I = \iint_{\Omega} A(x, y) dx dy = \iint_{\Omega'} A(r, \theta) r dr d\theta \quad \text{Equation 1.}$$

Results and Discussion

In Figure 1 the inventory distributions from each of the selected sites can be seen. At location V2 (point of impact) and location P (4.01 km from the impact point) it can be seen that the inventory is highly dependent on the hot particles. In location V2 there are many particles but two particles dominates that represent peak 2 and 3 in the inventory distribution. Peak 4 at location V2 represents the distribution if both of these two particles are sampled. At location P there are 4 particles, but three of them are located in the same section, i.e. they can not be sampled in the same core, and furthermore, they are grouped in two activity intervals. Peak 2 represents the distribution around the first lower interval and peak 3 the higher interval. Peak 4 is the distribution of one from the higher interval with one or the other of the two particles from the lower activity interval. Peak 5 is the distribution when both of the two particles in the higher activity group are sampled. At location Ny-3 most of the sub-samples were below the detection limit and only two samples were detectable. Earlier determinations of the inventory are in the lowest distribution peaks for location V2 and P and within the distributions for the other locations.

In Figure 2 the total simulated inventory distribution for the Bylot Sound can be seen. The median value is 9.47 TBq of $^{239,240}\text{Pu}$ which can be compared with the last estimate of 1.8 TBq (Eriksson et al., 1999). This is a significantly different result. The reason for the present higher estimate may be

explained by two factors. At first, earlier activity determinations have mainly omitted hot particles. and secondly by using an in-sufficient chemical separation method that does not totally dissolve plutonium in an oxide form. With the gamma measurements it is possible to include the hot particles, and there are no solubility problems. Another explanation can be that in this work a bi-exponential function was fitted to the data to calculate the total inventory rather than a single exponential function. The higher inventories calculated in this work might be somewhat biased as the calibration assumes constant ratio of $^{241}\text{Am}/^{239,240}\text{Pu}$ in the debris, however recently published data (Ikäheimonen et al., 2000) show that the ratio is not constant in the debris.

Future work will be to study which equation should be used as the best fit for the measured data and to analyse more samples for the “calibration” between gamma measurements of ^{241}Am and alpha measurements of $^{239,240}\text{Pu}$.

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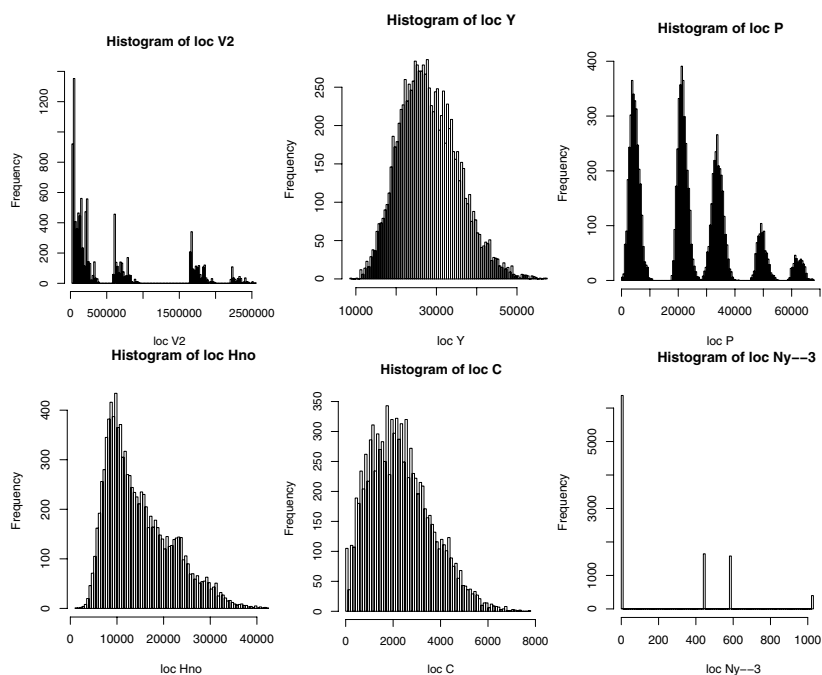


Figure 1. The inventory distributions at the six locations are shown as the result output from the simulation program. The scale on the x-axis is Bq m⁻². The explanation for the distributions at location V2 and P is that the hot particles are dominating the inventory distributions.

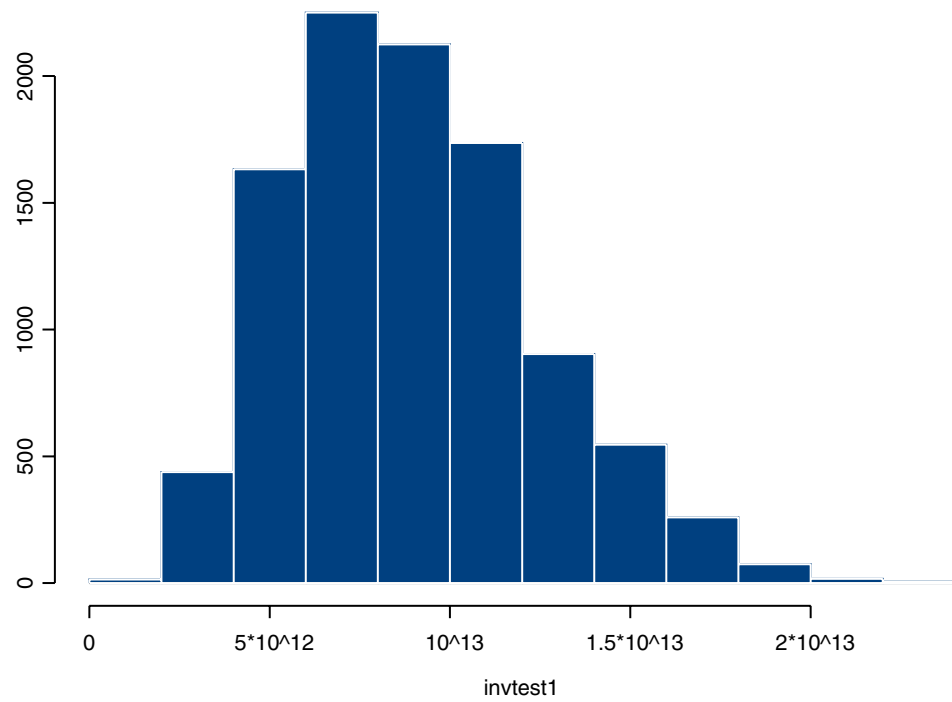


Figure 2. The distribution of the calculated inventories as a result of integration a bi-exponential function. The mean value is 9.48 TBq and the median value is 9.47 TBq.

Monitoring of radioactivity at the Russian nuclear submarine Kursk

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Introduction

In the morning of August 12th 2000, a Russian submarine accident occurred in international waters east of Rybatschi Peninsula in the Barents Sea about 250 km from Norway. The submarine, a Russian Oscar class II attack submarine, sunk to 116 meters depth at the position 69° 36,99N, 37° 34,50E (figure 1). The submarine “Kursk” is 154 meters long, equipped with two pressurised water reactors and the submerged displacement is 24000 tons. Each reactor has a thermal effect of 190 megawatt, or less than 10% of a typical nuclear power plant reactor. The submarines in Oscar-II class is one of the largest and most capable in the Russian Northern Fleet.

Kursk is a double hull construction with 10 watertight compartments separated with hatches. The outer hydrodynamic hull is made of 8 mm steel plates covered with up to 80 mm rubber. The purpose of the rubber is to prevent that other submarines or surface vessels recognise the submarine by eliminating the echo from sonar signals. The inner pressure hull is made of 50 mm steel plates and the distance between the two hulls varies from about 1-2 meters.



Figure 1. Location of Kursk (AMAP data centre)

Expeditions to Kursk

A rescue operation took place during the period 17th – 22nd August 2000 with the vessel “Seaway Eagle”, owned by the Norwegian company Stolt Offshore. The main purpose of the expedition was to open the rescue hatch in compartment no. IX in an attempt to rescue parts of the crew, if anyone were still alive. This work should be done by use of special educated Norwegian and British deep-

water divers. From the Russian side the whole operation was administered by the Russian Navy; The Northern Fleet. In the night between the 20th and 21st August, they managed to open the rescue hatch. Some air was coming out when opening the hatch. However, compartment IX was flooded with water.

After the expedition with the Seaway Eagle, the Russian officials started to plan a second expedition to gain access for divers to enter the interior of Kursk. The main purpose was to recover the bodies of the casualties. However, this expedition would also give possibilities for looking into documents and instrumentation to seek the reason for the catastrophe. The expedition took place during the period 20th October to 7th November 2000, this time the American firm "Halliburton" and their vessel MSV "Regalia" took part in the operation. At both expeditions NRPA were asked to assist the operators with regards to radiation safety for the divers and the general crew. Three experts joined the first expedition and two experts joined the second expedition. A total of twelve casualties were brought out of the submarine (from compartment IX) and on to Regalia. Debris from the seabed and documents from the control room were also brought up.

Onboard both Seaway Eagle and Regalia, a mobile radiation-monitoring laboratory was set up. In addition to different types of dose rate equipment, it contained two types of gamma spectroscopy devices, the high resolution (2.0 keV for ¹³⁷Cs) germanium detector (HpGe) and the sodium iodine detector (NaI) with lower resolution (58 keV for ¹³⁷Cs) but higher efficiency. Two types of NaI equipment were used; a 2" x 2" detector with an EasySpec multi-channel analyser and a 3" x 3" detector with a Canberra series 10 multi-channel analyser.

Sampling and monitoring

Different types of sampling and monitoring activities were performed at the two expeditions, which can be categorised as follows:

- Dose rate measurements
- Sediment sampling
- Water sampling
- Air and air filter measurements

At both expeditions, an underwater remote operating vehicle (ROV) did an initial survey around Kursk with a dose rate meter mounted in a water- and pressure proof transparent box. A camera at the ROV was pointing on the display and transferred the readings up to the surface vessel. The purpose of this survey was to control the radiation levels to make sure that the working conditions were safe for the divers. At the first expedition, the ROV were only allowed to survey the stern part of the submarine, at the position of the reactor compartment (compartment no. VI) and backwards. At the October recovery expedition the ROV went all around and on top of the submarine. The distance from the dose rate meter to the submarine hull was estimated to 50-100 cm.

At the August expedition a dose-rate reading was performed after opening the rescue hatch. At the recovery operation in October, dose rate readings were performed each time a piece of the outer or inner hull was cut out to gain access to the interior of Kursk. When pieces of the inner hull (pressure hull) were taken out, measurements were performed at the edge of the hole and also inside the submarine by mounting the meter, and a camera, on a stick.

Measurements were performed on bits and pieces from Kursk which were brought on to the main deck on Regalia (figure 2). Such pieces were debris lying on the seabed, pieces of the hull, pipe-

work from between the two hulls and pieces from the interior of Kursk. All environmental samples did also undergo dose rate measurements on the main deck before they were brought to the mobile laboratory onboard.

All dose rate measurements by the ROVs, from both expeditions, showed normal background levels in the range 0.0-0.1 $\mu\text{Sv}/\text{hour}$. The readings did not show any sign of leakage from the submarine. Neither readings outside the reactor compartment nor close to visible cracks in the submarine, showed any sign of increasing levels. Dose-rate measurements by the divers, and also measurements of bits and pieces on the main deck, showed no elevated levels of radiation. All readings were in the range 0.0 – 0.1 $\mu\text{Sv}/\text{hour}$. At both expeditions the divers used personal dosimeters during the whole operation. Readings from these batches showed low levels of radiation dose.



Figure 2. Left: Dose rate measurements at Regalia, of a piece of the inner hull from Kursk. Right: A NaI spectrum from a piece of the inner hull of Kursk (compartment VIII) is obtained.

A total of seventeen sediment samples were taken, three at the first expedition and 14 at the second. In August, Russian officials permitted sediment sampling only next to the rear part of Kursk, while samples were allowed to be taken all around the submarine in October. Six samples were taken on a straight line on each side of Kursk. Each sample was about 30 meters from each other and taken at a distance of approximately 3-6 meters from the hull of the submarine. The ROV took the sediment samples by use of a hydraulic titanium arm. A special steel corer device was made at Regalia, which made it possible for the ROV to pick up each corer from a rack of six corers. The diameter of the steel corer was 70 mm with a depth of 400 mm. The ROV picked the corer out of the rack, moved to the predefined sampling location and lowered it into the sediments once or twice to get bulk samples from the sediment surface. The sediment depth was estimated to about 10-30 cm. When the recovery operation was finalised two additional sediment samples were taken by use of a grab sampler.

Several water samples were taken in close vicinity of Kursk. These included samples of surface water, near surface water (16 m deep), bottom water and samples from inside the submarine. Surface and near surface sampling were performed by utilising the water intake of Seaway Eagle and Regalia. A Nansen water sampler device (5 litres) was used for bottom water sampling. Water from inside the submarine was taken from the bottom of the rescue hatch and from inside compartment IV and VIII. The sampling inside Kursk was performed by use of a manual drainage pump with two flexible tubes on each side. One of the tubes were lowered down into the submarine,

approximately 2 meters, while the other was placed in a 25 litres water can placed upside down. By pumping, the diver replaced the water in the can by water from inside the submarine.

Selected samples of water and sediments from “Kursk” have been analysed by gamma spectrometry. The results of these measurements did not show the presence of radionuclides leaked from the submarine and did not indicate activity levels above normal. Table 1 shows the concentrations of radionuclides in sediments, seawater and air filters after measurements by HPGe detectors at the laboratory at NRPA. Concentrations in the range 0.7 – 1.2 Bq/kg of ^{137}Cs were found in the sediment samples. These levels are “normal levels” in the Barents Sea and are attributed to fallout from the Chernobyl accident and from the bombs tests in the fifties and sixties (AMAP 1998; Grøttheim, 2000). Activity levels of ^{131}I , ^{134}Cs or ^{60}Co were not detected in any of the samples. Two NaI spectrums obtained from screening measurements onboard Regalia are shown below (figure 3). A spectrum from the screening of an air sample from inside Kursk (compartment IV) is shown together with a water sample originating from compartment IV inside the submarine. As shown in the figures, the dominating radionuclides were the naturally occurring Kalium-40 and Bismuth-214.

Table 1. Activity concentrations of samples taken in close vicinity of Kursk.

Air filters					
Sampling locality SeawayEagle; SE Regalia; REG SE	Period of measuring, date. in 2000	Concentrations in air (Bq/m ³)			
		I-131	Cs-137	Cs-134	Co-60
	20.08 – 21.08	< 0.0109 10 ⁻³	< 0.0109 10 ⁻³	< 0.0109 10 ⁻³	< 0.0109 10 ⁻³

Sediments					
Sample no.	Date	Concentrations in sediments (Bq/kg) d.w.			
		I-131	Cs-137	Cs-134	Co-60
Sed-1SE	20.08	< 0.7	0.7 +/- 38%	< 0.6	< 0.9
Sed-2SE	20.08	< 0.6	0.7 +/- 25%	< 0.6	< 0.9
Sed-3SE	22.08	< 0.3	0.7 +/- 11%	< 0.3	< 0.9
Sed-2REG	20.10		1.0 +/- 10%		
Sed-4REG	20.10		1.0 +/- 20%		
Sed-5REG	20.10		1.2 +/- 8%		
Sed-6REG	20.10		0.9 +/- 11%		
Sed-7REG	20.10		1.2 +/- 8%		

Water samples					
Sample no.	Date	Concentrations in water (Bq/l)			
		I-131	Cs-137	Cs-134	Co-60
Seawater-1-5SE	20.08	< 0.5	< 0.5	< 0.5	< 0.5

Three samples of air from the submarine were collected. A sample was obtained when the rescue hatch was opened in August, while two air samples were obtained from inside compartment no. VIII and IV in October. These samples were measured onboard by use of HPGe and NaI-detectors. No radionuclides originating from the reactors at Kursk were detected. An air sampling device, sucking 140 m³/hour through a Whatman GF/A glasfiber filter, was used at both expeditions, placed on deck. The filter (diameter 22 cm) was analysed by use of the HPGe detector. Again, no elevated levels of radioactivity were found.

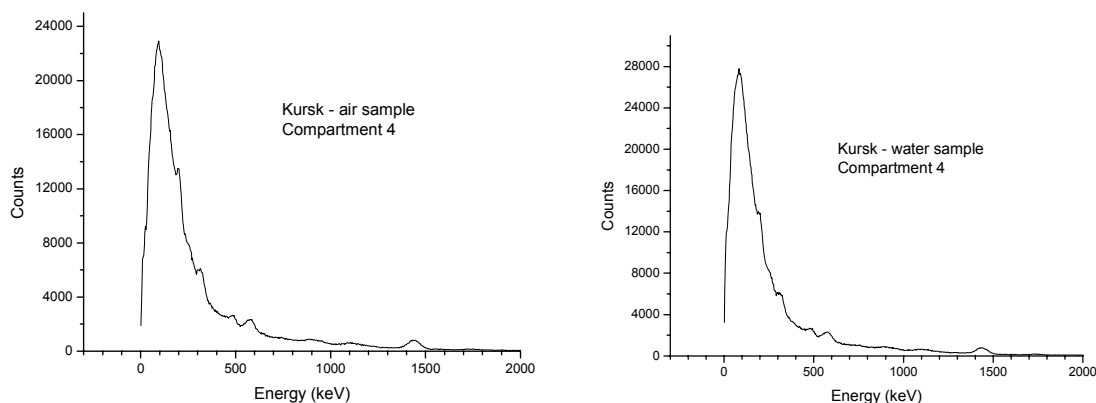


Figure 3. NaI energy spectrum of air sample (left) and water sample (right) from inside Kursk (compartment no. IV). Total number of counts is shown as a function of energy (keV).

Conclusions – future work

No indications of leakage from the submarine have so far been observed during these expeditions. Elevated levels of radioactivity have not been detected in any dose-rate readings or at any of the measurements of environmental samples taken close to Kursk. Furthermore, no increased levels were measured on bits and pieces from the submarine or from water sampled inside the submarine. A more comprehensive report covering experience and monitoring results from the two expeditions will soon be published (Amundsen et al., in prep.). It also includes estimations of inventory, source term and impact assessments of possible future releases from Kursk.

As long as Kursk is lying on the seabed it will be of importance to run a surveillance programme for controlling the activity levels in the environment in the area. It is also necessary to conduct modelling work on hypothetical leakage and spreading of radionuclides in the Barents Sea. Works on these tasks are now underway (Gerdes et al., subm.; Amundsen et al., in prep.). Norwegian and Russian authorities have agreed to co-operate on future expeditions in areas around Kursk for surveillance purposes. It was agreed to establish a common database on results of environmental monitoring in connection to the Kursk accident. In addition it will also be established a working group on performing an impact assessment as a consequence of a hypothetical radioactive release from the reactors in Kursk.

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Radiocaesium och polonium i säl från Östersjön, en preliminär studie

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Sammanfattning

Radiocaesium (^{137}Cs , $T_{1/2}=30$ år) och ^{210}Po ($T_{1/2}=138$ d) har studerats i muskel, lever och njure från 5 sälar (4 st gråsälar och en vikare) från Östersjön. Genom att anta att sälarna konsumerar 5% av sin egen kroppsvikt per dag och genom att utnyttja tidigare data för koncentrationerna i fisk har biologiska halveringstiden uppskattats till 20 dagar för radiocaesium och 35 dagar för polonium. De maximala årliga doserna beräknas till 76 μGy från radiocaesium till muskel och 3 500 μGy till njurarna från polonium. Dessa data är mycket preliminära eftersom antalet individer är lågt och det är svårt att uppskatta hur mycket annan föda än fisk som är av betydelse för intaget. Dessutom finns få data för koncentrationer i helfisk tillgängliga vilket speciellt påverkar resultaten för polonium.

Introduktion

Relativt få studier har gjorts vad gäller radioaktivitet i säl, speciellt vad gäller Östersjön, trots det är det innanhav som är mest kontaminerat av ^{137}Cs till följd av Chernobylolyckan. Även före Chernobylolyckan var koncentrationerna av ^{137}Cs och ^{90}Sr relativt mycket högre än för andra marina områden på samma latitud kontaminerade från kärnvapen fallout. Den speciella karaktären av Östersjön gör att lösta substanser har en lång uppehållstid. Saliniteten är varierande men låg och koncentrationsfaktorerna till alger och fisk är ofta betydligt högre än för normala marina områden vilket också borde vara fallet för säl. Sälarna i Östersjön utgör idag ingen föda för människor. Det är däremot ganska stora däggdjur som lever relativt länge och konsumerar stora mängder av fisk (5-10% av sin kroppsvikt per dag (1)). De kan därigenom utgöra en integrerande indikator på kontamineringsnivån i fisk. Sälarna utgör toppen av födoämneskedjan i Östersjön och i framtiden är det inte helt otänkbart att husbehovs och kommersiell jakt återupptas.

För att kunna utnyttja säl för detta ändamål är det viktigt att producera data ur vilka biologiska halveringstiden och koncentrationsfaktorerna i säl kan beräknas. Absorberade dosen kan därefter beräknas, mer av akademiskt intresse än att vi tror att populationen idag påverkas av de existerande dosraterna.

Radiocaesium är den radionuklid som gett högsta stråldosen till följd av Chernobyl olyckan i såväl akvatisk som terrest miljö. I Östersjön har radiocaesium haft ett dynamiskt förhållande. Som jämförelse har vi valt en naturlig radionuklid, ^{210}Po , som är dotterprodukt till radon och deponeras relativt lika år från år. Vi kan därför förvänta oss en jämvikt i systemet för denna radionuklid, bortsett från säsongsvariationer. Dessutom är det den radionuklid som ger människan den högsta dosen från alfa-strålare genom födointag.

Material och metoder

Prover av säl, muskel, lever, njure, från 1999 erhöles genom Naturhistoriska Riksmuséets försorg och utgjordes av de sälar som förolyckats och levereras till Muséet. Tabellen nedan anger data för proverna.

Proverna torkades, maldes och analyserades för radiocaesium genom gamma-spektrometri. En fraktion av provet våtinskades med perklor-syra och salpetersyra, varefter spontan deponeering av polonium gjordes på nickel. Mätning av polonium gjordes med alfaspektrometri. Polonium-209 användes som radiokemisk utbytes-bestämmare. Proverna analyserades någon till några månader efter insamling. Eftersom ^{210}Po är dotterprodukt till ^{210}Pb har vi antagit att aktivitetskoncentrationerna av ^{210}Po är mycket större än de för ^{210}Pb . Korrektion för sönderfall och uppbyggnad av ^{210}Po har i denna preliminärstudie gjorts för vissa uppskattade aktivitetsförhållanden.

Tabell 1. Provtagning

Art	Kön	Dödsdatum	Vikt (kg)	Lokal
Gråsäl	Hane	990620	142	Mellan Söderhamn och Gävle
Vikare	Hane	990504	50	Hanöbukten
Gråsäl	Hane	990222	60	Gransö/Flatö, Östergötland
Gråsäl	Hane	990728	60	Oxhällan, Medelpad
Gråsäl	Hane	990530	årsunge	Lacka, Sörmland

Resultat och konklusioner

Resultaten för aktivitetskoncentrationerna i sälarna framgår av Tabell 2. Det visar sig att det är ganska stor spridning. Möjligtvis kan detta spegla olika födoval vilket inte gör det lättare att beräkna biologiska halveringstiden. Vid beräkningarna har aktivitetsförhållandet $^{210}\text{Po}/^{210}\text{Pb}$ i levande säl antas vara 15 för muskel och 9 för lever och njure. Aktivitetsförhållandet har baserat sig för de man finner i ren men kan vara ännu högre för säl p.g.a. att förhållandet är c:a 1 i födan (lav) för ren (2) men större än 1 i födan (fisk) för säl.

Effektiva halveringstiden har beräknats enligt följande varefter den biologiska lätt kan härledas.

$$dC/dt = W_f A_f - \lambda F W_m C$$

där dC/dt är förändringen per tidsenhet av kroppsinnehållet i säl som antas vara noll (steady state).

W_f = dagliga intaget av fisk (kg våtvikt) som antas vara 5% av kroppsvikten

A_f = koncentrationen av radionukliden i fisk Bq kg våtvikt

$\lambda = \ln 2 / T_{1/2(\text{eff})}$ där $T_{1/2(\text{eff})}$ är effektiva halveringstiden

F är den fraktion av djuret som innehåller Cs, s.k. lean body weight = 0,6 (1)

W_m är totala vikten på sälen

Aktivitetskoncentrationen i fiskfilé (torsk och strömming) kan som medelvärde, 1999, beräknas till 15 Bq kg^{-1} våtvikt för ^{137}Cs och $0,68 \text{ Bq kg}^{-1}$ för ^{210}Po (3). För caesium antas koncentrationerna i filé motsvara den för hela fisken. För polonium anges så också vara fallet för torsk medan strömming kan beräknas på helkroppsbasis ha 5 gånger så hög koncentration som i filé (4). För polonium gissas grovt att koncentrationen i helfisk i medeltal är 3 Bq kg^{-1} .

Tabell 2. Aktivitetskoncentrationer, biologiska halveringstiden, koncentrationsfaktorer och doser för radiocaesium och polonium i säl från Östersjön.

Matrix	¹³⁷ Cs	¹³⁷ Cs	²¹⁰ Po	²¹⁰ Po
	Bq kg ⁻¹ t.v. (spridning)	Koncentrations- faktor	Bq kg ⁻¹ t.v. (spridning)	Koncentrations- faktor
muskel	122 (76-170)	2 400	40 (12-77)	80 000
lever	81 (54-127)	1 600	93 (21-191)	180 000
njure	96 (65-165)	1 900	188 (41-526)	360 000
Biologisk halveringstid, dagar	20±3		35±14	
Maximal dos, µGy år ⁻¹	76 (muskel)		3 500 (njure)	

Koncentrationsfaktorn (på torrviktsbasis, t.v., i tabellen) har beräknats för en vattenkoncentration på 50 mBq l⁻¹ för ¹³⁷Cs och 0,5 mBq l⁻¹ för ²¹⁰Po. Faktorerna för radiocaesium till muskel, 2 400 (c:a 670 på våtviktsbasis, v.v.) är, som förväntat, som regel högre än de som publicerats för andra områden; 13-70 (v.v.) (Kara havet)(5), 73 (t.v.) Barentshavet), (6), 290-1400 (v.v.) (Skottland), 196-424 (v.v.), (England och Irland) (1). För polonium finns få data att jämföra med. Kring Antarktiska halvön var koncentrationsfaktorn för polonium till säl c:a 8 000 för muskel (v.v.), 95 000, för lever (v.v.), och 91 000 för njure (v.v.) (7).

Den beräknade biologiska halveringstiden, 20 dagar, för ¹³⁷Cs är av samma storleks-ordning som man funnit vid England och Irland, 28 dagar (1).

Doserna har beräknats i µGy per år. Man skulle givetvis kunna anta samma kvalitets- och viktningsfaktorer som för människan, men vi tror inte på att sådana begrepp som sälSievert har någon relevans. Som jämförelse uppskattades dosen till 120 µSv år⁻¹ i ett extremfall där koncentrationen av ¹³⁷Cs i muskelvävanden var 66,6 Bq v.v. men mer generellt vid Englands och Irlands kuster till < 14 µSv år⁻¹ (1). Några jämförande data för polonium har inte gått att finna.

Konklusionen är den att preliminära data visar att koncentrationerna och koncentrationsfaktorerna speglar de högre koncentrationer av radiocaesium som finns i vatten och fisk i Östersjön samt den lägre saliniteten. Polonium verkar inte ha någon salinitetseffekt för koncentrationer i fisk (3). Tvärtom borde koncentrationsfaktorerna i säl härvid tänkas vara högre för polonium vid högre salinitet genom att räkor och skaldjur utgör en större del av födan.

Studien visar att säl på toppen av födoämnescykeln kan utgöra en indikator på radioaktiviteten i fisk och bör utökas med ett större antal djur samt också omfatta Svenska västkusten. Även plutonium och technetium bör ingå i ett selekterat program.

Erkännande

Detta arbete har haft ekonomiskt stöd av Statens Strålskyddsinstitut

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Chernobyl radiocesium in freshwater fish: Long-term dynamics and sources of variation

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Background

The radioactive cesium isotopes ^{134}Cs and ^{137}Cs are artificial products of nuclear reactions and are not occurring naturally in the biosphere. Nevertheless, they are today globally distributed contaminants due to atmospheric nuclear bomb detonations, nuclear power plant accidents and controlled release from nuclear industry. Cesium is a very reactive and therefore found in nature only as monovalent ions, Cs^+ . Because of the similar physicochemical properties of Cs^+ and K^+ radiocesium accumulates in biota, which implies potential health hazards for humans. For this reason radiocesium, especially the long-lived ^{137}Cs (half-life 30.17 years), has for decades attracted both radioecologists and health physicists. From another point of view, since ^{137}Cs is inert, not essential, has a simple chemistry and is able to measure at low levels, it has been suggested as a suitable tracer and tool for scientific research. Common applications for ^{137}Cs are the dating of recent sediments and studies of soil erosion. To a more limited extent radiocesium has been used in ecology, but has high potential in studies of bioenergetics, trophic transfer and migration in aquatic ecosystems (Davis & Foster, 1958; Kevern, 1966; Kolehmainen, 1974; Meili, 1994; Odum & Golley, 1963; Rasmussen & Lassen, 1994; Rowan & Rasmussen, 1996; Storebakken, Austreng & Steenberg, 1981; Tucker *et al.*, 1999; Tucker & Rasmussen, 1999).

When the radioactive fallout from the Chernobyl nuclear plant accident in 1986 was detected over large areas of Europe, radioecological research activities had been diminishing since the era of atmospheric nuclear bomb tests. Naturally the new and alarming situation led to the initiation of numerous research and sampling programs. Although much of the old work was substantial and of high quality it was not readily applicable to the post-Chernobyl situation. Unlike the global and more or less continuous fallout during years the 1950's and 1960's, selected areas received the nuclides from Chernobyl as a distinct pulse. As a consequence, affected ecosystems were in strong disequilibria with regard to long-lived radionuclides, such as ^{137}Cs . A dynamic equilibrium situation could no longer be assumed or justified; radioecologists and modelers were forced to focus more on dynamic situations. This proved not to be a simple task, since the equilibration rates differ substantially between different ecosystem compartments and depend much on local conditions. Taking radiocesium in freshwater fish as an example, lake chemistry, morphology and hydrological regime, the catchment's vegetation, soil types and land use, as well as climate and fish ecology, are among the factors that have been considered in post-Chernobyl dynamic models (e.g. Bergström, Sundblad & Nordlinder, 1994; Håkanson *et al.*, 1996a; Håkanson *et al.*, 1996b). In retrospect, the early attempts to predict the elimination of radiocesium from lake ecosystems were too optimistic. Recent studies report that levels in fish remain high and decline very slowly (Jonsson, Forseth & Ugedal, 1999; Saxén & Koskelainen, 1996).

There are two other problems that face both the radioecologist and the modeler that wishes to learn more about or predict radiocesium accumulation in fish. First, the shortage of data, especially from

the early 1990's and later – when ^{137}Cs concentrations in fish were declining again in the late eighties, many monitoring projects were discontinued. Also the quality of data has not been sufficient. The large scale national monitoring projects often acquired samples from a multitude of sources, and little information was given about catch conditions, fish size, sex, condition or diet. Second, within lakes and even at the same sampling occasion ^{137}Cs concentrations in fish display a very high variability, often two-fold or more (e.g. Håkanson, 1999). Knowledge about the magnitude and sources of this variation is important for the reliability of data, the design of sampling routines and accurate predictive modeling of radiocesium in fish. Furthermore, for applications that use ^{137}Cs as tracer for fish bioenergetic processes (He & Stewart, 1997; Kevern, 1966; Kolehmainen, 1974; Rowan *et al.*, 1995; Sherwood *et al.*, 2000), understanding of the factors responsible for individual variation is of crucial importance.

The aim of this thesis was to investigate both the long-term temporal pattern and sources of individual variation for radiocesium in freshwater fish. The basis for the study is time series of ^{137}Cs activity concentrations in fish from three lakes in the area North-west of Uppsala, Sweden that received considerable amounts of ^{137}Cs from Chernobyl in may 1986. The lakes were Lake Ekholmssjön, Lake Flatsjön and Lake Siggeforasjön, all small forest lakes, but with different morphometrical and chemical characteristics. The data were collected regularly, usually several times per year, during 1986-2000, using consistent methods. More than 7600 fish individuals from 7 species covering wide size ranges and feeding habits were analysed for ^{137}Cs . For each fish was the length, weight, sex, and often the stomach content recorded. The evaluation on long-term trends were based on data from all three lakes, while the study on sources of variation evaluated data from Lake Flatsjön only.

Long term trends

After the Chernobyl pulse deposition of radiocesium, the maximum activity concentrations in lake water were attained almost immediately. The following removal of ^{137}Cs from the water column was first very rapid and then gradually became slower (Saxén, Jaakkola & Rantavaara, 1996). The fast removal phase was largely dependent on lake dilution and particle setting rates (Smith, Comans & Elder, 1999). Later on, as the concentrations leveled off, secondary input from the catchment area, remobilization from sediments by diffusion (Comans *et al.*, 1989; Smith & Comans, 1996) or resuspension (Broberg, Malmgren & Jansson, 1995; Meili, Braf & Konitzer, 1997) have been pointed out as secondary sources of ^{137}Cs to lake water, thereby slowing down the net removal of ^{137}Cs . The slow sorption ^{137}Cs to clay minerals has been suggested to be the ultimate controlling factor for the long term decline of ^{137}Cs (Comans & Hockley, 1992; Hilton *et al.*, 1993; Kudelsky *et al.*, 1996).

Since fish take up cesium predominantly from the food, direct uptake being negligible, the pulse of ^{137}Cs levels in fish is significantly delayed compared to that lake water. The typical pattern is a fast increase until maximum levels are attained, followed by a relatively fast decrease and thereafter the concentrations level off along a plateau formation, probably indicating a gradual equilibration with the slowly declining concentration in water. Although there are many reports on the temporal trend of Chernobyl ^{137}Cs in fish there is no consensus on how the long-term pattern varies across species and different types of lakes. This is because most published studies covered only the first years after fallout, used small samples sizes or possessed little information of the size and diet of each fish.

In this study, a non-linear model was developed and fitted to the time series of ^{137}Cs in fish. The fish was first grouped into species and for abundant species also into different size classes. ^{137}Cs

concentrations were corrected for radioactive decay since May 1, 1986. The main benefit of the model was that it enabled an objective and consistent method for extracting the parameters of interest. These were: (1) maximum activity concentration (C_{max}); (2) the time to reach maximum activity concentration (t_{max}); (3) the maximum rate of decline (or minimum ecological half-lives); (4) the near-steady state level (Base) and (5) the long-term ecological half-life.

The ¹³⁷Cs dynamics during the initial years, as well as the leveling off during the following years, were related to the type of fish. Both t_{max} and minimum half-life increased significantly with trophic level and weight. The effect of trophic level on t_{max} was stronger than the effect of weight, suggesting that retardation by the food chain was largely responsible for the delayed ¹³⁷Cs peak in piscivorous fish. The opposite was found for the fast-decline phase. Fish weight had stronger effect on the minimum ecological half-life than did trophic level. Small fish entered the near-steady state phase already in 1888-1989, significantly earlier than larger fish. Clearly, during this phase the biological half-life was more important than the food chain retardation, probably because the change in concentration of ¹³⁷Cs in water was slower after the peak than earlier.

The maximum levels increased with trophic level but decreased with weight. As mentioned above, large fish reached maximum levels later than smaller fish, which implies that their food contained less ¹³⁷Cs due to the overall elimination of available ¹³⁷Cs from the lake. The positive effect of trophic level, however, indicates the importance of biomagnification of radiocesium. This is also reflected by the near-steady state levels (Base), which increased markedly with trophic level, especially in the transition to piscivory. The effect of weight on Base was smaller. Both C_{max} and Base were significantly higher for perch (*Perca fluviatilis*) than the other species. Even small non-piscivorous perch had considerably higher levels than other fish on comparable trophic positions.

Although the absolute levels varied among lakes, the relationships between the model parameters and trophic level or weight did not differ significantly. The general dynamic pattern of ¹³⁷Cs in lacustrine biota following a pulse contamination is thus to a large extent governed by the fish population and food web characteristics. This inertia of the food web implies that cesium dynamics may be partly uncoupled from other processes and would, if properly described and quantified, simplify the task of cross system modelers.

Long term ecological half-lives were of the same magnitude as the physical decay rate (30.17 years) and were not influenced by the fish type. In addition, the estimates of ecological half-life increased over the period, apparently approaching infinity, suggesting a gradual shift in the processes regulating the radiocesium levels in fish: From physiological, over ecological and geochemical factors, to finally radioactive decay. Ecological half-lives were long and increased over the period. If estimated over the period 1996-2000, the median ecological half-life was 20 years in Ekholmssjön, 50 years in Siggeforasjön and 80 years in Lake Flatsjön. This means that in practice the radioactive decay will determine the elimination of ¹³⁷Cs from these lake ecosystems. Large perch in Siggeforasjön and both perch and pike in Flatsjön had average levels above the radiation protection guidelines. Thus, fishermen in many lakes in this area ought to wait another 50-60 years before to eat or sell their catches.

Within-lake variation

During the period 1996-1999 the fishing was intensified in Lake Flatsjön. The aim was to study what factors that were responsible for the high individual variability that was observed for all species of fish. Linear models (ANOVA, ANCOVA) were used to detect the variables that could explain most of the variation. Variables of interest were species, size, condition factor, food choice, gonadal- and gastro-somatic indices, flesh water, potassium and lipid concentrations and seasonal variability. In addition, the variability that could be generated by individual variability in bioenergetic factors was calculated and compared with the observed variation.

The total variation was 22-fold among the 1338 individuals of seven fish species. Individual variation was high, across species CV:s ranged from 0.16 to 0.69 with an average of 0.35. Seasonal variation accounted for only 7% of total individual variation. The amount of the variability that could be explained by the combined effects of season (sampling date) and fish length is varied among species. R²-values ranged between 0.07 and 0.59, which means that 41-93% of the variability must have been generated by other factors. Fish species, diet composition, fish size and condition factor were significant predictors of ¹³⁷Cs levels. Perch and pike (*Esox lucius*) had generally higher levels than cyprinid fish. ¹³⁷Cs levels usually increased linearly with fish length, except for perch, where levels increased with length following a more complex or stepwise pattern. The average proportion of fish in perch diet was well correlated with this pattern. Comparison of the observed variation with the variation expected from potential individual differences in bioenergetics indicated that additional factors were important for most species. Although contamination levels decreased with increasing condition factor, neither muscular lipid, potassium concentrations, sex nor gastro- and gonado-somatic indices contributed significantly to the observed variation. For perch and pike, however, individual food choice based on stomach content analyses could explain some of the exceptionally high ¹³⁷Cs variation in this species. Accordingly, total variation was higher for species or size classes with wide diet preferences. The results suggest that, while basic trophic structure and physiology determine the baseline ¹³⁷Cs levels in fish within a lake, the high variation among similar individuals often found in ¹³⁷Cs-contaminated lakes is to a large extent caused by individual food choice.

The average biomagnification factor between piscivores and the most common prey fish was 4.13 for large perch and 2.17 for pike. The pooled estimate of 3.17 is similar to the average piscivore biomagnification factor of 3.69 obtained by Rowan & Rasmussen (1998). Although the trophic relationships within the zooplankton and benthic communities are complex because of detritivory and microbial food webs there are more carnivorous taxa within the littoral benthos than in the water column. Assuming that biomagnification occurs also in the invertebrate food webs, it would be expected that benthivores had higher ¹³⁷Cs levels than zooplanktivores, which in turn would have higher levels than herbivores. Carnivorous Odonata larvae were a common prey for small perch (10-15 cm), while herbivorous crustaceans dominated the diet of perch less than 10 cm. However, there was no difference in ¹³⁷Cs levels between the groups. Within the non-piscivore cyprinids there was only small differences between omnivorous roach (*Rutilus rutilus*), mainly benthivorous bream (*Abramis brama*) and white bream (*Abramis blicca*), and largely herbivorous rudd (*Scardinius erythrophthalmus*). The results provide no evidence of a significant biomagnification of cesium along invertebrate food chain. One explanation for this may be that, due to small body size, direct uptake of ¹³⁷Cs is relatively more important for invertebrates than for fish, for which trophic uptake dominates. Furthermore, it has been argued that benthos and hence benthivores should accumulate more ¹³⁷Cs than zooplankton and zooplanktivores since they are in direct contact with the highly

contaminated sediment surface. Contrary to the results of Rowan & Rasmussen (1998) no such effect was found in Lake Flatsjön suggesting the sediment-bound ^{137}Cs is not readily bioavailable.

In conclusion, although seasonal, size, and bioenergetic effects together can explain much of variation, individual food choice is of importance for species with wide diet preferences. This ought to be considered in bioenergetic models that use cesium as a tool for estimating e.g. feeding rates *in situ*. The generally high variability in popular game fish should be considered when monitoring environmental radioactivity or assessing the health risks associated with freshwater fish consumption.

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Origin and fate of ^{129}I in Swedish lakes and soils

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Abstract

Recent measurements of iodine-129 using accelerator mass spectrometry (AMS) have revealed surprisingly high concentrations in Swedish lake waters: often more than one order of magnitude above present background levels in Arctic lakes, and 5 orders of magnitude above natural levels. Patterns along a steep local gradient of Chernobyl fallout are first to show that present emissions from the nuclear fuel reprocessing facilities at La Hague (France) and Sellafield (UK) are the dominant source of ^{129}I not only in the oceans but also in remote freshwaters, even at distances of over 1400 km. Comparison of $^{129}\text{I}/^{127}\text{I}$ isotope ratios suggests that most of the ^{129}I is supplied by a large-scale dispersal of ongoing emissions to the atmosphere, rather than by volatilisation of ^{129}I from polluted seas. Calculations based on simultaneous monitoring of ^{129}I in precipitation indicate contributions from both wet and net dry deposition. Comparison of lakes with different watershed properties suggests an efficient retention of ^{129}I in boreal forest soils. All these findings point at the relevance of studying the dispersal and fate of European ^{129}I emissions not only in marine waters but also in freshwater and terrestrial environments.

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

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Abstract

Seaweed samples were collected at 28 stations along the Finnish coast on purpose to study, whether Tc-99 could be detected in the coastal waters of Finland. Prior to the project, Tc-99 had not been analysed at the STUK; thus one of the purposes was to bring an analysis method for Tc-99 in daily use at the laboratory.

A new modification of the method for analysing Tc-99 in marine samples was developed (Ikäheimonen et al., 2001). The method is based on extraction chromatography using *TEVA resin* as reagent. After pre-treatment, Tc is extracted into the resin, which is measured with liquid scintillation analyser. Yield measurements were made using gamma-spectrometry and a short-lived Tc-99m tracer. The method is suitable for various kinds of environmental samples, though the pre-treatments are different. The effectiveness of the measurement of Tc-99 is about 85% and the theoretical detection limit is 0.003 Bq per sample.

Small amounts of Tc-99 were observed in all the *Fucus* samples collected; the activity concentrations ranged from 1.6 to 11.6 Bq kg⁻¹ d.w. In the samples collected from the Gulf of Finland, the average concentration was 3.4 Bq kg⁻¹ d.w., and in those collected from the Archipelago Sea and Åland Islands 2.9 Bq kg⁻¹ d.w. In the samples collected from the Bothnian Sea the average concentration was 5.3 Bq kg⁻¹ d.w..

The activity concentrations of Tc-99 were clearly highest (11.4 - 11.6 Bq kg⁻¹ d.w.) in two samples taken from the northernmost stations located in the Quark (passage between the Bothnian Sea and the Bothnian Bay). This kind of distribution pattern was probably due to biological factors. Due to the low salinity of water, *Fucus* lives in the Quark in extreme environmental circumstances and at the northernmost border of its distribution range in the Baltic Sea. The thalli of *Fucus* are there slowly growing and small-sized, and this probably increases the uptake of radionuclides to the seaweed.

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an unabridged article will be published in Boreal Environmental Research

Miljöövervakning runt kärntekniska anläggningar i Sverige – utvärdering av 20 års data

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Introduktion

Miljön runt de kärntekniska anläggningarna i Sverige kontrolleras genom ett särskilt miljöövervakningsprogram. Det övergripande syftet med detta program är att mäta koncentrationen av radioaktiva ämnen kring anläggningarna. SSI gjorde förra året en mer omfattande utvärdering av programmet som baseras på närmare 20 års erfarenheter och mätdata. Huvudsakliga syftet med utvärderingen var att göra en mer utförlig analys av mätvärdena och ta fram förslag på hur provtagningsprogrammet skulle kunna förbättras. Några av slutsatserna sammanfattas här. En utförligare beskrivning av utvärderingen har publicerats i SSI-rapport 2000:13 (Wallberg och Moberg 2000).

Övervakningsprogrammet omfattar ett grundprogram och ett intensivprogram vid kärnkraftverken i Ringhals, Barsebäck, Oskarshamn och Forsmark samt vid forskningsanläggningen i Studsvik. Grundprogrammet utförs varje år under vår och höst, i huvudsak före och efter revisionsperioden då underhåll av verken sker. Förutom provtagningarna vår och höst tas vissa prover dessutom vecko-, månads- eller kvartalsvis. Intensivprogrammet genomförs vart fjärde år vid respektive anläggning och omfattar ett större geografiskt område än grundprogrammet.

Övervakningen omfattar mätning av radioaktiva ämnen i biologiska material (växter, djur och livsmedel) och andra provslag såsom vatten, sediment och rötslam (Tabell 1). Dessa mätvärden skall kunna användas för

- att testa beräkningsmodeller som används för att bedöma utsläppens påverkan på människa, både vad det gäller stråldos till de som kan väntas få de högsta stråldoserna och kollektivdos
- att detektera större oregistrerade utsläpp och utsläpp som eventuellt sker diffust
- att bedöma eventuell påverkan på biologiskt liv i recipienten
- att ge en bild av långsiktiga förändringar av radionuklidhalter i miljön
- att ge underlag för information till allmänheten
- att ge underlag för internationell rapportering och övrig samverkan inom miljöområdet

Med utgångspunkt från dessa mål har vissa arter och provslag valts ut på grundval av att de uppfyller ett eller flera av följande kriterier.

- Anrikar radioaktiva ämnen.
- Väl representerar området.
- Kan lätt insamlas varje år i tillräcklig mängd.
- Är stationär i området.
- Förekommer vid flera anläggningar.

- Används som livsmedel eller ingår i människans näringskedja.

Miljön kring anläggningarna skiljer sig på grund av att salthalten varierar längs kusten och att fördelningen mellan jordbruks- och skogsmark är olika. I vissa fall innebär detta att en art som inte finns på alla lokaler har fått ersättas av en annan som har motsvarande funktion i ekosystemet (Tabell 1).

Resultat

De radioaktiva ämnen som kan detekteras i proverna och som kan härledas från verken är främst ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$ och ^{137}Cs . Efter Tjernobylyolyckan 1986 detekterades i alla kontrollområden förhöjda koncentrationer och en förändring i sammansättningen av radioaktiva ämnen i många provslag. Framför allt märktes högre koncentrationer av ^{137}Cs , men även ^{134}Cs och ^{144}Ce var mer vanligt förekommande under några år efter nedfallet. I många provslag var koncentrationerna av ^{137}Cs fortfarande något högre 1999 än vad den var före Tjernobylyolyckan. Under 1999 uppgick dosen från anläggningarna till den mest utsatta gruppen av människor till 1-7 % av gränsvärdet (SSI-rapport 2000:19). De högsta utsläppen kom från kärnkraftverket i Ringhals.

I vattenmiljön visar en jämförelse mellan blåstång, grönslick/tarmtång, snäckor (*Radix*, *Theodoxus* och *Littorina*), Östersjömussla, blåmussla och fisk att halterna av ^{54}Mn , ^{58}Co , ^{60}Co , ^{65}Zn , $^{110\text{m}}\text{Ag}$ och ^{137}Cs är högst i blåstång. I snäcka uppmättes $^{110\text{m}}\text{Ag}$ i fler prov än i något annat provslag och resultat från Studsvik tyder på att ^{152}Eu detekteras oftare i Östersjömussla än i blåstång och blåmussla. Ligger provtagningslokalen nära utsläppspunkten kan en korrelation mellan halter av ^{60}Co i utsläppet och i blåstång påvisas (Figur 1). Vid stationer belägna längst bort från utsläppspunkten är koncentrationerna lägre men blåstång har de högsta halterna och största antalet detekterade radioaktiva ämnen jämfört med alla andra provslag.

Liksom blåstång visar även sedimentprover en spridningsgradient med minskande koncentrationer längre bort från utsläppspunkten. I sedimenten beror fördelning av radioaktiva ämnen inom spridningsområdet på strömmar och sedimentationsförhållanden. Stationer som ligger på utpräglade ackumulationsområden tenderar att ha högre koncentrationer än stationer på erosionsbottnar. En och samma station visar dock ofta stora variationer i halter från år till år. Det kan bero på faktorer som t.ex. sedimentationsförhållandena och svårigheten att upprepa provtagningen vid exakt samma position. Av de nuklider som förekommer i utsläppen detekteras ^{60}Co och ^{137}Cs i de flesta prov från det översta sedimentskiktet (0-2 cm). Av övriga radioaktiva ämnen som släpps ut kan låga halter av främst ^{54}Mn , ^{58}Co , ^{65}Zn och $^{110\text{m}}\text{Ag}$ detekteras i sedimentprov tagna i omedelbar närhet av utsläppen. Efter Tjernobylyolyckan dominerar den totala gamma-aktiviteten i sedimentproverna av ^{137}Cs . Halterna av ^{60}Co är låga med undantag för Hamnefjärden vid Oskarshamnsverket och i Biotestsjön vid Forsmarksverket där högre koncentrationer kan uppmätas. Några enstaka stationer nära utsläppen kan tyda på en anrikning av ^{60}Co . Vid stationer närmast utsläppet kan i vissa fall förändringar av halter i utsläppet återspeglas i sedimenten. Vid stationer som är belägna längst bort, men i den dominerande strömriktningen från utsläppspunkten, detekteras sporadiskt radioaktiva ämnen som kan härledas till verken.

I fisk är ^{137}Cs vanligt förekommande vilket väsentligen beror på Tjernobylnedfallet. ^{60}Co och ^{65}Zn detekteras sporadiskt. Koncentrationen av radioaktiva ämnen tenderar att vara högre och mer vanligt förekommande i arter som är bottenlevande och, som t.ex. rötsimpa, än icke bottenlevande fisk (Figur 2).

I landmiljön detekteras få radioaktiva ämnen och koncentrationerna ligger nära detektionsgränsen. Detta beror på att en stor del av utsläppen utgörs av ädelgaser som inte deponeras på marken. Den radionuklid som detekteras i alla provslag är ^{137}Cs och i många omgivningsprov kan även ^{60}Co sporadiskt detekteras. Framför allt i mossor, men även i lav, återfinns flest antal och de högsta halterna av radioaktiva ämnen. I rådjur och älg är koncentrationerna ofta högre än i nöt och får.

Diskussion

Miljöövervakningsprogrammet omfattar ett relativt stort antal olika provslag. Utvärdering av data har visat att i vattenmiljön har blåstång generellt högre halter av olika radioaktiva ämnen, med undantag för $^{110\text{m}}\text{Ag}$ och ^{152}Eu , än andra provslag. I vattenmiljön skulle man kunna överväga att ta bort vissa provslag som vi nu vet har ett lägre upptag av radioaktiva ämnen, för att i stället ta fler prover av t.ex. blåstång. Genom att ha fler än en station nära utsläppspunkten skulle möjligheten öka att detektera koncentrationsförändringar. Man skulle även kunna ha ytterligare stationer på längre avstånd ifrån utsläppspunkten i spridningsriktningen som skulle möjliggöra en tydligare avgränsning av spridningsområdet.

I många provslag från landmiljön, men också för t.ex. torsk och sik, finns det inga detekterbara halter av radioaktiva ämnen som kan härledas till utsläpp från anläggningarna. Det har ibland ifrågasatts om man skall fortsätta att använda provslag i landmiljön som år efter år inte visar några detekterbara halter. Erfarenheterna efter Tjernobylyolyckan har dock visat betydelsen av att i en beredskapssituation känna till koncentrationen i olika provslag för att kunna påvisa vilka områden som är påverkade och hur lång tid det tar innan halterna återgår till ursprungsnivån. Det är också av stort allmänintresse att veta om detekterbara koncentrationer förekommer i framför allt livsmedel men också i miljön. Ett resultat som visar att koncentrationen av olika radioaktiva ämnen ligger under detektionsgränsen är naturligtvis det mest önskvärda både för människan och för miljön.

Erkännanden

Tack till Carl-Magnus Larsson, Maria Lüning, Lena Wallberg och Helene Wijk för att ni har bidragit med diskussioner och värdefulla kommentarer på manuskriptet. Tack till Göran Lund, Barsebäck, Rose-Marie Svensson, Forsmark, och Jan Andersson, Oskarshamn, för mer detaljerad information om provtagningsförhållandena vid verken.

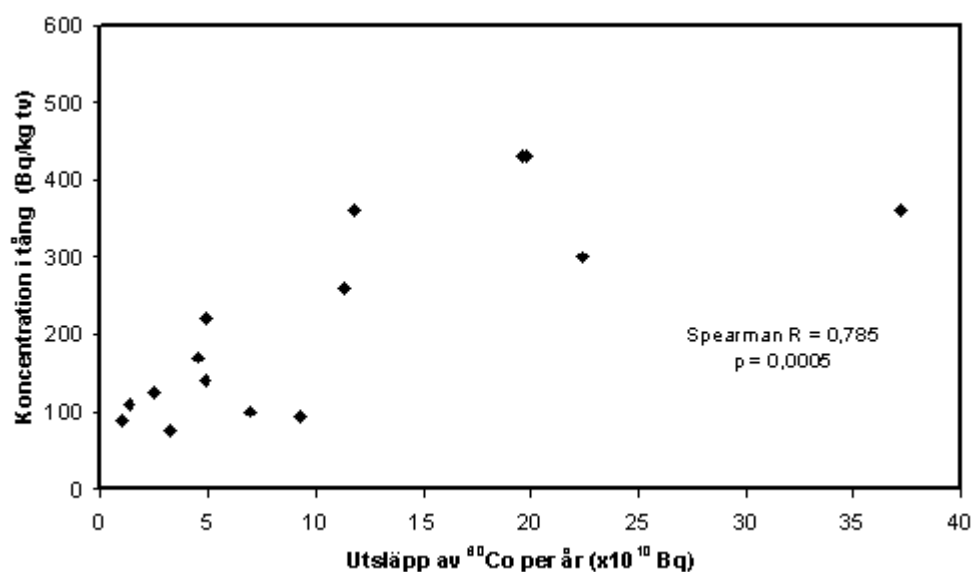
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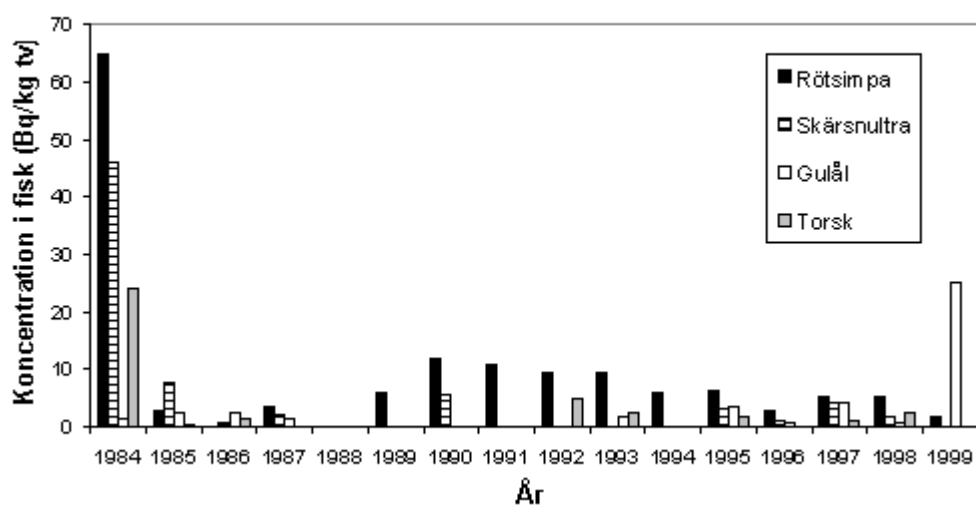
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Tabell 1. Arter som ingår i miljöövervakningen

	Ringhals	Barsebäck	Oskarshamn	Studsvik	Forsmark
Vattenmiljön					
Gulål	X	X	X		X
Abborre			X	X	X
Skärsnulta	X				
Gädda			X	X	X
Skarpsill/strömming		X	X	X	X
Sik					X
Torsk	X	X	X		X
Skrubbskädda		X		X	
Rötsimpa	X	X			
Krabbtaska	X				
Hummer	X				
Blåmussla	X	X	X	X	X
Östersjömussla			X	X	X
<i>Radix/Theoduxus/Littorina</i>	X		X		X
<i>a</i>					
Blåstång	X	X	X	X	X
Grönslick/Tarmtång	X	X	X	X	X
Sediment	X	X	X	X	X
Landmiljön					
Mossa	X	X	X	X	X
Lav	X		X	X	X
Ormbunke	X	X	X		X
Strandgräs	X	X	X		X
Sallad	X	X	X	X	X
Betesvall	X	X	X	X	X
Spannmål	X	X	X	X	X
Älg/Hjort/Rådjur	X	X	X	X	X
Får				X	X
Nötboskap	X	X	X	X	X
Fasan		X			
Mjölk	X	X	X	X	X
Rötslam	X	X	X	X	X



Figur 1. Sambandet mellan utsläpp av ^{60}Co och koncentrationen i blåstång nära utflödet från Biotesjön, Forsmark. Prover insamlade åren 1983-1999.



Figur 2. Koncentrationen av ^{60}Co i olika fiskarter fångade utanför vattenutsläppet från Ringhals under åren 1984-1999.

Radioecology in the Arctic: Activities of the new environmental protection unit of the NRPA in Tromsø

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Introduction

Radioactivity in the Arctic is a central topic within environmental pollution issues (AMAP 1998, Strand 2001). Sources, both actual and potential, for radioactive pollution in the Arctic are marine and atmospheric long-range transported radionuclides, originating from nuclear power and reprocessing plants, dumped radioactive waste, accidents with nuclear powered/ armed ships and submarines and nuclear test explosions (for an overview on sources see e.g. Aarkrog, 1993).

In August 1999, the Norwegian Radiation Protection Authority (Statens strålevern) established an Environmental Protection Unit in the Polar Environmental Centre (Fig. 1) in Tromsø, northern Norway. Major tasks of the three persons at the unit include monitoring and research in both marine and terrestrial environments in northern Eurasian areas and the Arctic. This includes field and laboratory work, collaboration with the other institutions within the Polar Environmental Centre (e.g. the Norwegian Polar Institute), and integrating studies within the AMAP programme (AMAP, 1998). The Environmental Protection Unit was recently involved in fieldwork in Kongsfjorden (Svalbard), an international cruise to the Fram Strait, a technetium monitoring program in the Western Barents Sea, and radiation measurements on the site in the Barents Sea where the Russian submarine “Kursk” sunk (see Amundsen & Lind, 2001).



Figure 1. Polar Environmental Centre and Polaria in Tromsø, Northern Norway (from <http://www.polarenvironment.no/>).

Kongsfjorden, Svalbard

Western Svalbard is characterised by its high Arctic environment with a relatively mild climate, arising from the nearby passage of the West Spitsbergen Current, a branch of the North Atlantic Current. The determination of radionuclide levels, their temporal and spatial variations and investigations concerning Arctic-specific processes provides insight into radionuclide behaviour and long-range transport of contaminants to Svalbard. At Ny-Ålesund/Kongsfjorden (Fig. 2),

fieldwork was performed in May 2000. In the fjord, surface seawater (^{137}Cs , Pu/Am isotopes, ^{99}Tc ; Fig. 3), sea ice (^{137}Cs , total γ -analysis), different types of seaweed (^{99}Tc), and seafloor sediments (total γ -analysis, Pu/Am isotopes) were sampled, whereas on land, plants and soil samples (total γ -analysis) were taken. Radionuclide measurements are currently on the way.

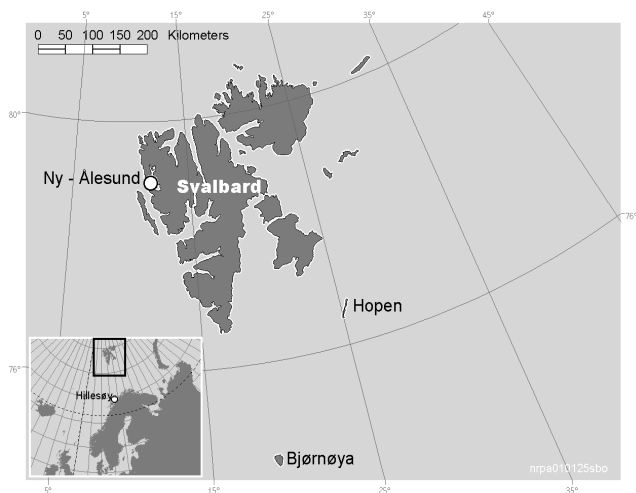


Figure 2. Map of Svalbard with Ny-Ålesund (Kongsfjorden), and the islands of Hopen and Bjørnøya. In the inset the location of the sampling station Hillesøy at the Norwegian coast is marked.

Investigation of ^{99}Tc concentration factors on seaweed showed large variations depending on seaweed types. In the mid-90s, a dramatic increase in ^{99}Tc discharges from Sellafield began, the effect of which was subsequently registered at distant locations in the Norwegian Sea where elevated environmental concentrations of ^{99}Tc could be measured (Brown et al., 1999; Kolstad & Rudjord, 2000). It is of interest to speculate as to the spatial extent of contamination arising from the releases and how the levels have changed over time. Preliminary results of our samples from May 2000 indicate similar levels of ^{99}Tc in seawater to those measured in the western Barents Sea in 1999 (Kolstad & Rudjord, 2000). Measurements on seawater west of Svalbard in 1995, undertaken before the increase of ^{99}Tc releases from the mid 90s could have reached the Arctic, show values about four times lower (Kershaw et al., 1999). However, before concluding from this evidence that a connection with the newer releases at Sellafield exists, we want to study ^{99}Tc levels and changes with time in the Svalbard area more thoroughly.



Figure 3. Surface water sampling from a rubber boat for ^{137}Cs measurements in Kongsfjorden in May 2000. At the same location, sea ice was sampled and melted for corresponding analyses (Photo: S. Gerland).

Fram Strait

Most of the sea ice which forms on the shelf and estuary areas off Siberia and Alaska leaves the Arctic Ocean through the Fram Strait, the area between the Arctic Ocean and Northern Atlantic in the north and south, and Greenland and Svalbard in the west and east. With participation of two researchers of the NRPA, water, ice, sediments and biota were sampled for radionuclide measurements along a transect at 79° N on board of the RV “Polarstern” in August 2000 (Gerland & Grøttheim, in press). By following this transect, water profiles in the East Greenland Current, coming from the north, and West Spitsbergen Current, coming from the south, were obtained. In addition, sediments from “dirty” multi-year sea ice were recovered (Fig. 4). Results from measurements on these long-distance transported sediments are going to be used also within numerical modelling work on ice transport by Iosjpe (2001).



Figure 4. Sampling of “dirty ice” with helicopter on a multi-year sea ice floe in the Fram Strait during the RV “Polarstern” cruise in August 2000 (Photo: S. Gerland).

Technetium-99 monitoring

Technetium-99 is an artificial radionuclide that was and still is released into the sea at European nuclear reprocessing plants (e.g. Sellafield, U.K.). A monitoring program for establishing time series of ⁹⁹Tc concentrations in the Barents Sea and Svalbard areas was started in October 2000 with continuing monthly sampling of sea water at the islands of Bjørnøya and Hopen (Fig. 2). In addition to monthly measurements from Hillesøy at the Norwegian coast (Kolstad & Rudjord, 2000) and occasional sampling at other coastal sites in Norway, these two sampling locations should give information on the spreading of radionuclides from European nuclear reprocessing plants to the Arctic. The measurements on the first samples from this initiative are currently on the way. For March 2001 we plan to extend the network further including also Ny-Ålesund at Kongsfjorden and the island of Jan Mayen east of Greenland to the set of stations with monthly sampling.

Sunken submarine “Kursk”

On 12 August 2000, the Russian nuclear powered submarine Kursk sunk north of the Kola Peninsula in the Barents Sea. The Environmental Protection Unit participated actively in monitoring of the environment and performed radiation protection for the divers during work inside and close to the submarine’s hull. On-site measurements on environmental samples and debris were performed using NaI and Hp-Ge detectors as well as dose rate meters. Collected samples (seawater and sediments) were brought to the NRPA laboratory at Østerås for more accurate analyses. No

increased levels above normal were registered. Details on this activity and results are given by Amundsen & Lind (2001).

Outlook

Aims for 2001 include installation of laboratory facilities with HP-Ge detector and technetium β -radiation detector equipment, extension of collaboration within the Polar Environmental Centre (active participation in its "centre programs") and external institutions, as well as follow up of field activities in northern areas. Time series from the mentioned monitoring sites should give indications on sources of radionuclides. Another aim will be to give contributions regarding the question of how arctic climate is affecting transport and effects in radioecology compared to known processes in milder conditions.

Acknowledgements

I. Amundsen and S. Grøttheim (both NRPA) participated in fieldwork presented here. We are further grateful to A.K. Kolstad and A.-L. Brungot (both NRPA), who did substantial laboratory work on samples in the context of this contribution. S. Borghuis (NRPA) provided the map (Fig. 2). J. Brown (NRPA) helped with discussions and by reading a draft of this contribution. Personnel of the Norwegian Meteorological Institute (DNMI) in Tromsø and on Bjørnøya and Hopen are helping with monthly sampling at these islands. The macroalgae group of C. Wiencke (Alfred Wegener Institute (AWI) for Polar and Marine Research, Bremerhaven, Germany) sampled seaweed while diving. H. Nies (Federal Maritime and Hydrographic Agency of Germany, Hamburg), U. Schauer (AWI) and J-G. Winther (Norwegian Polar Institute, Tromsø), loaned equipment to us and supported us in the field.

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Box modelling approach for evaluation of influence of ice transport of radionuclides for doses to man

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Introduction

Modelling of the ice transport of radionuclides, which is a unique pathway in the Arctic ocean and adjacent sea areas (Pfirman *et al.*, 1995; Strand *et al.*, 1996; Pfirman *et al.*, 1997), is limited by necessity to describe complete processes of incorporation of radioactivity into ice and ice sediment. These potential pathways for incorporation of radioactivity into ice are widely discussed in AMAP (1998).

Freezing / melting processes and transport of “clean” ice can be described with a good accuracy for relatively short time scale on the basis of the present level of modelling, but detailed description of the sediment entrainment into ice based on the Reynolds equations with attention to coagulation processes (Eidsvik, 1998) is limited by low concentration of particles (grease ice cannot be described) and time scale up to $5 \cdot 10^{-2}$ s ($1 \cdot 10^{-9}$ y) what is not available for large time scale and ice masses. Adding the radioactivity incorporation into the ice with following description of transport and fate of radionuclides will lead to further increasing of the complexity of the modelling.

Therefore, it is necessary to develop an alternative approach for purposes of radiological assessment on the basis of the box modelling (Iospje & Strand, 1998) to describe the incorporation of radioactivity into ice and ice sediment, transport of radioactivity by ice and incorporation of radioactivity into sea areas through melting processes.

Ice module parameters

Present development of the ice module is based on the following approach for incorporation of additional parameters: (i) to use parameters everywhere where possible, which are already involved in the box marine model and (ii) to use parameters, which are available for experimental measurements or model/expert evaluation.

This approach enables to develop the ice module without over-complexity of the model and therefore without the over-increase of uncertainty of the results of the calculations. The other achievement is a potential possibility for sensitivity and uncertainty analysis of model output.

Following additional parameters are included into the ice module:

$V_i^{(I)}$	is the ice volume of the ice box i [m^3],
$R^{(iw)}$	is the ice-water transfer factor [1], corresponding to the fraction of radioactivity, which is transferred from the liquid phase of the sea water box to the ice box during the freezing process,
$SL_i^{(I)}$	is the total ice sediment load for the ice box i [$\text{t} \cdot \text{m}^{-3}$],
$\varphi_i = \frac{SL_{ij}^{(I)}}{SL_i^{(I)}}$	is a fraction of ice sediment load $SL_{ij}^{(I)}$ from water/sediment compartment j in the total ice sediment load $SL_i^{(I)}$ in the box i ($\sum_i \varphi_i = 1$) [1],
$f_i^{(f)}$	is a freezing rate for the ice box i [$\text{m}^3 \cdot \text{y}^{-1}$],
$f_i^{(m)}$	is a melting rate for the ice box i [$\text{m}^3 \cdot \text{y}^{-1}$],
$t_{ij}^{(r)}$	is the ice flux from the ice box i to the ice box j [$\text{m}^3 \cdot \text{y}^{-1}$]

Expressions for transfer of radioactivity

With this set of parameters the transfer of radioactivity A_i from the liquid phase of the water column of the water box i with sediment distribution coefficient K_d suspended sediment load SSL_i to the ice box can be described as

$$A_i \frac{R^{(iw)} \cdot f_i^{(f)}}{1 + K_d \cdot SSL_i}, \quad (1)$$

The expression (4) is written on the assumption that transfer of radioactivity varies directly as freezing process.

The transfer of radioactivity from the suspended sediment in the water column to the ice box can be described as

$$A_i \frac{K_d \cdot SL_i^{(I)}}{1 + K_d \cdot SSL_i} \cdot \varphi_i^{(ss)} \cdot f_i^{(f)}, \quad (2)$$

where $\varphi_i^{(ss)}$ is a fraction of suspended sediment in water column of the water box i in sediment of the ice box. In a similar manner as expression (1), it is assumed here that transfer of radioactivity varies directly as freezing process.

The transfer of radioactivity from the sediment box i to the ice box can be described as

$$A_i^{(s)} \frac{K_d \cdot SL_i^{(I)}}{\omega + (1 - \omega) \cdot \rho_s K_d} \cdot \varphi_i^{(s)} \cdot f_i^{(f)}, \quad (3)$$

where $A_i^{(s)}$ is activity (Bq) in the sediment box i , $\varphi_i^{(s)}$ is a fraction of sediment from the sediment box i in sediment of the ice box, ω is the porosity and ρ_s is a sediment density. It is necessary to note that parameters ω and ρ_s correspond to the box model. As previously, it is assumed here that transfer of radioactivity varies directly as freezing process.

The transfer of radioactivity from the ice box i to the ice box j can be described as

$$A_i^{(I)} \cdot t_{ij}^{(r)} \quad (4)$$

where $A_i^{(I)}$ is radioactivity in the ice box i .

The transfer of radioactivity through melting process from the ice box i to the box j of the marine box model, which underlies the ice box i can be described as

$$A_i^{(I)} \cdot (f_j^{(m)} - \sum_k t_{ik}^{(r)}). \quad (5)$$

It is assumed here that all radioactivity in the ice box i , which will not transfer to other ice boxes $\{k\}$ will be transferred to the box j of the marine model.

It should be recorded that parameters $f_i^{(f)}$, $f_i^{(m)}$ and $t_{ij}^{(r)}$ must satisfy the expression

$$f_i^{(f)} - f_i^{(m)} - \sum_k t_{ik}^{(r)} = 0 \quad (6)$$

for each ice box i .

Results and discussion

Potential significance of ice transport for dispersion of radionuclides in oceanic space and some additional effects will be illustrated by the example of the transport of radionuclides from the Kara Sea to the Fram Strait through the Arctic Ocean after releases of radionuclides into the Obkaya Guba.

Figures 1 and 2 correspond to incorporation of radionuclides in the Kara Sea ice box, transport through the Arctic Ocean and melting in the Fram Strait. Sea water concentrations for different radionuclides in Figure 2 indicates a significant influence of the sediment distribution coefficient K_d on results of calculations and high K_d values lead to high concentration in the sea water during the ice melting phase.

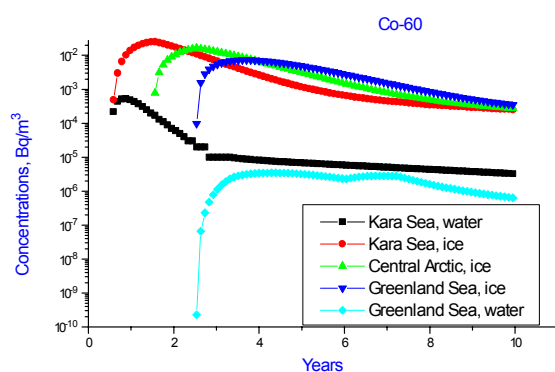


Figure 1. Comparison between concentrations of ^{60}Co in ice and sea water.

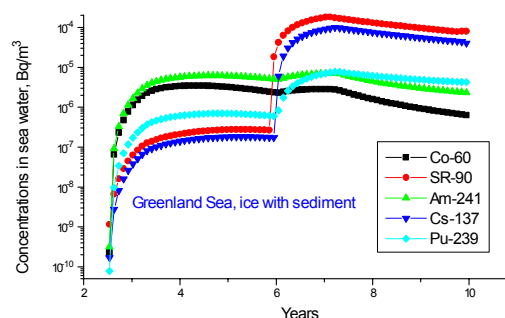


Figure 2. Influence of incorporation of radioactivity into ice sediment for the Greenland Sea.

The effect of radionuclides redistributing by ice transport is illustrated in Table 1, where it is compared dose impact of the Greenland Sea. Calculations correspond to 1 TBq discharges of radionuclides into the Ob Bay.

The effect of redistributing of radionuclides by ice transport for doses to man corresponding to the whole of the model is shown in Table 2.

Table 1. The effect of the ice transport of radionuclides for the impact of the Greenland Sea to doses to man. Scenarios A and B correspond to the calculations of doses with and without the ice module ($D^{(A)}$ and $D^{(B)}$), respectively. The effect is calculated as ratio between doses ($D^{(A)}/D^{(B)}$) for 10 years.

Radionuclide	Time (y)	Scenario A (manSv)	Scenario B (manSv)	$D^{(A)}/D^{(B)}$
^{90}Sr	10	$2.80 \cdot 10^{-6}$	$2.78 \cdot 10^{-6}$	1.0
^{137}Cs	10	$1.71 \cdot 10^{-5}$	$1.69 \cdot 10^{-5}$	1.0
^{239}Pu	10	$4.64 \cdot 10^{-5}$	$3.03 \cdot 10^{-5}$	1.5
^{241}Am	10	$2.09 \cdot 10^{-4}$	$8.97 \cdot 10^{-6}$	32
^{60}Co	10	$8.91 \cdot 10^{-6}$	$2.80 \cdot 10^{-7}$	32

Table 2. The effect of the ice transport of radionuclides for the doses to man for the whole oceanic space. Scenarios A and B correspond to the calculations with and without the ice module, respectively. the effect is calculated as ratio between doses ($D_t^{(A)}/D_t^{(B)}$) for 10 and 1000 years.

Radionuclide	Time (y)	Scenario A (manSv)	Scenario B (manSv)	$D_t^{(A)}/D_t^{(B)}$
⁹⁰ Sr	10	$7.40 \cdot 10^{-5}$	$7.40 \cdot 10^{-5}$	1.0
	1000	$2.01 \cdot 10^{-4}$	$2.01 \cdot 10^{-4}$	1.0
¹³⁷ Cs	10	$4.76 \cdot 10^{-4}$	$4.76 \cdot 10^{-4}$	1.0
	1000	$1.41 \cdot 10^{-3}$	$1.41 \cdot 10^{-3}$	1.0
²³⁹ Pu	10	$6.63 \cdot 10^{-4}$	$6.34 \cdot 10^{-4}$	1.0
	1000	$2.30 \cdot 10^{-1}$	$2.28 \cdot 10^{-1}$	1.0
²⁴¹ Am	10	$4.66 \cdot 10^{-4}$	$6.45 \cdot 10^{-5}$	7.2
	1000	$8.14 \cdot 10^{-2}$	$5.84 \cdot 10^{-2}$	1.4
⁶⁰ Co	10	$1.47 \cdot 10^{-5}$	$4.27 \cdot 10^{-6}$	3.4
	1000	$2.22 \cdot 10^{-5}$	$7.97 \cdot 10^{-6}$	2.8

According to the results from Figures 1-2 and Tables 1-2, effect of the ice transport of radionuclides to the distribution of radioactivity differs widely and can significantly increase the concentration of radionuclides in sea water, as well as, doses distribution for the radionuclides with high K_d , with this, the dose impact from some sea areas can increase up to factor three.

In summary, it is shown that the ice transport of radionuclides can be a significant factor for some scenarios and radionuclides. The influence of the ice transport increases with increasing K_d values for radionuclides.

It is necessary to note that the content and structure of the sediment load in ice vary within wide limits, and therefore, sensitivity and uncertainty analysis can improve the possibility to represent model results satisfactorily.

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Datasheet based countermeasure evaluation for radioactively contaminated Nordic food-producing areas

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Abstract

A Nordic expert group has identified and critically evaluated the countermeasures that may potentially be implemented in connection with major nuclear accident situations contaminating Nordic food-producing areas. This paper demonstrates how the derived technical information can be applied by decision-makers to identify practicable and cost-effective means for mitigation of the impact of a contamination.

Introduction

In the event of a major nuclear accident leading to airborne contamination of large Nordic food-producing areas it is imperative in advance to have identified and described potentially applicable countermeasures in terms of benefits and constraints, so that an optimised strategy for handling the situation can be developed as early as possible. The important factors forming the basis for the evaluation of strategical options for remediation of a contaminated food-producing area include the potentially averted dose, local method practicability, availability of equipment and workers with required skills, monetary costs, possible waste problems and other technical limitations. In a handbook recently published by NKS (Andersson et al., 2000), these technical factors have been evaluated for a total of 37 countermeasures, which are considered to be relevant to the Nordic context. It is these technical factors and their application in differential cost benefit analysis as a basis for countermeasure strategy formation, which are highlighted in this paper.

Methods/background

All important technical data in relation to each of the 37 countermeasures that are considered potentially feasible and practicable in radioactively contaminated Nordic food-producing areas (including food processing industry and domestic households) have been presented in a single data sheet, facilitating intercomparison of method features. Only methods influencing ingestion doses are considered. Methods for reduction of external doses are described elsewhere (e.g., Andersson & Roed, 1999). However, where methods designed for reduction of consumption doses (e.g., ploughing procedures) also affect external doses this is mentioned in the data sheets. The data sheets are systematised in the handbook according to the time stage following an accident and the

food chain for which they are relevant, and guidance/ recommendations concerning their application has been provided. An extended internet-based version of the handbook is currently under development.

An example of a data sheet is given in Table 1: removal of contaminated vegetation shortly after contaminant deposition.

Table 1. Example of technical descriptions in a countermeasure data sheet (from Andersson et al., 2000).

Method	Early removal of vegetation
Description	Removal of growing crops from contaminated fields reduces ground contamination; on grass land also contamination of stubble and grass sward.
Target surface / product	Fields with rather dense vegetation, where the radionuclides are mainly retained in growing crops (dry deposited).
Time of application (number of days after deposition, season, etc.)	Removal of crops as soon as possible after fallout. Removal before the 1st rain shower after fallout is the optimal situation. Transfer of radionuclides from vegetation to soil has a half-life of about 15 days. Rainfall increases this transfer.
Expected effect: DF (decontamination factor), DRF (dose reduction factor) - internal DRF depends on diet	Reduction of external dose on contaminated fields and reduction of uptake of radionuclides in subsequently grown crops may be substantial (reduction by a factor of up to 20).
Personnel requirements and costs (method time consumption)	Typically: 1 operator, ca. 0.2-0.3 man-days ha ⁻¹ for crop removal. Additionally: Loading, transport and waste disposal.
Equipment and other remedies - costs	Normally harvest equipment is available on the farms. For arable crops: anything from forage and swath harvesters (ca. 10,000 to 20,000 EURO) to combines (ca. 200,000 EURO). For grass crops: tractors (ca. 50,000 EURO) with mowers (ca. 4,000 EURO). Petrol of the order of ca. 10 l ha ⁻¹ .
Practicability	Achievable in a large scale. The limit is the number of workers and equipment locally available over a short period of time. The possibility to remove fallout from fields is usually best on grassland.
Waste	Removed crop material may, depending on contamination level, be used (after storage and/or preparation) or disposed of in repository. Costs of transportation/storage of waste within 20 km : ca. 20 EURO m ⁻³ .
Benefits	Reduction of external dose on treated fields may be substantial. Lower root uptake of nuclides due to less contamination of soils in following years.
Constraints	Sites for waste disposal and safe storage.
Remarks	Respiratory protection of workers may in some cases be radiologically necessitated.
References	CEC report EUR 12554 EN, ISBN 92-826-3272-5, 1991. NKS report NKS(97)18, BER6, ISBN 87-7893-017-0, 1997.

Method evaluation example/results

In the following a simplified example is given to illustrate how the data sheet shown in Table 1 could be used in an emergency situation. Consider a situation where airborne dry contamination occurs to a potato field in the month of July, and the contamination level of the main contaminant, ^{137}Cs , has been measured to be 10 MBq m^{-2} .

The predominant soil type in the area is known to be sandy loam (ca. 10 % clay), and at this time of the year the potato plants cover the ground well, so it is estimated that more than 90 % of the contamination in the field occurs to the rough plant surface. The weather forecast says dry weather followed by heavy rain in 2-3 days. This means that a removal of the plants within few days would be expected to reduce the contamination level in the field by a factor of ca. 10. According to Eriksson (1997) the soil-to-potato transfer factor for this type of soil is about $1.4 \cdot 10^{-3} \text{ m}^2 \text{ kg}^{-1} \text{ (dw)}$. Assume that an average Nordic person consumes ca. 0.3 kg d^{-1} of potatoes with a dry matter content of 20 %. According to IAEA (1994) ordinary peeling and boiling will reduce the contamination level to ca. 60 %.

According to ICRP (1995), each Bq of ^{137}Cs consumed by an adult will result in a dose of $1.3 \cdot 10^{-8} \text{ Sv}$ (slightly less for children). This means that the averted dose to one consumer over the 1st season after deposition (assuming that a very large area has been contaminated and alternative food can not be imported in sufficient amounts) can be approximated as follows:

$$D = 10 \text{ MBq m}^{-2} * 90 \% \text{ (early harvest)} * 1.4 \cdot 10^{-3} \text{ m}^2 \text{ kg}^{-1} * 20 \% * 60 \% * 0.3 \text{ kg d}^{-1} * 365 \text{ d y}^{-1} * 1.3 \cdot 10^{-2} \text{ Sv MBq}^{-1} = 2.2 \text{ mSv y}^{-1}.$$

Taking into account the radiological half-life of ^{137}Cs and assuming that the transfer factor will decrease somewhat with time (if deposited particles are more or less readily soluble), a rough estimate of the individual averted life-time dose could be of the order of 50 mSv. The average annual yield from a potato field is in Denmark currently ca. 36 t ha^{-1} . Assuming a moderate waste fraction of the potatoes, it can be deduced that probably some 200 people can have their supply covered from a 1ha potato field. This means that early harvesting in 1 ha of potato field can result in an averted collective life-time dose of some 10 man-Sv.

A value of an averted man-Sv may, e.g., be derived from figures currently applied by Danish Ministries in connection with cost analyses of other types of accidents, for instance casualties in traffic. If such a monetary 'casualty' figure were multiplied by 0.05 Sv^{-1} (the current ICRP estimate of the probability of developing fatal, radiation-induced cancer), the result would be an estimated value of an averted man-Sv of about 40,000 EURO. This would make the dose averted by treatment of a ha worth some 400,000 EURO over a life-time.

From Table 1 it follows that the harvesting of leafy material from 1 ha can be accomplished in less than 3 hours by one operator with a harvester. With current salaries, the operator costs, including expenditures to have the equipment brought to the site and mounted, would on this background be estimated to about 150 EURO ha^{-1} .

Further, the petrol requirements would according to Table 1 be ca. 15 EURO, whereas the equipment discount corresponding to 3 hours use would amount to ca. 70 EURO. As indicated in Table 1, waste transport and storage costs would be estimated to some 20 EURO m^{-3} , if a simple repository can be constructed within 20 km distance (this may however not be feasible or legal in

the particular area). From one ha, there may be some 100 m³ of waste, corresponding to a cost of ca. 2,000 EURO. The total direct method costs are thus of the order of 2,300 EURO ha⁻¹. As mentioned in the NKS report, contaminated bio-mass may e.g. alternatively be applied as fuel for power plants, substantially reducing the amount of waste for disposal.

This means that on this background, the technique would be considered very cost-effective for the particular scenario, even without considering the reduction in external dose. However, it should be stressed that a number of other, often politically determined, factors should be considered by decision-makers prior to implementation of countermeasures. These include psychology, public acceptability, social implications, communication routes and ethical considerations.

Conclusion

A series of countermeasures for handling emergency situations involving nuclear contamination of Nordic food-producing areas have been investigated and described in a standardised datasheet format. An example is given of how the information from a datasheet may be applied by decision-makers to identify practicable and cost-effective countermeasure strategies. The example highlights how technical aspects may be considered, particularly in relation to direct method cost-effectiveness evaluation, and stresses the need for incorporation in strategy formation of other, often politically governed considerations.

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Samples of 24-hour-meals in monitoring of dietary intake

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Introduction

The foodstuff surveillance has been an integral part of studying environmental radioactivity in Finland for about four decades. Of particular interest have been collective ingestion doses from the long-lived fallout nuclides ^{90}Sr and ^{137}Cs . The frequency and volume of nation-wide sampling have been adjusted according to the temporal changes in activity concentrations in foodstuffs. The nation-wide foodstuff programme has also been a framework for studies related to dietary assessment and radioecological conditions in domestic agricultural and wild food chains.

The parameters needed in assessment of ingestion doses from foodstuff data represent the whole chain from origin of agricultural or wild food products to edible fractions of food, and consumption rates nation-wide and by regions and groups of population. Reliability of all this data influences precision and accuracy of dose estimates obtained. For better consideration of also other than radioactivity data for instance food processing, using domestic methods and food types, has been investigated (Rantavaara 1990). Consumption of wild foodstuffs was studied for finding also regional consumption patterns and the species of mushrooms, berries, fishes and game animals with significant contribution to dietary intake in Finland (Markkula and Rantavaara).

Since the late 1990's structural changes in agriculture and in foodstuff logistics have complicated the search of reliable data for annual assessment of ingestion doses from activities in regionally collected foodstuff samples. The surveillance of activity contents of relevant foodstuff samples and collection of a variety of background information needs much human resources. Another approach based on sampling food portions consumed daily, a survey of 24-hour-meals, was tested in 1996. The long-term sampling and analysis of foodstuffs continued alongside the survey and offered possibility for comparison of results. The survey gave dietary intakes measured directly from daily meal samples, and information on activity contents through foodstuff surveillance, on production, processing and consumption of foodstuffs gave independent intake estimates.

The objective of our study was to analyse the usefulness of 24-hour meal samples in monitoring of ingestion doses. Our aim was also to learn more about the catered food services from radiological assessments' point of view. Long-lived environmental contaminants ^{137}Cs and ^{90}Sr were analysed. The relative annual changes in activity concentrations of foodstuffs were very small during our study, ten years after the Chernobyl accident.

Material and methods

Samples

Intakes of ^{137}Cs and ^{90}Sr through 24-hour meals were surveyed in co-operation with a catering service company. Meal samples were collected daily in an institutional kitchen using catering service of the city of Helsinki. The sampling period of six weeks covered the full circle of varying recipes in October - November 1996. The collected daily meals represented one person's daily food and drink, which was adjusted to contain 8.4 MJ (2000 kcal) energy. The normal energy supplement was bread. The amount of bread varied between two and nine pieces per day. Milk, juice, coffee and tea were included in the drink samples. The foods and drinks were collected daily in separate containers. The caterer also provided recipes of the meals for comparative assessments. Salad (100 g) containing season's vegetables was served daily with lunch, but the recipe was not recorded.

Sample analysis

All the food portions except for drinks were bulked to form daily samples. The samples were dried (105 °C), homogenised and ashed (400 °C). After measurement of the sample volume the daily drink samples were evaporated under infra-red thermal lamps, combined to represent weekly samples, and ashed.

Activity concentrations of ^{137}Cs and ^{40}K in sample ash were determined using cylindrical sample geometry and low-background gamma spectrometers, equipped with high purity germanium (HPGe) detectors. The software and measurement instructions were developed for environmental samples (Klemola and Leppänen). The measurement uncertainty for ^{137}Cs varied between 3 and 12%, and was mostly <7%. For ^{40}K the uncertainty was 3%.

For ^{90}Sr determination the ashes of drink samples were combined in two mixtures representing three weeks each. The food samples were combined according to similar level of activity contents of ^{137}Cs in five bulked samples. ^{90}Sr was determined on drink and food samples with a method based on chemical separation of strontium and calcium with nitrate precipitation (Bryant et al.), added by analyses of strontium and calcium concentration with atomic absorption spectrometry. Beta activities were measured with a low-background anti-coincidence counter. Measurement uncertainty for ^{90}Sr was 4 – 5% and the total uncertainty of the determination < 10%.

Analysis of data

The measured activity concentrations resulting from food and drink sample analysis were converted to daily intakes in Bq d^{-1} . For comparison, intakes of ^{90}Sr and ^{137}Cs were estimated with a calculation method developed earlier for nation-wide foodstuff assessments (Rantavaara 1991).

Results

The quantities of daily food samples show a systematic carrying out of sampling (Figure 1), and the data for contents of potassium in daily meals confirm the same (Figure 2). The observed daily intakes of ^{137}Cs in food differed manifold only on such days when wild berries were used in some of the meals. This finding was based on comparison of ^{137}Cs activities and recipe cards. Wild berries were the only wild food item used by the caterer. Agricultural produce caused the rest of the intake through food, being a rather constant low activity contributor to ^{137}Cs intake (Figure 2).

Contents of ^{90}Sr in the daily food and drink samples gave a total intake of 0.15 Bq d^{-1} on average. The drinks contributed one fifth to the intake. Intake of ^{90}Sr showed very slight or no association with the radiocaesium activity concentration in bulked samples. The finding is consistent with a low intake estimate for ^{90}Sr through all wild foodstuffs, a group, which largely explains the variation in intake of the ^{137}Cs (Table 1).

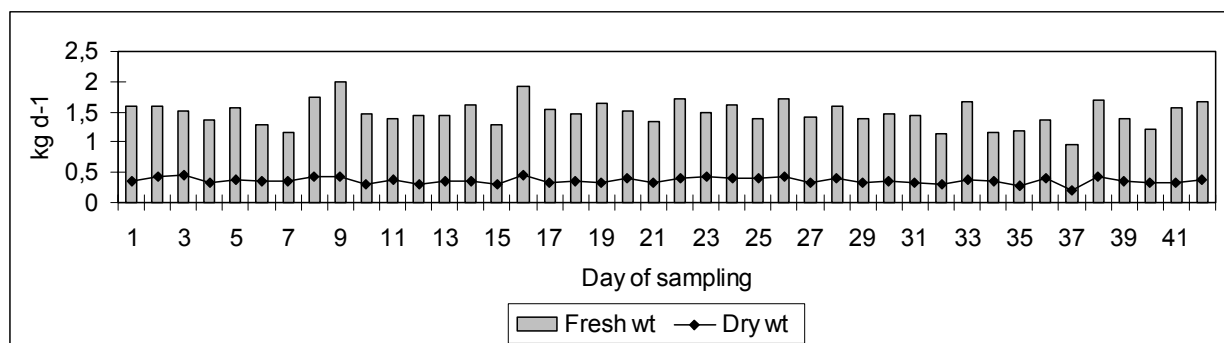


Figure 1. Daily food intake in the survey of 24-hour meals. Drinks, including milk, added roughly one litre per day.

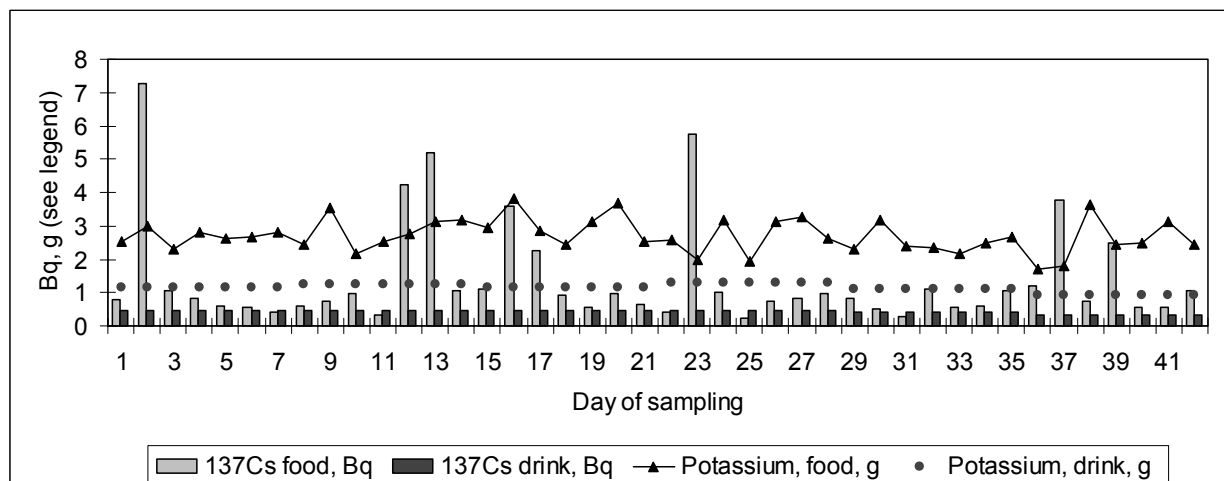


Figure 2. Daily intake of ^{137}Cs and potassium through food and drink.

Discussion

Comparison of survey results with intakes from foodstuff surveillance

A comparative assessment of dietary radiocaesium and -strontium was made using radioactivity data from nation-wide foodstuff surveillance in 1996, consumption rates from food balance sheets for 1996 (Yearbook of Farm Statistics 1997) and processing loss corrections analysed earlier for Finnish cooking methods (Rantavaara 1990).

The contributions of different food types to dietary ^{137}Cs exceed those of ^{90}Sr manifold in all groups of foods (Figure 3). Consumption, intake of ^{90}Sr and intake of ^{137}Cs fell within the range of two to three decades each, when the main types of foods were grouped for comparison.

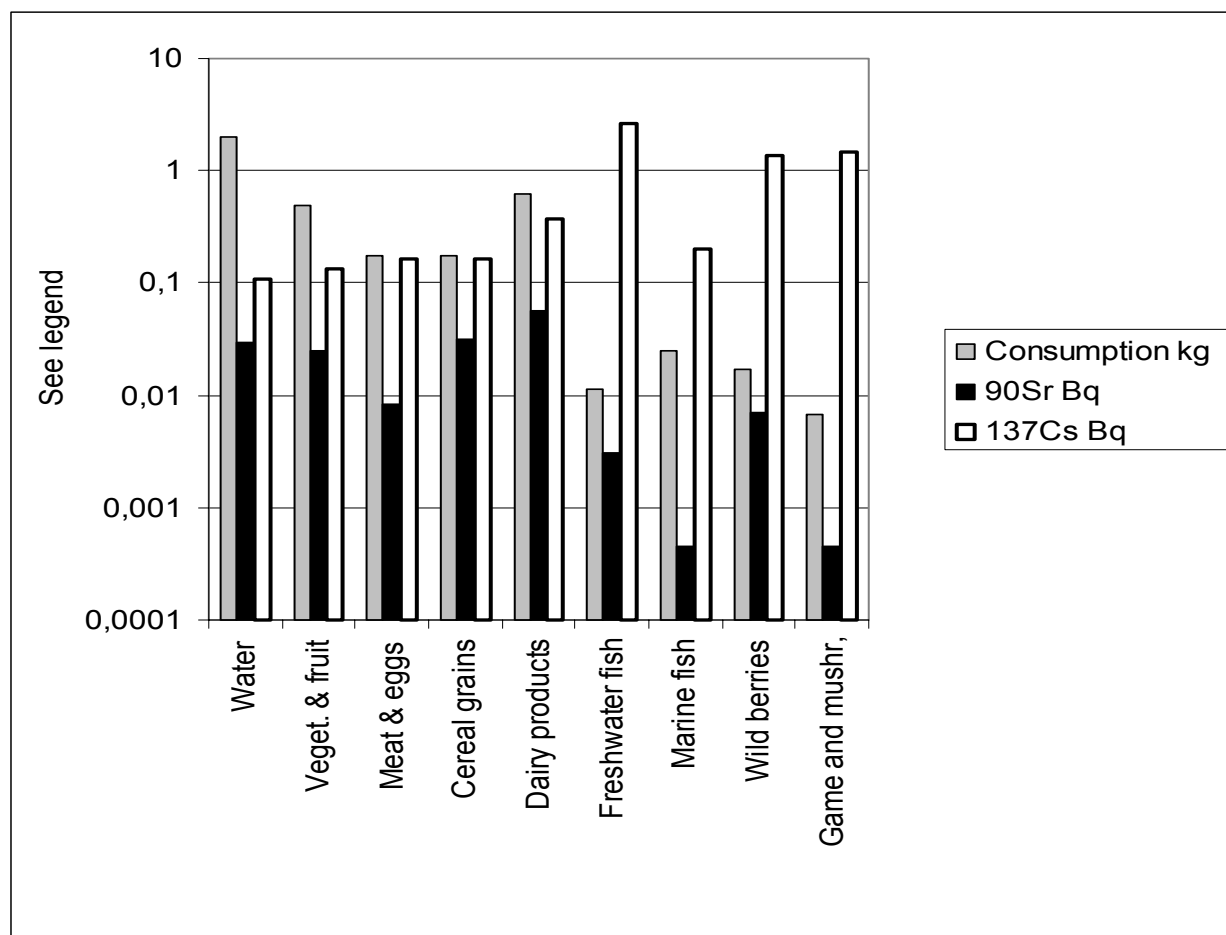


Figure 3. Daily intake of ^{137}Cs and ^{90}Sr through average diet of Finns. Activity concentrations of foodstuffs were from the nation-wide surveillance programme of STUK.

For practical reasons the 24-hour-meal samples were measured in two parts, foods and drinks. Rather distinct baseline of ^{137}Cs activity in food samples, the assumed non-wild food component (Figure 2), suggested defining the excess of activity in daily samples tracing from wild berries on such days when they were a constituent of the diet. Those days coincided well with observations. Thereby a rough estimate for the average share of berries to daily intake was derived. This figure

was only 50% of the nation-wide intake estimate through wild berries (Table 1). The quantities of wild berries in catered meals were lower than nation-wide mean consumption, and their ^{137}Cs contents varied referring to origin in regions of widely differing ^{137}Cs fallout.

Estimated intakes shown in Figure 3 were partially regrouped in Table 1 to show how well the measured intakes and surveillance-based estimates agree. For ^{90}Sr the bulked samples limited comparison. Naturally there are many possible sources of differences in a rather complex assessment, not least the uncertainties of both the survey results and the use of data for estimation. The quantities of foodstuffs in daily diets did not fully correspond to consumption rates in the food balance sheets for 1996.

Table 1. Comparison of measured daily intakes of ^{90}Sr and ^{137}Cs (in Bq d^{-1}) with independent estimates based on surveillance of foodstuffs. For measured intakes through daily food and drink samples only a limited number of diet fractions were possible to quantify.

Group of foods	Measured ^{90}Sr	Estimated ^{90}Sr	Measured ^{137}Cs	Estimated ^{137}Cs
1. Non-wild foods (no drinks)	–	0.09	0.7 ^a	0.5
2. Milk and other drinks	0.3	0.07	0.4	0.5
3. Wild berries	–	0.007	0.7 ^a	1.4
4. All wild products (incl. berries)	–	0.01	–	5.6
5. Non-wild foods and drinks (1+2)	–	0.16	1.2	0.9
6. Non-wild food and wild berries (1+3)	0.2	0.10	1.4	1.9
7. Non-wild foods, drinks and wild berries (1+2+3)	0.15	0.016	1.8	2.3
8. Whole diet (1+2+4)	–	0.017	–	6.6

^a) Derived from data series for surveyed foods as explained in the text.

Usefulness of the survey method

Our survey comprised of a variety of recipes of meals including foodstuffs of different origin, and revealed the importance of separating foods of agricultural and wild origin. The activity contents of long-lived ^{90}Sr and ^{137}Cs in wild food products follow different dynamics with time, by food types and environmental conditions, thereby referring to the necessity for different kinds of surveys.

There were practical reasons for missing of some wild foods from sampled meals. Freshwater fish has been considered too laborious to handle in routine catering in general. Wild mushrooms have been available only intermittently, and were therefore not included in the meals. Domestic wild berries were used in cooking, but their proportion during sampling period was below the mean consumption rate in Finland. Surveying wild foods is emphasised also for their increasing contribution to dietary intake of radiocaesium in the late phase of accidental fallout. If included in non-frequent sampling of daily meals they would increase random uncertainty of the results.

Observing also intake rates of food and minerals can confirm the correctness of sampling to some extent as was shown in Figures 1 and 2. Contents of calcium, strontium and potassium in daily food and drink may refer to the fraction of some food constituent rich in these elements in the sample. Differences in normally constant intakes of minerals may sometimes refer to exceptional composition of meals or inadequate or erroneous sampling. In drink samples both calcium and potassium content would relate with quantities of milk. The other drinks tea, coffee and juices are known to contain these minerals significantly less than milk.

Equal cooking methods improve comparability of meal samples from different locations. The food is of similar type as food cooked at home, as shown by the recipes used and comparative intake calculations based on foodstuffs. Therefore information on dietary intake can well be collected through daily meal samples also during unexpected contamination situations.

In early phase of accidental radioactive fallout surveying daily meals is useful for the follow-up of changes in contamination levels of actually consumed food. Such information is needed when the logistics of food distribution is highly unpredictable due to varying availability of acceptable foodstuffs by regions. The flux of foodstuffs through institutional kitchens is fast due to short storage times. The edible portion of food gives a realistic view on the activity levels of foodstuffs marketed commercially. Of course, daily meal samples cannot replace all other methods of food surveillance after a large scale environmental contamination.

In a late phase after radioactive fallout the 24-hour meal samples give relevant information on other than wild food contamination. Optimisation of surveillance resources is achieved through replacing several sample types of heterogeneous foodstuff groups with more representative samples of daily foods and drinks. Also in an acute fallout situation the 24-hour meal samples have the same advantages as during our study, but then also measurements of individual temporarily significant foodstuffs of agricultural origin have to be analysed for control of food safety.

Practical viewpoints

For production of representative intake data with surveillance of 24 h meal samples it is advisable to

- Use the survey for non-wild foods only.
- Use the recipe information in planning of sampling to achieve representative composition of foods and drinks.
- Record the courses included in daily samples
- Assure the possibility to call for clarification of sample information immediately if obvious deviation from known sampling design is found in the laboratory.
- Inform the persons in charge of sampling and analysis about the content and goals of the dietary survey.

Conclusions

The availability of 24 h meals for surveillance of collective ingestion doses is mostly an underused opportunity, and worth careful consideration. The analysis of daily meal samples is a fast and economic way of surveying food actually consumed. If correctly designed, the samples represent foodstuffs in realistic proportions. In fact, the catering practices today offer chances for an advanced sampling.

The method is independent on the availability of data for radioactivity of foodstuffs, human consumption data, correction factors for processing losses, and the delays in distribution of foodstuffs. It does not need consideration of import of foodstuffs. The accuracy of intake estimates is improved through reduction of the number of calculation parameters. The benefits of the method vary by radiation situation and the structure of food consumption, but often some parts of foodstuff surveillance can be replaced or complemented by analysis of daily meals. Relevant selection of sampling sites must be provided. In all, the surveying of daily meals can be an element in optimised use of laboratory resources.

With low sampling frequencies extremes of sample contamination (low or high) can hide a real trend. Therefore, in an acute fallout situation foodstuffs should be surveyed with a design arising from the actual demands of controlling food safety. Sampling locations should give a realistic view on regional differences. Also radioecological factors in production of foodstuffs and different consumption patterns are important. Of concern are also then such groups of population that are being exposed more than people on average. Wild foods, which have greatly variable, relatively high ^{137}Cs activities compared to agricultural produce, need special emphasis and separate surveys by food type..

The design of sampling and analysis of food is at best collaboration of experts on different parts of the dietary assessment. The catering service is a well-developed industry in Finland, and the popularity of warm dishes eaten outside home on weekdays (some 700 000 lunches per day) confirms its high standard. The analytical approach in planning catered meals today also gives chances for further development of surveying ingested radionuclides.

After carrying out this study STUK has included survey of daily meals from three locations in its regular surveillance of environmental radioactivity since 1999 (Mustonen 2000, 2001).

Acknowledgements

The plan and delivery of samples were designed and carried out in co-operation with the Helsinki Catering and Health department of Helsinki/Maria Hospital; the persons in charge were Airi Rintamäki and Regina Häihänen. At STUK the samples were received and analysed by Tuula Korttinen and Ulla-Maija Hanste. Ritva Saxén provided the activity concentrations in tap water, and Tarja Ikäheimonen the data for Baltic fish. The authors appreciate and kindly acknowledge the work and support for the study by all these persons.

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13 years of ^{137}Cs monitoring in meat and milk during the outdoor grazing period in Norway

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Introduction

After the Chernobyl accident, it was soon realised that the consequence of the radioactive fallout in Norway was more severe for animals grazing unimproved pastures than for other farm animals. High radiocaesium levels were registered in mutton, goat's milk and cow's milk. Due to this fact, monitoring of ^{137}Cs in meat and milk during the outdoor grazing period (June-September) was initiated for selected livestock. The research programme started in 1988 and is still ongoing. It includes monitoring of live sheep and lambs, and milk from goats and cows in regions moderately to heavily affected by the Chernobyl fallout. In years abundant in fungi, like 1988, 1991, 1997 and 2000, the ^{137}Cs levels in the autumn have been markedly higher for sheep and milk than expected from natural decay. Some of the time series from the summer monitoring is presented here.

Materials and methods

Long time series of ^{137}Cs contamination in meat and milk have been established by monitoring the same livestock every year. Monitoring of live sheep and lambs are performed every week in Tjøtta in Nordland county (until 1998) and every month in Baklia in Oppland county during the outdoor grazing period. A portable, battery operated Canberra Series 10 or 10+ MCA with a 3"x3" NaI detector is placed on the animals hind part for 1 minute. The detector is protected from environmental conditions in a cylindrical plastic shield, and is calibrated for live sheep monitoring in advance (Brynildsen et al., 1994).

Samples of goat's milk and cow's milk are taken every week from the same livestock during the grazing season. Some livestock have been monitored since 1988 while others have been monitored for shorter periods. Samples are measured every week in the laboratories of the local Food Control Authority units with the same type of instrument as described above, with the detector placed in a lead shielding.

Results and discussion

Goat's milk

The summer monitoring of goat's milk is performed in several areas of Norway. The monitoring has two aims: first to look at long time series, and secondly to be an early warning of high abundance of fungi in the pastures. In the summer of 2000 an increase in ^{137}Cs concentration was

registered in goat's milk from the beginning of August. An example from Ål in Hallingdal, where 50 goats graze unimproved pastures without access to Prussian blue¹, is presented in figure 1. It soon became clear that goat's milk in certain areas of Norway showed substantially higher values of ¹³⁷Cs than in the two previous years due to a high quantity of fungi. Such early warning can predict the extent of clean feeding needed for lambs prior to slaughtering later that same autumn.

A goat flock in Valdres showed a similar trend in the summer of 2000. The flock is divided in two: 10 of the goats have no access to AFCF while the rest of them are fed concentrate with AFCF when they are stalled in the evening. The results for the goats with no access to AFCF are presented in figure 2 for the years 1988 to 2000. Years abundant in fungi are highlighted, showing a large increase in the end of the grazing season (August-September). Figure 3 shows the comparison of ¹³⁷Cs activity concentrations in 2000 for the goats with and without access to concentrates with AFCF. The reduction in ¹³⁷Cs activity concentrations due to AFCF distribution ranged between 20% and 80% during the season, with a resulting goat's milk concentration below the ¹³⁷Cs intervention limit (370 Bq/kg) for all weeks except one.

Cow's milk

The summer monitoring also includes cow's milk. The ¹³⁷Cs levels are generally much lower than in goat's milk. Nevertheless, in the most contaminated parts of Norway the ¹³⁷Cs levels in cow's milk were even in year 2000 just above the intervention limit at the end of the grazing period, when no AFCF was applied. Even though fungi can contribute to higher values of ¹³⁷Cs in cow's milk, too, the influence is less significant than for goat's milk.

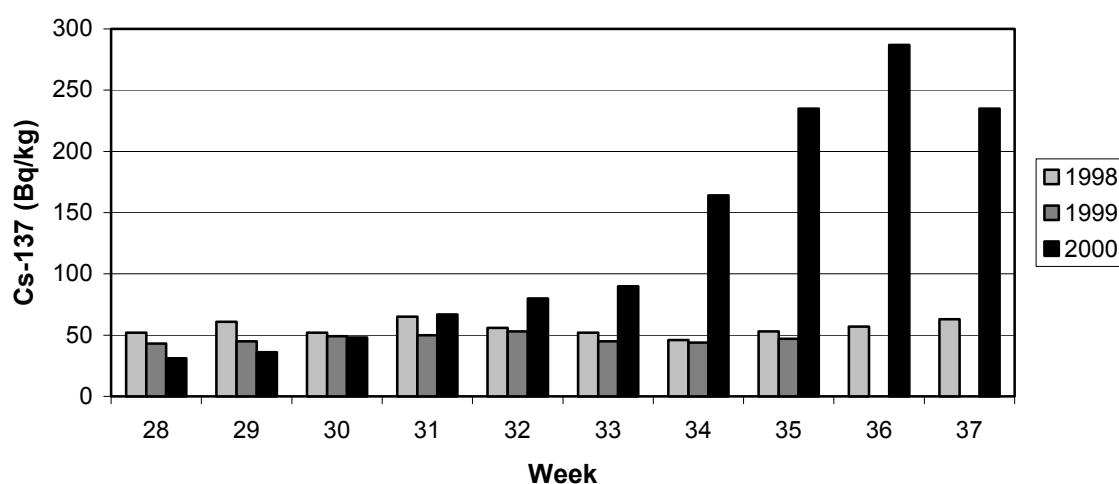


Figure 1. ¹³⁷Cs concentration in goat's milk in Ål for week 28-37 in 1998 to 2000.

¹ In Norway mainly in the form AFCF; ammoniumferricyanoferrate

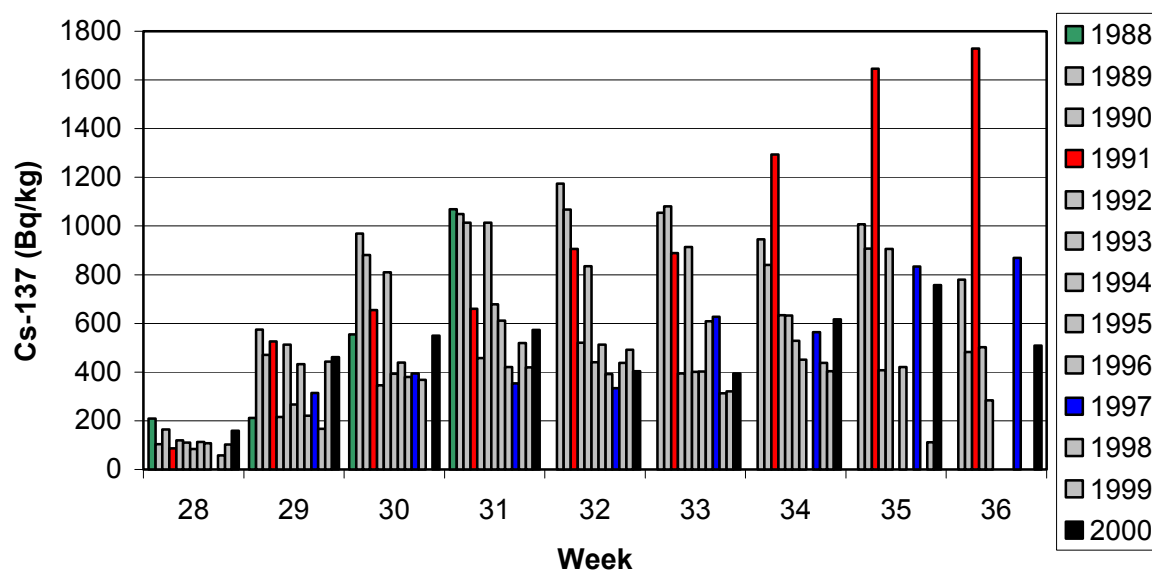


Figure 2. ^{137}Cs concentration in goat's milk in Valdres for week 28-36 in 1988 to 2000.

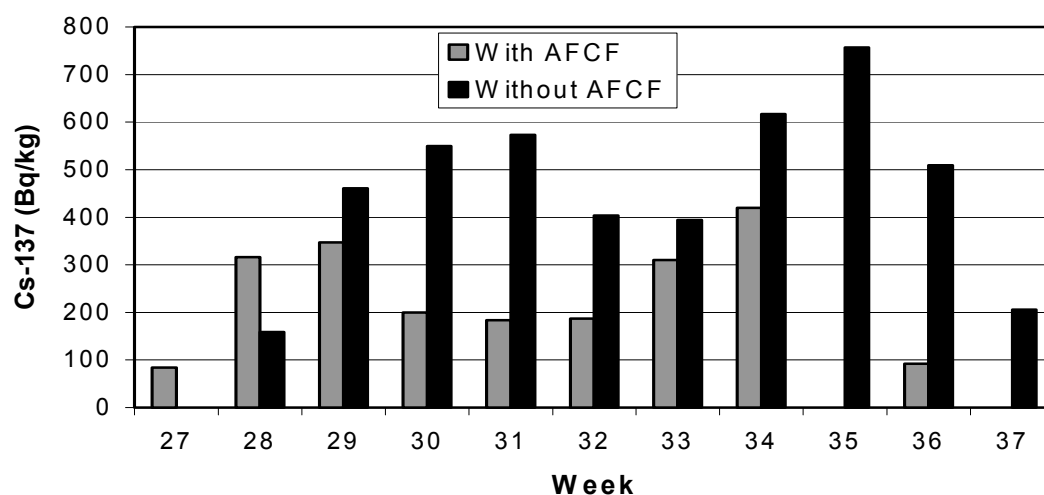


Figure 3: Comparison of milk from goats in Valdres with and without access to concentrates with AFCF during the grazing season 2000.

Live monitoring

Live monitoring of animals is performed every summer in some observation areas to predict the ^{137}Cs levels to be expected in sheep and lambs at the autumn slaughtering.

In Tjøtta (approx. deposition 35 kBq/m^2 of ^{137}Cs) 5 ewes and 10 lambs have been grazing the same area of 0.4 ha every year from 1988 to 1998. No countermeasures have been implemented. The average ^{137}Cs activity concentration after 8-12 weeks of outdoor grazing (presumed equilibrium state reached) is shown for lambs in figure 4. The results were particularly high in 1991, while 1995

had the lowest values in the given time period. An exponential decay has been fitted to the results, giving an effective ecological half-life of 10 years for lambs in the period 1988 to 1998. For ewes the calculations gave a half-life of 7 years for the same time period (as reported by Amundsen to the NKS BOK-2 status report).

In Baklia (approx. deposition 32 kBq/m² of ¹³⁷Cs) sheep and lambs are grazing in the mountains between Valdres and Hemsedal where they have access to saltlicks with AFCF. A total of 10-20 ewes and 20-30 lambs are measured in July, August and September every year. Figure 5 shows the average ¹³⁷Cs activity concentration in August and September from 1988 to 2000. The trend is very variable during the 13 year time period, so fitting an exponential decay to the data is not obvious. Depending on the time period chosen and on the month of measurement selected, an effective ecological half-life between 3 and 9 years is estimated.

The time trends in the two livestock are quite different. The results from Baklia show increased values in 1988, 1991, 1997 and 2000, corresponding to years abundant in fungi in that area. Fungi have very low occurrence in the grazing area at Tjøtta, and can thus not explain the time variation there. Fungi are known to show high ¹³⁷Cs concentrations and a very long ecological half-life (Amundsen et al., 1996). Calculating effective ecological half-lives in lambs is thus not straightforward for areas where fungi are present on the pastures. The variation from year to year is more dependent on the abundance of fungi than on other factors (Mehli et al., 1996).

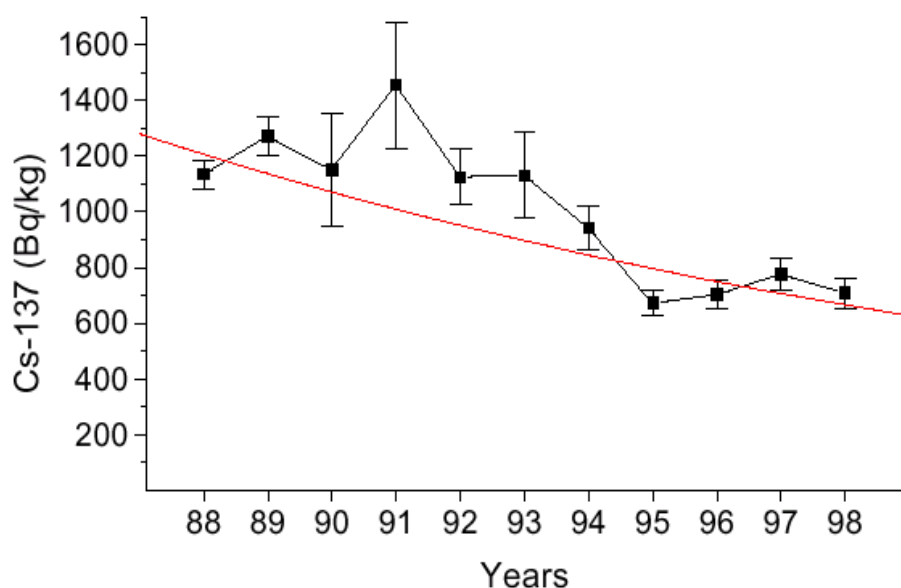


Figure 4. Cs-137 in lambs from 1988 to 1998 at Tjøtta after 8-12 weeks of outdoor grazing. Averages \pm standard error are given.

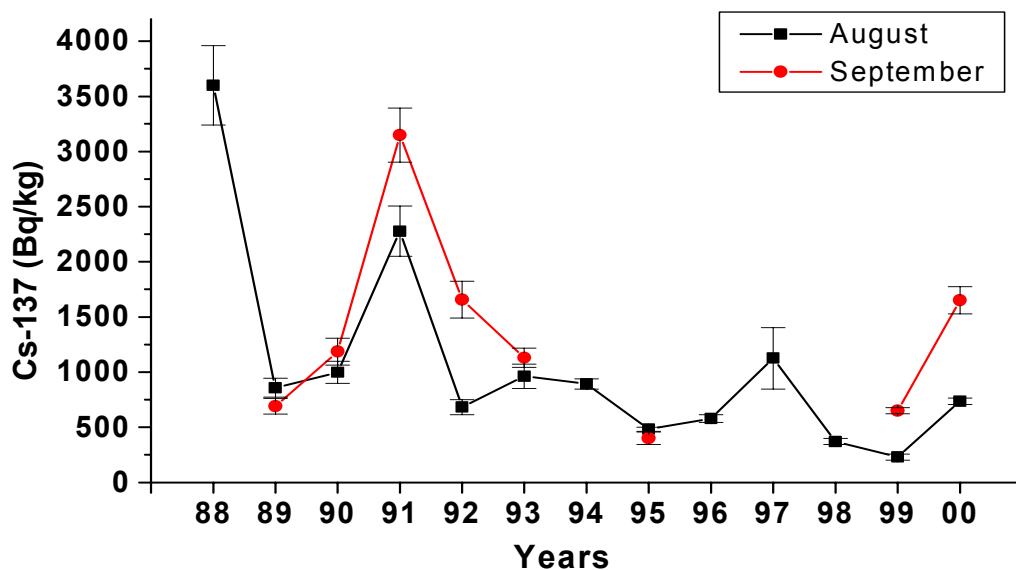


Figure 5. Cs-137 in lambs from 1988 to 2000 at Baklia in August and September. Averages \pm standard error are given.

Conclusion

The consequences of the Chernobyl accident had a serious impact on Norwegian extensive farming. The ^{137}Cs levels in goat's milk are still well above the intervention limits when AFCF is not distributed to the animals through saltlicks, boli or concentrates. For cow's milk the ^{137}Cs levels are only above the intervention limit in the most heavily affected areas of Norway. The annual variation of ^{137}Cs in mutton is dependent on the abundance of fungi on the pastures making it difficult to predict effective ecological half-lives in areas where fungi constitute part of the animals' diets in certain years. The contamination of sheep and lambs has decreased gradually since the Chernobyl accident both due to natural decay and the use of countermeasures such as distribution of AFCF. Nevertheless, the ^{137}Cs levels are still significant. Maximum values of about 4000 Bq/kg were reported in 2000. Up to eight weeks of clean feeding were imposed in 55 municipalities in Norway in the autumn of 2000, comprising a total of about 30 000 sheep, in order to comply with the intervention limit of 600 Bq/kg for mutton. For more results from the livestock monitoring, please see Liland et al., 2001.

Acknowledgement

The monitoring programme is funded by the Ministry of Agriculture through the Norwegian Food Control Authority.

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Radioecological sensitivity in the Faroe Islands estimated from modeling long-term variation of radioactivity

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Abstract

The Faroese environment has received radioactive debris from the nuclear weapons tests in the 1950'ies and 1960'ies and from the Chernobyl accident 26 April 1986. The paper presents results from modeling the relation between ^{137}Cs and ^{90}Sr in precipitation and the radioactivity from these nuclides in selected foodstuffs, using available data from the last four decades. The model relates the concentration of a radionuclide in a sample from a given year to the deposition rate of the radionuclide in the given year and in the year before, and to the accumulated deposition two years before. The effective ecological half-life of the radionuclides in the selected foodstuffs is estimated, and model-calculated sensitivities defined as time-integrated radionuclide concentration in an environmental sample from a unit ground deposition, as e.g. (Bq/kg)y per (kBq/m²), are presented.

Introduction

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

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

Material and methods

The available data for the modeling are summarised in Table 1. The data have been obtained from annual Risø-reports (e.g. Aarkrog *et al.*, 1997) and from personal communication with Sven P. Nielsen at Risø. Precipitation and cow milk have been sampled at three different locations: Klaksvík in the north of the country, the capital Tórshavn in the middle and Tvøroyri in the south. Lamb meat and total diet derive from countrywide samplings. The Faroese drinking water is obtained from surface water, and the samples have been collected as tap water in Tórshavn. It should be considered that Faroese total diet does not only contain locally produced foods. All fruits and cereals are imported, and other foods have been imported to various degrees during the years,

mainly from Denmark. Faroese lamb meat is mostly used for local specialities, and lamb meat is therefore also imported, mainly from Iceland and New Zealand. The composition of the total diet in this context is according to estimates referred to by Aarkrog *et al.* (1963) and Aarkrog (1979).

Table 1. Available data series for the modeling.

Samples	Fallout rates (3 localities)	Cow milk (3 localities)	Lamb meat	Total diet	Drinking water
Isotopes	^{137}Cs	^{137}Cs	^{137}Cs	^{137}Cs	^{90}Sr
Time period	1962-96	1962-98	1962-93	1962-93	1964-93

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

$$C_i = b_1 \cdot d_i + b_2 \cdot d_{i-1} + b_3 \cdot \sum_{k=1}^{k=\infty} d_{i-1-k} \cdot e^{-\lambda \cdot k} + b_4 \cdot \sum_{m=1}^{m=\infty} d_{i-1-m} \cdot e^{-\mu \cdot m} \quad (1)$$

The modeling is done in two versions, with emphasis on a version with $b_4=0$, often called the UNSCEAR model. C_i is the concentration of a given radionuclide in a sample from year (i), and d_i and d_{i-1} are the deposition rates (kBq/m^2) in the years (i) and (i-1), respectively. Index $i=1$ represents the year 1950, taken as the first year with radioactive fallout. The first sum on the right hand side represents the accumulated deposition up to year (i-1) assuming an effective radiological half-life of T years, corresponding to a decay factor $\lambda = \ln(2)/T$. The second summation, analogue to first one, assumes an effective half-life equal to the physical half-life, R, of the given radionuclide with μ given as $\mu = \ln(2)/R$. The estimated ecological half-life of a radionuclide in a given foodstuff is the value of T that gives the best fit between model and observations. It is presumed that all fallout is deposited by the start of a given year. This will underestimate the accumulated deposition, but it is immaterial for this study of long-lived radionuclides.

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

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

$$S = b_1 + b_2 + b_3 \cdot e^{-\lambda}/(1-e^{-\lambda}) + b_4 \cdot e^{-\mu}/(1-e^{-\mu}) \quad (2)$$

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

Results and discussion

The model with $b_4=0$ in Eqs. (1) and (2) has been run with all available data taken into account and with only data before 1986 taken into account (Table 2). Graphic presentations of the results for milk in Tórshavn are e.g. given in Figs. 1 and 2, where the “fit”-curve represents the model results.

The square regression coefficient, R^2 , is found to be high in all cases except for lamb meat. The lamb meat samples have been collected countrywide from few animals each year, and this may be an explaining factor for the relative weak correlation between observations and model in the case of lamb meat. It is well documented that the ^{137}Cs concentration in lamb meat varies significantly between animals even from a single pasture (Joensen, 1999).

The model is found to predict the ^{137}Cs and ^{90}Sr concentrations in the selected foodstuffs fairly well, although it generally gives higher concentrations after 1985 than observed, as e.g. can be noted from Figs. 1 and 2. All available observations are used for the modelling in Fig. 1, while only pre-Chernobyl observations are used in Fig. 2, making the curve after 1985 a prediction curve in Fig. 2. It should be noted that the model results stop in 1996 because no deposition data were available for this study after 1996. The correlation between model and observations is found to be higher when only pre-Chernobyl observations are taken into account in the regression (Table 2). Model estimates of the effective radioecological half-lives of the ^{137}Cs concentrations are 3-5 years for milk, 4.0 years for lamb meat and 4.5 years for total diet, while it is 5.5 years for the ^{90}Sr concentration in drinking water from Tórshavn.

The results for cow milk indicate geographical variation of the radioecological sensitivity, with highest value in the south (Tvøroyri). The reason for the geographical variation has not been further studied in this context, but possible differences in the soil characteristics at the localities would partly explain the variation.

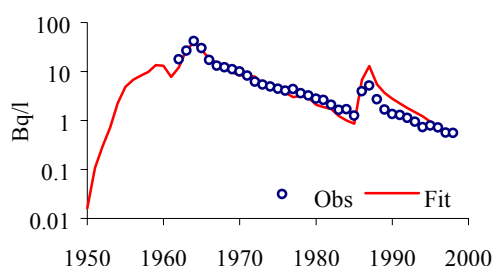


Fig. 1. Cs-137 in milk from Tórshavn.

Regression: 1962-96; $R^2=0.954$.

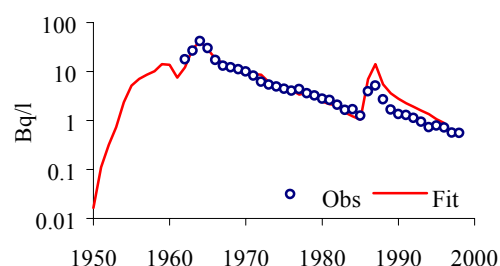


Fig. 2. Cs-137 in milk from Tórshavn.

Regression: 1962-85; $R^2=0.977$.

The calculated sensitivities are high compared to other countries (UNSCEAR, 1977; Aarkrog, 1979; Nielsen and Øhlenslæger, 1999). UNSCEAR (1977; p.143) reports sensitivities for milk in different countries around the world with the unit $\text{pCi}(\text{gK})^{-1}$ per mCi km^{-2} (Table 4). The highest value of 27.51 is reported for the Faroe Islands followed by 15.48 for Norway. The lowest value of 3.23 was reported for Denmark.

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

	b_1	b_2	b_3	T	R^2	Sens. S
^{90}Sr in drink. water; Tórshavn 1964-93	0.0205 (0.486)	0.0397 (0.018)	0.0114 (<0.001)	5.5	0.908	0.145 (Bq/l)y per kBq/m ²
^{137}Cs in total diet. Faroes 1962-93	0.0120 (<0.001)	0.0055 (0.001)	0.0031 (<0.001)	4.5	0.914	13.3 Bq/cap per Bq/m ²
^{137}Cs in lamb meat; Faroes 1962-93	0.1487 (<0.001)	0.0138 (0.594)	0.0168 (0.003)	5.5	0.675	289.0 (Bq/kg ww)y per kBq/m ²
^{137}Cs in milk from Klaksvík 1962-96	0.0075 (<0.001)	0.0086 (<0.001)	0.0040 (<0.001)	3.0	0.903	31.3 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tórshavn 1962-96	0.0093 (<0.001)	0.0171 (<0.001)	0.0041 (<0.001)	3.0	0.954	42.0 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tvøroyri 1964-93	0.0112 (<0.001)	0.0186 (<0.001)	0.0055 (<0.001)	4.5	0.982	62.6 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Klaksvík 1962-85	0.0080 (<0.001)	0.0111 (<0.001)	0.0030 (<0.001)	4.0	0.968	35.2 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tórshavn 1962-85	0.0094 (<0.001)	0.0189 (<0.001)	0.0036 (<0.001)	3.5	0.977	44.7 (Bq/l)y per kBq/m ²
^{137}Cs in milk from Tvøroyri 1964-85	0.0073 (0.062)	0.0222 (<0.001)	0.0050 (<0.001)	5.0	0.986	63.3 (Bq/l)y per kBq/m ²

Results with $b_4 \neq 0$ in the model equations are presented in Table 3. The results for ^{137}Cs are practically the same as in Table 2, and the simpler model with $b_4=0$ is therefore preferable.

Table 3. Results concerning ^{137}Cs , using the model without predefining b_4 to be zero. R^2 , T and S as defined in Table 2. All available data are used in the regression. Model coefficients are not presented.

Milk, Klaksvík			Milk, Tórshavn			Milk, Tvøroyri			Faroe lamb meat			Faroe total diet		
R^2	T	S	R^2	T	S	R^2	T	S	R^2	T	S	R^2	T	S
0.89 3	2.0	29.7	0.946	2.0	39.5	0.983	5.0	57.4	0.676	4.0	329.0	0.914	5.0	12.5
^{90}Sr in drinking water from Tórshavn: $R^2=0.912$, $T=4.5$ years, $S=0.204$														

Conclusion

The long-term variation of radioactivity in the selected foodstuffs is reproduced fairly well by the proposed regression models. The simpler model without the “ b_4 -term” in the model equations is found to be satisfactory for modeling ^{137}Cs and ^{90}Sr in the foodstuffs.

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

The sensitivities in the Faroe Island are high compared to other countries, indicating relatively high individual doses from ingestion of the foodstuffs in the Faroe Islands. Integrated transfer factors, like the sensitivity, provide a comprehensive assessment of transfer over the long term of ^{137}Cs and ^{90}Sr from deposition to foodstuffs.

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

Aknowledgement

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

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Sources of cesium-137 in the diet of reindeer herder families

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For radiation protection purposes it is important to know the intake of radionuclides in fallout situations. The intake can be estimated by indirect methods one of which is the dietary survey or by the direct method of whole-body counting. In an emergency situation there is a need for rapid recommendations on countermeasures. A lot of knowledge on the behaviour of radionuclides in foodchains is needed to give the proper recommendations taking into account all aspects. In Finland the Cs-137 body contents of about 100- 200 reindeer herders in Lapland have been followed since 1962. In connection with the whole-body counting measurements also dietary surveys have been performed, foodstuff samples collected and Cs- 137 concentrations determined.

The first investigation of the diet of Finnish Lapps was performed during the first half of the 60's. It was noted that in the foodstuffs produced in Lapland the amount of long-lived radionuclides, especially of Cs-137, was many times higher than in the corresponding produce of southern Finland. The interview method was used to obtain a general picture of the food consumption of the Lapps and a weighing method to check the results obtained in the interview survey and also to discover the effect of the season on the consumption of different foodstuffs. The following dietary surveys were done in 1971 and 1976 in connection with whole-body counting measurements. A simplified questionnaire was designed to be used in years when no dietary expert was present. The reindeer herder group was followed by whole-body counting for determination of the body content of Cs-137. To compare the intake values obtained by whole-body counting and by dietary investigations a new detailed dietary survey was done in 1997.

In 1997, 102 persons were interviewed by a dietary expert. The results were compared with those from the 60's. The results show that the consumption of reindeer meat has decreased since the earlier investigations in the 1960's and 1970's. The consumption of reindeer meat of male reindeer herders decreased from 323 g in the 1960's to 244 g in 1997 in Inari and from 396g to 159 g in Utsjoki. The groups of females were small in this investigation but for all women the consumption of reindeer meat did not decrease as it did for males. In the 60's the consumption of women from Inari varied from 60 to 116 g per day and in the 70's from 50 to 123 g . In 1997, the consumption varied from 126 to 158 g per day. This could be a consequence of new ways of reindeer-herding. Today men come home from the forest in the evening using a motor sleigh not staying in the forest even for weeks as was the case still in the 1970's. Today the whole family sits together at the table having dinner often eating reindeer meat.

The weighted mean of reindeer meat consumption was 205 g per day in the investigation of Jokelainen in 1962 compared to the value of 182 g per day in 1997. The median was 155 g/day. Still the occupational reindeer herders consume almost twice the amount of reindeer meat compared to people from other occupational groups.

Estimating ^{137}Cs ingestion doses to Saamis in Kautokeino (Norway) using whole body counting vs. dietary survey results and food samples

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Introduction

Following the findings of elevated concentrations of radioactive caesium (i.e., ^{137}Cs) in humans consuming reindeer meat in the early 1960's, Norwegian authorities initiated a monitoring programme among reindeer keeping Saamis in Kautokeino (69°N , 23°E) in 1965. Results of the studies for the period 1965-1983 were summarised by Westerlund et al. (1987). Updates on the results of the whole body monitoring (up to 1999) and analyses of reindeer meat samples (to 1996) were presented by Skuterud et al. (1999).

From 1965 to 1990 whole body measurements were carried out on an annual basis. Since then, 3-year cycles have been followed. In most years, the reindeer keepers have provided samples of reindeer meat for radiocaesium analysis. In 1989-1990 and 1999 dietary surveys were performed in conjunction with the whole-body monitoring. Earlier diet information is available from a separate study in 1963.

Rough estimates of the radiocaesium intake by the studied population in Kautokeino have indicated that the dietary surveys have overestimated the radiocaesium intake. The aim of the present study (a part of the NKS-BOK-2.1.2 project) was to evaluate the available information from Kautokeino, and to derive some conclusions regarding the reindeer meat consumption by today's reindeer keepers, and what ^{137}Cs ingestion doses they are exposed to.

Materials and methods

Every year the monitoring has taken place in end March or early April, as this is assumed to give results representative of the annual mean whole body activity concentrations (Berteig et al. 1972).

The dietary surveys carried out in Kautokeino in 1989 and 1990 focused both on household procurement and individual consumption (with the aid of pictures with portion sizes, frequency of intake etc.) during personal interviews (Bøe and Andersson Sørli 1991).

In 1999 food consumption was studied using a questionnaire derived from a nation wide dietary survey. The questionnaire was in two parts, the first focusing on household's procurement of traditional foods. The second part focused on individual consumption of most products potentially consumed (using a 69 points long detailed list), not only the traditional. Rough estimates of

individual consumption were also made from the information received on household procurement (by correcting for edible fractions, number of persons (incl. children and adults)) in the family. The respondents were left to fill in the questionnaire themselves, with little involvement of the survey personnel.

Studies of dietary habits in Kautokeino were also conducted in February-March of 1963 (Solvang and Øgrim 1967), not related to studies of radionuclide intake. In that study, consumption was estimated based on weighed and recorded food during one week per household.

Results and discussion

Table 1 presents some results of the dietary surveys among reindeer keepers in Kautokeino. The quantities of reindeer meat consumed in 1989-90 were found to be similar to the values found in 1963. Note that the 1963 survey recorded consumption during winter, while the later surveys have estimated the average annual consumption. The results estimated from the two parts of the questionnaire in 1999, indicate lower reindeer meat consumption. About half of the respondents (47 %) answered that there are seasonal differences in their dietary habits, with generally lower reindeer meat consumption during summer.

Estimates of whole body activity concentrations can be made using results of the dietary surveys and measurements of reindeer meat, by applying ICRP's model for caesium retention in "reference man" (ICRP Publication 56). ICRP acknowledge that the long-term biological half-life ($T_2 = 110$ d) may be conservative, especially for females. No measurements have been done of the biological half-life of caesium in the reindeer keepers in Norway. The average body masses of the female and male reindeer keepers in Kautokeino in 1999 were 68 ± 12 and 82 ± 11 kg, respectively.

Table 1. Consumption of foodstuffs by adult reindeer keepers in Kautokeino in 1963 (Solvang and Øgrim 1967), 1989 and 1990 (Bøe and Andersson Sørli 1991¹), and 1999 (based both on household procurement and individual consumption). Average values (\pm SE in 1999), g d⁻¹.

Product	Females				Males		Average ²		
	1963 n=26	1989 n=26	1990 n=21	1999 ³ n=9	1963 n=16	1989 n=19	1990 n=22	1999 ³ n=9	1999 n=19
Reindeer meat	200	258	266	52 ± 13	380	433	384	68 ± 19	192 ± 29
Other meat	6.0	25	25	62 ± 16	6.8	52	36	77 ± 20	-
Reindeer entrails	-	-	-	13.0 ± 7.5	-	-	-	5.3 ± 3.3	-
Milk	170	433	430	160 ± 60	220	778	690	450 ± 160	-
Freshwater fish	-	14	11	9.1 ± 4.8	-	14	16	41 ± 30	13.7 ± 5.2
Wild berries	-	38	41	30.7 ± 8.8	-	36	22	59 ± 22	41.3 ± 9.2

¹ Most of the interviewees participated in the survey both years

² Estimated from the household procurement part of the questionnaire

³ Estimated from the individual consumption part of the questionnaire. One respondent did not state his gender and the results on individual consumption are therefore for 18 persons totally.

The use of ICRP's caesium model requires some assumptions relating to the state of equilibrium of radiocaesium intake and excretion. Radiocaesium activity concentrations in reindeer exhibit a pronounced seasonal variation. In addition there are changes in the Saami diet during the year. An early study in Finland showed that the annual average consumption among male Saamis was 410 g d⁻¹, while being 720 g d⁻¹ in the period November-April (Miettinen et al. 1963). The seasonal

monitoring of reindeer keepers in Kautokeino in 1970-1971 showed that the concentrations measured in July were about 14 and 25 % higher than in March/April for males and females, respectively, due to a longer period of consumption of winter slaughtered reindeer (Berteig et al. 1972). Using the model and the Finnish seasonal diet information, the average activity concentration in reindeer keepers at the time of the monitoring in Kautokeino is estimated to 1.8-2.4 Bq kg⁻¹ per Bq d⁻¹ intake.

Table 2 compares the results of estimating the average ¹³⁷Cs whole body activity concentration in male reindeer keepers using the ICRP caesium model with the observed values for the years when dietary surveys were performed. Also given is an estimate using constant intake and the revised caesium retention model proposed by Melo et al. (1997), which gives an equilibrium caesium activity concentration (normalised to the daily intake) in 80 kg males of about 1.4 d kg⁻¹. The last column in Table 2 also gives the ratio between estimated and observed concentrations in females, as Melo et al. (1997) gives a separate value for T₂ = 65 d for females. This gives a normalised equilibrium caesium activity concentration in an average 70 kg female of about 1.16 d kg⁻¹.

The results show considerable overestimated ¹³⁷Cs whole body activity concentrations most years, especially for the calculations using the ICRP model.

Results of other comparisons of estimated and measured ¹³⁷Cs whole body contents generally show a discrepancy of a factor 2-6 or even higher (Andrási 1994; Tracy et al. 1993). Caesium is lost during most food processing and preparation (IAEA 1994) and this can probably explain the general tendency of overestimating the caesium whole body contents. Therefore it may be concluded that the lowest ratio in Table 4 is due to underestimated individual reindeer meat consumption.

The main reason for the overestimated intakes and body concentrations in Table 2 is probably that reindeer meat traditionally is being boiled, with only limited consumption of broth (Solvang and Øgrim 1967). According to IAEA (1994), boiling meat removes about 60 % of the radiocaesium, and may thus explain about a ratio of 2.5 between estimated and observed ¹³⁷Cs body concentrations in Table 2.

Table 2. Observed and estimated ¹³⁷Cs whole body activity concentrations in adult male reindeer keepers in Kautokeino. Estimates were made using the caesium models of both ICRP and Melo et al. (1997).

Year	¹³⁷ Cs in reindeer meat, Bq kg ⁻¹ (mean ± SE)	¹³⁷ Cs in male reindeer keepers, Bq kg ⁻¹ (mean ± SE)	Estimated ¹³⁷ Cs concentrations in male reindeer keepers, Bq kg ⁻¹ (range)		Ratio estimated/observed	
			ICRP	Melo et al.	ICRP	Melo et al.
1967 ¹	2270 ± 130	473 ± 27	1550-2070	1210	3.3-4.4	2.6 (2.6) ²
1990	340 ± 30	45.9 ± 3.0	230-310	180	5.1-6.8	3.9 (3.2)
			55-73 ³	43	2.0-2.7	1.6 (2.1)
1999	160 ± 27	26.7 ± 2.9	62-83 ⁴	48	2.3-3.1	1.8 (1.8)
			21-28 ⁵	16	0.8-1.0	0.6 (0.7)

¹ 1967 was the first year when several reindeer meat samples were obtained. The consumption values from the 1963 survey were used in the estimate for this year.

² Number in brackets is the corresponding ratio for females.

³ Estimates for 1999 based on household procurement (average consumption for females and males).

⁴ Estimate based on household procurement, but adjusted to male consumption by assuming that males consume 1.3 times more reindeer meat than females (Table 1)

⁵ Estimate using the male consumption values in Table 1.

Using only measured activity concentrations in winter-slaughtered animals in Table 2 will also contribute to overestimated whole body concentrations, as about half the reindeer meat is obtained during August-December. There is no information available on the ^{137}Cs contamination in reindeer in Kautokeino in autumn. In Lovozero (Russia) the contamination of reindeer meat was 50 % higher in winter than in autumn in 1998-1999 (Travnikova et al. in press). A similar difference in Kautokeino would give 30 % overestimated whole body concentrations on the basis of winter samples.

Studies of the gastrointestinal tract (GIT) uptake of caesium from caribou meat showed that it was 'virtually 100 %' (Tracy et al. 1993). Similar results with venison showed an average uptake of about 80 % (Henrichs et al. 1989). Thus, probably the retention model is of more significance for the whole body concentration estimates than the assumption of 100 % caesium uptake in the GI tract, but some overestimated body concentrations may also be attributed to lower GIT uptake.

Accepting that the meat consumed on average has lost 50 % of its radiocaesium activity due to preparation, and that applying results of winter slaughtered reindeer gives a 30 % overestimate, it may be concluded that a ratio around 2.6 between estimated and observed ^{137}Cs whole body activity concentrations is acceptable. This is equal to the results for 1967 in Table 2 using the model by Melo et al. (1967), assuming that the whole body concentrations have attained equilibrium. Using a ratio of 2.6 suggests that the reindeer meat consumption in 1999 was about 240 and 310 g d⁻¹ for females and males respectively. This is in agreement with an average consumption of 192 g d⁻¹, if the population consume twice as much reindeer meat during November-April as during May-October, and assuming that male meat consumption is 30 % higher than female. A ratio higher than 2.6 is possible with a larger difference between summer and winter consumption. Thus, assuming that the meat still is mainly prepared as in 1963, it seems that the average consumption of reindeer meat has decreased in the period 1963-1999, and that the decline is due to reduced consumption by the males mainly. This is also supported by the observed differences in ^{137}Cs whole body activity concentrations, which in 1965-1967 was a factor of 2-2.3, and in 1993-1999 about 1.6. Probably there is also a decrease for females (and a larger decline for males), hidden in changed reindeer meat preparation, which give less loss of ^{137}Cs (e.g. use of casseroles).

The estimates in Table 2 are based on intake of ^{137}Cs via reindeer meat only. By including other products the ^{137}Cs intake estimates would increase. Bøe and Andersson Sørli (1991) found that reindeer meat consumption contributed some more than 90 % of the total radiocaesium intake. Following from the estimates and discussion above, it appears that this intake is overestimated. Thus, reindeer meat might not contribute as much as 90 % of the intake. However, it is likely that Bøe and Andersson Sørli (1991) also overestimated the ^{137}Cs intake via other products. The recent results by Travnikova et al. (in press) suggest that reindeer meat consumption contributes about 83-88 % of the annual ^{137}Cs intake by the reindeer keepers in Lovozero (Russia). Wild mushroom consumption appear to be more common among Russian reindeer keepers than Norwegian, and it is therefore likely that reindeer meat consumption plays an even more predominant role as ^{137}Cs source for reindeer keepers in Kautokeino.

In conclusion, the analyses presented support the apprehension that the reindeer meat consumption by the reindeer keepers and their families has been declining during the last 30-40 years. Annual average values of 170 and 220 g reindeer meat (incl. entrails) per day for females and males, respectively, are probably upper estimates of today's average consumption.

Applying a dose conversion factor of 2.5 mSv y⁻¹ per Bq kg⁻¹ (UNSCEAR 1988) the whole body activity concentrations in 1999 corresponds to an annual average effective dose of about 0.055 mSv.

Maximum concentrations were observed in 1965, corresponding to an annual average effective dose of 1.3 mSv.

Acknowledgement

The studies in Kautokeino have been funded by different sources during the years: The support by the Directorate of Health during first years, later the Ministry of Agriculture through the Norwegian Food Control Authority, the Norwegian Research Council (project nos. 121441/720 and 134118/720), and the Nordic Nuclear Safety Research program (project NKS-BOK-2.1) is gratefully acknowledged. Thanks also to nutritionist Nina Øverby, who analysed the dietary survey results in 1999. Last but not least: Thanks to the participants of the studies during all these years.

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^{242}Pu as tracer for simultaneous determination of ^{237}Np and $^{239,240}\text{Pu}$ in environmental samples

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Abstract

A procedure has been developed using ^{242}Pu as tracer for simultaneous determination of ^{237}Np and $^{239,240}\text{Pu}$ in environmental samples by ICPMS and by α -Spectrometry. The paper describes a suitable chemical procedure for the simultaneous determination of Np and Pu including quantitative pre-concentration of ^{237}Np and $^{239,240}\text{Pu}$. The key concept is to maintain a strict valency control by transforming neptunium and plutonium into the +4 state: $\text{Np}(\text{NO}_3)_6^{2-}$ and $\text{Pu}(\text{NO}_3)_6^{2-}$. Under those conditions, Np and Pu are behaving chemically similar.

The ratio (R) of $^{237}\text{Np}/^{242}\text{Pu}$ (or $^{237}\text{Np}/^{239}\text{Pu}$) before and after the procedure has been determined using 10 g soil as a matrix as $R_{\text{before}}/R_{\text{after}}=1.004\pm3.3\%$ (S.D., $n=20$) and in 1 litre seawater as $R_{\text{before}}/R_{\text{after}}=1.019\pm1.9\%$ (S.D., $n=12$).

Results are presented from the IAEA intercomparison samples IAEA-135 and IAEA-381, from Palomares sediment samples, from sediment and seawater from Sellafield, and from 100 litre Danish seawater samples.

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

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Introduction

This work is part of an investigation of the mobility of plutonium in freshwater systems containing humic substances. A well-defined bog-stream system located in the catchment area of a subalpine lake, Øvre Heimdalsvatn, Norway, is being studied. The area has been thoroughly investigated with respect to other radionuclides (i.e. ^{137}Cs and ^{90}Sr) in several studies after the Chernobyl accident [1,2,3].

During the summer of 1999, six water samples were collected from the tributary stream Lektorbekken and the lake itself (Figure 1). However, the analyses showed that the plutonium concentration was below the detection limit in all the samples. Therefore renewed sampling at the same sites was carried out in August 2000.

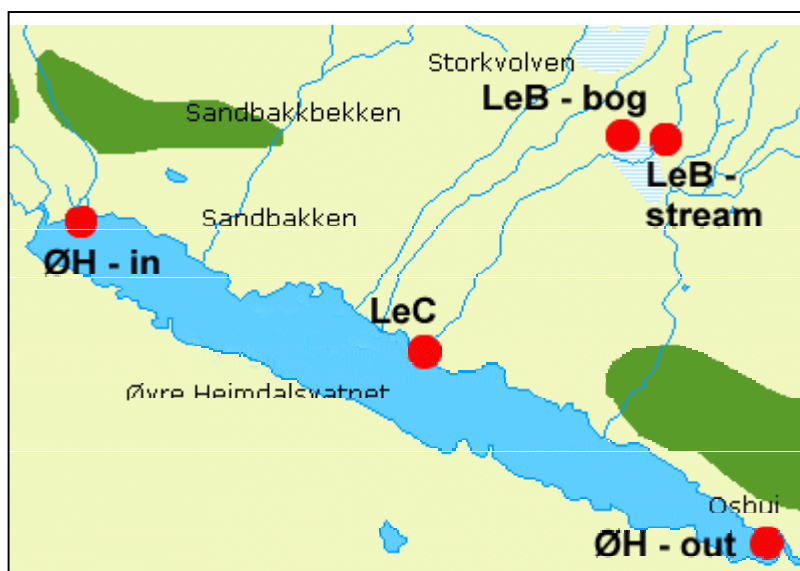


Figure 1. Map of sampling locations.

Fieldwork

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

Table 1. List of samples collected from Øvre Heimdalsvatn and Lektorbekken, August 2000.

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

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

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



Figure 2. Sampling set-up.

Laboratory analyses and results

The samples were brought to IFE's laboratory. Concentrated HCl was added to the samples to dissolve the precipitate. The samples were then analysed for $^{239,240}\text{Pu}$ using a controlled valence procedure, and counted by alpha spectrometry [4].

Only two of the five analyses were completed at the time of writing. The results are shown in Table 2.

Table 2. Results of analyses.

Sample	$^{239,240}\text{Pu}$ [Bq/l]
ØH – out	n. c.
ØH – in	$3.3 \cdot 10^{-6} \pm 0.7 \cdot 10^{-6}$
LeC	n. c.
LeB – bog	n. c.
LeB – stream	$1.4 \cdot 10^{-6} \pm 0.6 \cdot 10^{-6}$

n. c.: analysis not completed

Discussion

Analyses of water samples collected in Heimdalen in 1991 showed that the concentration of $^{239,240}\text{Pu}$ was in the range $5\text{--}8 \cdot 10^{-6}$ Bq/l [5]. Our results are in agreement with these concentrations.

In 1996, samples of stream and surface water were collected in Himdalen near Kjeller, close to the then planned repository for low and intermediate radioactive waste in Norway. The purpose of the work was to establish background values for the concentration of different radionuclides in the area before operation of the repository. The results showed $^{239,240}\text{Pu}$ concentrations in stream water in the range $0.0002\text{--}0.0005$ Bq/l [6], a factor 100 higher than found in Heimdalen. The concentration in surface water was a little lower, $0.00009\text{--}0.0002$ Bq/l. Both Himdalen and Heimdalen have received Pu from fallout, mostly from the atmospheric nuclear weapon tests in the 1950s and 60s.

Water samples are collected annually as a part of the environmental surveillance program for the nuclear activity of the Institute for Energy Technology at Kjeller. In 1999, the concentration of $^{239,240}\text{Pu}$ in water from the Nitelva River close to the institute was between 0.000025 and 0.00033 Bq/l. These results are also higher than what was found in Heimdalen.

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

During eight months of the year, the Heimdalen area is covered with snow. Consequently, major amounts of fallout plutonium have been deposited directly on snow. During the spring flood, this plutonium has possibly been washed out with the melt water. In the lowlands, the ground will be covered with snow for a period of 4-6 months, consequently the plutonium will have had more time to adsorb to soil particles. The soil will therefore, to a higher degree than in the mountains, be a continuing source of remobilised plutonium to the lakes and rivers.

Conclusion

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

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Sampling methods for pasture, soil and deposition used in the Nordic countries

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Abstract

The aim of this work was to survey sampling techniques used for radioactivity measurements in emergency preparedness in the Nordic countries. The survey was a part of the NKS BOK-1.1 project *Laboratory measurements and quality assurance*. The types of samples were restricted to pasture, soil and deposition. The basis of the survey was a questionnaire about sampling methods, sampling equipment, sample preparation, measurement methods, sample storage and reporting of results. This questionnaire was sent to radiation protection authorities and research departments in the Nordic countries and all of the five Nordic countries are represented in the study.

It was found that the participating laboratories apply similar sampling procedures for pasture with a cutting height of between 1 and 5 cm above the ground and sampled areas about 1 m². The sampling plots on the investigated area (field) are usually randomly positioned but the number of plots on each area varies. The analysis is made with NaI or high-resolution gamma spectrometry on fresh and/or dried samples, depending on the fallout situation.

Soil samples are generally taken by some sort of corer of varying diameter. At each sampling site 3 to 20 cores are taken in various patterns and the cores are usually sliced. The analysis is made with high-resolution gamma spectrometry on fresh or dried samples, depending on the expected radionuclides present. For comparison with field-gamma spectrometry and determination of soil water content both fresh weight and dry weight are registered.

To determine the activity concentration in the precipitation for deposition sampling, precipitation collectors of a range of sizes are used. The analysis is made with high-resolution gamma spectrometry, either on ion exchange resins or by direct measurements of a water sample.

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Dose assessment considering evolution of the biosphere

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Introduction

Swedish Nuclear Fuel and Waste Management AB (SKB) is presently updating the safety assessment for SFR (Final repository for radioactive operational waste) in Sweden. The biospheric part of the analysis is performed by Studsvik Eco & Safety AB (Karlsson et al., 2001) and the model system used is based on models set up earlier (Bergström et al., 1999). According to the regulations (SSI, 1998) the safety of the repository has to be accounted for different possible courses of the development of the biosphere. A number of studies have been carried out during the past years to investigate and document the biosphere in the area surrounding the repository. Modelling of shore-level displacement by land uplift, coastal water exchange and sedimentation have provided data for prediction of the evolution of the area (Brunberg & Blomqvist, 1999 and 2000, Brydsten, 1999a and 1999b, Enqvist & Andrejev, 1999 and 2000, Jerling et al., 2001, Kautsky et al., 1999, Kumblad, 1999 and 2001), see Figure 1. The prediction is done without considering a future change in climatic conditions.

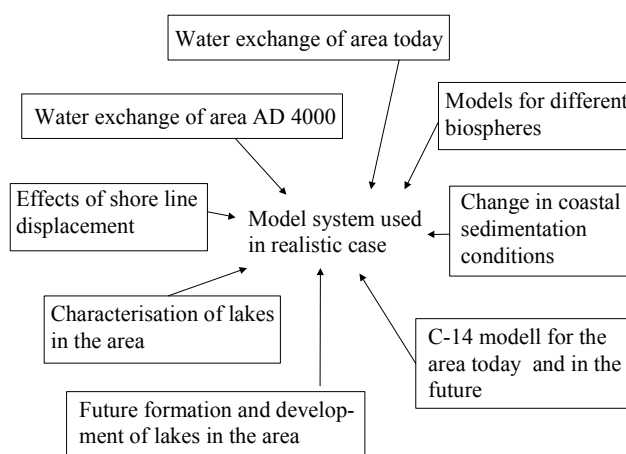


Figure 1. Studies utilised when creating the model system.

Evolution of the SFR-area

SFR is situated 50 m below the bottom of the strait Öregrundsgrepen in northern Uppland. The area is relatively shallow and is strongly influenced by land rise - 0.6 cm/y (Påsse, 1997) - which is shown on maps in Brydsten (1999a), causing continuous changes of the landscape resulting in altered biotopes (Wallström & Persson, 1997, Jerling et al., 2001). The simulation of the shoreline displacement in the area over time shows that the southern part of the strait will close at about 4000 AD and a coastal bay will be formed (Brydsten, 1999a). The distribution between brackish and

fresh water in the area will change over time and a large lake and several smaller ones will be formed in the area about 5000 AD. The large lake is relatively shallow (maximum depth 4.1 m, Brunberg & Blomqvist, 2000). Because of this, the lake will relatively soon be transformed into a mire. Another possible scenario is that part of the lake may be drained and the gained area used for agricultural purposes. The rest of the area is stony and relatively poor. The only area which may be used for cultivation to any larger extent is the lake sediments according to the prediction made (Jerling et al., 2001).

If radionuclides leak from the repository they may reach the biosphere through transport with groundwater. The groundwater passing the repository discharges north of it. Today this point is situated within Öregrundsgrepen. As the shoreline moves towards north-east with time the outflow point will follow. When eventually the large lake is formed the groundwater outflow will take place into this basin.

Implementation in the safety assessment

How has all this been implemented in the safety analysis? In the biospheric part of the assessment the description of the evolution of the area has been a base for the construction of a realistic case for the calculations. The gradual change of the biosphere has not been modelled, instead four different stages have been identified which each one represents different time periods; coast 1 (present conditions), coast 2, lake and agricultural land (see Figure 2). When performing dose assessments the emphasis is put on finding events or processes which may lead to particularly high exposure situations. One such aspect, identified when setting up the realistic case, is that contaminated sediments in the future may be used for agricultural purposes. Radionuclides which reach the biosphere during its coastal or lake stages may sorb to the sediments and accumulate. This is simulated in the realistic case.

For comparison, two other cases were studied: a coastal case and a mire case with constant biospheric conditions for the whole time period (2000 – 12000 AD). The latter case was set up because it resulted in the highest dose rates for most radionuclides in a previous study (Bergström et al., 1999).

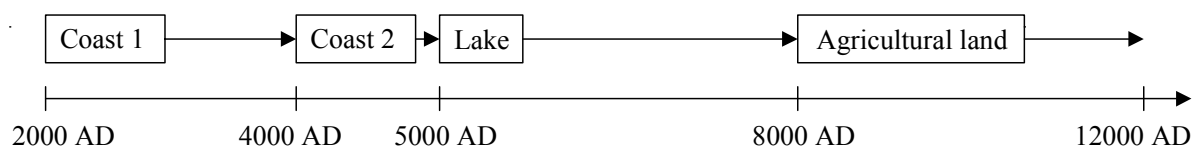


Figure 2. The various models used during different time periods.

Comparison of results from the different cases

The models were run for a continuous release of 1 Bq/year for two radionuclides; Cl-36 and Pu-239. These two nuclides were chosen because they are both long-lived (half-life Cl-36 301 000 years and Pu-239 24 065 years) and behave very differently from one another in the environment. Chlorine is very mobile in surface ecosystems whereas plutonium sorbs to particles in soils, water and sediments. The unit release rate was chosen to simplify the analysis of results. It is more difficult to interpret the behaviour of the models with a source term which changes with time.

The dose rates for the three cases, realistic, coastal and mire, are shown in Figure 3. The realistic case shows an increase in dose rate with time as the different ecosystems replace each other. The increase depends i.a. on diminishing dilution as the water volume decreases when the coastal area develops into a lake. In the latest stage, when the area is assumed to be drained and used for agricultural purposes, the dose rates increase even more.



Figure 3. Dose rates (Sv/year) for the realistic, coastal and mire cases, Cl-36 and Pu-239, respectively.

For the coastal case equilibrium concentrations are rapidly established whereas it takes a couple of years (about 30 for Cl-36 and about 100 for Pu-239) to reach equilibrium in the mire case. The dose rates for the coastal case coincide with those from the first stage of the realistic case (the same model and parameter values were used) whereas the dose rates for the mire case is about two orders of magnitude higher than those in the end of the realistic case (agricultural land).

A realistic case without an accumulation in sediments was also run to determine the importance of an earlier stage accumulation of radionuclides. The difference was most evident in the beginning of the agricultural stage (about eleven and four orders of magnitude higher dose rates for Pu-239 and Cl-36, respectively) and decreased with time as a consequence of i.a. soil mixing and erosion. It should be remembered that this is valid for a continuous release of radionuclides. In case of a scenario where a large accumulation of radionuclides occur during the coastal and lake stages and the source term decreases during the agricultural land period this effect may otherwise be even more evident.

Conclusions

The results from this study show that accumulation of radionuclides in sediments is an important process to simulate when performing dose assessments covering biosphere evolution. The dose calculated for the first years of the period with agricultural use of the contaminated sediments may be severely underestimated in a scenario with large accumulation in coastal and lake stages.

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Dosen i en bastu eller in sauna veritas et corpore sano

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Sammanfattning

Doskonversionskoefficienten (DCC, mSv år⁻¹/Bq kg⁻¹) för årliga expositionen från ¹³⁷Cs och ⁹⁰Sr i trä som konstruktionsmaterial har beräknats för en bastu. Den största skillnaden jämfört med ett timmerhus är ockupationsfaktorn, 100 timmar jämfört med 4 500 timmar per år. En skillnad är dock att huddosen från betapartiklar från ¹³⁷Cs och ⁹⁰Sr/⁹⁰Y kan tas med i beräkningen för en bastu.

Uteslutningsnivåerna för trä beror på vilken dosrestriktion som tillämpas. Med vår kunskap om aggregerade transferfaktorer i skogsekosystemet; (Bq kg⁻¹(ved)/Bq m⁻²) kan en uteslutningsnivå för markkontaminering också beräknas.

Den mest extrema dosbegränsning som kan tänkas är 0,01 mSv a⁻¹ (trivialgränsen) vilket skulle resultera i en uteslutningsnivå på 1,3x10⁻⁴ Bq kg⁻¹ och en mark-kontaminering motsvarande 4,8x10⁶ Bq m⁻². Sådana kontamineringsnivåer finns endast i ytterst begränsade områden till följd av Chernobylolyckan. Sett ur strålskyddssynpunkt spelar andra scenarier än bastuproblematiken en större roll inom skogsindustrin och för skogsprodukter.

Inledning

Timmer och skogsprodukter produceras i många länder både för internt behov och för export till andra länder. Till följd av kärnvapentester och olyckor har radio-nuklider deponerats i regioner där det finns intensiv skogsindustri. Det mest kända exemplet är skogar kontaminerade med ¹³⁷Cs, ¹³⁴Cs och ⁹⁰Sr till följd av Chernobylolyckan. Koncentrationerna i ved före Chernobylolyckan kan i Sverige beräknats ha varit 2-5 Bq kg⁻¹ och är nu efter olyckan 10 - 50 Bq kg⁻¹ (1). Nivåerna i ved från länder inom f.d. Sovjetunionen kan vara en storleksordning högre.

Det finns ett allmänt intresse att bestämma vilka koncentrationer i ved som skall tillåtas för import och export. En generell nivå beskriver inte nödvändigtvis den radiologiska expositionen utan denna beror på radionukliden och användningen av veden. Det finns en möjlighet att till exempel koncentrationerna av ¹³⁷Cs i ved är sådana att det begränsar såväl inhemskt bruk som produktexport till vissa länder, beroende på den radiologiska standarden i ett land. Detta har olika ekonomiska och sociala verkningar för de berörda länderna.

En metodik kan utarbetas som bestämmer den radionuklidkoncentration som resulterar i en dos till allmänheten som inte överstiger den radiologiska standarden som tillämpas i landet. Expositionen blir en funktion av användningen av råvaran och olika scenarier kan utarbetas och doskonversionskoefficienter (mSv per år/Bq per kg) härledas(2). Vi har valt bastuscenariot, som kanske är det mest exotiska.

Exposition i ett timmerhus och bastu

Absorberade dosraten, Sv s^{-1} , från primärstrålning kan beräknas som:

$$\dot{D} = \frac{n_{\gamma} A_m E_{\gamma} \left(\frac{\mu_{en}}{\rho} \right)_{H_2O}}{4\pi} \int_{-X}^X \int_{-Y}^Y \int_0^Z \frac{e^{-\left(\mu_{wood} z + \mu_{air} h \right) \frac{\sqrt{z^2 + y^2 + (h+z)^2}}{h+z}}}{x^2 + y^2 + (h+z)^2} dx dy dz$$

där, η är antalet fotoner per sönderfall, A_m är aktiviteten i Bq per m^3 , ρ är densiteten av trä, kg m^{-3} , E_{γ} är fotonenergin, eV konverterade till J, μ_{en}/ρ är massenergi-absorptionskoefficienten, $\text{m}^2 \text{kg}^{-1}$, μ_{wood} är lineära attenueringskoefficienten för trä, m^{-1} , μ_{air} är lineära attenueringskoefficienten för luft, z är vägg tjockleken m, x är väggens längd, y är väggens bredd och h är kortaste avståndet från väggen till mätpunkten.

Absorberade dosfaktorn beräknades först för gammastrålning i centrum av en hypotetisk träbyggnad. Dimensionerna var $7 \times 7 \times 3$ m. Väggar, golv och tak ansågs konstruerade av trä med tjockleken 0,2m, 0,045 m, och 0,0225 m respektive. Densiteten på trä är 430 kg m^{-3} , ett värde representativt för torr gran och fur. Ockupationsfaktorn för en bastu har satts till 100 timmar per år. Resultaten framgår av Tabell 1. Aggregerade transfer-faktorer för Cs och Sr har uppskattats till $2,5 \times 10^{-3}$ och 5×10^{-3} respektive (3).

Dosraten i en bastu blir liknande den i ett timmerhus. Bastun har mindre dimensioner och personen är närmare källan. Dimensionerna för en bastu kan t.ex. anses vara $4 \times 3 \times 2,5$ m. Dosraten är proportionell mot $1/r^2$ om r är avståndet från källan. Detta kompenseras för genom att totala källstyrkan ökar approximativt med kvadraten av dimensionen i en byggnad. Vi har gjort en korrektion för luftattenuering motsvarande 4% högre dos i bastun.

För betastrålare som ^{90}Sr , blir högsta dosen på ytan av en träbänk. Dosen från betastrålare är mer relevant i en bastu eftersom kläder normalt utgör ett strålskydd för betapartiklar. För den effektiva dosen till huden har antagits att 10% av hudytan exponeras och att viktningsfaktorn är 0,01.

Räckvidden av beta partiklar med energier högre än 0,3 MeV kan approximeras med:

$$R = 0,52E_x - 0,09/\rho$$

där E_x är betaenergin i MeV, R är räckvidden i cm, och ρ är densiteten i g cm^{-3} .

En betapartikel på 1 MeV kommer att ha en räckvidd av 4,3 mm i vatten. För ^{90}Sr kommer dosen från summan av ^{90}Sr och dess kortlivade dotter, ^{90}Y . Medelenergierna för ^{90}Sr och ^{90}Y är 196 keV och 934 keV respektive. För en bastu skall också betapartiklarna från ^{137}Cs , medelenergi 188 keV, tas med i beräkningen.

Dosraten på en träbänk från betapartiklarna blir.

$$\dot{D} = E_{\beta, \text{mean}} \times A_m \times 1,6 \times 10^{-19} / 2 \text{ Gy s}^{-1}$$

där $E_{\beta, \text{mean}}$ är medelenergin av betapartiklarna, eV, och A_m är aktivitetsskoncentrationen i trä, Bq kg^{-1} .

Tabell 1. Doskonversionskoefficienter i en bastu, maximala koncentrationsnivåer i trä och maximala ytkontamineringar för dosrestriktionerna 0,01 och 0,1 mSv a⁻¹.

Beräkningsgrund	¹³⁷ Cs	⁹⁰ Sr/ ⁹⁰ Y
Gammastrålning, DCC, mSv a ⁻¹ Bq ⁻¹ kg	7,8 x10 ⁻⁷	-----
Betastrålning, DCC, mSv a ⁻¹ Bq ⁻¹ kg	0,6x10 ⁻⁸	3x10 ⁻⁸
Uteslutningsnivå i trä, Bq kg ⁻¹ vid 0,01 mSv a ⁻¹	1,2x10 ⁴	3x10 ⁵
Uteslutningsnivå i trä, Bq kg ⁻¹ vid 1 mSv a ⁻¹	1,2x10 ⁶	6x10 ⁷
Uteslutningsnivå för markdeposition, Bq m ⁻² vid 0,01 mSv a ⁻¹	4,8x10 ⁶	6x10 ⁷
Uteslutningsnivå för markdeposition Bq m ⁻² vid 1 mSv a ⁻¹	4,8x10 ⁸	6x10 ⁹

Konklusioner

Dosen från en normal svensk eller finsk bastu kan beräknas till 0,02 µSv per år om koncentrationen av ¹³⁷Cs i konstruktionsmaterialet är 25 Bq kg⁻¹ eller kommer från ett område med markdepositionen 10 000 Bq m⁻² och ockupationsfaktorn är 100 timmar per år.

På basis av resultaten kommer man fram till att det är högst osannolikt att en bastu skulle vara konstruerad av råmaterial som i sin helhet är kontaminerat så att det skulle innebära exposition över dosbegränsningarna. Trivialgränsen, 0,01 mSv per år, skulle innebära att råvara från, t.ex. Bryansk regionen, skulle överstiga uteslutningsnivån endast i några mindre områden av den mest kontaminerade zonen på 310 km². För dosnivån 1 mSv finns inga sådana områden.

Alltså "in sauna veritas et corpore sano". Det finns andra scenarier för skogsindustri och skogsprodukter som kommer mer i fokus från radiologisk synpunkt vid dosrestriktioner. Dessa vill vi återkomma till senare.

Erkännande

Detta utgör ett delprojekt som har ekonomiskt stöd av Nordiskt Kontaktorgan för Kärnsäkerhet, NKS.

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Airborne cesium-137 in Northern Finland in the early 1960's based on the measurement of archived air filter samples

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The Finnish Meteorological Institute (FMI) started to collect weekly aerosol filter samples in Northern Finland in the end of 1962. The sampling location was first at Ivalo airport (68°36'N, 27°25'E, h = 140 m a.s.l. [above sea level]) but in October 1964 the site was moved to Kevo research station (69°45'N, 27°02'E, h = 98 m a.s.l.). The filter samples were obtained from the instruments monitoring continuously aerosol beta activity. The filter material was paper (Whatman 40) and the weekly air volume some 1000 m³. The samples were sent to the FMI's laboratory where they were measured for long-lived beta activity five days after the end of sampling. This long-lived beta activity consisted of lead-210 and artificial beta emitters. After the measurement the samples were archived. In 1997-1998 the oldest filters were retrieved from the archive and analysed for ¹³⁷Cs with two scintillation gamma spectrometers. The detectors were 4"x 2" NaI(Tl) crystals. The detection limit was about 30 µBq/m³.

The observed weekly ¹³⁷Cs activity concentrations in the air are depicted in Figure 1. The values below detection limit were plotted as equal to the detection limit (= 30 µBq/m³). The weekly concentration values decreased with an efficient half-life of 19 months. Eisenbud and Gesell give a value of 6 to 12 months for the half-life of artificial radioactivity in the stratosphere [1]. The result of this study is, however, affected by fresh releases of ¹³⁷Cs. The average April-May concentration decreased an order of magnitude from 2340 µBq/m³ in 1963 to 420 µBq/m³ in 1967. For comparison, during the same time period the deposition of ¹³⁷Cs decreased from 90 to 6 Bq/m² in Helsinki [2].

The ratio of total beta activity to ¹³⁷Cs at Ivalo and Kevo is presented in Figure 2. The ratio decreases with a half-life of 11 months, that is more rapidly than the concentration of ¹³⁷Cs. This is due to the decay of fallout nuclides with a shorter half-life. The increase in the ratio in December 1966 is related to the atmospheric release of fresh fission products from a leaking underground nuclear test explosion at Semipalatinsk, Kazakhstan (former Soviet Union) [3].

The results of the present study contain two possible error sources not encountered in current atmospheric monitoring programmes. The sampling technology, especially the air flow measurement, during the sample collection was not as good as it is nowadays. In addition, the aerosol particles collected onto filters might be affected by the over 30 years long storage. Still, the results are reasonable and in agreement with other studies. The present study also points out the importance of long-term collection and storage of environmental samples as in most cases a retroactive sample collection is impossible.

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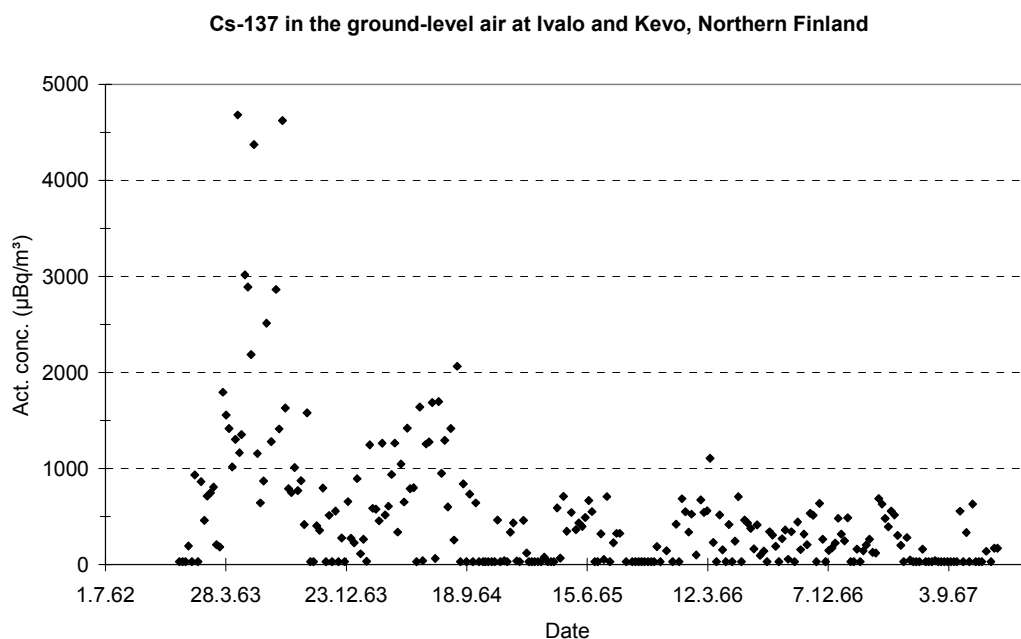


Figure 1. Weekly concentrations of ^{137}Cs ($\mu\text{Bq}/\text{m}^3$) in ground-level air at Ivalo and Kevo, Northern Finland.

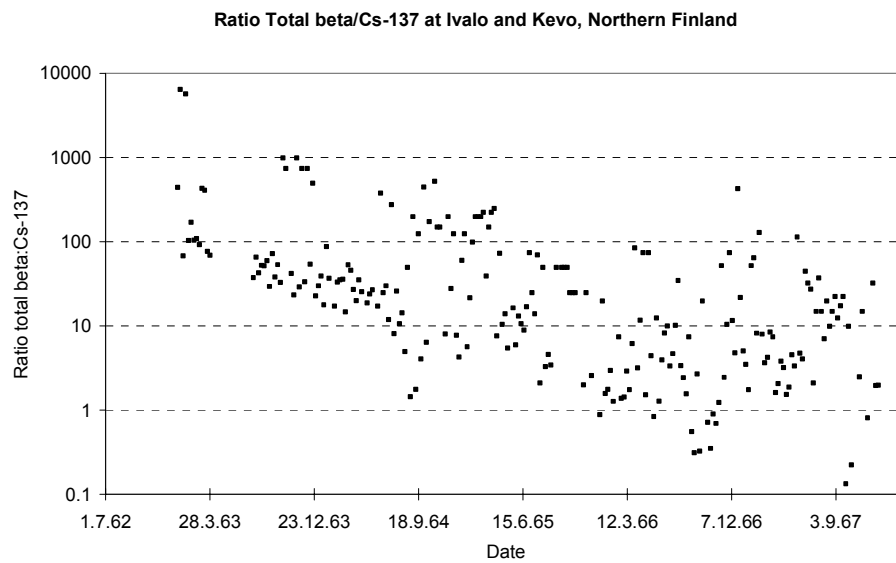


Figure 2. Weekly values of total beta activity/ ^{137}Cs ratios in ground-level air at Ivalo and Kevo, Northern Finland.

Radiocaesium (Cs-137) fallout in Iceland and its behaviour in subarctic volcanic soils

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In the autumn of 2000 the Icelandic Radiation Protection Institute began systematic analysis of radiocaesium in Icelandic soils. The main objectives of the study are to investigate the spatial variation of radiocaesium concentration in Iceland and make estimates of the retention of Cs-137 in major soil types. Soil samples from 17 sampling sites were collected (see Table 1). Soils were sampled with a 17 mm sampling probe. Twenty cores were collected at each site at even intervals along a 20 m long line (see Isaksson 2000). The concentration of Cs-137 was measured using HPGe spectrometry. Results are reported as Bq/kg and Bq/m².

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

Icelandic soils have mainly formed in aeolian and tephra deposits overlying the glacial till and lava bedrock. They classify as Andosols. Andosols constitute 70-80 % of the soils of Iceland. Among their main characteristics are high CEC, considerable organic carbon content (1-10 %) and high clay content (20-40 %) (Arnalds 1999, Arnalds *et al.* 1995, Wada *et al.* 1992). All these factors influence the ability of soil to bind Cs-137.

The Cs-137 retention for three depth intervals (0-5 cm, 5-10 cm and >10 cm) is compared with texture of the soils in Figure 1. The soils group into four different classes, loamy sand, sandy loam, loam and organic material (peat). Significant differences are observed in Cs retention between the soil texture classes indicating that binding of Cs is highly dependant on soil properties. A deeper penetration of Cs-137 is observed for sandy soils than finer loams.

The main results gained so far are the following:

- Total activity per unit area of Cs-137 was found to be 900-4700 Bq/m².
- In all cases > 85 % of Cs-137 in soils is fixed in the uppermost 15 cm of the soil cover (see Figure 1).
- In loam and sandy loam 60-95 % of Cs-137 is fixed in the top layer of the soil (i.e. 0-5 cm interval). In sandy loam and loamy sand (Vitric Andosols) considerable amount of Cs is fixed in deeper layers, i.e. below 5 cm depth, indicating that mobility of Cs-137 is dependent on grain

size distribution and clay content of the soil. Former studies indicate that Cs is rather mobile in Icelandic soils (Oughton *et al.* 1997).

- d) In peat (Histosols) 80-85 % of Cs-137 is bound in the top layer of the soil (0-5 cm) and 10-15 % within the 5-10 cm depth range.
- e) Comparison of samples obtained from undisturbed hayfield (MYRa in Figure 1) and uncultivated land at the same site (i.e. MYR) showed lower concentration of Cs-137 in the top layer of the hayfield but on the other hand higher total activity per unit area. Sample from GRI, collected in a hayfield, shows similar behaviour of Cs-137 as in MYRa.
- f) Good correlation was obtained between precipitation and total activity of radiocaesium in soils (see paper by Pálsson *et al.* 2001)

Table 1. Soil sampling in Iceland 2000.

Sampling site/ abbreviation	Date of samplin g	Location- GPS	Soil type	Vegetation	Bedrock ¹	Ann. Precip. in mm ²
Rjúpnahæð RJU	18.09.00	N64 05.093 W21 50.450	Andosols	Grass	Lavas/sedim. < 0.8 m.y.	870
Keflavík KEF	19.09.00	N64 01.591 W22 35.129	Andosols	Willow, moss, sedge	Lavas/sedim. < 0.8 m.y.	960
Grindavík GRI	19.09.00	N63 49.236 W22 31.224	Andosols (vitric)	Grass, hayfield	Lavas, postglacial	972
Stykkishólmur STH	20.09.00	N65 02.211 W22 46.105	Histosols (fibric)	Heath, moss	Lavas, Upper Tertiary	611
Barkarstaðir BAR	20.09.00	N65 12.387 W20 46.763	Histosols	Sedge, moss	Lavas, Upper Tertiary	437
Nautabú NAU	20.09.00	N65 27.533 W19 22.030	Andosols	Grass, hayfield	Lavas, Upper Tertiary	435
Mýri MYR	21.09.00	N65 22.602 W17 22.802	Andosols	Grass, infield	Lavas/sedim. 0.8-3.3 m.y.	381
Mánárbakki MAN	21.09.00	N66 12.001 W17 05.916	Andosols	Heath, moss, sedge	Lavas/sedim. 0.8-3.3 m.y.	514
Grímsstaðir GRF	21.09.00	N65 39.876 W16 05.789	Andosols	Grass, hayfield	Lavas/sedim. < 0.8 m.y.	363
Grímsárvirkjun GRV	22.09.00	N65 08.552 W14 31.132	Andosols	Heath, moss, sedge	Lavas, Upper Tertiary	643
Teigarhorn TEI	22.09.00	N64 39.191 W14 18.547	Andosols	Moss, sedge	Lavas, Upper Tertiary	1070
Hólar HOL	22.09.00	N64 18.003 W15 11.685	Andosols	Grass	Lavas, Upper Tertiary	1417
Fagurhólmýri FAG	22.09.00	N63 53.081 W16 37.703	Andosols/ Arenosols	Grass/sedge	Holocene sediments	1840
Kirkjubæjarklaustur KIR	23.09.00	N63 46.833 W18 04.939	Andosols	Sedge, moss	Lavas/sedim. 3.3-0.8 m.y.	1589
Vík in Mýrdal VIK	23.09.00	N63 24.635 W19 01.742	Andosols	Moss, sedge	Hyaloclastics < 0.8 m.y.	2030
Hella HEL	23.09.00	N63 51.291 W20 23.940	Andosols	Heath, sedge, moss	Lavas/sedim. < 0.8 m.y.	1052
Stóra-Ármót ARM	02.10.00	N63 59.632 W20 56.729	Andosols	Heath	Holocene lava	1300

¹ Ref: Jóhannesson and Saemundsson 1998

² Precipitation data from 1960-1967. The Icelandic Meteorological Office.

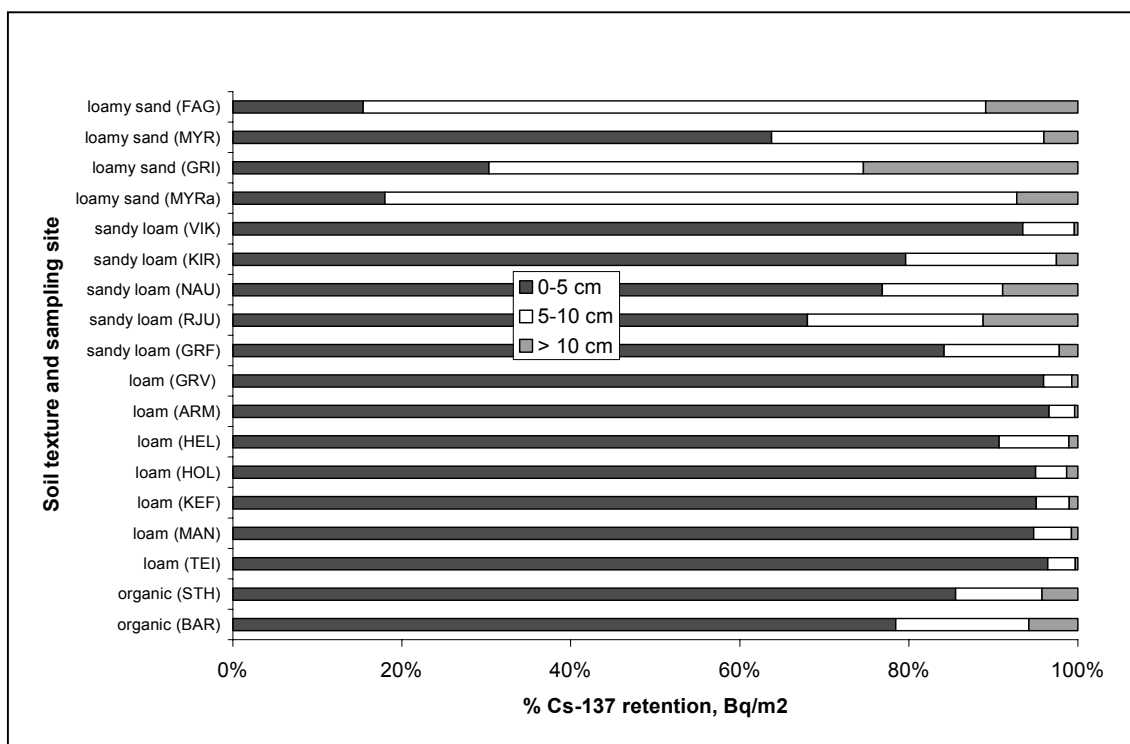


Figure 1. Proportional retention of Cs-137 within depth intervals versus soil texture.

Future plans involve further soil sampling, which is vital for getting a more complete picture of fallout Cs-137 in Iceland. Further studies on the binding ability of Andosols have been planned. Measurements of RIP (Radiocaesium Interception Potential) and organic carbon are in process. The project is carried out in co-operation between the Icelandic Radiation Protection Institute and the Agricultural Research Institute.

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Observations of atmospheric lead-210 over the Atlantic Ocean and Antarctica during the FINNARP-1999/2000 expedition

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Physical and chemical properties of the atmosphere were measured in November-December 1999 onboard the Russian research vessel Akademik Fedorov over the Atlantic Ocean between the English Channel and the coast of Antarctica. After the cruise the measurements were continued in January 2000 at the Finnish research station Aboa (73°03'S, 13°25'W, 470 m above sea level) in Dronning Maud Land, Antarctica. The location of the station is about 170 km inland from the coast of Weddell Sea. The study is part of the Finnish Antarctic Research Programme (FINNARP) financed by the Academy of Finland.

Daily high-volume (~3000 m³) aerosol particle samples were collected onto quartz-fibre filters. The lead-210 contents of the samples were assayed six months after the sampling by counting the alpha particle emissions of the in-grown polonium-210. The detection limit for ²¹⁰Pb depends on the counting time and air volume but is usually about ten µBq/m³. Main ionic components of the particulate matter, e.g. sulphate, nitrate and ammonium, were determined with ion chromatography. Elemental and organic carbon were determined with an automatic EC/OC analyser based on thermal-optical method. Aerosol optical thickness (AOT) was measured with a sun photometer and light scattering coefficients in three wavelengths with an integrating nephelometer. The high-volume samples were analysed also for methanesulfonic acid (MSA). Comparison to results from parallel size-segregated samples indicate that the high-volume sampler collects efficiently accumulation mode (submicron) aerosol particles. Earlier studies show that most of the airborne lead-210 activity is attached to the submicron particles [1].

The observed ²¹⁰Pb activity concentrations in the air varied between 24 and 790 µBq/m³ during the cruise between the English Channel and Cape town. The highest values were recorded close (<300-400 km) to the West-African coast between Senegal and Liberia. This coincides with the observations of highest aerosol optical thickness and light scattering coefficients. The latter observation can be attributed to two factors, biomass burning in Western Africa or resuspension of soil in the Sahara desert. Because anthropogenic activities have a negligible effect to the amount of lead-210 in the air [2], the resuspension in the Sahara desert is a more plausible explanation. Between Cape Town and the coast of Antarctica the observed ²¹⁰Pb activity concentrations varied between <10 and 51 µBq/m³.

At the research station Aboa the observed values varied between <10 and 37 µBq/m³. Between 6 and 11 January 2000 the air masses at Aboa originated from central regions of Antarctica according to the trajectory calculations made with the NOAA Hysplit model [3]. The concentrations were below detection limit between 11 and 14 January. These samples were connected to the air masses originating from the South Atlantic Ocean. Between 14 and 20 January air masses came to Aboa mainly from easterly direction from coastal and inland regions of the Antarctica.

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Table 1. Sampling periods and activity concentrations of ^{210}Pb ($\mu\text{Bq}/\text{m}^3$) in the ground-level air over the Atlantic Ocean and Antarctica during the FINNARP-1999/2000 expedition.

Start	Stop	$\mu\text{Bq}/\text{m}^3$
8.11.99	9.11.99	60
9.11.99	10.11.99	82
10.11.99	11.11.99	180
11.11.99	12.11.99	170
12.11.99	13.11.99	210
13.11.99	14.11.99	170
14.11.99	15.11.99	300
15.11.99	16.11.99	590
16.11.99	17.11.99	790
17.11.99	18.11.99	480
18.11.99	19.11.99	370
19.11.99	20.11.99	290
20.11.99	21.11.99	170
21.11.99	22.11.99	72
22.11.99	23.11.99	83
23.11.99	24.11.99	24
24.11.99	25.11.99	24
1.12.99	2.12.99	51
2.12.99	3.12.99	21
3.12.99	4.12.99	10
4.12.99	5.12.99	<10
5.12.99	6.12.99	18
8.12.99	8.12.99	12
6.1.00	7.1.00	22
7.1.00	8.1.00	22
8.1.00	9.1.00	18
9.1.00	10.1.00	37
10.1.00	11.1.00	20
11.1.00	12.1.00	<10
12.1.00	13.1.00	<10
13.1.00	14.1.00	<10
14.1.00	16.1.00	14
16.1.00	17.1.00	12
17.1.00	18.1.00	23
18.1.00	19.1.00	23
19.1.00	20.1.00	16

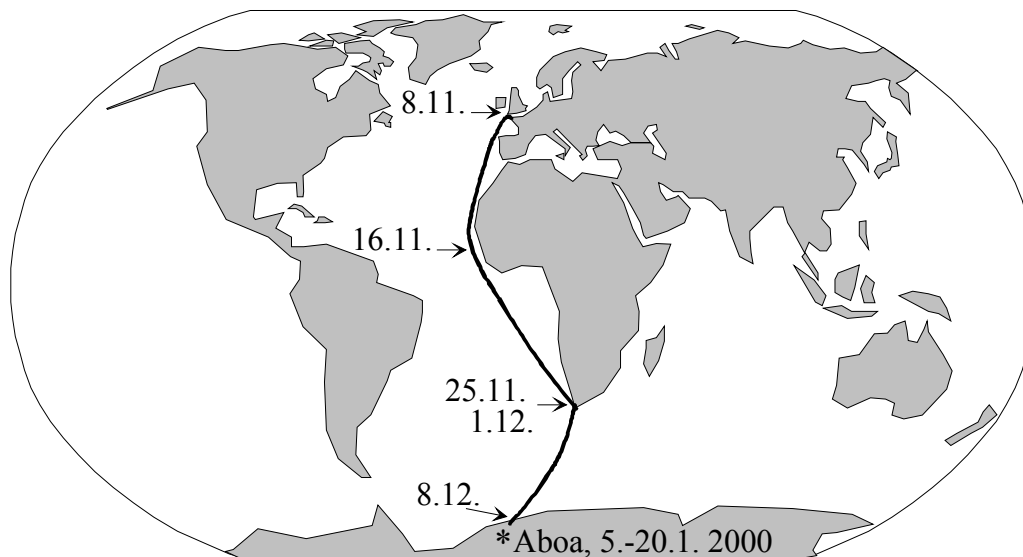


Figure 1. The route of R/V Akademik Fedorov in November-December 1999 and the location of the Finnish research station Aboa.

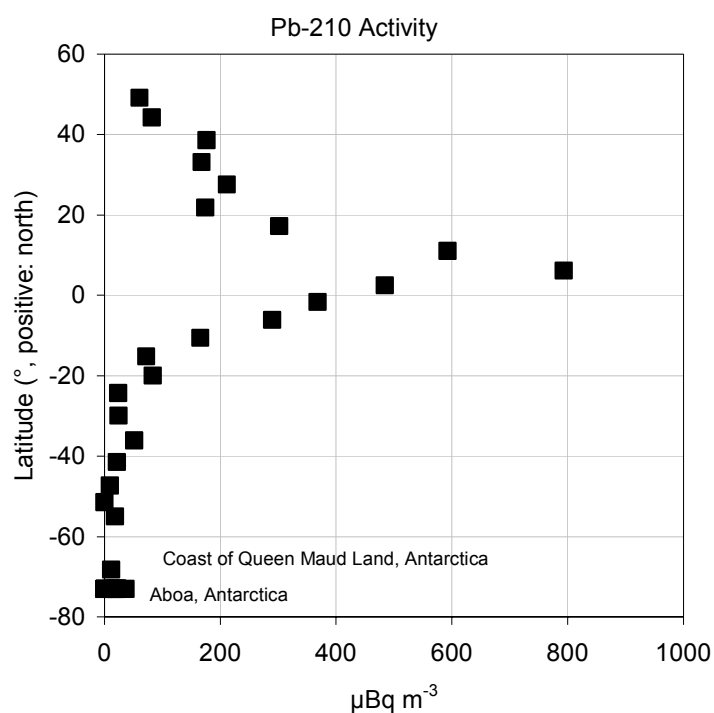


Figure 2. Activity concentrations of ^{210}Pb ($\mu\text{Bq}/\text{m}^3$) in the ground-level air over the Atlantic Ocean and Antarctica during the FINNARP-1999/2000 expedition as a function of latitude.

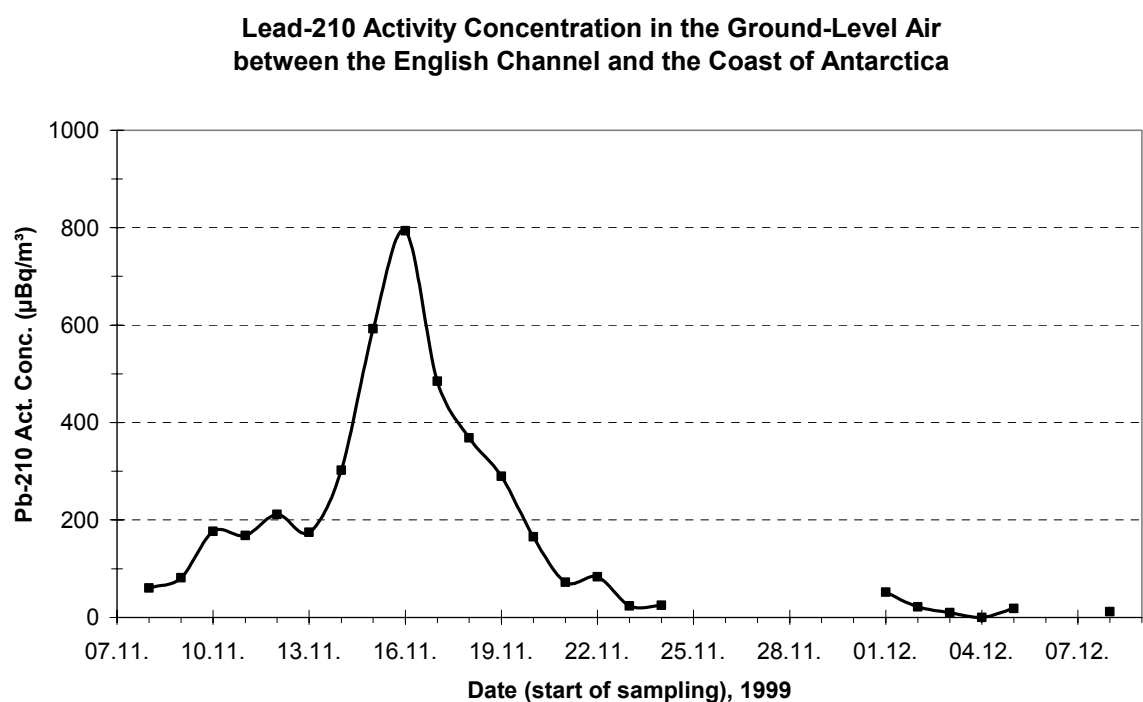


Figure 3. Activity concentrations of ^{210}Pb ($\mu\text{Bq}/\text{m}^3$) in the ground-level air over the Atlantic Ocean in November-December 1999.

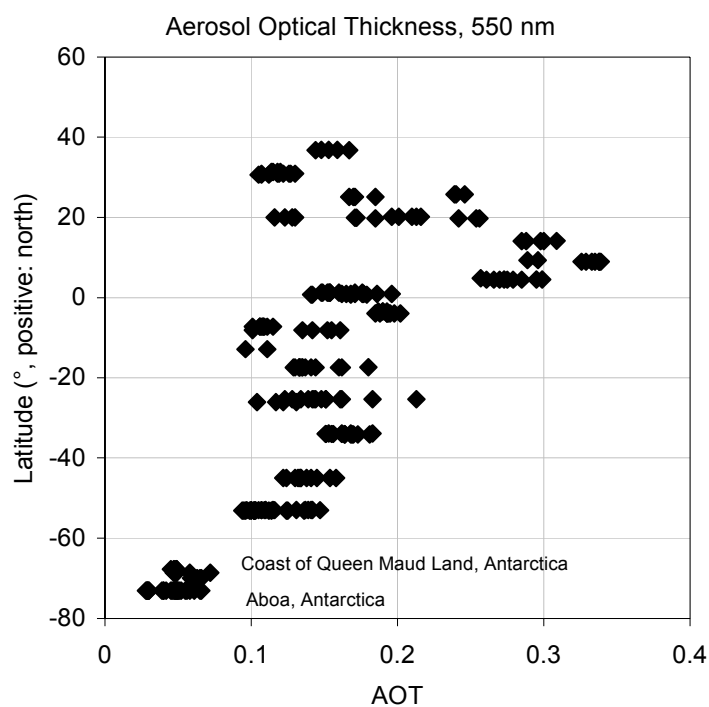


Figure 4. Aerosol optical thickness (AOT, $\lambda = 550$ nm) over the Atlantic Ocean in November-December 1999.

Extracted fractions of ^{137}Cs in Arctic soil as a function of pollution load

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Introduction

The aim of study was to determine the easily available fraction of ^{137}Cs in soil as a function of pollution load as well as the association of ^{137}Cs in organic and mineral soil constituents using sequential extraction technique.

Material and methods

The soil samples were taken from a reference site in Finnish Lapland (REF) and from four polluted sites (A-D) at increasing distance from the Monchegorsk copper-nickel smelter in the Kola Peninsula, NW Russia. The sampling sites were at distances of 7, 16, 21, 28 and 152 km from the smelter. The main pollutants emitted from smelter were SO_2 , Cu and Ni. Total concentrations of Ni and Cu in the organic layer increase from about 10 ppm at REF to about 5000 ppm at the site A.

At each site soil samples were collected from five plots and they were divided into organic layers (Of and Oh) and mineral layers (E1,E2,B1,B2). Combined organic (O) and E1 layers were subjected to sequential extractions according to the procedure described by Bunzl et al.(1997). The sequential extraction procedure was used to extract the fractions: I) Easily exchangeable: Extraction with 1M NH_4Ac ; II) Acid reducible (Fe-Mn oxides-bound): Extraction with 0.04 M $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25% (v/v) HAc; III) Acid oxidizable (organic-bound): Extraction with 30% H_2O_2 ; IV) Residual.

Gamma-spectrometric analyses for ^{137}Cs were performed using high purity germanium detectors. The measuring times varied between 600 and 1000 minutes.

Conclusion

The readily exchangeable fraction of ^{137}Cs decreases from 50% in site REF to about 25% at site D and 15 % at site A with increasing pollution load in the organic layer. Most of the ^{137}Cs (35% – 47%) was persistently bound at the Russian sites but only 13 % at REF site. In the mineral E1 layers most of ^{137}Cs (39% - 50%) was in the persistently bound fraction at all sites including the reference site.

There was noted a clear positive correlation between the distance from the smelter and the extracted percent of ^{137}Cs in readily exchangeable fraction (Spearman correlation $r_{\text{sp}}=0.7805$, $p=0.0001$) and an inverse correlation in the persistently bound fraction ($r_{\text{sp}}=-0.7650$, $p=0.0001$) and a lower correlation in fraction 'bound to organic matter' ($r_{\text{sp}}=-0.3707$, $p=0.0681$) in organic layers. In E1

layers no correlation was noticed between the ^{137}Cs percentage in different extraction steps and the distance from the smelter.

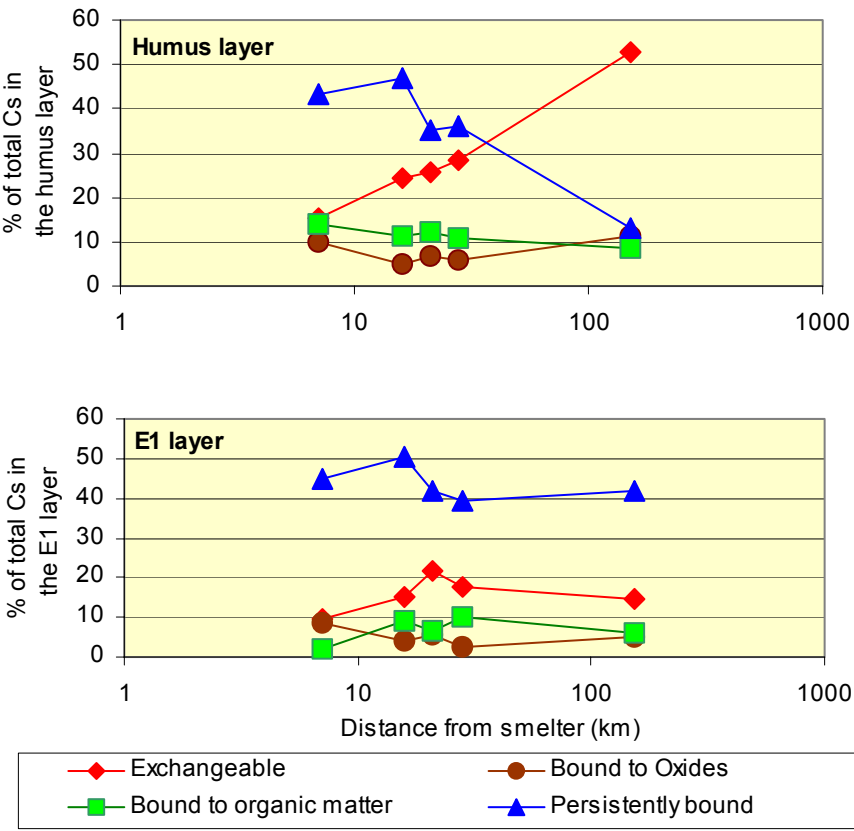


Fig.1 Influence of heavy metal loads in the soil organic and the upper mineral layers on the ^{137}Cs fractions as determined through sequential extraction

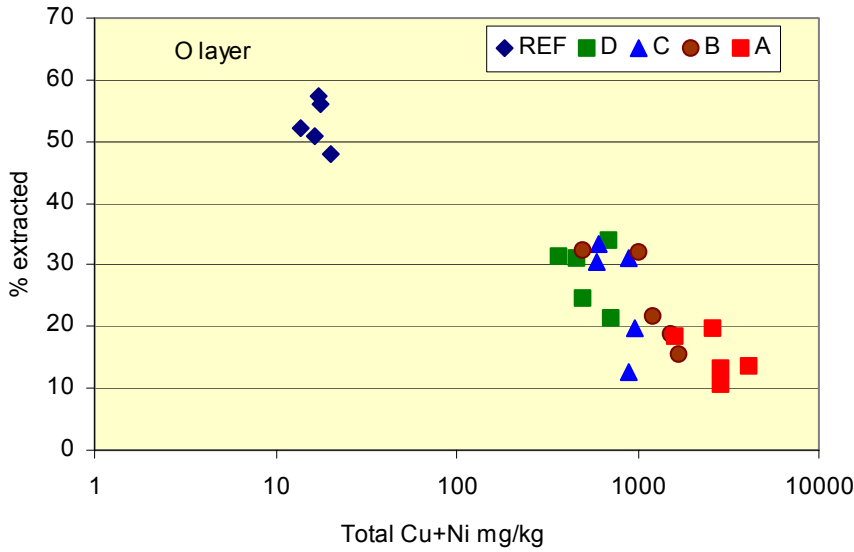


Fig 2. Percentage of readily exchangeable ^{137}Cs as a function of the concentration of Cu and Ni in the O layer

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Plutonium in coniferous forests

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

Soil

Soil, trees, dwarfs, berries and mushroom samples were collected at a pine-dominated site in Central Finland (Fig.1, site 1). Litter and humus, and 0-2 cm mineral soil layers were analysed for ^{238}Pu and $^{239,240}\text{Pu}$ in five profile samples (Taipale, Tuomainen, 1985). The layers 2-5 cm and 5-10 cm were studied in two of these core samples (Fig.2). In litter, humus and mineral soil layer 0-2 cm the total $^{239,240}\text{Pu}$ activities per surface area were almost equal (Fig.2). In deeper mineral soil layers additional Pu was found, and a total of about 40 Bq m^{-2} was estimated for the site no 1.

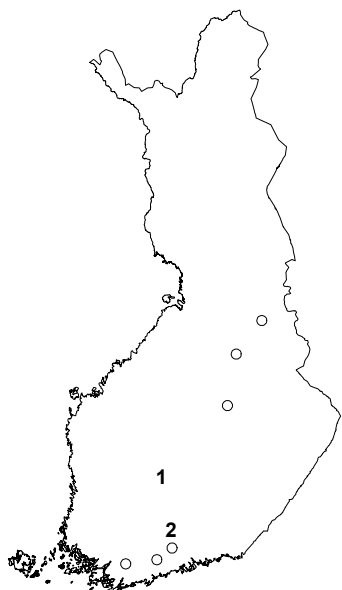


Figure 1. Sampling sites 1 and 2 representing wet deposition in 1986 and dry deposition locations for moose sampling.

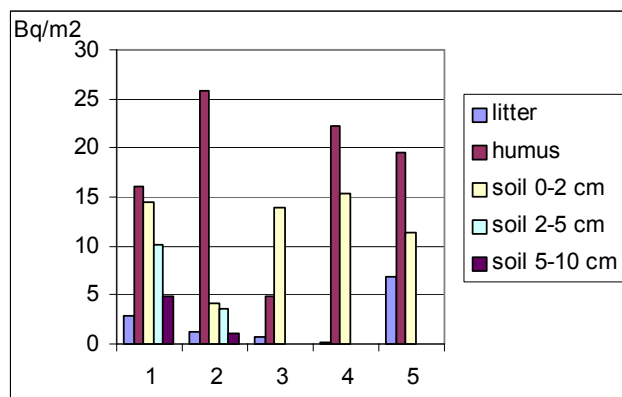


Figure 2. $^{239,240}\text{Pu}$ (Bq m^{-2}) in five soil profiles from site 1.

The maximum activity ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$ in litter, 0.20, was closest to the values found for plutonium in the Chernobyl fallout, 0.47 suggested by Holm et al. (1992), and 0.55 by Paatero and Jaakkola (1998) (Fig. 3). In humus almost the same ratio was occasionally measured, whereas in the mineral soil the ratio was equal to 0.03, estimated for nuclear weapons fallout by Holm et al. (1992).

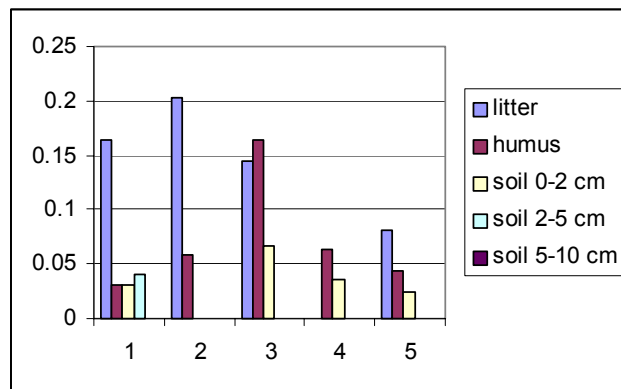


Figure 3. Activity ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$ in soil samples from the site 1.

Trees

A 20-year old Norway spruce (*Picea abies*) was sectioned by age of needles and branches. The stem was divided in three parts. Wood and bark of the two uppermost stem parts were studied. $^{239,240}\text{Pu}$ was found in the bark of the middle part of the stem, 18 mBq kg⁻¹ dw and in the sample of dead branches and needles, 27 mBq kg⁻¹ dw. Detection limit for other tree fractions varied between 3 and 20 mBq kg⁻¹ dw.

Needles from the two uppermost quarters of five Scots pine canopies (*Pinus sylvestris*) were analysed. $^{239,240}\text{Pu}$ was not detected, the range of the detection limits was 2 - 5 mBq kg⁻¹ dw. Neither did $^{239,240}\text{Pu}$ activities in stem wood or bark exceed the detection limit 2 mBq kg⁻¹ dw.

Wild foods and dwarfs

Mixtures of moose meat (*Alces alces*) from site 2 and from locations of dry deposition in spring 1986 were analysed for both adult animals and calves (Fig.1). Dwarfs and berries of *Vaccinium vitis-idea* and berries of *Vaccinium myrtillus*, as well as mushrooms *Cantharellus cibarius* and *Cantharellus tubaeformis* were analysed. In dwarfs the detection limits of the activity concentration of $^{239,240}\text{Pu}$ ranged from 10 to 20 mBq kg⁻¹ dw.

Plutonium was not found in wild food samples. The detection limit of $^{239,240}\text{Pu}$ ranged between 0.7-3 mBq kg⁻¹ fw. The transfer of Pu from soil to wild foods was of the order of 10⁻⁵ or less (m² kg⁻¹ fw), comparable with the findings by Suomela et al, 1999. The ingestion dose estimate for adults through average consumption (Markkula, Rantavaara, 1996) of wild foods was less than 4 nSv a⁻¹. Per unit deposition this upper limit corresponds to 0.1 nSv a⁻¹ per Bq m⁻².

Conclusions

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

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Detection of radionuclides originating from Loviisa nuclear power plant in a municipal sewage sludge

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Abstract

Sewage sludge is a sensitive indicator used in studying radioactive materials transported into sewage via various routes. Radionuclides in sewage sludge from municipal wastewater treatment plant, Vårdö, in the town of Loviisa close to Loviisa nuclear power plant have been measured to find out which radionuclides originating from discharges from nuclear power plant can be detected. The main source is radionuclide discharges of the NPP into the atmosphere, but workers may transmit small amounts through their clothes or skin, or from internal contamination. Radioactive discharges into the air from the Loviisa NPP in 1998 - 2000 were dominated by ^{58}Co , ^{60}Co , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{54}Mn and ^{51}Cr . ^{60}Co and $^{110\text{m}}\text{Ag}$ were also among the most abundantly detected nuclides in sludge from Vårdö during the maintenance, refuelling and shutdown periods. Other nuclides typically found in sludge and probably originating from the Loviisa NPP were ^{54}Mn , ^{58}Co , and ^{124}Sb .

Introduction

Sewage sludge is a sensitive indicator used in studying radioactive materials transported into sewage via various routes (1-5). Radionuclides in sewage sludge from a wastewater treatment plant close to Loviisa nuclear power plant (NPP) have been measured to find out which radionuclides originating from discharges from the nuclear power plant can be detected. The main source is radionuclide discharges of the NPP into the atmosphere, but workers may transmit small amounts through their clothes or skin, or from internal contamination.

Material and methods

Sludge samples for radioactivity measurements were taken in the municipal wastewater treatment plant, Vårdö, in the town of Loviisa during maintenance refuelling and shutdown periods of the Loviisa NPP. The town of Loviisa with 8000 inhabitants is situated about 7 km from the Loviisa NPP. The drinking water is from ground water. In Loviisa there are no hospitals and institutes using radionuclides. Many workers of the NPP live in Loviisa in areas with a municipal sewage system. Some rain and run-off waters also flow into the wastewater treatment plant, and some sludge is transported to the treatment plant from septic tanks of private homes outside the municipal sewage system. The dry material content of the sludge is 15-20% and the amounts of the sludge about 20-25 m³ per week.

Loviisa NPP, on the south coast of Finland, has two 488 MW pressurised water reactors. Cooling water is taken from and released into the sea. Liquid effluents are discharged to the sea and can't be detected in the municipal sewage water. The routes of radionuclides from the NPP into the municipal sewage is via air from fallout and via people.

Results

Data on radionuclide discharge of the NPP were collected and compared with results of the radionuclide measurements in sludge to obtain information on the radionuclide transport from the NPP to sludge.

Radioactive discharges into the air from the Loviisa NPP in 1998 - 2000 were dominated by ^{58}Co , ^{60}Co , $^{110\text{m}}\text{Ag}$, ^{124}Sb , ^{54}Mn and ^{51}Cr . ^{60}Co and $^{110\text{m}}\text{Ag}$ were also among the most abundantly detected nuclides in sludge from Vårdö during the maintenance, refuelling and shutdown periods. Other nuclides typically found in sludge and probably originated from the Loviisa NPP were ^{54}Mn , ^{58}Co , and ^{124}Sb . Figure 1 shows reported weekly discharges of ^{60}Co , $^{110\text{m}}\text{Ag}$ and ^{124}Sb into the air from the Loviisa NPP in 1998-2000 and the concentrations of nuclides found in sewage sludge in the Vårdö wastewater treatment plant. The annual maintenance periods for the two reactors are shown with a horizontal line.

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Table 1. Artificial gamma-emitting radionuclides in sewage sludge at the Vårdö wastewater treatment plant in the town of Loviisa.

Date	⁵⁴ Mn	⁵⁸ Co	⁶⁰ Co	Bq kg ⁻¹ dry weight		¹²⁴ Sb	¹³¹ I	¹³⁷ Cs
				⁹⁵ Nb	^{110m} Ag			
3.8.1998	0	0	0	0	0	0	0	35
11.8.1998	0	0	4.3	0	0	0	0	35
17.8.1998	0	0	4.0	0	0	3.4	0	26
25.8.1998	0	0	4.1	0	0	2.5	0	55
1.9.1998	0	0	2.4	0	0	3.1	0	65
7.9.1998	0	0	6.8	2.9	0	4.5	0	44
15.9.1998	0	3.0	6.6	8.0	1.6	2.0	0	28
21.9.1998	1	2.4	7.3	0	0	0	0	49
29.9.1998	5	3.4	11	0	0	0	0	38
2.11.1998	3.5	0	5.4	0	0	0	690	53
3.8.1999	0	0	0.8	0	0	0	4.2	21
10.8.1999	0	0	1.5	0	0	0	2.6	20
16.8.1999	0	0	2.6	0	3.3	0	3.1	17
25.8.1999	0	0	1.0	0	3.3	0	380	37
31.8.1999	1.7	0	5.0	0	2.0	0	150	31
6.9.1999	0	1	1.9	0	1.7	0	240	23
4.10.1999	2.0	0	5.1	0	0	0	0	39
11.7.2000	0	0	1.2	0	0	0	22	15
24.7.2000	0	0	0	0	0	0	9.4	17
7.8.2000	2.2	0.7	3.3	5.0	7.1	2.4	2.4	23
22.8.2000	59	2.9	15	0	8.9	1.7	1	25
5.9.2000	1.0	7.9	2.5	0	7.9	1.1	0	19
18.9.2000	2.7	4	3.2	0	4.0	1.4	0	22

0=not detected

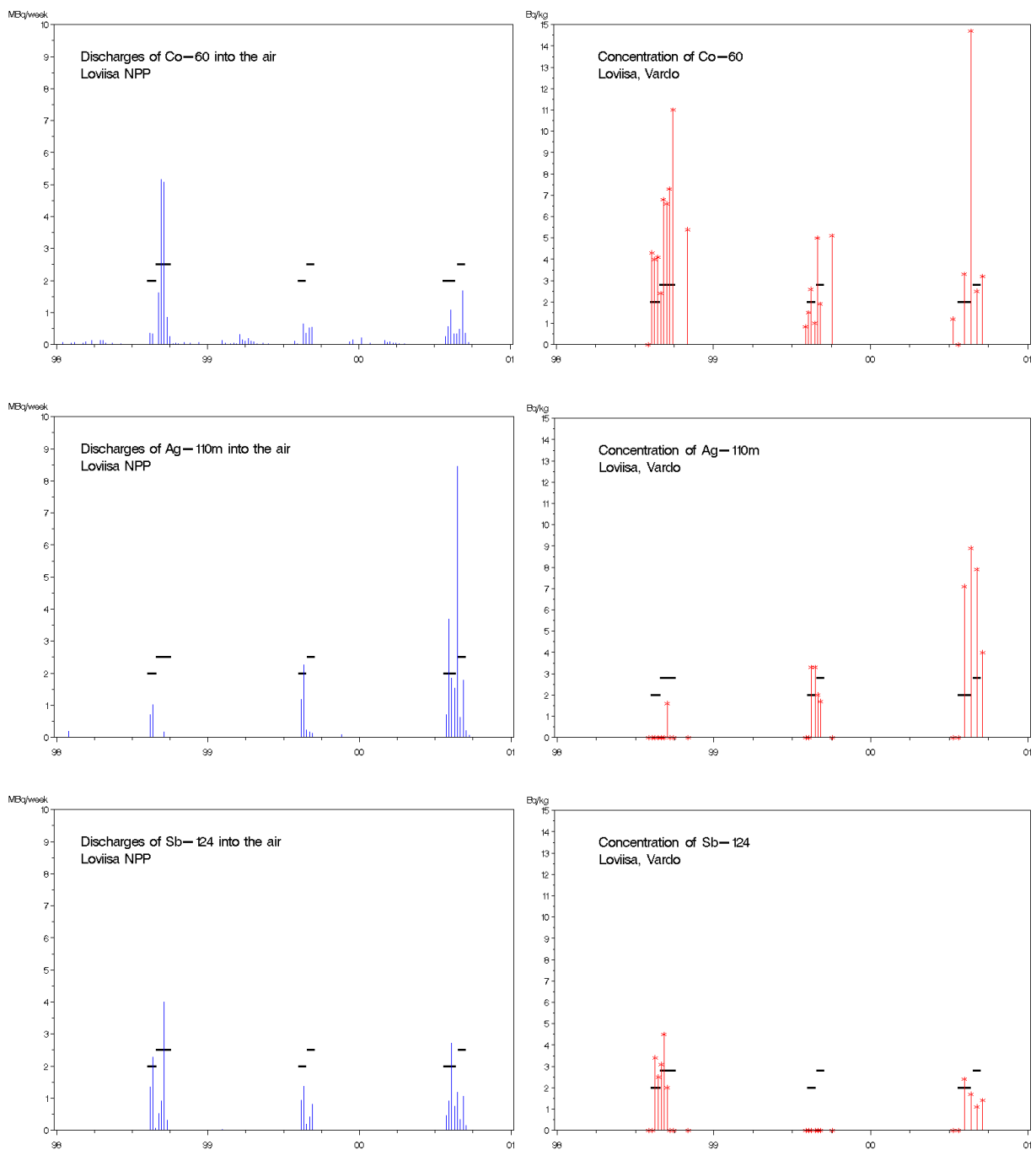


Figure 1. Reported weekly discharges (MBq week^{-1}) into the air from the Loviisa NPPs and the concentration of these nuclides (Bq kg^{-1} dry weight) in sewage sludge in the Vårdö wastewater treatment plant in the town of Loviisa in 1998-2000. Annual maintenance periods for reactors are shown with horizontal lines.

Removal of radionuclides at a waterworks

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Abstract

A waterworks, providing several large cities in the province of Scania with drinking-water, with an average production rate of $1.3 \text{ m}^3 \cdot \text{s}^{-1}$ has been studied regarding its removal capacity for several natural and antropogenic radionuclides. The raw water is surface water from lake Bolmen which is transported through an 80 km long tunnel in the bedrock before it enters the waterworks. The method used for purification is a combination of precipitation and filtration in sand filters. Two different purification lines are at the moment in use, one using $\text{Al}_2(\text{SO}_4)_3$ as a coagulant and one using FeCl_3 . After coagulation and flocculation the precipitation is removed and the water is passed through two different sand filters (rapid-filtration and slow-filtration).

Water samples have been collected at the lake, the inlet at the waterworks, after each of the flocculation basins ($\text{Al}_2(\text{SO}_4)_3$ and FeCl_3), after rapid-filtration and from the municipal distribution net. The samples have been analysed with respect to its content of uranium, thorium, polonium, radium, plutonium and caesium.

Introduction

The two main sources for potable water are surface water and groundwater. Groundwater has naturally been purified by filtration through its passage in the soil, which demands less treatment than surface water before distribution to consumers. Surface water has a higher content of organic substances and is more exposed to different pollutants which requires a higher degree of purification before distribution. In Sweden about half of the production of potable water comes from surface water and half from groundwater or artificial groundwater (VAV, 1994).

A common purification method is a combination of chemical precipitation and filtration in a series of sandbeds. A widely spread method is the use of $\text{Al}_2(\text{SO}_4)_3$ as a coagulant which forms a $\text{Al}(\text{OH})_3$ precipitation when the $\text{Al}_2(\text{SO}_4)_3$ reacts with, mainly, HCO_3^- in the water (VAV, 1992). The precipitate captures suspended particles, colloids and dissolved molecular compounds which makes it suitable for water purification. At the waterworks in this study, FeCl_3 is also used as a coagulant, which instead gives a $\text{Fe}(\text{OH})_3$ precipitation.

Scenario

Several communities in the western part of the province of Scania (southern part of Sweden) are supplied with drinking water from lake Bolmen (area 184 km^2 , max depth 37 m), located about 130 km NNW of the city of Lund. The water is transported from the lake through an 80 km long tunnel in the bedrock (red and grey gneiss), to a reservoir near the village Äktaboden where the water is

lead through a 25 km long pipeline to the waterworks. The transportation time for water in the tunnel is about seven days. The average flow of water to the waterworks is $1.3 \text{ m}^3 \cdot \text{s}^{-1}$, of which about 10 % consists of groundwater which infiltrates into the tunnel from cracks in the bedrock.

When the raw water enters the waterworks it is led to large flocculation basins where the pH is adjusted and a coagulant is added. The waterworks is equipped with two different purification lines, one using FeCl_3 as a coagulant and one using $\text{Al}_2(\text{SO}_4)_3$. When FeCl_3 is used, $40\text{-}50 \text{ g} \cdot \text{m}^{-3}$ is added to the water, and after the pH has been adjusted, flocculation occurs at $\text{pH}=5.2$. In the basins where $\text{Al}_2(\text{SO}_4)_3$ is used, $35\text{-}40 \text{ g} \cdot \text{m}^{-3}$ is added and flocculation occurs at $\text{pH}=6$. Occasionally sodium silicate is added to facilitate the flocculation. The flocculation time (the volume of the flocculation basin divided by the flow) for the process is 45 min. After the flocculations have been removed by sedimentation, the pH is raised to 7.2-7.4 (by addition of lime) and the water is passed through a sand filter (rapid-filtration). The sand bed (grain-size 0.8-1.2 mm) has a thickness of 1 m and the water is passed at a speed of 3 meters per hour. The sand filter is back-flushed with pure water every 24 hours in order to remove impurities trapped in the filter. After rapid-filtration the pH is raised once more to 7.8 before it is passed through the next sand filter (slow-filtration) at a speed of 25 cm per hour. The thickness of this sand-filter is about 1 m and the top layer of the sandbed is cleaned 2-4 times per year. Slow-filtration takes place outdoors in large basins where water from both purification lines are mixed. Altogether the purification process takes about 9 hours to complete. Before the water is distributed to the water mains the water is disinfected and the pH is raised to 8.8.

The precipitation that is removed in the process is further treated in order to reduce its water content. The annual production of untreated precipitation is 36000 m^3 , of which 97 % consists of water.

Several physical and chemical parameters of the incoming and outgoing water are measured at regular intervals to ensure that the quality of the water is acceptable. The analyses are performed by an external accredited laboratory and some of the results valid for the time of the sampling campaign for this study are presented in Table 1.

Table 1. Water quality of incoming and outgoing water at the waterworks.

Parameter	Incoming water	Outgoing water
Turbidity [FNU]	0.80	0.28
Color [mg/l Pt]	50	<5
COD _{Mn} [mg/l O ₂]	9.0	1.3
Ca [mg/l]	8	22
Mg [mg/l]	2	2
Fe [μg /l]	390	<5
K [mg/l]	1	1
Mn [mg/l]	<0.03	<0.03
SO ₄ ²⁻	9	13

Material and Methods

Water samples (unfiltered) with volumes in the range of 5-350 litres were collected at lake Bolmen, at the waterworks and from the distribution net. Several preconcentration methods were used *in situ* to facilitate the handling of the samples. Thorium and plutonium were coprecipitated with $\text{Fe}(\text{OH})_3$ after addition of radiochemical yield determinants (^{229}Th and ^{242}Pu) and sodium disulfite. For caesium, $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ impregnated cotton filters were used (Roos *et al.* 1994).

Uranium was, like plutonium and thorium, coprecipitated with $\text{Fe}(\text{OH})_3$ (^{232}U was used as a radiochemical yield determinant). Polonium and radium were coprecipitated with MnO_2 after the samples had been spiked with ^{209}Po or ^{133}Ba .

After standard radiochemical separation methods, plutonium, thorium and uranium activities were, after electrodeposition (Hallstadius, 1984), determined by alpha-spectrometry. Polonium was spontaneously deposited on nickel discs and also measured by alpha-spectrometry. Radium was measured from its daughter ^{222}Rn by the emanation method (Rushing *et al.* 1964). For gamma-spectrometry a HPGe-detector was used.

Results

In tables 2-5 the results from the activity measurements are listed. The result is presented as the mean value and the standard deviation of duplicate samples. Errors originating from counting statistics were in most cases below 5 % at the 95 % confidence level. For the samples with the lowest activity concentration the errors from counting statistics could be up to 40 % at the 95 % confidence level. In Tables 2-5, these results are marked with an asterisk.

Table 2. Activity concentration in water [mBq/l] and isotope ratios for ^{234}U and ^{238}U .

<i>Uranium</i>							
	Lake Bolmen	Lake Bolmen (filtrated)	Intake	After $\text{Al}(\text{OH})_3$ precipitation	After $\text{Fe}(\text{OH})_3$ precipitation	After rapid-filtration (Al-line)	After slow-filtration
^{238}U	3.0 ± 0.2	1.34	1.18 ± 0.01	0.14 ± 0.01	0.26 ± 0.01	$0.08^* \pm 0.01$	0.16 ± 0.01
^{234}U	3.0 ± 0.3	1.57	1.79 ± 0.03	0.24 ± 0.03	0.42 ± 0.02	$0.17^* \pm 0.02$	0.26 ± 0.03
$^{234}\text{U}/^{238}\text{U}$	1.0 ± 0.1	1.17	1.5 ± 0.04	1.7 ± 0.30	1.6 ± 0.1	2.0 ± 0.4	1.6 ± 0.3

Table 3. Activity concentration in water [mBq/l]. For $^{239/240}\text{Pu}$ the activity concentration is expressed in $\mu\text{Bq/l}$. nd = not determined.

<i>Thorium, Caesium, Plutonium, Polonium and Radium</i>						
	Lake Bolmen	Intake	After $\text{Al}(\text{OH})_3$ precipitation	After $\text{Fe}(\text{OH})_3$ precipitation	After rapid-filtration	After slow-filtration
^{232}Th	0.32 ± 0.02	0.12 ± 0.01	0.013 ± 0.005	0.016 ± 0.001	0.004 ± 0.001	0.004 ± 0.001
^{230}Th	0.25 ± 0.03	0.15 ± 0.01	0.025 ± 0.001	0.037 ± 0.001	0.006 ± 0.001	0.013 ± 0.001
^{228}Th	0.97 ± 0.06	2.55 ± 0.11	0.20 ± 0.01	0.34 ± 0.01	0.12 ± 0.01	0.11 ± 0.03
^{137}Cs	4.66 ± 0.10	4.55 ± 0.06	4.50 ± 0.02	4.38 ± 0.14	4.28 ± 0.01	4.25 ± 0.03
$^{239/240}\text{Pu}$	43 ± 3	20 ± 0.3	$0.8^* \pm 0.1$	$1.8^* \pm 0.1$	$0.7^* \pm 0.2$	$0.65^* \pm 0.04$
^{210}Po	2.7 ± 0.5	1.6 ± 0.2	$0.10^* \pm 0.09$	< 0.01	< 0.01	0.28 ± 0.04
^{226}Ra	1.2 ± 0.2	4.3 ± 0.3	3.0 ± 0.6	5.1 ± 0.6	nd	4.0 ± 0.6

Table 4. Activity concentration in precipitations [Bq/kg dry weight].

<i>Al(OH)₃ and Fe(OH)₃ sludge</i>						
	^{239/240} Pu	²³² Th	²³⁴ U	²³⁸ U	²¹⁰ Pb	⁷ Be
Al(OH) ₃ sludge	0.86 ± 0.08	4.53 ± 0.03	45.0 ± 0.7	61.8 ± 4.5	230 ± 3	280
Fe(OH) ₃ sludge	0.72 ± 0.07	4.54 ± 1.42	43.7 ± 0.3	62.8 ± 3.5	368 ± 20	353

Table 5. Fraction of radioactivity passing the purification process, calculated as activity concentration in water at the intake divided by the activity concentration after slow-filtration.

<i>Fraction of activity passing the total purification process</i>							
¹³⁷ Cs	^{239/240} Pu	²³² Th	²³⁰ Th	²²⁸ Th	²²⁶ Ra	²³⁴ U	²³⁸ U
0.93 ± 0.01	0.03 ± 0.01	0.03 ± 0.01	0.08 ± 0.01	0.04 ± 0.01	0.93 ± 0.20	0.15 ± 0.02	0.14 ± 0.01

Discussion and Conclusions

From Tables 3 and 4 above one can see that the activity concentration in the water, after the passage in the tunnel is decreasing for thorium (except ²²⁸Th), uranium, plutonium and polonium. An explanation for this could be that the activity is associated with particles that are removed from the water by sedimentation. Another possible explanation is that Fe(HCO₃)₂ in the groundwater is converted to Fe(OH)₃ when it comes in contact with oxygen rich surface water: 4Fe(HCO₃)₂ + O₂ + 2H₂O ⇌ 4 Fe(OH)₃ + 8CO₂ (Vattenteknik, 1992). Coprecipitation with the Fe(OH)₃ could then explain the decrease of activity in the water.

For one uranium sample a pre-filter was used which showed that roughly 50 % of the uranium activity in the lake water was associated with particles larger than 1 µm. For caesium on the other hand, it seem like most is present in ionic form or associated with colloids with a low sedimentation rate and small enough not to be removed in the following precipitations and sand filters.

Except sedimentation, infiltration of groundwater also influence the activity concentration. For antropogenic nuclides the contribution of groundwater leads to dilution, while for ²²⁶Ra and ²²⁸Th (grand-daughter of ²²⁸Ra) a noticeable increase in the activity concentrations were observed, which can be explained by the comparatively high solubility of radium (Osmond and Ivanovich, 1992). The infiltration of groundwater also leads to an increased ²³⁴U/²³⁸U ratio due to alpha-recoil, where ²³⁴Th (grand-parent of ²³⁴U) recoils into the water medium, which is usually more significant in ground-water than surface water (Osmond and Ivanovich, 1992).

For most radionuclides studied, except for caesium and radium, most of the activity is removed by the two different precipitations (see table 5). A general trend for uranium and thorium is that the Fe(OH)₃ precipitation has a slightly less removal efficiency than the Al(OH)₃ precipitation. For plutonium and polonium the uncertainties in the activity concentration after the flocculation basins due to counting statistics are too large to draw this conclusion with any certainty. The increase of the polonium concentration after slow-filtration compared to the concentration after the flocculation basins could be explained by atmospheric deposition of polonium in the outdoor basins where slow-filtration takes place. Radium has approximately the same activity concentration in the incoming water as in the purified water distributed in the water mains, which shows that this purification process is not effective for radium-rich water.

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Radiocesium bioaccumulation in freshwater plankton: Influences of cation concentrations (K^+ and Na^+) on direct uptake of ^{137}Cs in *Chlamydomonas*, *Scenedesmus* and *Daphnia*. Food-chain transfer of ^{137}Cs from *Chlamydomonas* to *Daphnia* at different K^+ concentrations.

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Abstract

The influences of cation concentrations (K^+ and Na^+) on radiocesium (^{137}Cs) bioaccumulation in two freshwater phytoplankton species (*Scenedesmus quadricauda* and *Chlamydomonas noctigama*) were systematically investigated in batch-cultures monitored during two weeks. Both species were cultured at $9 \mu E m^{-2} s^{-1}$ constant illumination at $20^\circ C$. The exponential growth phase lasted for more than 100 hours ($\mu \approx 0.02 h^{-1}$ for *C. noctigama* and $0.03 h^{-1}$ for *S. quadricauda*). Over cation concentration ranges encountered in natural freshwaters ($[K^+]$ from $0.1 \mu M$ to $3 mM$, $[Na^+]$ from $20 \mu M$ to $3 mM$), a more than three order of magnitude variation was found for both uptake rate and observed bioconcentration factors (BCF) at apparent steady-state (from less than 10^3 to $10^6 L (kg C)^{-1}$). For both species, the major effector on BCF and uptake rate was external $[K^+]$, which was inversely proportional to these parameters over wide ranges ($1\text{--}1000 \mu M$ for *S. quadricauda* and 0.1 to $300 \mu M$ for *C. noctigama*). At concentrations above these ranges K^+ still reduced ^{137}Cs biouptake, but less effectively. A minor influence of external $[Na^+]$ on ^{137}Cs bioaccumulation was indicated for *S. quadricauda*, whereas no such influence was significant for *C. noctigama*. A biphasic pattern for ^{137}Cs bioaccumulation was discovered in *C. noctigama*. A rapid “quasi-steady-state” with an effective equilibration time of less than 100 hours was approached during the exponential growth phase. A surge in the uptake occurred when exponential growth ceased, and this pattern was consistent over the range $30 \mu M$ to $1.4 mM$ external $[K^+]$. Since depletion of external $[K^+]$ was not detected for these treatments, this pattern can only be explained if there are at least two different cellular compartments involved. Although less certain, a second steady-state BCF appeared within two weeks, which seems to be up to one order of magnitude higher than the first. Microcosm experiments with the freshwater zooplankter *Daphnia magna* were performed in order to address the influence of $[K^+]$ (ranging from 4.6 to $300 \mu M$) on the direct uptake of ^{137}Cs from the water in the absence of contaminated food. In other experiments (performed at 4.6 and $30 \mu M K^+$), non-contaminated individuals of *D. magna* were transferred to *Chlamydomonas* cultures that had approached steady-state BCF in order to address the relative importance of the different uptake routes of ^{137}Cs in freshwater zooplankton. Both direct uptake and food consumption contributed significantly to radiocesium bioaccumulation in *D. magna*, and observed steady-state bioaccumulation factors (BAF) were generally 2 –10 times higher than *Chlamydomonas* BCFs at corresponding external $[K^+]$.

Introduction

The ongoing contamination with radiocesium (^{134}Cs and ^{137}Cs) in the freshwater environments calls for a deeper comprehension of the biotic and physicochemical factors controlling the fate of these radionuclides in freshwater biota. In the Nordic countries, most of the recent ^{137}Cs in the freshwater environment originates from the Chernobyl accident in 1986, when considerable quantities of this radionuclide were deposited over northern Europe.

Since cesium is an alkali metal, it is highly soluble in the water, and exists almost exclusively as the monovalent cation Cs^+ in aqueous solution (1). These properties of cesium dictate a high degree of mobility and bioavailability in freshwater systems, in particular since the dissolved form is chemically similar to the potassium (K^+), sodium (Na^+) and ammonium (NH_4^+) ions (2-4). Despite its chemical relativeness to the essential cations K^+ and Na^+ , there exists no known biological function for Cs^+ (4).

It is well-established that the biouptake of ^{137}Cs is constituted by two major processes with different predominance depending on the trophic position of the organism, i.e. the direct uptake from the water (bioconcentration) and the uptake from contaminated food (5). Together, these processes contribute to the net bioaccumulation in organisms at all trophic positions of the pelagic foodwebs.

The bioaccumulation of ^{137}Cs in pelagic foodwebs is highly dependent on the ability of the primary producers to accumulate this radionuclide via bioconcentration from the water. Organisms at higher trophic levels (fish) accumulate ^{137}Cs mainly from the food, whereas zooplankton, smaller invertebrates, and juvenile fish are suggested to represent an intermediate position, where both bioconcentration and food consumption are important for the net bioaccumulation.

The steady-state net accumulation of ^{137}Cs in aquatic organisms can be described in terms of a bioaccumulation factor (BAF), defined as the ratio of the activity concentration in the organism to the ambient activity concentration. One of the advantages in using BAFs over absolute values of ^{137}Cs activity concentrations in organisms is that different geographical locations (lakes) become comparable even if the ambient ^{137}Cs activity concentrations are significantly different.

Among phytoplankton and other autotrophs, BAF is equivalent to the bioconcentration factor (BCF), since the net bioaccumulation in these organisms is a result from bioconcentration only. Consequently, BCFs for organisms at higher trophic positions cannot be obtained directly from *in situ* observations. Instead, these must be determined from controlled experimental studies.

Another interesting feature of radiocesium is that it has been reported to undergo biomagnification, i.e. the steady-state activity concentrations of ^{137}Cs in organisms increase with the trophic position in the aquatic food webs (6-9), and this standpoint has apparently most supporters at present. Very few are supporters of the opposite scenario, i.e. "biodiminution" (10). In contrast to many organic contaminants for which food chain biomagnification at presumed steady-state is a common phenomenon, this does normally not occur for inorganic metal ions.

Data from field studies (11-14) show that ^{137}Cs BAF for a given organism varies significantly among different lakes, and several *in situ* investigations have shown that this variation is significantly related to conductivity, cationic strength and water hardness (11-13, 15). In particular, it has been shown that among freshwater lakes ^{137}Cs BAF is inversely related to the K^+ concentration in the water. However, little information has been available about the influence of other major cations, which is

difficult to resolve from field data since $[K^+]$ in freshwaters usually is strongly correlated with that of other major cations.

The causal explanation to this pattern is so far attributed to the “nonisotopic carrier hypothesis” (16, 17), which suggests an inverse relationship between BAF of the radionuclide (e.g. ^{137}Cs) and the dissolved concentration of a chemically similar bioavailable nonisotopic analogue (other alkali cations). This holds only if the concentration of the chemically similar analogue is several orders of magnitude higher than the radionuclide, and that the analogue is homeostatically regulated in the organism (e.g. K^+ and Na^+) to a relatively constant concentration and thus relatively independent of the external concentration. This theory is particularly relevant to the bioconcentration process, and therefore its validity should be tested on phytoplankton and other micro-organisms only capable to accumulate the radionuclide and its bioavailable analogues directly from the water.

The intracellular concentration of K^+ in freshwater organisms is homeostatically regulated to about 100 - 150 mM (18-20). Since the $[K^+]$ in freshwaters typically ranges between 1 μM and 1 mM on a global scale, this means that intracellular $[K^+]$ can be 100- to 100000-fold higher than the ambient. In Swedish lakes $[K^+]$ is generally somewhat lower (Table 1). In freshwaters, Na^+ is the “second” cation with respect to intracellular concentrations, typically ranging between 5 -15 mM (21), and intracellular $[Na^+]/[K^+] \ll$ external $[Na^+]/[K^+]$ over these wide ranges.

Table 1. Typical ranges, medians and means of $[K^+]$ and $[Na^+]$ in μM covering about 95 % of more than 4100 Swedish lakes investigated during the year 1995 (Swedish University of Agricultural Sciences, RI-95).

cation	range (95 %)	median (μM)	mean (μM)
$[Na^+]$	18 - 790	82	183
$[K^+]$	3 - 88	12	22

The concentration of stable (nonanthropogenic) cesium in Swedish lakes, i.e. $[^{133}\text{Cs}^+]$, ranges between 0.02 and 0.2 nM (M. Meili, pers. comm).

Furthermore, two major different transport mechanisms operate in the uptake of K^+ by phytoplankton as well as other aquatic plants. At external $[K^+]$ in the micromolar range (below 200 μM), thermodynamically “active” uptake dominates, and this process is controlled by distinct transport systems, also referred to as the high-affinity K^+ transport systems (HAKT). These are energised by either H^+ -ATPases or Na^+ -ATPases which both pump protons or Na^+ out from the cell via ATP-hydrolysis. The HAKT is highly selective for K^+ and seems to discriminate strongly against other cations (also Cs^+ and Na^+). The second major transport mechanism is characterised by “passive” K^+ uptake via less specific K^+ channels, also known as low-affinity K^+ transport systems, operating mainly at $[K^+]$ above 1 mM. This system discriminates less effectively against other monovalent cations than the HAKT. Both major transport systems seem to operate in the range 200 to 1000 μM (22, 23), with a gradual transition from high-affinity to low-affinity upon increasing external $[K^+]$. Taken altogether, this implies that the ATP-driven, highly selective HAKT should be the dominant one in the freshwater environment.

Given these precautions, the “nonisotopic carrier” hypothesis can be expressed, provided that K^+ is the only significant analogue to ^{137}Cs , and that $[K^+]_w \gg [^{137}\text{Cs}]_w$:

$$\frac{[^{137}\text{Cs}]_w}{[K^+]_w} = S_{\text{Cs/K}} \frac{\text{steady-state } [^{137}\text{Cs}]_{\text{organism}}}{[K^+]_{\text{organism}}} \quad (\text{eq. 1})$$

where $S_{Cs/K}$ denotes the discrimination constant or selectivity coefficient (Polikarpov, 1966) between Cs^+ and K^+ , which should be constant as far as the K^+ transport systems are identical over a given range of external $[K^+]$. This can be expected true at $[K^+]$ well below 1000 μM , since only the HAKT should operate in this range. Since it is further assumed that also $[K^+]_{organism}$ is constant over this range due to homeostasis, rearrangement and log-transformation of (2) gives:

$$\log(^{Cs} BCF) = \log\left(\frac{\text{steady-state } [^{137}Cs]_{organism}}{[^{137}Cs]_w}\right) = \text{constant} - \log [K^+]_w \quad (\text{eq. 2})$$

This theoretical expression (3) may be an oversimplification, not only because it ignores other abundant cations (such as Na^+), but also for the reason that there are several other environmental parameters, which can further influence the ^{137}Cs bioconcentration. Additionally, the Cs/K selectivity may depend on the type of transport system, which in turn depends on external $[K^+]$, implying that $S_{Cs/K}$ may not be constant over the wide range of $[K^+]$ encountered in freshwaters.

The main scope of this experimental study is to determine to which extent the major monovalent metal cations K^+ and Na^+ influence the bioavailability of ^{137}Cs in plankton organisms. Another aspect of this study is to be able to distinguish between the relative contributions from bioconcentration and food uptake of ^{137}Cs in zooplankton, which also gives a clue to whether or not ^{137}Cs should undergo biomagnification in the first link of the pelagic food-chain. To achieve the knowledge to all this, the experimental study was designed to understand the influences of varying ambient $[K^+]$ and $[Na^+]$ on the bioconcentration kinetics in two freshwater phytoplankters (*Chlamydomonas noctigama* Korsh. and *Scenedesmus quadricauda* Turp.) and the bioaccumulation kinetics in a freshwater zooplankter (*Daphnia magna* Strauss).

Experimental Section

Three types of experiments were conducted in order to study the:

1. Bioconcentration of ^{137}Cs by the microalgae *Scenedesmus quadricauda* and *Chlamydomonas noctigama* under the influence of varying ambient $[K^+]$ and $[Na^+]$
2. Bioconcentration (only direct uptake) of ^{137}Cs by the crustacean *Daphnia magna* under the influence of varying ambient $[K^+]$ in the absence of contaminated food
3. Bioaccumulation of ^{137}Cs by *Daphnia magna* under the influence of varying ambient $[K^+]$ in the presence of contaminated food (*Chlamydomonas noctigama*)

1. Phytoplankton experiments

Axenic batch-cultures of *Scenedesmus quadricauda* (Turp.) and *Chlamydomonas noctigama* (Korsh.) were spiked with ^{137}Cs to a total activity concentration of about 100 kBq L^{-1} .

The subsequent cellular accumulation resulting from direct uptake of ^{137}Cs during the experiment time (2 weeks) was described as the concentration ratio between the organism and the external environment (BCR). From the temporal trend of BCR, the steady-state bioconcentration factor (BCF) and rate constants for uptake, elimination and equilibration were evaluated.

Both BCF and BCR are here expressed on the basis of *particulate organic carbon* (POC), equivalent to the frequently used K_{OC} , with units of $L (kg POC)^{-1}$. According to Olrik et al. (24), the

carbon content of the wet biomass among chlorophycean microalgae is approximately 16%, which roughly would yield a factor of 6 between K_{OC} and a wet-weight based BCF. However, for microalgae in general, a more conservative estimator of the wet weight based carbon content is 12% (25), which instead implies a conversion factor of 8 between K_{OC} and a wet-weight based BCF.

Experimental design. Uptake kinetics and bioconcentration ratios of ^{137}Cs and major cations were studied along combined gradients of $[\text{K}^+]$ and $[\text{Na}^+]$ in the culture medium. The *Chlamydomonas* experiments consisted of:

- A. Eight treatments where the sum of K^+ and Na^+ was 1.4 mM. The K^+ concentrations ranged from 0.5 μM to 1.4 mM, and Na^+ concentrations ranged from 20 μM to 1.4 mM.
- B. Three treatments where Na^+ was kept at 1.4 mM μM while K^+ concentrations were 4.5, 30 and 300 μM .
- C. Three treatments where K^+ was kept at 1.4 mM while Na^+ concentrations were 20, 60 and 300 μM .

The *Scenedesmus* experiments consisted of:

- A. Four treatments where the Na^+ concentration was maintained at 2.9 mM, while K^+ ranged from 0.1 μM to 1 mM.
- B. One treatment where the K^+ concentration was 2.9 mM while the Na^+ concentration was reduced to 40 μM .

This strategy was chosen to distinguish the relative effects of sodium and potassium without substantially changing the ionic strength of monovalent cations in the medium.

The inoculated biomass was between 0.5 - 2 mg POC L^{-1} , in order to allow for algal growth while minimising ionic depletion of the dissolved phase. All ionic gradients were obtained by modification of Lindström's (26) algal culture medium 5•L16. All treatments were set up at least in triplicates.

Algal cultures. Inoculated batch cultures of *S. quadricauda* and *C. noctigama* were allowed to equilibrate with the culture medium for at least 24 hours in the laboratory before addition of ^{137}Cs . Both species were cultured at 9 $\mu\text{E m}^{-2} \text{s}^{-1}$ constant illumination under fluorescent white light tubes (Philips TLD 18W). The temperature was about 20°C in all preincubations and experiments.

Estimation of algal biomass. Measurements of algal biomass concentration in controls prior to incubation, and at all later sampling occasions, were performed on a Hitachi U-2000 double-beam spectrophotometer, using the absorbance at 680 and 420 nm. From a standard curve of the relationship between absorbance and mg algal POC L^{-1} , the biomass in controls could be estimated and expressed as mg POC L^{-1} .

Sampling. The first aliquots were taken shortly after incubation (0-4 hours). At each sampling occasion 10 mL were taken with a pipette from incubates and controls. Prior to sampling, the culture flasks were irregularly shaken for 15 seconds in order to obtain homogenous cultures. The aliquots were first transferred to polyethylene scintillation vials and measured with respect to total radioactivity, then transferred to 15 mL styrene centrifuge tubes and centrifuged at 5000 rpm (4620 g) for 5 min. After that, the supernatant was removed and the pellet resuspended in clean medium, thereafter centrifuged as above. This procedure was repeated once more to ensure that no remaining radioactivity was present in the solution, as determined from pilot experiments. The pellet was then resuspended in 10 mL and transferred to polyethylene scintillation vials. After allowing sedimentation of the pellet to increase detector efficiency, the radioactivity was measured.

2. Zooplankton experiments

2A. ^{137}Cs bioconcentration in *D. magna*

Experimental design. To study the influence of $[\text{K}^+]$ on ^{137}Cs bioconcentration in *D. magna*, three experimental treatments were performed, where the K^+ concentrations were 4.5, 30, and 300 μM respectively. The Na^+ concentration was kept constant at 1.4 mM.

Experimental procedures. Non-contaminated individuals of *D. magna* were transferred to sterile spiked medium (100 kBq L^{-1}) and the subsequent accumulation resulting from direct uptake was recorded during 24 hours. Samplings for measurement of radioactivity and carbon biomass of the animals were conducted at 1.5, 3, 6, 12, and 24 hours of exposure to ^{137}Cs . The activity concentration in the culture medium was also determined at each sampling occasion. The experiments were performed in 1000 mL glass beakers with 200 mL culture medium. Each treatment consisted of three replicates and each replicate originally consisted of 30 animals (150 ind. L^{-1}).

Animals. The animals used in these experiments were about one week old (about 1 mm in length), and were selected such that the distribution of their individual size was as small as possible prior to the experiment start-up.

2B. ^{137}Cs bioaccumulation in *D. magna*

Experimental design. To study the influence of $[\text{K}^+]$ on the ^{137}Cs bioaccumulation in *D. magna*, two triplicate treatments were performed, where the K^+ concentrations were 4.5 and 30 μM respectively. The $[\text{Na}^+]$ was 1.4 mM in both treatments (cf section 2A).

Experimental procedures. Non-contaminated individuals of *D. magna* were transferred to dilute (3.5 mg POC L^{-1}) and ^{137}Cs contaminated (100 kBq L^{-1}) *Chlamydomonas* cultures, growing at 9 $\mu\text{E m}^{-2} \text{s}^{-1}$ constant illumination ($\mu \approx 0.02 \text{ h}^{-1}$). The algal ^{137}Cs activity concentration was in steady-state with the activity concentration in the medium at the experimental start-up, i.e. at the time the daphnids were added to the culture. The subsequent bioaccumulation resulting from direct uptake and feeding on contaminated algae was monitored during 12 days and described as a bioaccumulation ratio (BAR; L (kg C)^{-1}) in analogy with the term BCR. Control replicates of the *Chlamydomonas* cultures with no animals added, but otherwise treated in the same way (inoculation, light conditions and experimental volume), were used to estimate the initial growth rate and to compare the temporal trend of ^{137}Cs BCR with the grazed experimental cultures. Samplings for measurement of radioactivity and carbon biomass of the animals during these 12 days were conducted after 3, 6, 12, 24, 48, 96, 192, and 288 hours of exposure to ^{137}Cs .

Algal cultures. The algal cultures used in these experiments were prepared by inoculating the spiked 4.5 $\mu\text{M K}^+$ and 30 $\mu\text{M K}^+$ media with cells from exponentially growing *Chlamydomonas* cultures (9 $\mu\text{E m}^{-2} \text{s}^{-1}$ constant illumination; $\mu \approx 0.02 \text{ h}^{-1}$) that had accumulated ^{137}Cs for four days in the corresponding media. The cellular ^{137}Cs activity concentration was near steady-state at the time of inoculation. The algal biomass in controls and animal batches was measured spectrophotometrically at each sampling occasion according to the schedule above. Samplings for measurement of algal ^{137}Cs activity concentrations were performed at all timepoints except at 6 and 12 hours.

Animals. The animals used in these experiments were one day old and relatively uniform in size (about 0.5 mm in length) at the experiment start-up.

2C. Methods common to all zooplankton experiments

Sampling. The individuals were taken from the experimental vessels by pipette and placed into a small sieve, allowing the ^{137}Cs solution to run off. The specimen was washed by sequentially dipping the sieve into six beakers containing clean medium. Then the sample was placed in a small tin capsule using two pinches and dried at 95 °C for 48 hours. After drying, the tin capsules were placed in scintillation vials for measurement of ^{137}Cs radioactivity.

Estimation of carbon biomass. The carbon content in the Sn-capsules was determined in an elemental analyser (LECO CHNS-932) after measurement of ^{137}Cs radioactivity. For estimation of growth rate it is necessary to estimate the carbon biomass of the individuals ($\mu\text{g C ind}^{-1}$), which was done by dividing the amount of carbon capsule by the number of individuals in the tin capsule.

Methods common to all phytoplankton and *Daphnia* experiments

Addition of ^{137}Cs . For the addition of ^{137}Cs to the experimental media, 20 μL of a stock solution (Amersham) with a specific activity of about 1 kBq (ng Cs) $^{-1}$ (34 $\mu\text{g CsCl mL}^{-1}$, 33 MBq mL^{-1}) were transferred to 180 mL ultrapure deionized water (24 M Ω), resulting in a handy stock solution of 3.7 MBq L^{-1} . At incubation, 2.5 mL of prepared ^{137}Cs solution was added to each culture flask, resulting in a total [Cs^+] of 100 ng L^{-1} ($8 \cdot 10^{-4}$ μM), or about 100 kBq L^{-1} (31 ng L^{-1}) of ^{137}Cs .

Measurement of ^{137}Cs activity concentrations. ^{137}Cs activity concentrations were measured with an Intertek Model CG-4000 gamma counter equipped with a 3•3 inch NaI crystal well detector and an autosampler. Counting thresholds were set to $2 \cdot 10^4$ counts or 40 minutes of counting time, although 100 minutes were usually set for samples presumed to contain very small amounts of ^{137}Cs . Background measurements were made by using blanks for at least every tenth measurement to correct for background radiation. Long-term time series of blank measurements (100 min. each) were also made to gain accurate statistics of the background activities and to ensure that temporal variation was negligible. The resulting counting error was usually 1-5%. Readings were corrected for sample geometry and converted to ^{137}Cs activities by comparison with readings from standard solutions.

Analysis of dissolved cations in the culture medium. After addition of stabilising agents (LaCl_3 for Mg^{2+} and Ca^{2+} ; CsCl for K^+ and Na^+), the dissolved concentrations of K^+ , Ca^{2+} , Na^+ and Mg^{2+} were analysed on a Shimadzu AA-670 AAS/FES spectrophotometer (AAS: Ca^{2+} , Mg^{2+} ; FES: Na^+ , K^+) with reference to appropriate standard solutions.

Results

^{137}Cs bioconcentration in *C. noctigama* and *S. quadricauda*

The overall results strongly indicate a biphasic ^{137}Cs bioaccumulation in *C. noctigama* related to the transition from exponential growth to the stationary growth phase. The exponential growth phase ($\mu \approx 0.02 \text{ h}^{-1}$) lasted for about 100 hours in *C. noctigama* cultures, and during this timespan ^{137}Cs bioaccumulation approached a "quasi-steady-state", having an effective equilibration time of about

24 to 100 hours. The second phase of Cs accumulation was initiated after 100 hours, coincident with the gradual decline in specific growth rate. A second steady-state of Cs accumulation, which was about one order of magnitude higher than the first, seems to be approached within 16 days when the specific growth rate was close to zero (Figure 1).

In the treatments where $[K^+]$ was initially below 10 μM , it is obvious that depletion of dissolved K^+ in the medium as a result from biomass growth, results in an increasing uptake rate of Cs during accumulation.

However, the biphasic pattern cannot be explained by depletion of K^+ since it occurred in all treatments, and no significant depletion of K^+ was found for treatments above 10 μM initial $[K^+]$. The biphasic pattern of Cs accumulation was thus independent of ambient $[K^+]$ at initial $[K^+] > 10 \mu\text{M}$.

During the exponential growth phase, i.e. the first 100 hours, *S. quadricauda* accumulated about 3-5 times more Cs than *C. noctigama* when comparing similar treatments at external $[K^+]$ below 1000 μM . However, at 288 hours levels of Cs accumulation in *C. noctigama* were of the same order of magnitude or higher than those in *S. quadricauda* at 100 hours. Also *S. quadricauda* seems to exhibit a biphasic accumulation pattern of ^{137}Cs coincident with the transition from exponential growth ($\mu \approx 0.03 \text{ h}^{-1}$) to stationary growth, but the experimental duration was not long enough to reveal the second plateau of ^{137}Cs accumulation. The exponential growth phase of *S. quadricauda* cultures normally lasted for at least 170 hours, and a surge in the ^{137}Cs uptake occurred also in this case when growth rate declined after that timepoint. The effective equilibration time of ^{137}Cs bioaccumulation in *S. quadricauda* was about 100 hours (before μ declined) provided that $[K^+]$ did not change during this time.

Due to higher productivity in *Scenedesmus* cultures than in those of *Chlamydomonas* there was a more pronounced influence of declining $[K^+]$ on the temporal trend of Cs bioaccumulation in the *Scenedesmus* cultures. In particular the depletion of K^+ at initial $[K^+] \leq 5 \mu\text{M}$ was significant during the experimental time. Ambient $[K^+]$ remained virtually constant during the entire experimental timespan only at $K \geq 1 \text{ mM}$.

Net ^{137}Cs accumulation in *S. quadricauda* at 100 hours (near steady state) expressed as BCR was inversely related to ambient $[K^+]$ over the range 1 μM to 1 mM (Figure 2). External $[\text{Na}^+]$ might have an additional influence on ^{137}Cs accumulation in *S. quadricauda* since BCR (100 hours) was substantially higher at 2.9 mM $[K^+]$ ($[\text{Na}^+] = 40 \mu\text{M}$) than at 1000 μM K^+ ($[\text{Na}^+] = 2.9 \text{ mM}$).

In *C. noctigama*, net ^{137}Cs accumulation at 48 hours (the first plateau) expressed as BCR was inversely related to $[K^+]$ only over the range 0.1 μM to 300 μM (Figure 2). Above 300 μM K^+ still reduced ^{137}Cs biouptake, but less effectively. This pattern remains also at 288 hours when ^{137}Cs accumulation approached the second plateau with the only difference that BCR were one order of magnitude higher. No obvious influence from external $[\text{Na}^+]$ on Cs bioaccumulation in *C. noctigama* was found at any of these timepoints (Figure 3).

The experimental results given in Figure 2 indicate the possible transition from a high-affinity K^+ uptake mechanism to a low-affinity K^+ uptake mechanism with higher Cs/K selectivity when $[K^+]$ is increased above 300 μM (cf equation 2). Another, but less likely possibility is that the cells accumulate more K^+ above this concentration without changing the Cs/K selectivity.

¹³⁷Cs bioconcentration in *D. magna*

The ¹³⁷Cs bioconcentration in *D. magna* resulting from direct uptake during 24 hours was significantly reduced upon increasing ambient [K⁺] over the range 4.6 to 300 μM (Figure 4). However, the relationship between BCR and [K⁺] was far from an inverse, having a log-log slope of about -0.5. Again, this pattern might be explained in terms of changing Cs/K selectivity.

¹³⁷Cs bioaccumulation in *D. magna*

Steady-state ¹³⁷Cs bioaccumulation factors (BAF_D) were not obtained in these combined experiments since the algae showed an unexpected loss of cellular Cs (Figure 5). It was not until after the animals were added (48 hours after inoculation) to the experimental vessels that the algae began to release cellular Cs. The second inoculation with contaminated algae after 96 hours also failed to maintain constant cellular ¹³⁷Cs levels. However, the controls (with no animals added) maintained the original levels of cellular ¹³⁷Cs during the exponential growth phase in accordance with previous results.

Nevertheless, the experimental results suggest that the ¹³⁷Cs activity concentration in the food significantly influences the net bioaccumulation in *D. magna*. When comparing the short-term bioconcentration experiments with the combined experiments during the first 24 hours (Figure 6) it is obvious that the feeding animals accumulated faster than the starving in the beginning (BAR_D > BCR_D), but also that the rapid decline in algal BCR during this time finally reduced the difference. The most notable feature is the significant pulse in *Daphnia* BAR at 144 hours, i.e. after reinoculation with *C. noctigama* cells at 96 hours. After that timepoint BAR_D declines to the levels at 48 to 96 hours, while *Chlamydomonas* BCR fall to levels 10-30 times below the steady-states levels in the controls. Thus, BAR_D at 48, 96 and 288 hours are likely equivalent to BCF_D in the absence of food. Taken altogether, it is obvious that BAR_D closely tracks *Chlamydomonas* BCR, and it therefore seems that both uptake routes should be equally significant if the food has the same bioconcentration factor as *D. magna*.

For this reason a rough estimate of the biomagnification factor for *D. magna* (BMF_D) can be obtained, although neither of the organisms have experimental ¹³⁷Cs concentration factors in steady-state. The steady-state definition of BAF in *D. magna* is

$$\text{BAF}_D = \text{BCF}_D + (\text{BMF}_D \cdot \text{BCF}_C)$$

where the indexes D and C denote *Daphnia* and *Chlamydomonas*, respectively. By this definition it follows that

$$\text{BMF}_D = \frac{\text{BAF}_D - \text{BCF}_D}{\text{BCF}_C}$$

Since BCF_D is approached within 96 hours combined with the fact that BAR_D closely tracks BCR_C, the following approximation give a rough estimate of BMF:

$$\text{BMF}_D = \frac{\text{BAF}_D - \text{BCF}_D}{\text{BCF}_C} \approx \frac{\text{BAR}_D (144 \text{ h}) - \text{BCF}_D}{\text{BCR}_C (144 \text{ h})}$$

It is here assumed that BAR_D (48 h) and BAR_D (96 h) which are approximately equal, are mainly a result from bioconcentration only and hence that the average of these can be set to BCF_D .

This yields an estimated BMF ($\pm\text{SE}$) of 1.8 ± 1.4 at $4.6\ \mu\text{M K}^+$ and 2.4 ± 0.6 at $30\ \mu\text{M K}^+$. Consequently, if the *Chlamydomonas* cells would have maintained intracellular ^{137}Cs levels corresponding to the first steady-state, there would have been a significant contribution from the feeding route to the *Daphnia* bioaccumulation at all timepoints.

The experiments fail to maintain steady-state levels of *Chlamydomonas* BCR but yet the results confirm that Cs bioaccumulation in *D. magna* is significantly dependent on both uptake routes. The most important implication is that the bioconcentration factors should contribute with at least half of the net bioaccumulation factor in *D. magna*. On the other hand, availability to food should be an important ecological factor with profound influence on the net ^{137}Cs BAFs in zooplankton.

Conclusions

Microcosm experiments with the freshwater microalgae *Scenedesmus quadricauda* and *Chlamydomonas noctigama* suggest that the concentration factor of ^{137}Cs in freshwater phytoplankton is inversely related to ambient $[\text{K}^+]$ over the wide range ($1\ \mu\text{M}$ to $1\ \text{mM}$) covered by Scandinavian lakes. Over cation concentration ranges encountered in natural freshwaters ($[\text{K}^+]$ from $0.1\ \mu\text{M}$ to $3\ \text{mM}$, $[\text{Na}^+]$ from $20\ \mu\text{M}$ to $3\ \text{mM}$), a more than three order of magnitude variation was found for both uptake rate and observed bioconcentration factors (BCF) at apparent steady-state (from less than 10^3 to $10^6\ \text{L (kg C)}^{-1}$). Consequently, the bioconcentration factor of ^{137}Cs in phytoplankton can be expected to range three orders of magnitude among Scandinavian lakes.

For both species, the major effector on BCF and uptake rate was external $[\text{K}^+]$, which was inversely proportional to these parameters over wide ranges (1 to $1000\ \mu\text{M}$ for *S. quadricauda* and 0.1 to $300\ \mu\text{M}$ for *C. noctigama*). At concentrations above these ranges K^+ still reduced ^{137}Cs biouptake, but less effectively.

A minor influence of external $[\text{Na}^+]$ on ^{137}Cs bioaccumulation was indicated for *S. quadricauda*, whereas no such influence was significant for *C. noctigama*.

Concerning the first phase of accumulation, i.e. during the exponential growth phase, the algae exhibit substantial differences in ^{137}Cs bioconcentration kinetics. The effective equilibration time during the first phase of accumulation is about 48 hours for *C. noctigama* (at $[\text{K}^+] > 3\ \mu\text{M}$), and about 100 hours for *S. quadricauda* (at $[\text{K}^+] \geq 100\ \mu\text{M}$). A comparison between similar $[\text{K}^+]$ treatments also shows that *S. quadricauda* accumulated 3 – 5 times more ^{137}Cs than *C. noctigama* during the first 100 hours at $[\text{K}^+]$ below $1000\ \mu\text{M}$.

The results from these experiments, dealing only with the influence of cation chemistry on ^{137}Cs bioconcentration in phytoplankton, suggest that it is also very important to understand the influence of growth conditions on ^{137}Cs bioconcentration kinetics.

Microcosm experiments with the freshwater zooplankter *Daphnia magna* suggest that both direct uptake and ingestion of contaminated food contribute significantly to net bioaccumulation of ^{137}Cs . The ^{137}Cs bioconcentration factors in *D. magna* are in the same order of magnitude as the bioconcentration factors in the microalgae *S. quadricauda* and *C. noctigama*, ranging from $5\cdot10^3\ \text{L (kg C)}^{-1}$ to $5\cdot10^2\ \text{L (kg C)}^{-1}$ when K^+ ranges from $4.6\ \mu\text{M}$ to $300\ \mu\text{M}$. The effective equilibration

time appears to be about 48 hours. Steady-state ^{137}Cs BAFs in *D. magna*, resulting from food consumption and direct uptake can be expected to be about 2-fold higher than the BCFs resulting from only direct uptake if the food (phytoplankton) has similar bioconcentration factors.

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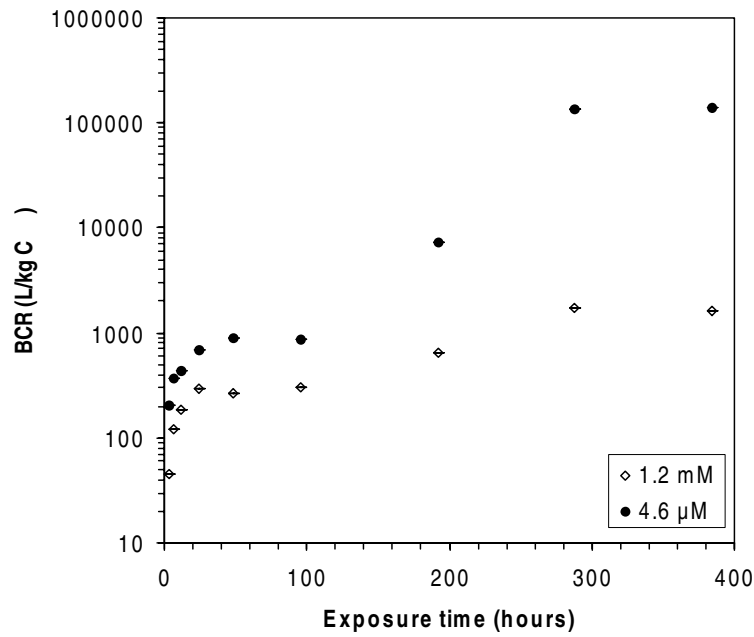


Figure 1. The temporal trend of bioconcentration ratios (L (kg C)^{-1} ; geometric mean \pm geometric SD; $n = 3$) for ^{137}Cs in the freshwater microalga *Chlamydomonas noctigama*. The biphasic ^{137}Cs accumulation is illustrated by two treatments ($4.6 \mu\text{M}$ and 1.2 mM K^+) covering the wide range of $[\text{K}^+]$ found in natural freshwaters.

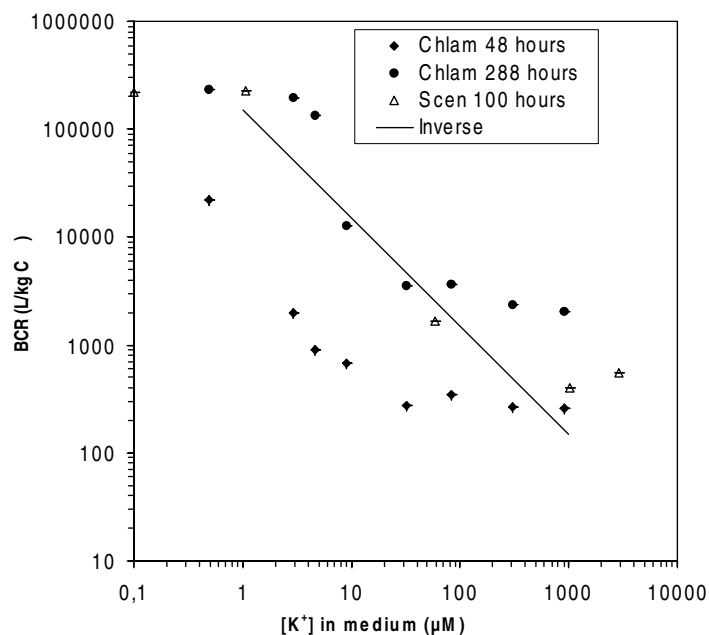


Figure 2. Bioconcentration ratios (L (kg C)^{-1} ; geometric mean \pm geometric SD; $n = 3$) for ^{137}Cs in the freshwater microalgae *Chlamydomonas noctigama* (48 and 288 hours) and *Scenedesmus quadricauda* (approx. 100 hours) at apparent steady-state versus ambient $[\text{K}^+]$ in the culture medium. Both phases are indicated for *C. noctigama*, i.e. the steady-state BCRs during the exponential growth phase ($\mu \approx 0.02 \text{ h}^{-1}$) at 48 hours and the BCRs at 288 hours ($\mu \approx 0 \text{ h}^{-1}$) approaching the second plateau. The solid line with a log-log slope -1 indicates the inverse proportionality between BCR and $[\text{K}^+]$ according to the nonisotopic carrier hypothesis. Note the deviations from this slope at $[\text{K}^+]$ outside the range $1 \mu\text{M}$ to $300 \mu\text{M}$. *Scenedesmus*: $[\text{Na}^+] = 2.9 \text{ mM}$ at all $[\text{K}^+]$ up to $1000 \mu\text{M}$ and reduced to $40 \mu\text{M}$ at the highest $[\text{K}^+]$ (2.9 mM). *Chlamydomonas*: $[\text{Na}^+] = 1 \text{ mM}$ at all $[\text{K}^+]$ up to $300 \mu\text{M}$, and reduced to $300 \mu\text{M}$ at the highest $[\text{K}^+]$ (1 mM).

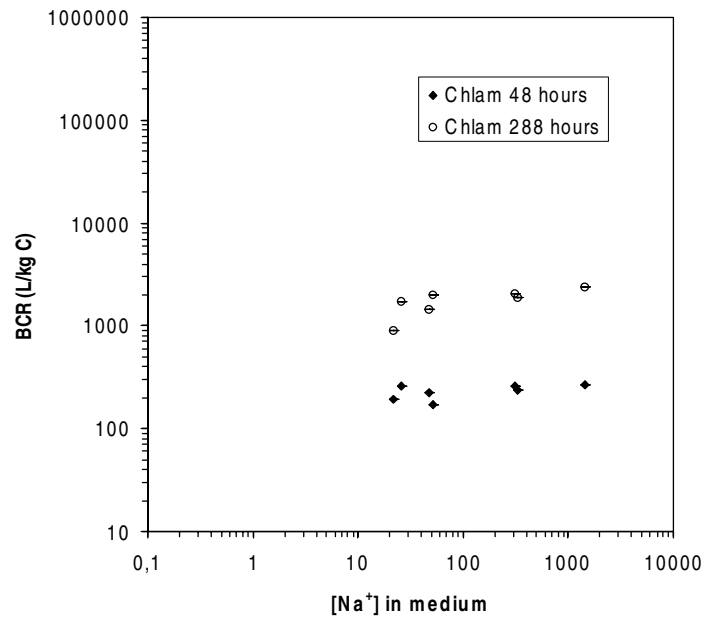


Figure 3. Bioconcentration ratios (L (kg C)^{-1} ; geometric mean \pm geometric SD; $n = 3$) for ^{137}Cs in the freshwater microalga *Chlamydomonas noctigama* (48 and 288 hours) at apparent steady-state versus ambient $[\text{Na}^+]$ in the culture medium. The K^+ concentration is about 1 mM in all treatments up to 300 μM $[\text{Na}^+]$. At the highest $[\text{Na}^+]$ (1 mM), the K^+ concentration is about 300 μM (which overlaps with Figure 2).

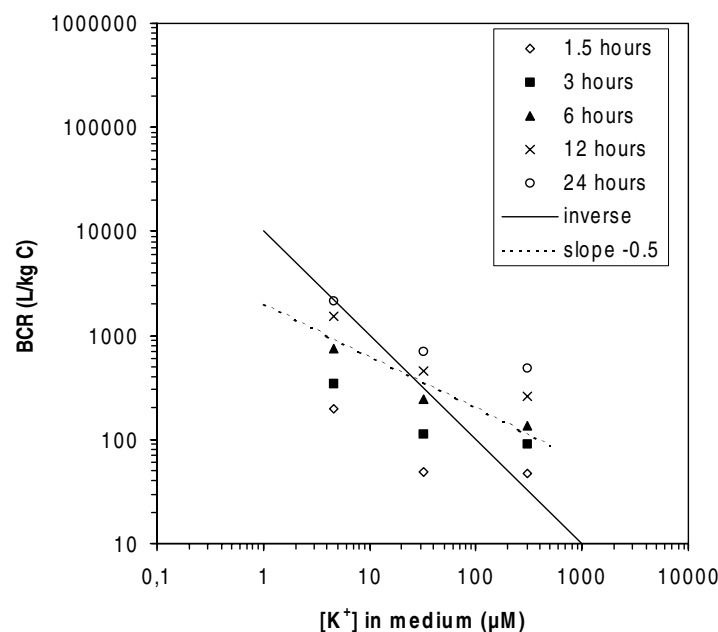


Figure 4. The temporal trend of bioconcentration ratios (L (kg C)^{-1} ; geometric means; $n = 3$) for ^{137}Cs in the freshwater zooplankter *Daphnia magna* during 24 hours of direct uptake versus ambient $[\text{K}^+]$ in the culture medium. Note the deviations from the solid line (cf Fig. 2).

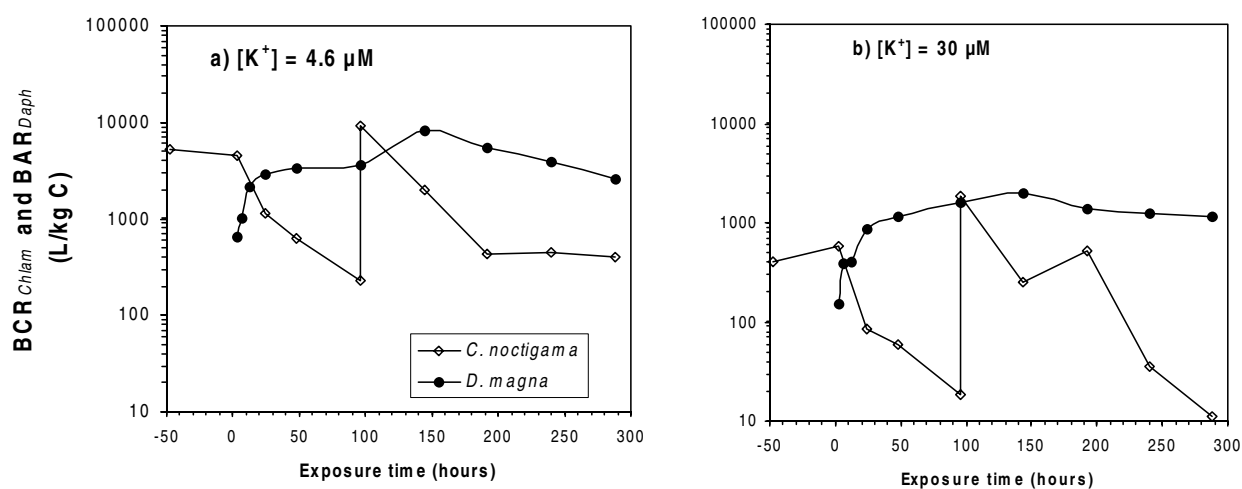


Figure 5. The temporal trend of ^{137}Cs bioaccumulation (BAR ; L (kg C)^{-1} ; geometric means; $n = 3$) in *D. magna* feeding on *C. noctigama* and the temporal trend of bioconcentration ratios (L (kg C)^{-1} ; geometric means; $n = 3$) for ^{137}Cs in *C. noctigama* at two different $[\text{K}^+]$. Note the unexpected decline in *C. noctigama* BCR after the animals were added. Also note the significant pulse in *D. magna* ^{137}Cs bioaccumulation at 144 hours after reinoculation at 96 hours.

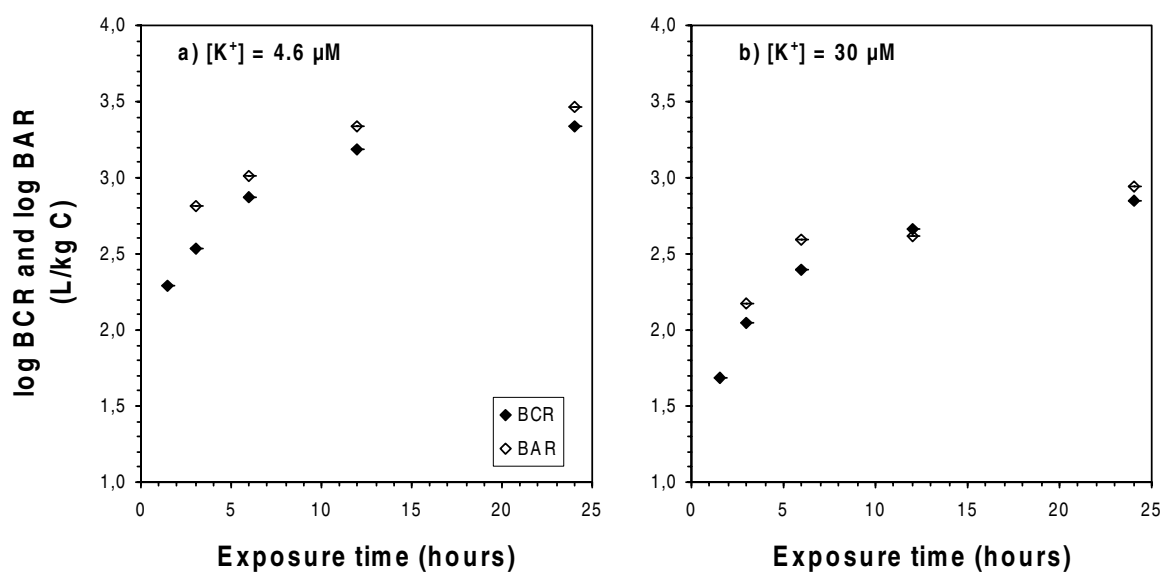


Figure 6. The temporal trend of logBCR ($L (kg C)^{-1}$; means; $n = 3$) in *D. magna* resulting from direct uptake only, and the temporal trend of logBAR ($L (kg C)^{-1}$; means; $n = 3$) in *D. magna* resulting from an additional consumption of contaminated *C. noctigama*. Note that the feeding animals accumulated faster than the starving in the beginning ($BAR > BCR$), i. e. before *C. noctigama* ^{137}Cs levels had declined too much to contribute to net bioaccumulation in the feeding animals (cf Fig. 5).

Preliminary report of radiocesium (Cs^{137}) in freshwater fish within Murmansk and Archangelsk region, Russia and the Pasvik River in Finnmark County, Norway

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Abstract

This report gives a preliminary status of the project "radiocesium in freshwater fish within Murmansk and Archangelsk region, Russia and the Pasvik river in Finnmark County, Norway". This is a co-operation between Norwegian College of Fisheries Science, (university of Tromsø), INEP Apatitt (Institute of the North Industrial Ecology Problems) in Russia and Norwegian Radiation Protection Authority.

Muscles of fish for radionuclide analyses have been collected since 1994 till 2000 within these areas. The analysed samples in this report come from the river Petchora and lake Imandra. In addition to these two areas there are expected more samples from Harlovka river, lake Endozero and the Pasvik river in Norway.

The Petchora is situated in the North-East district of European Russia, starting at the Western slopes of the Northern Ural and ending in the Petchora bay (in the Barents Sea) at Naryan Mar in the Nenets Autonom Area at 53° - 55° E and 68° N. The Imandra lake is the largest lake of Murmansk region situated in the centre of the Kola peninsula, located at 31° $30'$ - 33° $30'$ E and 67° $30'$ - 68° N. The Pasvik river in Finnmark County in Norway is located approx. at 33° E and 69° $30'$ N.

The results (radiocesium) from the about 40 samples are located in a database which also contains detailed information about the samples and the measurements. This report, however, describes the locality from where the samples are collected. This includes type of lake, biological / chemical conditions, ichthyofauna and species determination from the areas. The results from the different measurements are also present.

Distribution of ^{137}Cs and ^{90}Sr in various tissues and organs of freshwater fish in lakes from Finland

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Abstract

The degree of radiocontamination in fish is usually analysed to estimate doses to fish consumers, for which the radionuclide contents in the edible parts of fish are needed. Recently increasing attention has been focused on radiation protection in plants and animals. To estimate the effects of radiation on fish, the distribution of radionuclides in various organs and radiosensitivity of the organs must be known. A step towards this was taken when the Radiation and Nuclear Safety Authority - STUK of Finland determined the distribution of ^{137}Cs and ^{90}Sr in muscle tissue, skin+fins, and bones in perch samples, in muscle and bones or in muscle, skin+fins, and bones in pike samples and in muscle+skin+fins and bones in vendace samples. Accumulation of ^{137}Cs in fish liver, spawn and sperm was also studied and compared with respective values in the edible parts. The importance of skin and fins was evident as a source of ^{90}Sr to fish. Fish sperm showed somewhat higher activity concentrations of ^{137}Cs than muscle.

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Transuranic elements in fishes compared to ^{137}Cs in certain Finnish lakes

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Abstract

Transuranic elements from Chernobyl fallout were detected in freshwater fish sampled in Finland after the accident. Maximum concentrations of $^{239,240}\text{Pu}$, ^{241}Pu , ^{241}Am and ^{242}Cm were 3.3, 110, 2.6 and 340 mBq kg^{-1} , respectively. In certain samples, $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios were between 1.0 and 7.2 while the expected value was 0.5. This could indicate differences in environmental mobilization between ^{238}Pu and $^{239,240}\text{Pu}$. The concentration factor (CF) for Am from water to fish flesh, 230, was more than 2 times higher than the figure of 84 for Pu. The CF for Cm, 5100, was considerably higher than that of Am and does not confirm the presence of similar biological behaviour for Am and Cm. The average transfer factor (TF) from deposition to fish was highest for ^{137}Cs , $44 \times 10^{-3} \text{ m}^2 \text{ kg}^{-1}$. This was somewhat higher than the TF for Am but more than 10 times higher than that for Pu. Transuranic depositions that could cause the European Council regulation limit in freshwater fish to be exceeded are, Pu and Am respectively, 3000 and 1500 times higher than the highest fallout of Pu and Am recorded in Finland following the Chernobyl accident.

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High levels of ^{129}I in rivers of south Sweden

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Abstract

The concentration of iodine-129 was measured in water samples collected during summer 1999 from ten rivers located in southern Sweden. The results show ^{129}I concentrations ranging from 4×10^8 to 1.4×10^9 atoms per liter. This range is comparable to that observed in lake and river waters of central Sweden, but includes some of the highest values ever recorded in low-salinity freshwaters without direct discharge of ^{129}I from a nuclear installation. The globally most important source of ^{129}I is presently the discharges (marine and atmospheric) from the nuclear reprocessing facilities at La Hague (France) and Sellafield (UK). The marine discharges from these facilities have increased the level of ^{129}I in the North Sea by about 5 orders of magnitude above natural background. Our study indicates a similar increase also in remote European freshwaters, although concentrations are about 2 orders of magnitude lower than those in the North Sea. The data suggest a substantial atmospheric deposition of ^{129}I discharged from reprocessing facilities, which is supported by recent precipitation measurements in central Sweden. Furthermore, it appears that recent deposition is already evident in runoff waters. This opens the question of whether and when concentrations of ^{129}I in Nordic freshwaters will increase to levels comparable to those presently encountered in the North Sea.

French and British Emissions of Radioactive Iodine-129 in the Baltic Sea

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Iodine-129 (15.7 Ma) is a naturally occurring as well as a manmade radioisotope of iodine. The ratio of $^{129}\text{I}/^{127}\text{I}$ was estimated to be $\sim 10^{-12}$ in the ocean in the pre-nuclear era, releases from nuclear weapon tests have increased this ratio to $\sim 10^{-10}$. However, the greatest releases of ^{129}I are from two European reprocessing plants, especially in recent years. By 1998, 1860 kg and 760 kg ^{129}I have been discharged to the marine environment from La Hague (France) and Sellafield, respectively. This is an order of magnitude larger than the total ^{129}I inventory in the pre-nuclear ocean and from weapon test releases, and 3 orders of magnitude higher than the release from the Chernobyl accident. The discharges of ^{129}I from the reprocessing plants continue.

The radionuclides discharged into the English Channel from La Hague and to the Irish Sea from Sellafield reprocessing plants are transferred to the North Sea by the Atlantic Current, and further to the Norwegian coast and the Arctic by the Norwegian Coastal Current. Meanwhile, some of the discharge enters the Skagerrak along the European coast together with other anthropogenic pollutants from the European continent. By analysis of ^{129}I in the Kattegat and Baltic Sea and comparing with the data from other places, not only ^{129}I and other reprocessing derived radionuclides, but also other anthropogenic pollutants in this area can be identified.

Our recent analytical results show that the ^{129}I concentration in seawater at Kattegat has reached $66\sim 220 \times 10^{-13}$ g/L ($^{129}\text{I}/^{127}\text{I} = 29\sim 48 \times 10^{-8}$), at Møen in the Baltic, 9.6×10^{-13} g/L ($^{129}\text{I}/^{127}\text{I} = 6.3 \times 10^{-8}$) in 1999, which is much higher than the reported values from 1992 in this area. The $^{129}\text{I}/^{127}\text{I}$ ratio measured in seaweed samples collected from Bornholm in the Baltic Sea increases continuously from 1.5×10^{-8} in 1992 to 4.9×10^{-8} in 1999. All of these ratios are much higher than the pre-nuclear and weapon test fallout level. The possible anthropogenic sources of ^{129}I in the Baltic Sea will be discussed including:

1. The marine discharge from European reprocessing plants, which may be transferred to this area through the North Sea, Skagerrak and Kattegat.
2. Direct atmospheric emission from European reprocessing plants, which may be transferred by atmosphere and deposited to this area.
3. Atmospheric transport of ^{129}I from the North Sea, where a ratio of $^{129}\text{I}/^{127}\text{I}$ as high as $\sim 10^{-5}$ has been reported.
4. Direct fallout from the Chernobyl accident and runoff from surrounding continental areas.
5. Release from the nuclear power plants surrounding the Baltic Sea.

In addition, the radiation risk of this radionuclide and its utilisation as an environmental tracer for reconstruction of high radiation hazards from Iodine-131 will be discussed.

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

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Abstract

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

Introduction

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

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

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

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

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

Material and methods

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

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

Results and discussion

As can be seen in Fig. 1 the ^{99}Tc concentration in *Fucus serratus* varied with the Sellafield discharge. The activity concentration increased from about 10 Bq/kg in 1995 up to 230 Bq/kg in the year 2000. This indicates a transit time from Sellafield to the Swedish west coast of 4-5 years, which is in good agreement with earlier estimations by Dahlgaard, 1995.

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

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

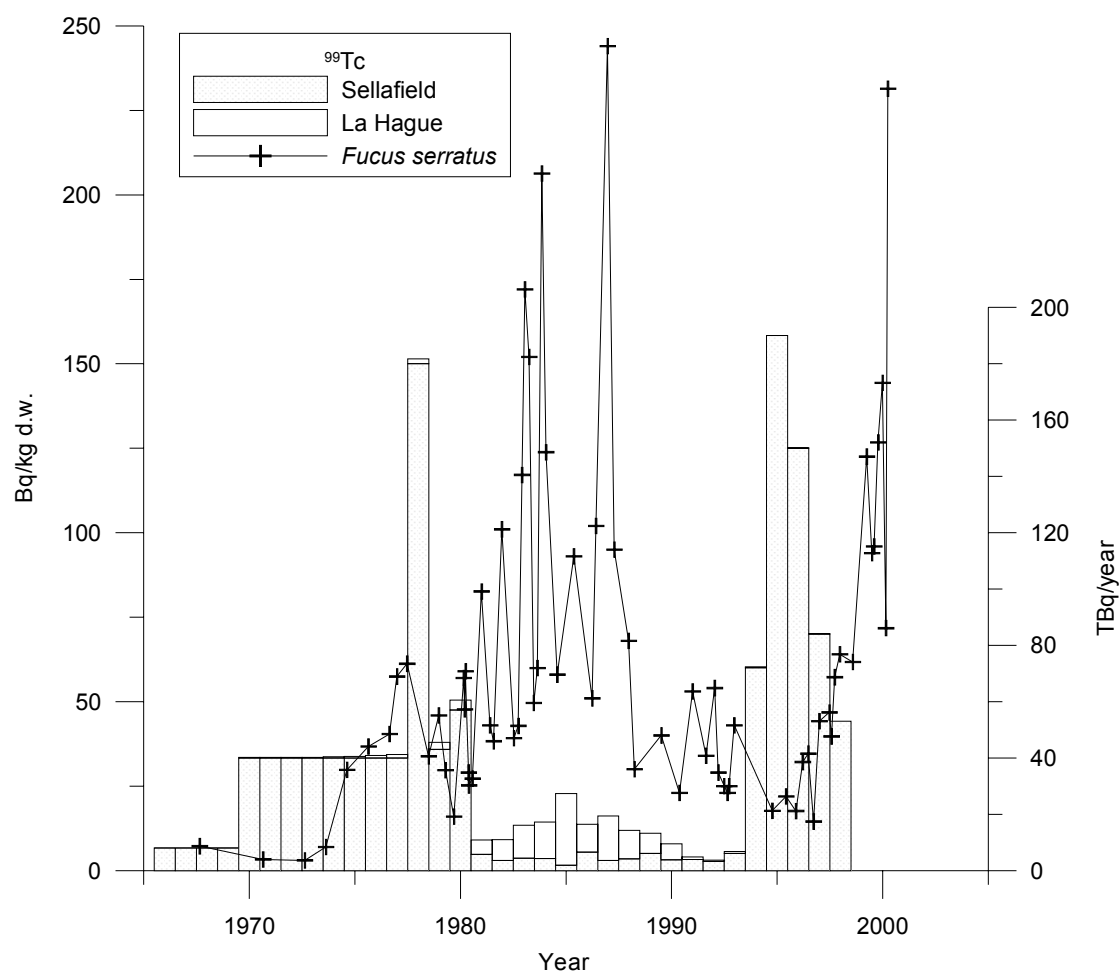


Figure 1. Activity concentration of ^{99}Tc (primary axis) in *Fucus serratus* from Särödal (56.76 °N, 12.63 °E) and annual ^{99}Tc discharge from Sellafield and La Hague (secondary axis).

Acknowledgement

The present work was partly supported by the NKS BOK-2 program.

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Timing of fluxes in the Nordic seas

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

Contaminants in the waters from Western Europe reach the eastern areas of the Nordic Seas from the south and the western areas from the north by the cyclonic current systems. The timing of this pathway from the North Sea area and around the Nordic Seas into North Icelandic waters was according to time-series on salinity as well as radionuclides in the 1970's and 1980's 4-5 years. Ongoing research on the fate of the recent technetium - 99 release in the latter half of the 1990's may reveal its appearance in North Icelandic waters in present or near-by times.

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On-line determination of the depth distribution of ^{137}Cs by means of gamma spectrometry of primary and forward scattered photons

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Abstract

Stationary and mobile field gamma spectrometry is a useful tool for rapid estimations of environmental radioactivity inventories in the ground. A weak point however, is that the vertical distribution of the activity in the ground must be known in order to calculate the activity per unit area or unit mass from an observed photon fluence rate. A promising method for converting incoming spectral data into both true area activity density and activity depth distribution in real time is the peak-to-valley method. It is based on an analysis of the ratio between count rates from primary and forward scattered photons. Here our plans to adopt the peak-to-valley method to carborne mobile gamma spectrometry using a large HPGe detector are described. Preliminary results from testing the method in the field, utilising a point source at different depths, are presented. The results show a useful decrease in the peak-to-valley ratio for an infinite area source at different depths in the soil.

Introduction

In the case of stationary measurements, the depth distribution in soil is commonly determined by analysing core samples by gamma spectrometry afterwards in the laboratory. In order to be representative, the number of samples to be analysed in the lab is roughly one hundred for each a $10 \times 10 \text{ m}^2$ surface and a 30 cm depth module monitored. During mobile field spectrometry, large areas are covered and on-line presentation of the result is given high priority. During such circumstances, traditional core sampling methods are not feasible and other more direct methods have to be used. One promising method for conversion of incoming spectral data in real time to both true area activity density and activity depth distribution is based on an analysis of the ratio of primary and forward (inelastically) scattered photons. This peak-to-valley concept was introduced by Zombori et al. (1992). The method has been proven to work in stationary *in-situ* gamma spectrometry (Tyler et al. 1995, Gering et al. 1998). The counting time for an accurate stationary *in-situ* determination of the peak-to-valley ratio is depending on the level of contamination and the efficiency of the detector. Gering *et al.* (1998) came to the conclusion that the contamination level should be at least $5\text{-}10 \text{ kBq}\cdot\text{m}^{-2}$ for a counting time of 1 hour, using a HPGe detector of 25% relative efficiency. In mobile gamma spectrometry the spatial resolution is important. 1 hour counting time at a speed of $50 \text{ km}\cdot\text{h}^{-1}$ results in a resolution of 50 km, which is not acceptable. A more reasonable resolution is about 5-10 km, thus needing a counting time of ca 5-10 minutes. Here our plans to adopt the peak-to-valley method to carborne mobile gamma spectrometry using a large

HPGe detector are described. The objective is to determine the minimal contamination level giving reasonable statistical uncertainty within a counting time of about 5 minutes.

Material and methods

The normal way to convert the pulse height distribution to from a gamma spectrometric measurement to activity density is to use the peak area method (Lowder *et al.* 1964, Beck *et al.* 1964). The method is based on the assumption that the measured area under the full energy absorption peak is proportional to the primary photon fluence at the detector. The proportionality (detector efficiency) is determined in advance in the laboratory. The activity density in the soil can then be calculated if an assumption is made about the angular distribution of the primary photon field. The angular distribution depends on the attenuation in the soil. The attenuation results from the depth distribution of the radionuclide in the soil and surface roughness.

In this work the method to estimate the attenuation is based on the peak-to-valley concept, *i.e.* ratio of primary and inelastically scattered photons. A HPGe-detector with an efficiency of 100% (relative to that of a 3" x3" NaI crystal for 1.33 MeV and for a source-detector distance of 25 cm) was used. The peak-to-valley method uses three energy regions in the spectrum: the peak, the valley and the background region; see Figure 1. The valley is a spectral region just below the full energy absorption peak, corresponding to photons with small energy losses. Here the interval from 615 to 655 keV is used. The total counts in the valley are not only caused by forward scattered photons but also by photons with higher primary energies and cosmic radiation, which should be subtracted first. To do this a background region just above the full energy absorption peak is used; here the interval is from 670 to 720 keV. The count rate in the background region, scaled to the same energy width as the valley, is subtracted from the valley region. If the measurements are performed in an area with high contamination level the background region might have to be corrected for pile-up. After the subtraction the remaining counts in the valley region consists of forward scattered photons and an additional background component, named B in Fig. 1. The additional background component is caused by the differences in the scattered radiation from photons with higher primary energies and the increase in detector intrinsic efficiency. To make an accurate determination of B the contribution in the valley region from each gamma line above the ^{137}Cs peak has to be known. This can be done with photon transport calculations or Monte Carlo simulations. An approximate way to determine B is to assume that the ratio between A+B and A is constant. The ratio must be measured in an environment free from forward scattered photons from ^{137}Cs . The determination of the ratio was performed in the basement of our laboratory.

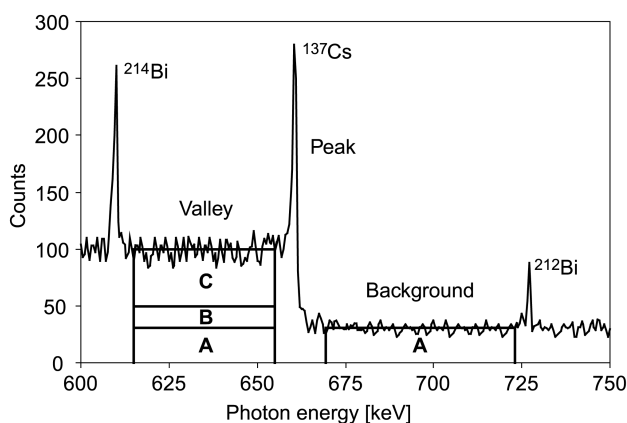


Figure 1. The regions of interest in the peak-to-valley method. Region A is the background component above the peak, corrected for possible pile-up. B is the additional background mainly caused by the differences in the scattered radiation from photons with higher primary energies. C is the in soil forward scattered photons from ^{137}Cs .

In the field the detector was horizontally mounted in a GMC van facing backwards, about 2 m above the ground surface. When using detectors with high efficiency pile-up can cause a problem even at moderate contamination levels. In the peak, pile-up does not become a problem until the count rate exceeds 1000 cps, which is reached with a 100% HPGe detector from soil with an activity concentration in the order of 1 MBq·m⁻². However pile-up necessitate significant corrections also at lower peak count rates. Since the count rate in the background region are much lower than the peak count rate it is easily affected by the pile-up tail at the right side of the peak. In the background region pile-up starts to be significant at count rates as low as 50-100 cps. In order to quantify the influence of pile-up in the background region measurements were performed in the laboratory. A 50 MBq ¹³⁷Cs point source was placed at various distances from the detector to obtain spectra with peak count rates up to 500 cps. A power function could then be fitted to the count rate in the background region caused by pile-up.

The approach to determine the peak-to-valley ratio for different depth distributions in the soil was field measurements using a ¹³⁷Cs point source and varying the depth in the soil and distances from the detector. Depths between 0 and 250 mm and distances between 5 and 30 m were measured. Background spectra were also obtained in order to separate the point source from caesium originating from weapons fallout and the Tchernobyl accident. Two sources were used with activities of 50 MBq and 1970 MBq, the stronger source for greater depths and distances. The peak-to-valley ratio for an infinite surface source at different depths was then calculated, assuming a constant attenuation coefficient in the soil.

Results and discussion

The resulting count rate in the background region due to pile-up, N_{pileup} , can be expressed as

$$N_{pileup} = 0.0006 \cdot (N_{peak})^{1.48} \quad [\text{cps}]$$

where N_{peak} is the net count rate in the full energy absorption peak for ¹³⁷Cs. The formula is valid for our detector system and count rates up to 500 cps.

In the field the count rates in the three regions for various depths and distances were determined and corrected for pile-up. Exponential functions were fitted to the peak and valley count rates with depth and distance. Integration over the distance then results in peak and valley count rates for an infinite area source as function of depth. The activity density normalised to 1 kBq·m⁻² gives

$$\begin{aligned} N_{peak} &= 0.994e^{-0.021d} & [\text{cps} \cdot \text{kBq}^{-1} \cdot \text{m}^2] \\ N_{valley} &= 0.105e^{-0.018d} & [\text{cps} \cdot \text{kBq}^{-1} \cdot \text{m}^2] \end{aligned}$$

where d is the soil depth in mm. The peak and valley count rates for an infinite area source as function of is shown in Figure 2.

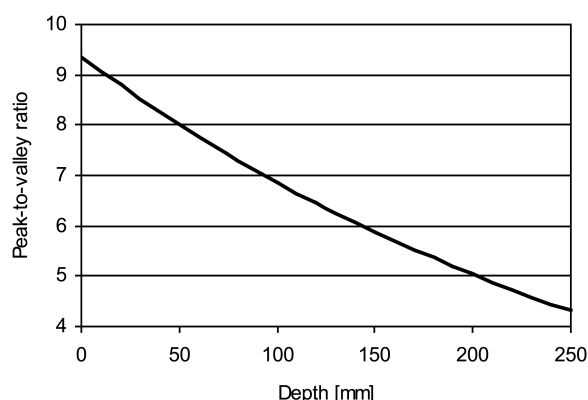


Figure 2. The PTV (peak-to-valley) ratio as function of depth in the soil for an infinite area source.

Future work is to determine the PTV ratio for different kind of depth distributions in the soil, *e.g.* uniform, exponential and Lorenz distributed sources. The count rates for the peak and valley as function of depth are to be integrated over depth and weighted with the depth distribution function. The crucial count rate, *i.e.* determines the minimum counting time, is the valley count rate. A 5 minute counting time results in valley counts in the order of 30 counts per $\text{kBq}\cdot\text{m}^{-2}$, which indicates that the peak-to-valley method is useful for carborne gamma spectrometry in areas where the contamination level is at least about $10 \text{ kBq}\cdot\text{m}^{-2}$.

Acknowledgement

The Swedish Radiation Protection Institute, SSI, supported this work, (project number 710/2846/98).

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The effect of x-ray summing in calibration of extended energy range GE detectors

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Introduction

Gammaspectrometric analyses of low-level samples are usually performed using close measuring geometry. In this measuring set-up the effect of the true coincidence summing (TCS) has to be taken into account. The detection of true coincidence sum pulse is possible when a nuclide emits two or more cascading photons within the resolving time of a spectrometer. The magnitude of summing depends strongly on the total and full energy peak efficiencies of the detector and on the decay scheme of the nuclide.

In addition to summing of gamma rays, other radiation, especially X-rays, can be in true coincidence with the gamma rays. X-rays in cascade with gamma rays can originate from electron capture, positron decay, or internal conversion. The summing effects with most of the X-rays can be ignored for the p-type Ge detectors having thick dead layer. On the contrary, the detectors with a thin entrance window, e.g. n-type detectors and new extended range p-type detectors, can experience severe summing of X-rays and gamma rays.

Material and methods

The effect of increased detection efficiency for low-energy photons was studied using the data obtained in the efficiency calibration of Canberra BEGe detector (model BE5030, area 50 cm², thickness 30 mm, nominal efficiency 50%). The detector, coded "B1", has a 0.5-mm carbon epoxy window and a thin germanium dead layer enabling the entrance of low-energy photons into the active volume of the germanium crystal. The calibration was performed by counting radionuclide standard source provided by NPL (National Physical Laboratory, UK) for the Proficiency Test Exercise for Radionuclide Laboratories Supporting the International Monitoring Stations. The planar source was a compressed Petrianov PVC air filter (diameter 70 mm, height 5 mm) in a plastic source holder. The mixed source contained the following 14 radionuclides: ⁵⁴Mn, ⁵⁷Co, ⁶⁰Co, ⁶⁵Zn, ⁸⁵Sr, ⁸⁸Y, ¹⁰⁹Cd, ¹¹³Sn, ¹³⁷Cs, ¹³⁹Ce, ¹⁴¹Ce, ²⁰³Hg, ²¹⁰Pb, ²⁴¹Am. Unfortunately most of these common calibration nuclides decay by electron capture with subsequent emission of characteristic X-rays of the daughter nuclide. The effect of X-ray summing can be decreased using a suitable absorber, e.g. copper. The Cu disc of 0.1 mm was chosen and the standard source was recounted with the absorber on top of the detector window.

To demonstrate the effect of X-ray summing also on qualitative analysis, a sample of ¹¹¹In was counted in close geometry by two detectors of different type. ¹¹¹In decays to ¹¹¹Cd by electron capture. The K_{α1} and K_{β1} energies of cadmium are 23.17 keV and 26.09 keV, correspondingly. The spectra were acquired by detectors B1 (above), without absorber, and a p-type HPGe detector "N2" (model Ortec GEM-80190, nominal efficiency 78%).

The results and conclusions

The both sets of measured efficiency values were plotted in the same graph (Fig 1.). The low-energy part of the efficiency curve is presented also as an insert using linear scales. The efficiency curve is always difficult to interpolate in the range 60-150 keV due to the rapid change in this region but without absorber it is almost impossible to obtain reliable curve, having only one peak free from potential summing (145.4 keV from ^{141}Ce decaying by β^-). Using the above-mentioned absorber the efficiency loss at 46.5 keV is only 40%, but most of the cascading X-rays are strongly attenuated and a more plausible fit can be achieved. It is interesting to note that the count rate of the 165.8 keV peak of ^{139}Ce is actually increased when using the absorber.

When fitting the linear part (in log-log scale) of the efficiency curve, only 'pure single-line' nuclides were used i.e. excluding the gamma energies of ^{60}Co , ^{88}Y , ^{113}Sn and ^{139}Ce . The values obtained with the peaks experiencing summing deviate from those given by the fitted curve and the amount of deviation provides the TCS correction coefficient.

Although the TCS concerning the calibration nuclides is decreased substantially by the absorber, other nuclides decaying with higher energy X-rays can still have significant summing. Especially high Z nuclides decaying by EC or having a high probability for internal K-conversion are prone to increased summing when measured by a detector with extended energy range.

As an example of multiple summing events, the comparison of two ^{111}In spectra is presented in Fig. 2. Considering that ^{111}In has practically only two gamma energies, the complexity of spectrum obtained by the detector B1 is striking. The multiple summing of gamma and X-ray peaks and additional Ge escape peaks complicate the peak identification considerably. This feature of the detectors with high efficiency for low-energy photons has perhaps not fully been realised.

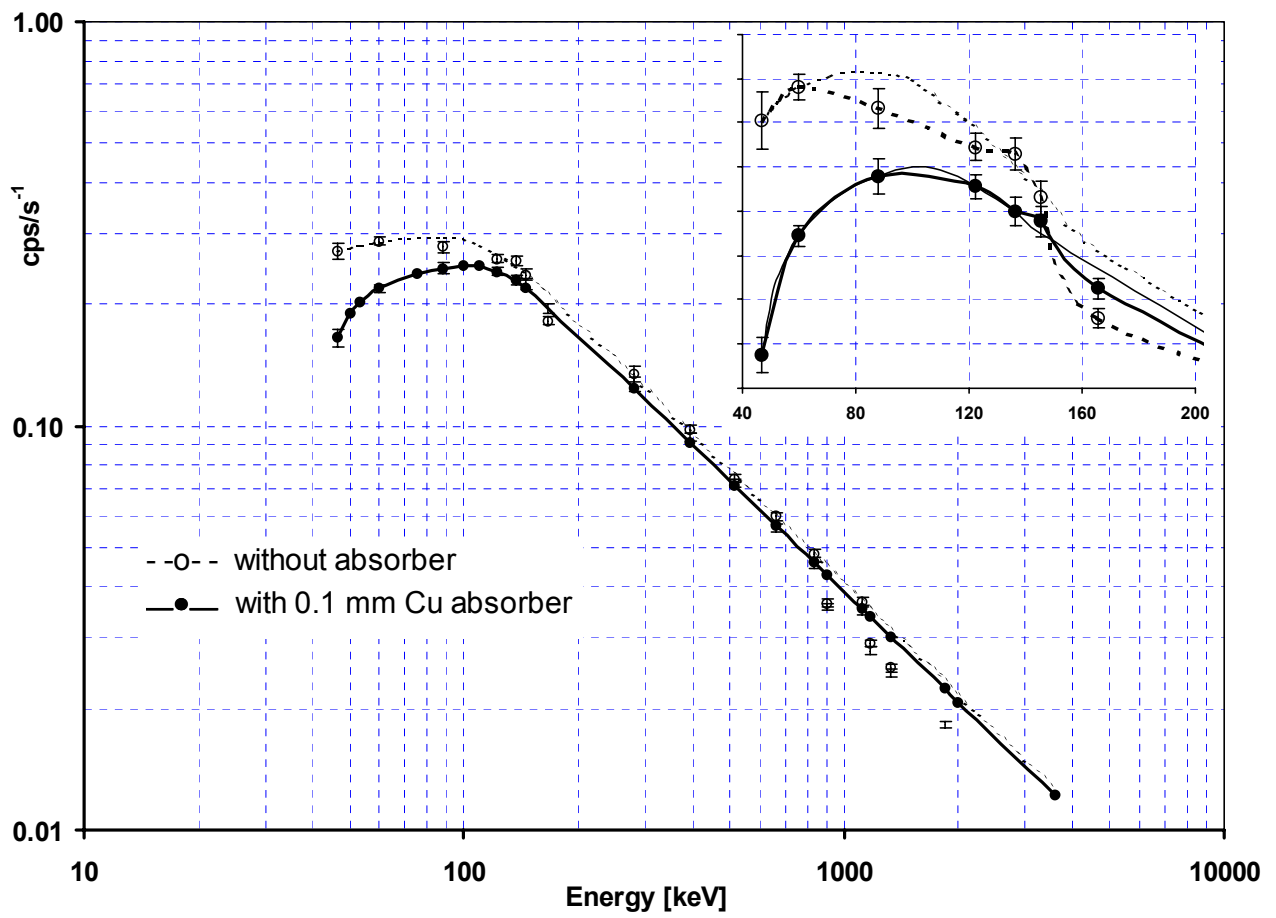


Fig. 1. The efficiency curves of the BEGe detector for the disc source on top of the detector window, with and without Cu-absorber.

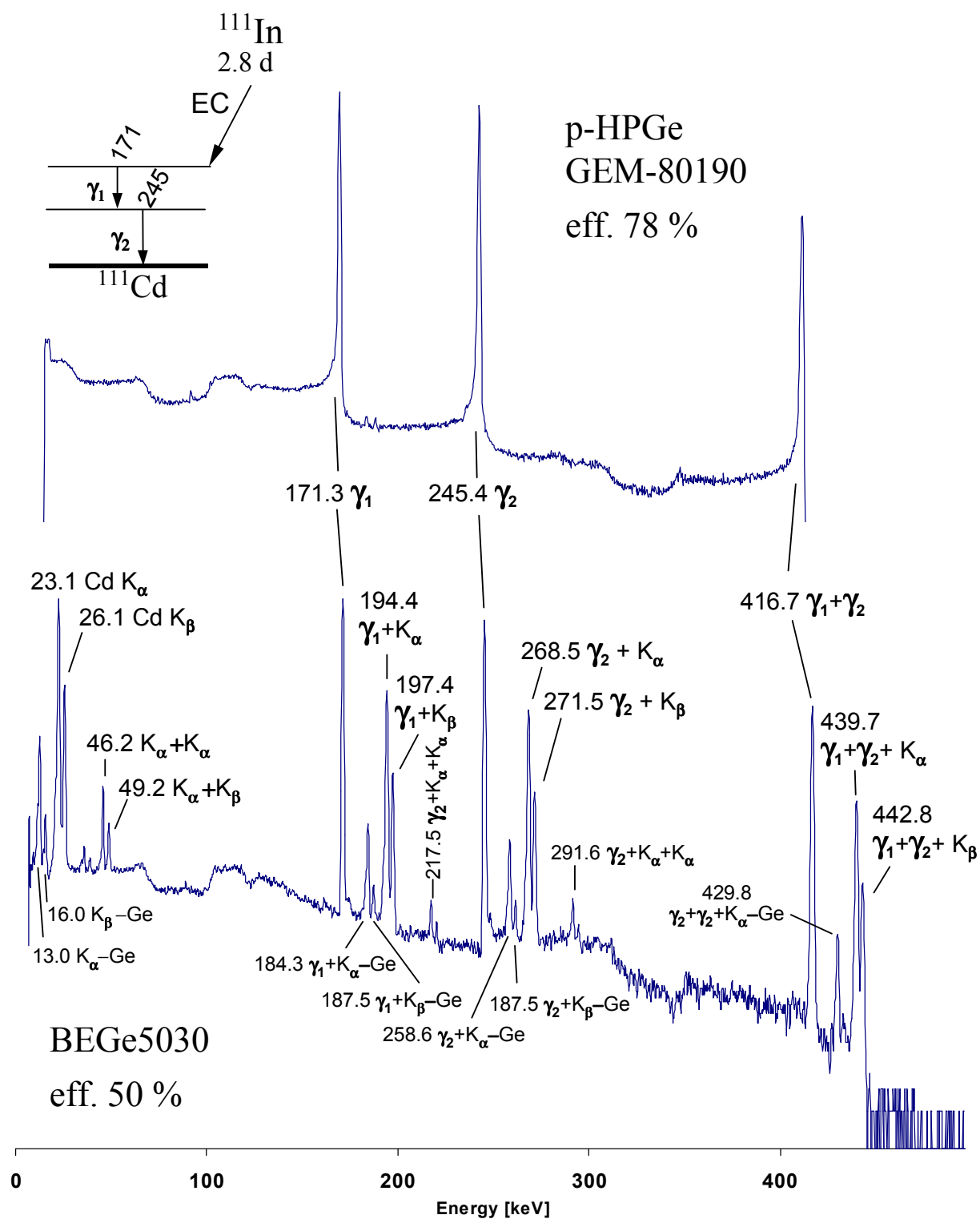


Fig. 2. Two spectra of ^{111}In acquired by two detectors of different type.

Testing of extraction chromatography for determination of actinides from environmental samples

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Introduction

The concentrations of transuranic elements in the environment are usually extremely low. Special procedures are needed for preconcentration and separation of these nuclides from various matrices and from other actinides. Traditional ion exchange methods have been used for separation of Pu and Am for decades. In recent years extraction chromatography methods based on new materials have been developed.^{1,2} These new methods are simpler and faster to use than the conventional methods based on ion exchange, especially for americium.

In this work application of Eichrom's extraction chromatography resins (UTEVA, TRU, TEVA) and an anion exchange resin (Dowex 1x4) were studied for the separation of Pu and Am from different natural matrices.

Experimental

The concentration levels of Pu and Am in our experiments were as follows: ^{239,240}Pu and ²⁴³Am in air ~ 10 nBq/m³, in grass ~5 mBq/kg, in organic soil and litter ~5 Bq/kg and in mineral soil ~100 mBq/kg. Because of low activity levels, large sample sizes are required.

Materials used for testing and the amount of materials were:

- Air filter: 20 000 m³ of air was collected on the air filter
- Grass: 100 g dry wt.
- Litter: 20 g
- Organic soil: 20 g
- Mineral soil (E and B layers): 30 g

For yield determination ²⁴²Pu and ²⁴³Am were added as tracers (0.025 Bq/sample).

Two different methods were tested. The separation schemes of the methods are shown in Fig 1.

Method A:

- 1) Samples which contain organic material (air filter, grass and organic soil layer) were ashed in an oven at 400 °C.

- 2) The ash/mineral soil sample was leached with conc. HNO_3 and HCl acids. For the air filters the leaching was carried out only with HNO_3 . Then the sample was filtered through a glass fiber filter and evaporated.
- 3) Oxalate coprecipitation was used to separate actinides from most other substances. The filtrate was diluted to 150 ml with deionized water. Ca carrier and oxalic acid were added and precipitation was carried out at pH 1.5. The sample was heated and precipitate was left to settle overnight. Sample was filtered through a Whatman 42 filter. Filtrate and the filter paper were ashed at 600°C overnight. For the air filters the oxalate coprecipitation was not needed.
- 4) Eichrom's UTEVA and TRU resins were used as in Eichrom's method of Americium, Plutonium and Uranium in water (ACW03). Americium was eluted from TRU with 4M HCl and plutonium with 0,1 M $\text{NH}_4\text{HC}_2\text{O}_4$. Pu was evaporated to dryness.
- 5) Plutonium was coprecipitated with NdF_3 . Pu fraction was dissolved in 1 M HNO_3 and 50 μg Nd carrier and 40 % HF were added to the sample. The precipitate was collected on 0.1 μm membrane filter.
- 6) Americium was loaded into Eichrom's TEVA column in 2 M NH_4SCN + 0.1 M formic acid. The column was washed with 1 M NH_4SCN + 0.1 M formic acid and americium was eluted with 2 M HNO_3 . Aqua regia was added to the eluate and it was evaporated to dryness. For the air filters TEVA was not needed because of the very low amounts of lanthanides present in the sample.
- 7) Am was coprecipitated with NdF_3 on 0.1 μm membrane filter.

Method B was modified from the method A.

- 1) Similar to step 1) in method A.
- 2) Similar to step 2) in method A.
- 3) The residue was dissolved in 8 M HNO_3 . NaNO_2 was added to the sample, which was then heated and allowed to cool. The sample was then loaded into a Dowex 1x4 column and the column was washed with 8 M HNO_3 . The load and wash solutions were collected for Am determination. The column was washed with conc. HCl and finally Pu was eluted with conc. HCl + 1 M NH_4I . The eluate was evaporated to dryness. Anion exchange was carried out twice for the samples with very low Pu concentration in order to remove traces of Th.
- 4) Pu was coprecipitated with NdF_3 on 0.1 μm membrane filter.
- 5) Am fraction in 8 M HNO_3 was evaporated and oxalate coprecipitation was carried out as explained in method A (step 3) except that the sample was diluted to 700 ml with deionized water to prevent coprecipitation of iron.
- 6) The residue was treated with conc. HNO_3 and dissolved in 3M HNO_3 + 0.5 M $\text{Al}(\text{NO}_3)_3$. In some cases precipitate formed (iron), which was dissolved by adding ascorbic acid to the sample. The sample was loaded into a TRU column and the column was washed with 2 M HNO_3 and 0.5 M HNO_3 . Am was eluted from the column with 9 M HCl and 4 M HCl .
- 7) Am was purified from lanthanides as explained in method A (step 6).
- 8) Am was coprecipitated with NdF_3 on 0.1 μm membrane filter.

The radioactivity of Pu and Am were measured with passivated implanted planar silicon (PIPS) detector. The alpha spectra indicated high radiochemical purity achieved by the separation methods used.

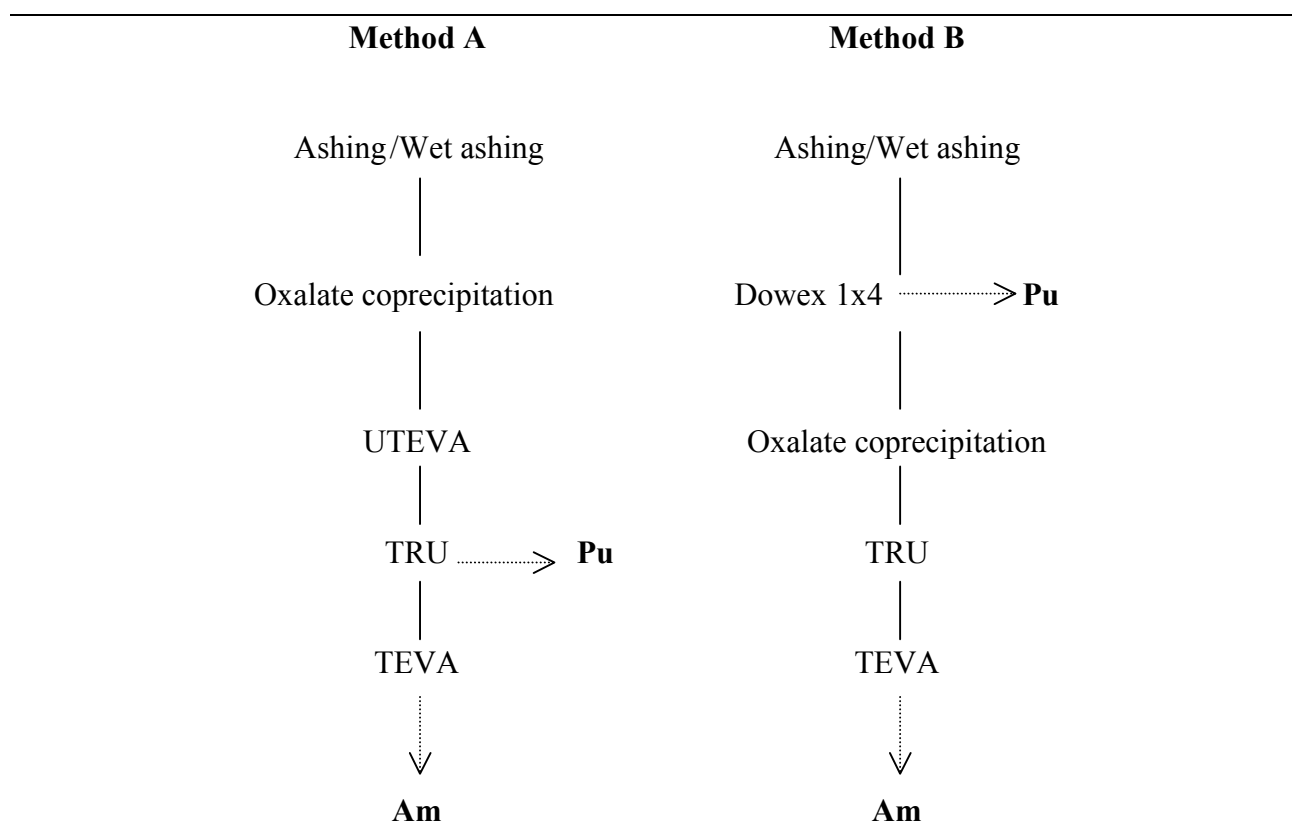


Figure 1. Separation schemes for Pu and Am from environmental samples

Results and discussion

In the analysis of Pu and Am from the air filter samples (method A) interference due to natural ^{210}Po was observed. The activity concentration of ^{210}Po in the air is approximately three orders of magnitude higher than those of the transuranic elements.³ The presence of ^{210}Po interferes the alpha measurements of $^{239,240}\text{Pu}$ and ^{243}Am , which is used as tracer. Po can be removed from Pu and Am fractions by using crushed silver. Po electrodeposits from 2M HCl on silver when the temperature is raised. From the americium fraction Po can also be removed by using TEVA resin. In method B, Po did not interfere since it was removed from the sample by anion exchange.

The method A gives also an opportunity to elute uranium from UTEVA with 0.01 M HCl.

Pu and Am yields are given in Table 1. In the method A, plutonium yields for air filters were high (63-100%), but for litter and soil samples yields were rather low (less than 50%). Americium yields were high for air filters, litter and upper soil layers (75-100%). However, yields for lower soil layers were only a few percent. The reason for low Am recoveries was coprecipitation of iron hydroxide in oxalate precipitation. This could be prevented by adding more oxalic acid and diluting the sample to higher volume with water.

Due to the low yield of Pu found for litter and soil samples, the method B was developed. In this method Pu was separated by anion exchange before the use of TRU and TEVA resins for the

separation of Am. By this method (method B), plutonium yields were high for all sample matrices (84-93%) and Am yields were between 47 and 93%.

The reliability and repeatability of these two methods could not yet be fully verified due to scarceness of data.

Table 1. Yields (%) for Pu and Am determined by two different methods: A) Both Pu and Am were separated by means of extraction chromatography and B) Pu was separated by anion exchange and Am by extraction chromatography. In the cases where several experiments for the same material were performed, the average value and the range of recoveries are given.

Matrices	Method A		Method B	
	Yield for Pu	Yield for Am	Yield for Pu	Yield for Am
Air filter ($2 \cdot 10^4 \text{ m}^3$)	87 (63-100)	73	n.t	n.t
Grass (100 g)	n.t	n.t	84	93
Litter (20 g)	33	93	100	47
Organic soil (30 g)	48 (42-54)	90 (80-100)	97	75
Mineral soil E-layer (30 g)	10 (2-18)	83 (75- 90)	100	87
Mineral soil B-layer (30 g)	21 (6-45)	2 (1-2)	98	53

n.t = not tested

Conclusions

In the method A, which was based only on extraction chromatography, rather high yields for Am in almost all tested matrices were obtained. However, Pu yield was high only for air filters. In the method B, in which Pu was separated with anion exchange and Am with extraction chromatography, high yields for Am and Pu for all tested matrices were obtained in generally. The results suggest that the method B is suitable for Pu and Am determinations from large environmental samples for various materials and method A for air filters.

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Helautomatiskt mobilt system för mätning av dosrater

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Inledning

I beredskapssammanhang är det ofta en fördel att kunna automatisera och förenkla mätförfaranden, huvudsakligen av två skäl. Dels behövs mätresultaten göras tillgängliga för andra snabbt, dels för att kunna minimera utbildning och träning av den oftast stora kader av mätoperatörer som är inblandade i beredskapssammanhang.

Nedan beskrivs ett sådant system, där operatören endast behöver placera systemet i ett fordon, sätta GPS-antennen på lämplig plats och koppla om en strömbrytare. Därefter sköter systemet sig självt.

Teknisk beskrivning

Systemet är byggt runt mobiltelefonen Nokia Communicator 9000. Denna telefon har en liten inbyggd dator med seriellt gränssnitt, vilket gör den lämplig för datafångst och datatransmission. Förutom telefonen ingår ett mätinstrument för dosrat (RNI 10 SR), en GPS-navigator (Garmin 12 XL), en seriell elektronisk omkopplare och ett batteri (3.6 Ah).

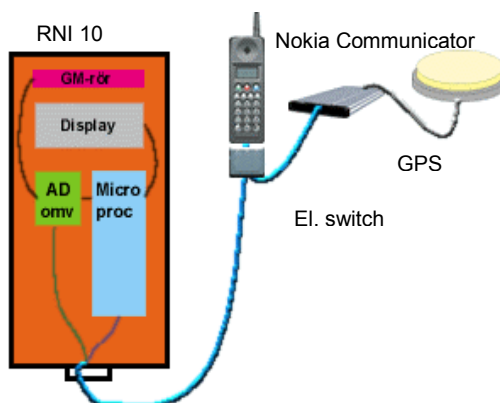


Med en omkopplare kan systemet ställas om från *stand-by* till operationsläge mobil mätning alternativt stationär mätning. Datorn läser, beroende på operationsläge, växelvis från dosratsinstrumentet och GPS:en. En textsträng (max 160 tecken) byggs nu upp med utvalda

data. När strängen är uppbyggd skickas den iväg som ett SMS-meddelande (Short Message Service). Om man befinner sig i radioskugga, lagras meddelandena automatiskt i telefonen och skickas när man får kontakt med GSM-nätet igen.

Meddelandena kan antingen tas emot direkt med en mobiltelefon kopplad till en dator eller skickas via e-post systemet och hämtas i en brevlåda. Efter mottagning avkodas textsträngen och stoppas i en databas. Mätningarna kan sedan visualiseras via en hemsida.

Med befintligt batteri blir operationstiden > 24 timmar.



Mätförfaranden

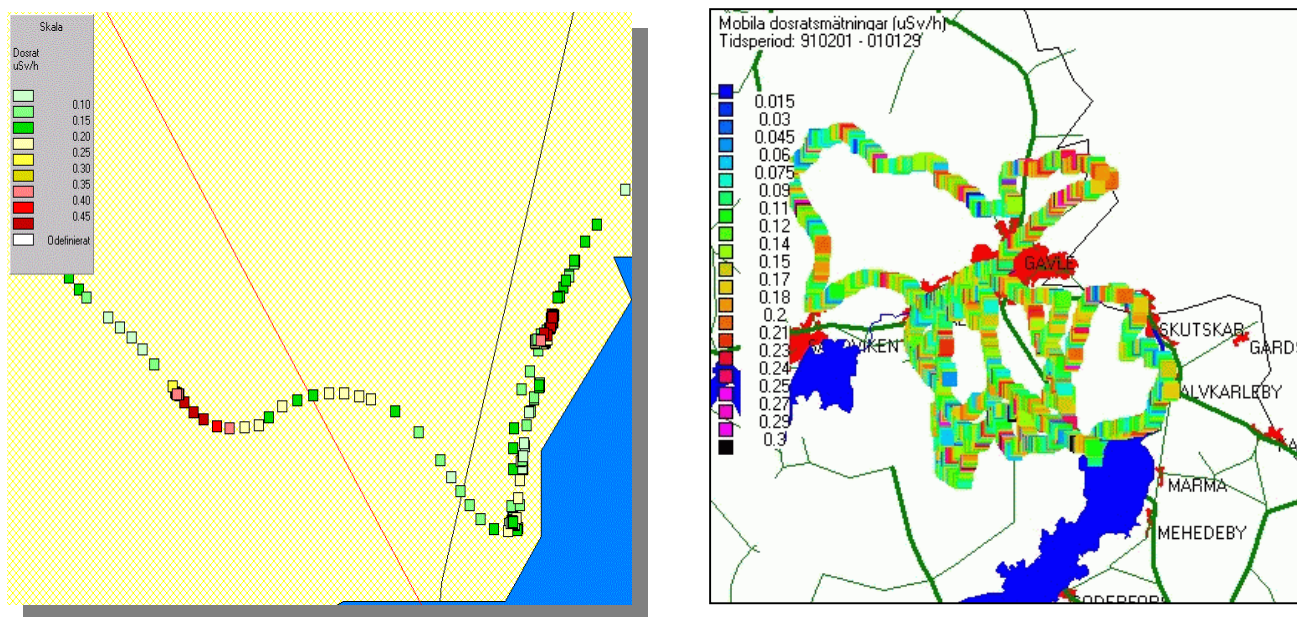
Systemet kan användas huvudsakligen på två sätt: antingen som mobil mätning eller som stationär mätning på en förutbestämd plats.

Mobil mätning

Den momentana dosraten avläses från mätinstrumentet under 5-6 sekunder, därefter kopplas läsningen över till GPS:en ca 2 sek och positionen avläses. I datorn skapas sedan en textsträng bestående av aktuell tid, position och ett medelvärde av dosraten. När 5 sådana strängar adderats, skickas den sammansatta strängen iväg.

Normalt lagras varje 8:a sekunders mätning i textsträngen, men systemet kan ställas in så att varannan, var tredje etc mätning lagras. Detta görs i första hand för att minska kostnaden (ca 1.50 SEK/meddelande) vid övningar m.m.

Ibland kan det vara önskvärt att kombinera den mobila mätningen med att mäta en längre tid på ett och samma ställe, t.ex. om man vill följa dosratens variation med tiden i ett passerande radioaktivt moln. Systemet kan konfigureras så att det känner självt av om det står stilla, och övergår då automatiskt i att registrera 5-min medelvärden. I detta fall registreras tid, position och 14 st dosratsvärden per SMS-meddelande. Så fort systemet börjar röra på sig, återgår det automatiskt till 8:a sekunders registreringar.



Bilden till vänster visar en detalj från en mobil mätning från Resumee-99 övningen i Gävle, Sverige, där två radioaktiva källor var utplacerade längs vägen. Bilden till höger visar samtliga mätningar med systemet under övningen.

Jämförelser med motsvarande mätningar med spektrala system (NaI och HPGE), som mäter dosraten selektivt från Cs-137, visar bra överensstämmelse där nedfallet är relativt stort. Systemet klarar helt naturligt inte av att särskilja bidraget från nedfallet där detta utgör en liten del av totala

dosraten, främst på grund av den korta medelvärdestiden. Systemet börjar fungera bra vid ca dubbla bakgrunden.

Stationär mätning på förutbestämd plats

Vid detta användningssätt kopplas satellitnavigatören helt bort, och systemet används nu som en utplaceringsbar gammastation. I stället för positionen lagras nu en för mätplatsen unik plastkod, som kan matas in av operatören. Systemet lagrar nu 15 st 5-min registreringar per meddelande.

Detta användningssätt av systemet kan vidareutvecklas. Eftersom man kan programmera i ett högnivå språk (C++ liknande), är det lätt att bygga in olika typer av larm och funktioner som beror av den aktuella dosratsnivån. Vid normala nivåer kan systemet sända långtids-medelvärden, för att vid förhöjda sådana övergå till t.ex. 5-min medelvärden.

Eftersom systemet drar lite ström, kan det kompletteras med ett större batteri och en solpanel och därmed bli helt oberoende av yttre anslutningar.

Sammanfattning och diskussion

I en beredskapssituation med färskt nedfall kan nuklidfördelningen vara komplicerat, och det är alltid viktigt att utföra en eller flera spektralmätningar för att fastställa denna. Nuklidfördelningen är dock oftast relativt konstant inom nedfallsområdet och dosrats mätningar är därför tillräckliga, i alla fall om dosraten från nedfallet är jämförbart med eller större än bakgrunden.

Den stora skillnaden gentemot motsvarande mobila mätningar med spektrala system (NaI och HPGE) är, förutom den mycket enkla hanteringen, kostnaden. Detta system kostar <15 kSEK mot det tiodubbla för de spektrala systemen.

Fördelarna jämfört med manuella dosratsmätningar, är främst möjligheten av att snabbt mäta av ett område och den automatiserade rapporteringen.

Nackdelen med systemet sett ur beredskapssynpunkt är att det bara fungerar i områden med utbyggd GSM. Användandet av SMS gör dock att det fungerar även i områden med svag radiokontakt. SMS-trafiken påverkas heller inte av ev. överbelastning, då den utnyttjar GSM-systemets s.k. servicekanaler och inte talkanalerna.

Representation of radioecological data on digital

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Introduction

The aim of this work was to create a database for the environmental samples collected by the Regional Laboratory in Northern Finland of STUK-Radiation and Nuclear Safety Authority in Russian Arctic Seas in 1993-1996 during expeditions organised by the Murmansk Marine Biological Institute. The database contains information about GPS-positioned sampling sites, the results of gamma-spectrometric analyses and radiochemical plutonium analyses etc. A map of the $^{239,240}\text{Pu}$ concentrations in surface sediments in the Russian Arctic Seas is presented as an example of the use of the database in visualising environmental data.

Material and methods

The Relational Database

The database was based on entity relationships (Fig.1). The entity **Sample Collection** includes GPS co-ordinates and the code number of the sampling location, and entity **Analysis Result** includes in this case the plutonium $^{239,240}\text{Pu}$ concentration: Bq/kg. In Fig. 1 the relationships between the entities are depicted using 1 and n. For example, one **Sample Collection** could include several **Environmental Samples**, but every **Environmental Sample** is connected to only one **Sample Collection** record.

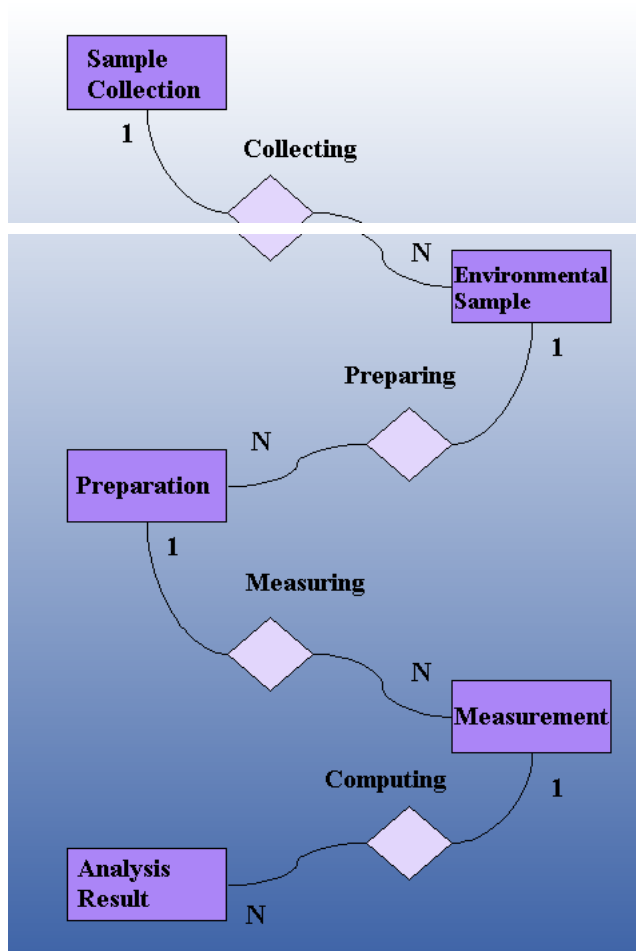


Figure 1. Entity Relationships.

Select Query Language (SQL)

The attributes of the data were searched from the database using the standard of the Select Query Language (SQL). The SQL -sentence was formed into a number of keywords: SELECT, FROM, WHERE (Fig. 2) [1]. For the representation (Fig. 5) the four attributes **Sample Collection** (x,y) and **Analysis Result** (nuclide, concentration) were needed. In addition, the INNER JOIN -clause was needed for the SQL -sentence.

```
SELECT Attribute  
FROM Entity  
WHERE Attribute=X
```

Figure 2. SQL –sentence structure.

Database Management System (DBMS)

The DBMS was connected in two sections: Data Description Language (DDL) and Data Manipulation Language (DML) [2] (Fig. 3). The connection between the applications, in this case MapInfo and database, was created by informing the source about the information in the DBMS -installation dialog. After installation of the DBMS, the measured data were retrieved and used.

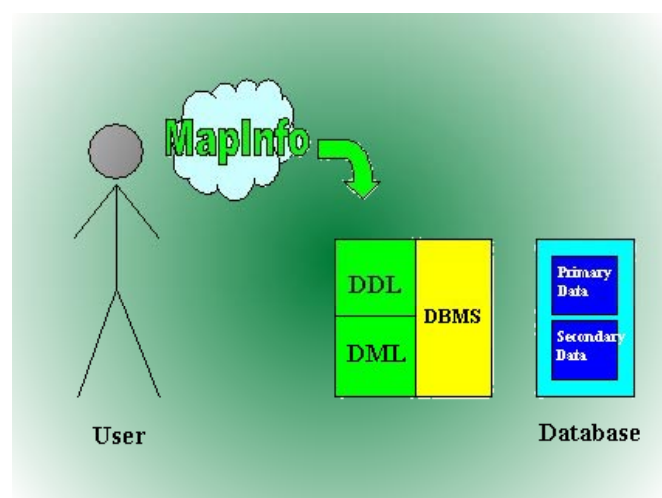


Figure 3. Operations model of the DBMS.

Map Catalog

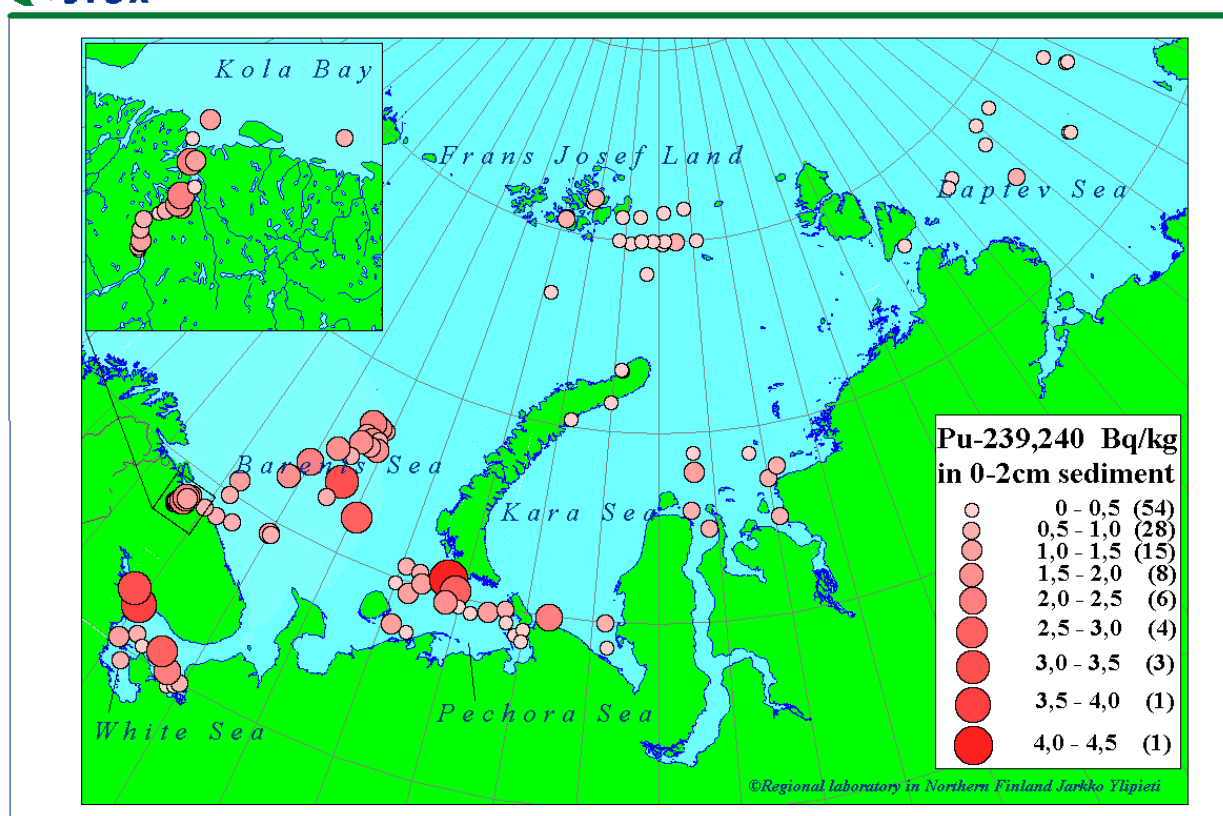
The Map Catalog was needed in the Database. The name of the column containing X- and Y- co-ordinates was specified in the Map Catalog [3]. The name of the table and present co-ordinate system was also described. The Map Catalog could also be created manually, but the format should be same as that in Fig. 4 [4].

MAPINFO_MAPCATALOG	
SPATIALTYPE	
TABLENAME	
OWNERNAME	
SPATIALCOLUMN	
DB_X_LL	
DB_Y_LL	
DB_X_UR	
DB_Y_UR	
COORDINATESYSTEM	
SYMBOL	
XCOLUMNNAME	
YCOLUMNNAME	

Figure 4. The MapInfo Map Catalog in The Microsoft Access Database.

Result

The results of the select-sentence were saved as a file and geocoded by desktop mapping software. Retrieval of the data is a mechanical task in which only one point in the wrong place could result in corruption of the data. On the other hand, the retrieved data were very useful in data processing. One of the representations made by retrieving data is shown in Fig.5. A digital map of ADC World Map, volume 2 Europe and Northern Asia, was used in the representation. The vector data map was digitised at a scale of 1:1 000 000. In the thematic map, the retrieved data are layered on the ADC World Map, each sample-point being geocoded on the basis of the GPS -positioned X- and Y -coordinates. The thematic layer of the 0-2cm surface sediment is shown in fig. 5 using circles of different size to representing the concentrations. The properties of the samples are indicated by the colour, as well as the size. Finally, the retrieved data were classified into concentrations ranging from zero to 4.5 Bq/kg in half -unit (0.5) intervals. The number of the samples in each class is given in the parentheses.



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Figure 5. The thematic map.

References

- [1] <http://www.aedu.kuopio.fi/opinnet/visbas/>
- [2] Elmasri/Navathe, Fundamentals of Database Systems, p. 29.
- [3] <http://testdrive.mapinfo.com/mapxhelp/map00107.htm>.
- [4] MapInfo Corporation, MapInfo Professional Owners Manual, p. 330.

¹³⁷Cs in Soil Profiles in NE Estonia

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Introduction

Radiocaesium contamination in soil was produced by two fallouts: a global fallout caused by nuclear weapons testing in the atmosphere (1950s to 1970s) and a regional one - during/after the Chernobyl accident (1986). The areal distribution of the ¹³⁷Cs fallout after the 1986 accident was extremely uneven in Estonia. Our previous studies [Realo et al., 1994, 1995] showed that about 60% of its total deposition with the countrywide mean of 2 kBq m⁻² occurred predominantly in the north-eastern part of Estonia. In this region the maximum deposition approached the values of 40 kBq m⁻². This is the reason why soil profiles in this region have been of interest. We have also determined the ¹³⁷Cs activity concentrations as a function of soil depth during 1991-2000. Attempts have been made to model the found depth-distributions of radiocaesium concentration and to find the possible time-dependent behaviour of these distributions in soil [Realo et al., 1994]. The migration of radiocaesium into soil is dependent on several factors: soil properties, vegetation, mode of deposition, etc. For this reason, averaged distribution parameters for both total and Chernobyl ¹³⁷Cs have been used for deriving general trends of the migration processes.

Sample collection and preparation

Depth distributions of the ¹³⁷Cs activity concentration were studied for undisturbed soil profile samples collected at more than 20 locations in NE Estonia in 1998 and 1999. Some of the sampling locations (including also sampling sites from 1991-1993) are listed in Table 1. In this report analysis data are presented together with our preliminary modelling results.

Soil profiles down to a depth of ~ 20 cm were collected by means of a 9 cm diameter steel corer. Soil cores were cut into 2-3 cm thick slices, dried, homogenised and put into metallic beakers. A low-background HPGe gamma spectrometer with a 42% efficiency and 1.7 keV resolution (1.3 MeV) was applied for the analysis of both ¹³⁴Cs (604 keV) and ¹³⁷Cs (662 keV). The Chernobyl fallout fractions in the total ¹³⁷Cs concentrations were calculated using the analysis data for ¹³⁴Cs.

Table 1. Sampling data of soil profiles.

Soil profile No	Location N/E	Short description	No of the soil profile	Location N/E	Short description
#87	59.202	Väike-	#121	59.264	Eesti SEJ
	27.343	Pungerja		27.903	
#90	59.382	Sinimäe	#122	59.275	Eesti SEJ
	27.84			27.914	
#94	59.003	Vasknarva	#123	59.281	Eesti SEJ
	27.722			27.911	
#98	59.448	Sillamäe	#124	59.298	Balti SEJ
	27.723			27.913	
#117	59.391	Olgina	#126	59.354	Balti SEJ
	28.081			28.071	
#118	59.329	Eesti SEJ	#129	59.372	Soldina
	27.932			28.104	
#119	59.405	Narva	#133	59.351	Balti SEJ
	28.173			28.138	

Results and Discussion

Typically the depth-dependent behaviour of ^{137}Cs activity concentration along an undisturbed soil profile is successfully described by using an exponential distribution of the following type [UNSCEAR, 2000]:

$$C(x) = C_0 \exp(-\alpha x), \quad (1)$$

where $C(x)$ is the radionuclide concentration (Bq/kg) at depth x (cm), C_0 is the surface concentration ($C(x=0)$) and α (cm^{-1}) is the characteristic coefficient of the distribution, the reciprocal of the relaxation length. Unfortunately, in many cases of long-term radionuclide transport the above simple distribution is a poor approximation to the real situation. When analysing our soil profiles, the same conclusion can be drawn. Considering the radionuclide transport in soil as a diffusion-convection process has lead to multiple models and their modifications, resulting in the depth-distributions of the Gaussian or lognormal types [Isaakson et al., 1998].

In the present report, we use the lognormal distribution of the form:

$$C(x) = \frac{A}{\sqrt{2\pi} w x} \exp\left[-\frac{(\ln x - \ln xc)^2}{2w^2}\right], \quad (2)$$

where its parameters are mean, $\ln xc$, and variance, w^2 , of the distribution. According to theory these parameters are time-dependent functions of the diffusion coefficient and the mass flow transport velocity, respectively [Konschin, 1992]. In principle, the actual dependence of these parameters on time can be derived from fitting of the depth-distributed activity concentrations in soil profiles collected at different times.

Some typical results on the depth distributions of the measured ^{137}Cs concentration in soil profiles collected in NE Estonia in 1991, 1993, 1998 and 1999 are presented in the Figures 1, 2, 3 and 4,

respectively. All activity concentrations are recalculated to the reference date of May 1, 1986. A notable site-specific variability is seen for depth-distributions in profiles collected in different locations.

In the profiles collected in 1991 - 1993 the maximum ^{137}Cs concentration was observed in the top-most layer and no sub-surface maximums were found. Radiocaesium concentrations decrease rather monotonously with increasing depth in these sampled soil profiles and the simple exponential model (1) described above was a satisfactory approximation [Realo et al., 1994, 1995]. The average mass relaxation length values determined for about 25 profiles all over Estonia (incl. those from the NE part) were $42 \pm 15 \text{ kg m}^{-2}$ ($\sim 3 \text{ cm}$) and $110 \pm 30 \text{ kg m}^{-2}$ ($\sim 10 \text{ cm}$) for the Chernobyl and the nuclear weapons test fallout, respectively.

In almost all profiles collected in 1998-1999 the highest concentrations of the total and Chernobyl radiocaesium were not observed in the top layer, but at the depth of 3 - 5 cm below ground surface. As the sub-surface maximums were found, this feature determined the search for a different model. Notable variations in the depth- dependent concentrations of different soil type, vegetation, water composition, etc. were also found. As a result, we choose the lognormal model (2) and together with the data for the profiles collected, revisited the former depth-distribution data collected in 1991-1993. Both total and Chernobyl ^{137}Cs concentration in profiles were fitted by using the SCIENTIST 2.0 software. As a rule, the lognormal distribution gave a reasonable fit to the measured data. The fitting parameters, x_c and w , were determined for each profile and their mean values for the samples collected during one year were calculated.

Figure 5 demonstrates time-dependence of the mean parameters values, x_c and w , for total ^{137}Cs in the measured profiles. The same dependence for the Chernobyl ^{137}Cs is presented in Figure 6. A rather weak increase in both parameter values with time is observed for both cases. That means a broadening of the distribution and a shift of its maximum to deeper soil layers in time. For total ^{137}Cs , where both contamination components coexist, a linear fit describes the observed time dependence of the yearly mean parameters in the profiles as follows: $x_c \sim 0.4 t$ and $w \sim 0.05 t$, where t is time (y) and x_c is given in cm. For the Chernobyl radiocaesium component a slightly weaker dependence of these parameters on time is obtained: $x_c \sim 0.24 t$ and $w \sim 0.029 t$ in the time interval of 1991 - 1999. It is seen that these time-dependencies have not been linear over the whole time period since 1986.

The observed weak time-dependences of the ^{137}Cs depth-distribution parameters demonstrate a rather slow migration rate of radiocaesium to deeper soil layers and its strong retention in the near-surface soil. This conclusion drawn for the neighbouring Nordic countries [Isaakson et al., 1999] supports our former assumption made on comparison of the nuclear weapons test and Chernobyl radiocaesium depth-distributions in Estonian soils [Realo, et al., 1995]. At the same time, shapes of the depth-dependencies in profiles collected in different times shows significant differences.

Our profile results can be further evaluated by means of the other mathematical models describing the time-dependent transport of radionuclides in soil. We have started the compartmental modelling approach, which has been successfully used by other authors for describing the time-dependent activity in different soil layers and predicting the future behaviour [Kirchner, 1998]. For this purpose a simple model has been compiled for modelling using SCIENTIST. In addition, in order to eliminate site-specific variability and to enable better comparison with the model new samplings in the former locations have been performed. At present both modelling attempts as well as analysis of these profiles are in progress.

Conclusions

Most sample profiles collected in NE Estonia in 1998-1999 show a different depth-dependent behaviour of the Chernobyl ^{137}Cs activity concentration in comparison with our results obtained in 1991-1993. Sub-surface maximums of the ^{137}Cs activity concentrations are characteristic for the soil profiles collected in later years, which can not be explained by a monoexponential model any more. Our preliminary modelling has demonstrated that a lognormal distribution can be satisfactorily used for fitting of the found ^{134}Cs / ^{137}Cs (Chernobyl) depth-dependencies and to derive the time-dependent migration parameters in Estonian soils. A weak time-dependence of the fitting parameters confirms slow effective migration rates and a strong retention of radiocaesium in upper soil layers. Additional samplings and analysis for different soil types and locations are in progress.

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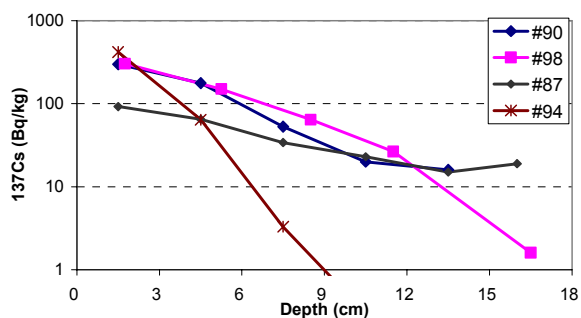


Fig. 1. The vertical distribution of ^{137}Cs in the soils in NE Estonia in 1991.

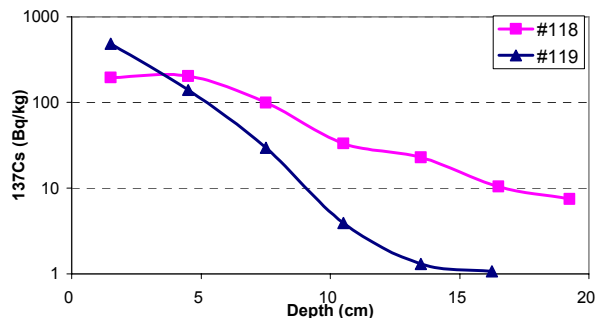


Fig. 2. The vertical distribution of ^{137}Cs in the soils in NE Estonia in 1993.

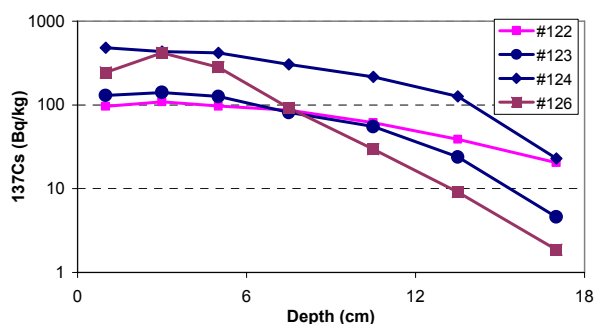


Fig. 3. The vertical distribution of ^{137}Cs in the soils NE Estonia in 1998.

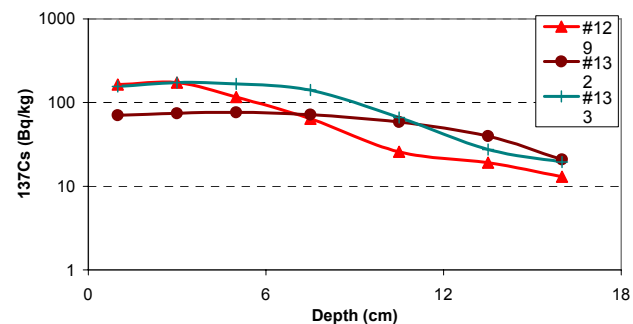


Fig. 4. The vertical distribution of ^{137}Cs in the soils NE Estonia in 1999.

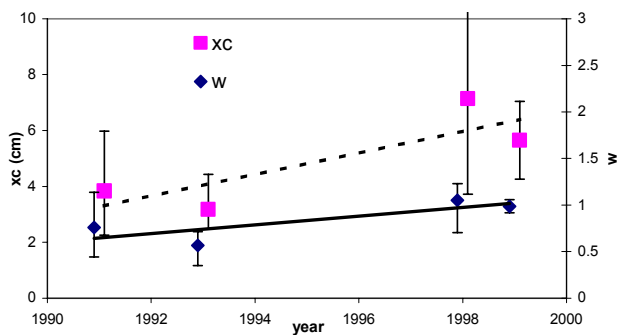


Fig. 5. Mean fitting parameters x_c and w for total ^{137}Cs in soil profiles.

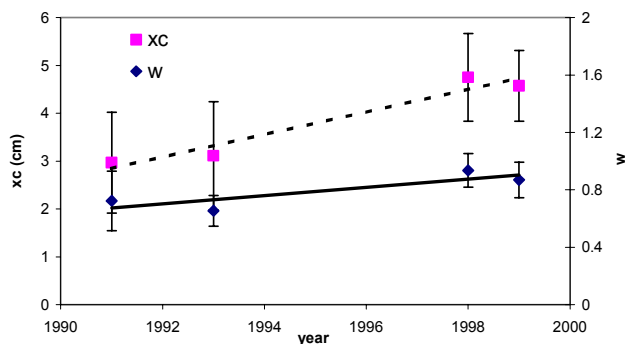


Fig. 6. Mean fitting parameters x_c and w for the Chernobyl ^{137}Cs (^{134}Cs) in soil profiles.

Regional probabilistic nuclear risk and vulnerability assessment by integration of mathematical modelling and GIS-analysis

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Abstract

The Kola Peninsula, Russian Arctic exceeds all other regions in the world in the number of nuclear reactors. The study was aimed at estimating possible radiation risks to the population in the Nordic countries in case of a severe accident in the Kola Peninsula. A new approach based on probabilistic analysis of modelled possible pathways of radionuclide transport and precipitation was developed. For the general population, Finland is at most risk with respect to the Kola NPP, because of: *high population density or proximity to the radiation-risk sites and *relatively high probability of an airflow trajectory there, and precipitation. After considering the critical group, northern counties in Norway, Finland and Sweden appear to be most vulnerable.

Key words: Kola Peninsula, Nordic countries, nuclear reactors, residential risk, vulnerability

Introduction

The Kola Peninsula of Russian Arctic exceeds all other regions and countries in the world in number of nuclear reactors. Bergman and Baklanov (1998) classified the Kola NPP and nuclear submarines at refuelling as high-risk objects regarding radiological consequences. For risk assessment close to a NPP zone, numerical modelling methods have been actively developed (PSA 1995, MACCS 1990). However, on the regional scale, and for complex assessments of risk, especially including social and regional aspects, such approaches are either under elaboration or cover only some aspects of risk. GIS-based analyses integrated with mathematical modelling allow to develop a common methodological approach for complex assessment of regional vulnerability and residential risk, by merging together separate aspects: modelling of consequences, probabilistic analysis of pathways, dose estimation etc.

The purpose of this study was to develop a methodology for complex nuclear risk and vulnerability assessment and to estimate possible radiation risk to the population in the Nordic countries in case of a severe accident at the Kola NPP.

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Materials and methods

Numerical modelling

Numerical modelling is widely used to study long-range airborne transport and deposition of radioactive matter after a hypothetical accident. Depending on scale and consequences of interest, different models can be used.

1. For probabilistic analysis (atmospheric transport pathways, airflow and precipitation probability), isentropic trajectory model (Merrill 1994) and cluster analysis technique (Mahura et al., 1999; Baklanov *et al.* 2000) were used.

For the airflow probability fields, more than 233000 5-day forward trajectories that originated over the nuclear accident region for the period 1991-1995 were calculated (Jaffe *et al.* 1997; Baklanov *et al.* 2000). These fields show geographical variations of airflow patterns from the accident site. To account for the contribution of the possibility of radionuclide wet removal during the transport of an air parcel, the value of relative humidity was calculated simultaneously for each trajectory point (i.e. at latitude, longitude and pressure level point) (Baklanov *et al.* 2000). Based on the calculated temporal and spatial distribution of the relative humidity, precipitation factor fields were constructed over the geographical areas.

2. For long-term consequences for population after an accident, the MACCS (MACCS 1990) and empirical models based on the Chernobyl effects on Scandinavia (OCDE 1987; Hägg 1990; Moberg 1991; Dahlgaard 1994) were applied.

Probabilistic social-geophysical risk

A vulnerability analysis identifies the geographic areas and populations susceptible to damage or injury in case of an accidental release (Lowry *et al.* 1995). In contrast to risk in the Probability Safety Assessment, in our probabilistic approach, ‘risk’ is some complex characteristic of vulnerability of different territories with respect to the Kola NPP or other nuclear risk sites.

For estimation of consequences of radiation impact to Man, we will proceed from the official conception of IAEA (1987). This conception supposes a non-threshold linear dependence between dose and effect. Any level of radiation is considered as harmful.

For assessment of risk/vulnerability we consider the social-geophysical *factors*, which depend on the location of the area of interest and its population: proximity to the radiation risk sites; population density in this area; presence of critical groups of population; and *probabilities*: probability of an accident of a certain severity at the radiation risk sites; probability of air transport pathways towards the area of interest from a risk object (from probabilistic trajectory modelling); probability of precipitation over this area during the transport of the plume along the trajectory (from probabilistic modelling).

For estimation of vulnerability/risk for different regions, let us introduce a risk function defined as a complex index of probability of risk for different factors. Let us define this risk function R_i from the Kola NPP for an administrative unit by two different methods. The first method supposes multiplication of different factors and probabilities (all P and F will be defined later):

$$R_i = P_{acc,i} P_{tr,i} P_{pr,i} F_{dem} F_{dis,i} F_{t,i} F_{cg} F_{soc} F_{ev}. \quad (1)$$

The second method doesn't have a clear physical interpretation, but is widely used in various risk/vulnerability studies (Lowry *et al.* 1995; Obee *et al.* 1998). It supposes a weighted sum of the above-mentioned factors of different nature. We suggest an alternative approach that involves the multiplication of a number of risk probabilities and a weighted sum of other factors. This allows for attributing different importance to the risk factors, yet introduce an element of uncertainty associated with the subjective choice of weights:

$$R_i = P_{acc,i} P_{tr,i} P_{pr,i} (a_1 F_{dem} + a_2 F_{dis,i} + a_3 F_{t,i} + a_4 F_{cg} + a_5 F_{soc} + a_6 F_{ev}). \quad (2)$$

In (1) and (2), P_{acc} is a function defining probability P_k of an accident of a certain class k and severity I_k : $P_{acc} = \sum_{k=1}^m P_k I_k$ (In this study, we, however, assume that an accident has happened, i.e. $P_{acc} = 1$).

P_{tr} is a probability that the trajectory of the accidental plume will reach a certain territory (area of interest). In our case we consider a probability of passing the trajectory through a territory of 2.5 degree x 2.5 degree size (Jaffe *et al.* 1997).

P_{pr} is a probability of precipitation over a certain territory during the plume pass. In a general case, wet deposition - being determined by precipitation - can be included as a factor controlled by the intensity of precipitation. In our case, the factor of wet deposition is determined by relative humidity during passing the air masses through a territory (Baklanov *et al.* 2000). So, it is advisable to consider it as a function of probability. Let us assume that the probability of precipitation, P_{pr} , is 0 for a relative humidity, q , less or equal to 50%, and 1 for $q = 100\%$.

F_{dis} is a factor, representing dispersion and dry deposition of the radioactive plume on its way from the accident site. Different long-range transport models (ADPIC, DERMA) or a simpler empirical function of distance can be used for its definition. Let us define F_{dis} as a function of distance and dispersion parameter σ according to the Gaussian equation at short distances (< 100 km) and as a polynomial on a basis of numerical experiments by the model DERMA (Sørensen 1998; Baklanov 1999) at a regional scale (> 100 km). And let us scale F_{dis} to 1, normalising it by the maximum concentration, found close to the accident site.

F_{dem} is a population factor for the general group. For method 2, let us define this factor as population density, scaled so that $F_{dem} = 1$ in the areas with the maximum population density and $F_{dem} = 0$ for inhabited areas. Since we use population density, we will classify the result as vulnerability of a territory with respect to the Kola NPP or other radiation risk site. In method 1 we use the number of people, rather than population density. Collective and individual risk is calculated analogous to the collective and individual doses.

F_t is a function defining risk connected to a quick transport of contamination, and it is inversely proportional to the time for reaching a certain territory by the plume. Let us initially set it to 1, because it is already indirectly included in F_{dis} . However, it is advisable to include this factor in the analysis for emergency preparedness to stress the quick impact.

F_{cg} is a factor defining vulnerability of the critical groups of the population to radioactive contamination. Let us define it as a function proportional to the population density of critical groups D_{cr} normalised by the population density for the general population D_g and a ratio of risk/criticality

for a critical group r_{cg} : $F_{eg} = r_{cg} D_{cr}/D_g$, where r_{cg} is defined as the ratio of the individual dose for a critical group to the individual dose for the general population based on experimental data of the Chernobyl effects on Scandinavia. In this study we consider only one critical group of the population - reindeer-herders, constituted generally by aboriginal Saami in Lapland. The ratio r_{cg} for different areas of the north of Norway, Finland, Sweden and the Murmansk region varies from 40 to 100 based on the Chernobyl data (AMAP 1998).

F_{soc} is a factor of social risk, which depends on risk perception, preparedness of safety measures, systems for quick reaction, economical and technical means, counteracting consequences of a possible accident etc. This factor can be defined for the administrative units in accordance with some scale by subjective estimation or a set of criteria. However, in this study it was not included.

F_{ev} is a factor defining ecological vulnerability of an area. Cumulative intake of radionuclides by Man can be estimated based on models of different complexity (Balonov *et al.* 1999) through effectiveness of transfer in the food chains within specific ecosystems and consumption of various food. In this case, it is necessary to include new GIS layers of soil, vegetation, consumption habits etc. into the database. Now we are not going into details with this aspect of risk, because it will be a task for further consequence analysis (Bergman and Ågren 1999). Methodical approach based on critical loads to recognise ecologically sensitive (to radioactive contamination) territories is actively developed within the bounds of the AMAP program.

The weight coefficients a_i vary depending on the relative importance of each of the factors. These weights introduce a main difficulty and an uncertainty in the analysis, because there is a summation of the factors of different nature.

The correctness or appropriateness of the suggested formulations is indirectly supported: considering fewer factors of the similar nature (i.e., in a limit or asymptotic case), we obtain the known and approved dependencies. I.e., the novelty of the suggested method is in generalisation of the known methods at the expense of the introduction of new factors of different nature and, from the other side, in simplification of the relations for each factor.

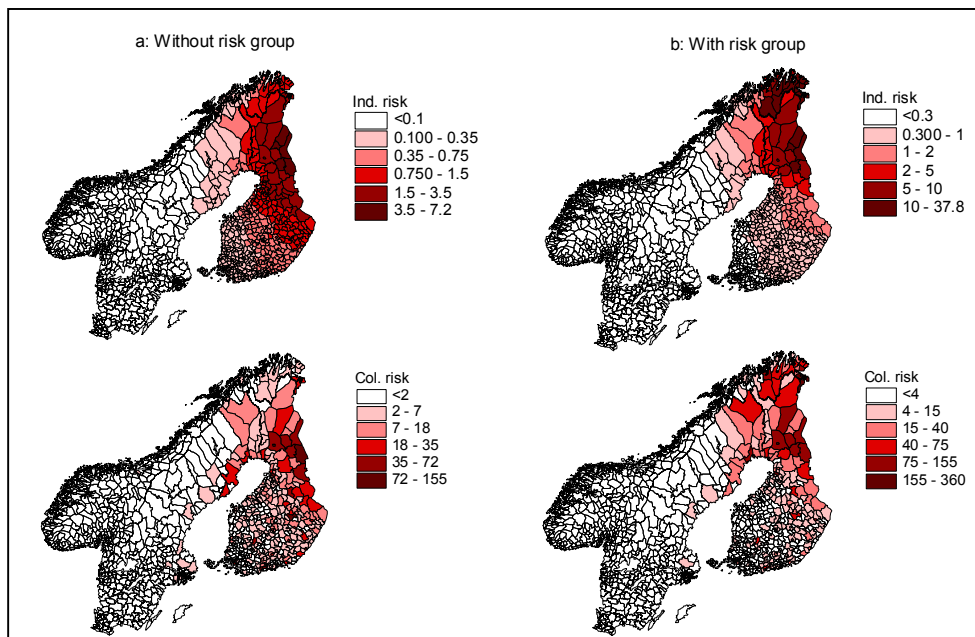


Figure 1. Map of probabilistic risk (relative units) from the Kola NPP to the population in the Nordic countries estimated by method 1.

GIS-analysis for risk and vulnerability assessment

A GIS provides a powerful tool for analysing the relationship among different factors in order to assess risk/vulnerability for the Nordic countries at administrative level (community, county, country). First, the modelling results showing probability for atmospheric pathways and precipitation during the transport were exported to the GIS format for producing several thematic layers. The population data at administrative level for the Nordic countries were obtained for 1996. Then each layer was georeferenced to the basic map (in geographic projection) by an affine transformation using tics.

The population density was derived for each administrative unit from the population attributive table by normalising the population number by the area. The thematic layers were joined with the administrative data by means of the Spatial Join module (within Arc/Info). As a result, new features such as the probabilities, and proximity to the nuclear site were assigned to each administrative unit. In case, when an administrative unit didn't fall within one class, the attribute was calculated as an area-weighted sum.

Composite Mapping Analysis (CMA) is based on the GIS overlay method, but applies adding, multiplying, scaling and weighting of the GIS layers. A GIS-based CMA integrates a wide range of risk-site-related and human-related factors that affect the territorial vulnerability and thus was used for probabilistic vulnerability assessment.

Results

Figure 1 shows maps of the collective and mean individual risks from the Kola NPP for the population of the Nordic countries by administrative unit, estimated by method 1. For general group of population, Finland appears to be at highest residential risk, because of either relatively high population density or proximity to the radioactive risk site, coupled with a relatively high probability of both airflow trajectories and precipitation. Figure 2 shows a map of the estimated by method 2 vulnerability of different administrative units in the Nordic countries with respect to the Kola NPP. Despite different approaches, both methods have revealed the same administrative units (counties) as the most vulnerable or the units where population is at most risk. In the 1st method, risk is divided in collective and mean individual risk, whereas in the 2nd method the population factor was accounted for by using a population density for each group.

Although the critical group - saami (reindeer-herders) is small, after taking it into account, the collective- and mean individual risk patterns have changed towards dominance of the northern counties, such as Finnmark in Norway, Lappland in Finland and Norbotten in Sweden. For example, in the most saami-dominated commune Finnmark in Norway, the mean individual risk increases from 0.59 to 10.08, whereas the collective risk elevates from 63 to 1090 after taking the critical group into account.

For the general population group, along with the northern counties, the county Uleåborg is also subject to high risk because of high population density.

In Figures 1 and 2, the scale for the risks should be understood in a relative way. For example, the county Uleåborg is at twice higher risk compared to the county Lappland with respect to summary collective risk (method 1, column 2), or Finland in general is at 10-times higher risk compared to Norway and Sweden (method 2, column 8). The absolute values of the estimated risk are not

important in this context. In reality, the absolute risk and severity of a situation will be defined by the parameters of the release. For example, in case of a release of 1 PBq, the deposition, dose and absolute risk (expressed, for example, in mortality) can be very low compared to a release of 60 PBq under the same other conditions, whereas the relative proportion between risks for Finland and Sweden, for example, will remain about the same.

Also, these estimations outline possible worst-case situation: a person can remain at home and thus will not get the highest dose, or special rescue measures will be undertaken in time (withdrawal radioactively contaminated food and goods from use, early warning, treatment with medicine, evacuation).

Estimation of the probabilistic complex risk can be illustrated for the limit cases, when certain probabilities are set to 1. If for a certain territory, for example, Västerbotten county in Sweden, we set the probability of trajectory there from the Kola NPP to 1, the probability of precipitation there is also equal to 1, then in case of a unit release (1 Bq) of ^{137}Cs the mean deposition of radionuclides will be equal to $1.5 \cdot 10^{-12} \text{ Bq/m}^2$. Correspondingly, in case of a release of 10 PBq, the deposition will be ca 15 kBq/m^2 . Taking the factors of population and critical groups for Västerbotten into account, the mean collective dose for population by unit area (km^2) will be $5.7 \cdot 10^{-4} \text{ manSv/km}^2$, and the mean individual dose will be 0.132 mSv, and total collective dose will form 34 manSv. This risk is comparable with the mean deposition of ^{137}Cs and the radiological consequences from the Chernobyl in this county of Sweden. This method can be used for verification of the optimal scaling and weighting coefficients for various factors.

Conclusion

1. For the general population group, a large part of Finland is subject to high risk with respect to the Kola NPP, because of high population density or proximity to the radiation risk sites coupled with a rather high probability of atmospheric pathways there with precipitation during the transport.
2. Although the critical group - saami (reindeer-herders) is small, taking it into account implies that the collective- and mean individual risk patterns change towards the dominance of the northern counties: Finnmark (No), Lappland (Fi) and Norbotten (Sw).
3. The integration of the GIS-analysis and mathematical modelling is a very successful and flexible tool for a complex risk assessment.

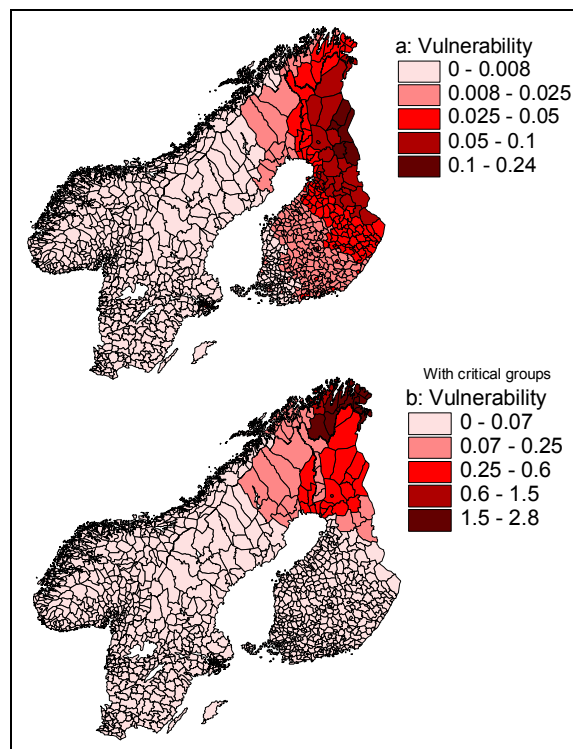


Figure 2. Map of vulnerability (relative units) by administrative unit in the Nordic countries in respect to the Kola NPP estimated by method 2.

Acknowledgement

The study was done within the bounds of ÖCB, INTAS and NARP projects. The authors are very grateful to Dr. L. Thaning and Prof. R. Bergman from FOA, Sweden, to A. Mahura from the University of Washington, USA, and to Ö. Pettersson from CERUM, Umeå University, Sweden.

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Title	Proceedings of the 8 th Nordic Seminar on Radioecology. 25-28 February 2001, Rovaniemi, Finland
Author(s)	Erkki Ilus (editor)
Affiliation(s)	STUK, Finland
ISBN	87-7893-126-6
Date	April 2002
Project	NKS/BOK-2
No. of pages	233
No. of tables	35
No. of illustrations	99
No. of references	-
Abstract	<p>This report contains proceedings of the 8th Nordic Seminar on Radioecology held on February 25-28, 2001 in Rovaniemi, Finland. The Seminar was arranged by STUK - Radiation and Nuclear Safety Authority of Finland and supported by the NKS. The Seminar was intended to be a “final forum” of the four-year NKS radioecology project BOK-2, <i>Radioecological and Environmental Consequences</i>, which was focused on the consequences of releases of man-made radionuclides into the environment. The programme of the Seminar consisted of 3 invited lectures, 31 oral presentations and 22 poster presentations dealing with marine, terrestrial and freshwater radioecology, methods, foodstuffs, models, whole-body counting and doses to man.</p>
Key words	Radioecology; marine radioecology, terrestrial radioecology, freshwater radioecology, methods, foodstuffs, modelling, doses to man