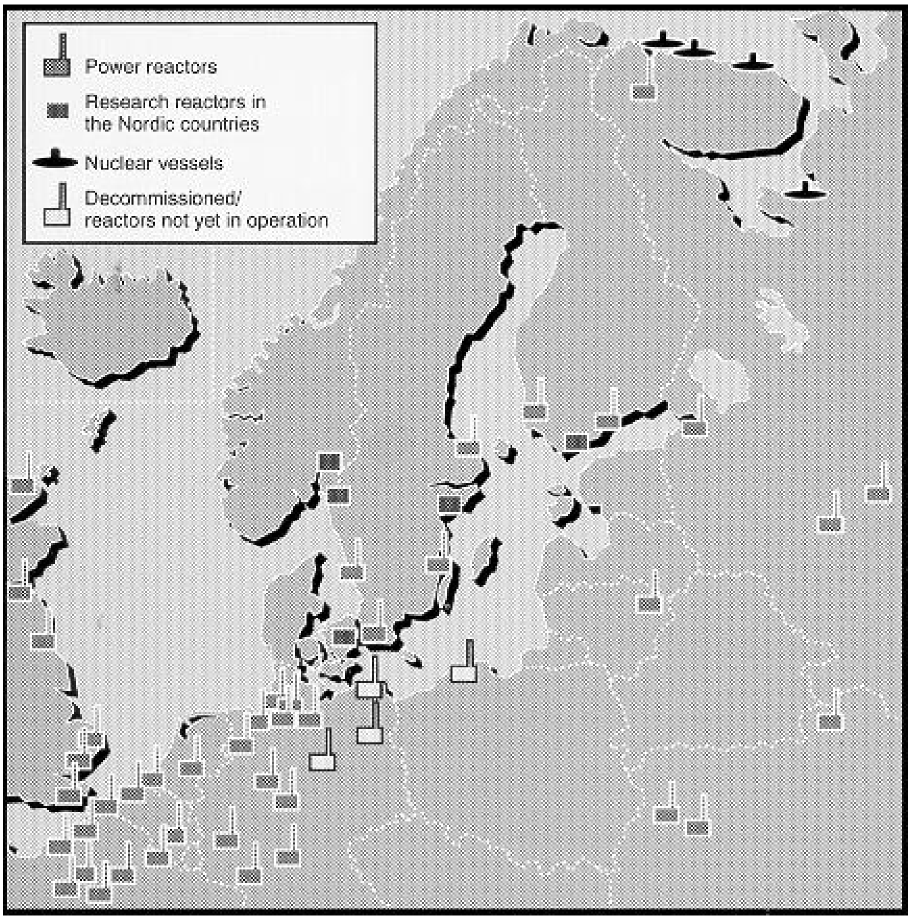
Marine Radioecology



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

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

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

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Final Report of the Nordic Nuclear Safety Research Project EKO-1

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June 1998

This is NKS

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

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

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

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

Additional financial support has been given by the following organizations:

In Finland: Ministry of the Interior; Imatran Voima Oy (IVO); Teollisuuden Voima Oy (TVO)
In Norway: Ministry of the Environment
In Sweden: Swedish Rescue Services Board; Sydkraft AB; Vattenfall AB; Swedish Nuclear Fuel and Waste Management Co. (SKB); Nuclear Training and Safety Center (KSU)

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

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

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Abstract

Results of the EKO-1 project for the period 1994 - 1997 are summarised in this report. The aim of the project was to make a joint Nordic study on radionuclides in sediment and water and the interaction between these two phases. Relatively less emphasis has been put on this factor compared to others in previous Nordic studies on marine radioecology. For some of the participating countries this work was the first of its kind undertaken. The project work involved field, laboratory and model studies. Results of the study have appeared in various scientific journal and it has formed the bases for two Ph.D. theses and two M.Sc. theses.

Key words

Radioecology, marine radioecology, sediments, distribution coefficient, sedimentation rate, radiocaesium, modelling, doses to man

EKO-1 Summary

In the original project plan for the NKS / EKO-1 project it was stated that:

> *The main aim of this project is to enable better and faster assessments to be made of the effects of releases of radionuclides to the marine environment, taking health and economic factors into account.*

The last sentence reflects that policy makers often need to consider sources which are of relatively minor importance as health risks, but which can be of considerable concern for the public (and thus have a major effect on society, e.g. economically).

Assessments are generally based on models describing the main processes influencing the behaviour of the radionuclides. In the marine ecosystem these main processes are:

1. water movement and mixing
2. sediment-water interaction
3. biological transfer (e.g. the uptake of radionuclides by fish)

Of these processes the interaction of sediments with water has been studied relatively less than the other main processes. It was therefore decided to focus the EKO-1 project work on radionuclides in sediments and water, and the interaction between them. Various site specific factors can affect this interaction, e.g. sedimentation rates. The ability of the sediment to bind radionuclides from sea water is also an important factor. This is described by the distribution coefficient (K_d) which gives the ratio of concentration of the radionuclide in sediments to that in water.

The EKO-1 project work was planned as follows:

1. *Model work* - Identifying, estimating and validating parameters of main interest
2. *Research*
    - 2a) *Field studies:*
        - 2a1) Environments typical for various Nordic regions
        - 2a2) Environments with special physical or chemical characteristics
    - 2b) *Laboratory studies*
3. *Dissemination of information* - Seminars, reports, articles

In the project work emphasis was also put on other aspects viewed to be important for the aim of the project:

- Quality assurance
- Use of internet technology for more efficient dissemination of information
- Maintaining a link with the related work done within NKS/EKO-2.3 on freshwater ecosystems

- Following what was being done internationally in a similar field and avoiding duplicate work
- Supporting developments of plans for a Nordic course on radioecology.

Increased Nordic competence and co-operation in marine radioecology

Maybe the most important outcome of the EKO-1 project is the increased Nordic competence and co-operation in marine radioecology, especially concerning the interaction of radionuclides with sediments. Only some of the Nordic countries had previously conducted their own research in this field and there had never been any joint Nordic research or compilation of sedimentation rates and levels of ^{137}Cs and $^{239+240}Pu$ in sediments. The quality of the research done is manifested in the scientific articles that have been published as a result of the project work. Furthermore the EKO-1 project work has formed the basis for one Ph.D. thesis finished in April 1997 and another will be finished in 1998. Two M.Sc. theses have also been finished based on the project work.

Model are a very important tool for assessing the (real or possible) consequences of releases of radionuclides to the environment. The most valuable resource for the decision maker are however the scientists who know both the models and their parameters, and what can be predicted and with what accuracy. Every model, however good, needs a skilled interpreter.

Model studies

The EKO-1 project supported models studies for the Baltic Sea area and the long term effects of reactors dumped in the Kara Sea and from the Komsomolets submarine.

The model studies predict that the collective dose to year 2050 to the population around the Baltic Sea is around 1400 manSv from Chernobyl fallout, 700 manSv from nuclear weapons fallout and 200 manSv from European reprocessing. Other sources such as nuclear installations contribute much less (1 manSv). The man-made radionuclides are however dwarfed by natural radionuclides such as ^{210}Po, which gives 20000 manSv for the same period.

Model studies were also used to estimate the long term effects of the marine reactors dumped in the Kara Sea by the former Soviet Union. The collective dose to the world population truncated at 10000 years give a total dose of about 1 manSv.

The effects of possible releases from the Komsomolets submarine were investigated using model studies. The calculation of the dose was truncated after

1000 years. The total collective dose due to ingestion of seafood was estimated at 8 manSv.

Additionally the reliability of the model was tested by comparions with measured values, sensitivity analysis and parameter uncertainty analysis. The analysis of uncertainty of collective doses showed that the predicted variability of the collective dose from dumping in Arctic waters is two orders of magnitude. Thus the collective dose quoted earlier is predicted at a level of 1 manSv and not to exceed 100 manSv.

Laboratory studies

The laboratory studies have helped to gain a better understanding of the water-sediment interaction process. They showed a variation in the distribution coefficient (K_d) with e.g. sediment type, salinity and contact time. The variation due to sediment types was an order of magnitude and the same types of sediments showed values of K_d that were up to two orders of magnitude higher in freshwater than seawater. This implies that e.g. floods moving contaminated sediments from freshwater systems to the sea could cause release of radionuclides from the sediments. The results from the laboratory studies are also important for model work where K_d is an important parameter. The studies provide a better understanding and quantification of the variability that can be expected in this important parameter.

Field studies

The field studies were divided into two main categories:

1. Environments with special physical or chemical characteristics (emphasis on processes)
2. Environments typical for various Nordic regions (emphasis on regional characteristics)

Environments with special physical or chemical characteristics (process studies)

The process studies focused mainly on the behaviour of plutonium in sediments and its interaction with water. For 20 years there have been speculations about the possible remobilisation of plutonium from sediments, but until now it had not been proven that it takes place. Special attention has been paid to possible releases under anoxic (oxygen deficient) conditions. Poor ventilation and organic releases causes anoxic conditions in considerable parts of the near bottom water in the Baltic Sea (about 19% of the total area). This situation can also occur in bays and fjords of Skagerak and Kattegat. A study at Framvaren Fjord, Norway, was the first to prove that remobilisation is occurring and a model explaining the behaviour of the plutonium was also constructed. The model fits well with the observed data.

The indication is that permanent anoxic conditions must persist for many years with little or no water renewal in order to produce suitable conditions for actinide mobilisation from sediments. The behaviour of plutonium under anoxic conditions was also the subject of a study in Hästholmsfjärden Bay, Finland. Radionuclides from the Loviisa Nuclear Power Plant enter this bay, which becomes anoxic during part of the year. Remobilisation of plutonium or caesium from the sediments to water could not be detected. This is however in agreement with the results of the study at Framvaren Bay which showed that the anoxic conditions must prevail for a long time for the remobilisation to occur. Plutonium can be transported by rivers to the marine ecosystem. A study of the processes in the mixing zone between fresh water and sea water was carried out at the mouth of the Kalix river, northern Sweden, where it flows into the Baltic Sea. Laminated sediments and suspended matter collected in the estuary of the Kalix river shows that the input of plutonium to the Baltic Sea from river runoff at present is of rather small importance but may in the long term perspective become one of the more important sources as the residence time of plutonium in the Baltic Sea water column is much shorter than the residence time in the river drainage basins. Drainage basins containing large percentage of mire and wetland will be of particular importance.

Environments typical for various Nordic regions (regional characteristics)

Field studies were carried out in various areas of concern for the Nordic countries. The study area spanned from Thule on the west coast of Greenland to the Arctic Seas north of Siberia. The Baltic Sea was included and parts of the Atlantic Ocean. In most cases the distribution in sediments of various radionuclides was determined as well as sedimentation rate and distribution coefficient (K_d). The studies helped to determine site specific parameters for the different areas. They also showed that the sedimentation rate could not in some cases be correctly determined by using just one method. In one study there was an extensive comparison of the different methods in the Baltic Sea. None of the methods was judged suitable for routine use and no systematic difference between methods could be seen. More than one method should be used whenever possible.

An investigation in the arctic seas gives no indication of any large extra sources for anthropogenic activity besides the well known fallout from atmospheric nuclear bombs test, discharges from European reprocessing plants and the Chernobyl accident releases. However smaller or local contributions from e.g. the dumped nuclear material in the Kara Sea and releases by the Siberian rivers from Russian nuclear facilities are not possible to exclude in this investigation.

Quality assurance

Quality assurance has been an important ingredient in the EKO-1 project work. Emphasis was put on the following two factors:

a) Sampling
b) Analysis

The design of a sediment sampler can have a crucial effect upon the results. A sampler that is well suited for certain conditions (or a certain type of study) might be less suitable in other conditions. A survey was made of the samplers in use and a report was written listing the results and discussing the advantages and disadvantages of each type of sampler.

An intercomparison was organised amongst Nordic laboratories participating in the NKS EKO projects. A few additional laboratories were invited to join (including some in the Baltic States). Two types of samples were sent to the participating laboratories for analysis, and the results were then compared, evaluated and the findings were published in a report. The results showed that many laboratories did not show satisfactory results (according to criteria described in the published report). This was especially true concerning the analysis of beta emitting radionuclides. The study showed also that the analysis of gamma emitting radionuclides such as Cs-137 can be improved considerably. Work on improving quality in gamma spectrometry has subsequently been taken up in the NKS EKO-3.2 project.

The EKO-1 seminars

Another important factor in increasing the Nordic competence in this field have been the EKO-1 seminars. Radioecology is a multi-disciplinary field. Much of the work done is related to work done on stable nuclides. The EKO-1 seminars have therefore been used to bring together on one hand participants in the EKO-1 project and other Nordic experts in radioecology and on the other hand Nordic experts in related fields and other international experts. During the project period two seminars have been held dealing with issues which were considered to be of major relevance for the project work:

- Kristineberg September 20th - 21st, 1995, *Sedimentation processes*
- Helsinki April 2-3, 1997, *Dating of sediments and determination of sedimentation rate*

Many of those who have participated in the EKO-1 project work have commented that for them personally these seminars were the most important aspects of the EKO-1 work.

Use of internet technology for the dissemination of information

The project period 1994-1997 has seen an explosive growth in the use of internet technology. The EKO-1 project has tried to make use of the increasing opportunities the Internet offers. Texts have been distributed within the EKO-1 work group, and World Wide Web (WWW) pages have been set up.

Contacts with the Baltic States

The Baltic States were invited to participate in the EKO-1 intercomparison for laboratory measurements. Subsequently most of the laboratories also participated in EKO-3.2 work on quality assurance in laboratory measurements. The participating laboratories have expressed their pleasure with being able to participate in this NKS work and expressed a wish for some form of continuation. The participation of the Baltic States in the EKO-1 intercomparison did not require any NKS funding.

Recommendations

The funding that the NKS provided for work in the current period was very valuable, even though it only covered part of the project cost. It provided the necessary stimulus to turn work in each of the participating countries into a joint Nordic undertaking. The EKO-1 project has resulted in that the Nordic countries are now in a much better position than before to assess the short and long term effects of radionuclides that have been (or might be) released and incorporated into sediments. The individual countries can now continue their own work within this field.

Marine radioactivity continues to provide topics of concern for the Nordic countries. The Baltic Sea is an important source of food for many of the Nordic countries. Even though the individual doses resulting from the consumption of sea food from the Baltic Sea are relatively low, they can nevertheless be considerable compared to other doses caused by man-made radionuclides. In Denmark the major part of doses to humans from man-made radionuclides comes from sea food caught in the Baltic Sea.

A project focusing on the Baltic Sea would be of considerable relevance for the Nordic countries. The project could also be linked with studies concerned with the short and long term effect of runoff from the catchment area and into the Baltic Sea.

Recently there has been considerable public concern in the Nordic countries over the possible effects of the increased release of ^{99}Tc from Sellafield. It is important for authorities in the Nordic countries to be able to make an assessment of the effect of this release. Furthermore the ^{99}Tc pulse can be a very useful tracer for studying processes in the Atlantic Ocean, the Arctic seas and the Baltic Sea.

The NKS has made a major contribution to the maintenance of competence in the field of environmental radioactivity in the Nordic countries. But it has also been an important factor in the build-up of competence. As a part of the previous project period (1990-1993) a course on radioecology was held in 1991 at Lund, Sweden.

Many of the students from that course have been active participants in the work within EKO-1 and EKO-2 in the present period. Now a new course has been scheduled for the spring of 1998. Support from the NKS for this course and other activities strengthening the build-up of competence would be valuable.

The Nordic countries have established a culture of co-operating together in a very close and informal manner. The NKS has created a network of competent people in the field of marine radioactivity in the Nordic countries. Other forms of international co-operation and projects cannot replace this network. It is important that work on marine radioactivity will continue to be supported by the NKS. Otherwise it would be much more difficult to deal with many issues of concern for the Nordic population in the marine environment, especially in case of a sudden release of radionuclides to the environment.

EKO-1 Sammenfatning

I den oprindelige projektplan for NKS/EKO-1 projektet fremgår følgende:

> *Hovedformålet med projektet er at gøre det muligt at udføre bedre og hurtigere vurderinger af sundhedsmæssige og økonomiske konsekvenser af udslip af radioaktive stoffer til havmiljøet.*

Dette udsagn afspejler, at man ofte er nødt til at tage hensyn til forureningskilder, der udgør forholdsvis små sundhedsmæssige risici, men som kan fremkalde betydelig bekymring i befolkningen og således være af stor (f.eks. økonomisk) betydning for samfundet.

Vurderinger af konsekvenser af radioaktiv forurening baserer sig ofte på modeller, der beskriver transport af radioaktive stoffer i miljøet. De vigtigste processer for transport og overførsel af radioaktive stoffer i det marine miljø er følgende:

1. havstrømme og opblanding af vandmasser
2. vekselvirkning mellem vand og sediment
3. biologisk optag (f.eks. optag af radioaktivitet i fisk)

Af disse processer er vekselvirkningen mellem vand og sedimenter blevet undersøgt forholdsvis mindre end de andre. Det blev derfor besluttet at fokusere EKO-1 projektarbejdet på radioaktive stoffer i sedimenter og vand og på vekselvirkningen mellem disse medier. Sedimenternes egenskab til at binde radioaktive stoffer fra havvandet er en vigtig faktor. Denne beskrives ved den såkaldte sediment fordelingskoefficient K_d, der udtrykker forholdet ved ligevægt af koncentrationen af radioaktivitet i sedimentet i forhold til koncentrationen i vandet.

Arbejdet i EKO-1 projektet blev planlagt som følger:

1. *Modelarbejde* - Identifikation, estimering og validering af de vigtigste parametre
2. *Undersøgelser*
   - 2a) *Feltstudier:*
     - 2a1) Typiske nordiske miljøer
     - 2a2) Miljøer med særlige fysiske eller kemiske forhold
   - 2b) *Laboratorieundersøgelser*
3. *Informationsarbejde* – Seminarer, rapporter, artikler

I projektarbejdet blev endvidere lagt vægt på andre aspekter, der var af betydning for projektets formål:

- Kvalitetssikring
- Brug af internettet for en mere effektiv informationsformidling
- Forbindelser til tilsvarende arbejde inden for NKS/EKO-2.3 projektet om ferskvandsøkosystemer
- Forbindelser til andet internationalt arbejde inden for marin radioøkologi for at undgå overlap
- Støtte til forberedelse og planlægning af et nyt nordisk kursus i radioøkologi.

Øget nordisk kompetence og samarbejde inden for marin radioøkologi

Blandt de vigtigste resultater af EKO-1 projektet er den øgede nordiske kompetence og det øgede samarbejde indenfor marin radioøkologi, især vedrørende vekselvirkningen mellem radioaktive stoffer og marine sedimenter. Kun enkelte af de nordiske lande har tidligere forsket inden for dette område, og der har ikke tidligere været gennemført fælles-nordiske forskningsprojekter f.eks. med sammenstilling af sedimentationsrater og niveauer for ^{137}Cs og $^{239+240}$Pu i sedimenter. Kvaliteten af det udførte arbejde er dokumenteret i de videnskabelige artikler, der er blevet offentliggjort som en følge af projektarbejdet. EKO-1 projektarbejdet har desuden bidraget til to ph.d. afhandlinger, den ene afsluttet i april 1997 og den anden i 1998. Desuden er gennemført to kandidatafhandlinger (M.Sc.) baseret på projektarbejdet.

Matematiske modeller er vigtige redskaber for at kunne vurdere de potentielle eller faktiske miljømæssige konsekvenser af radioaktiv forurening. Det er endvidere vigtigt med erfarne folk, der kender modellerne, ved hvad de kan benyttes til og resultaternes nøjagtighed. Det kræver erfaring at fortolke modelresultater uanset hvilken model, de stammer fra.

Modelundersøgelser

EKO-1 projektet har støttet modelstudier for Østersøen og for langtidskonsekvenserne af atomreaktorer dumpet i Karahavet og fra Komsomolets undervandsbåden.

Modelberegninger har vist, at kollektivdoser fremskrevet til år 2050 til befolkningen omkring Østersøen fra ^{137}Cs i fisk er omkring 1400 manSv fra radioaktivt nedfald fra Tjernobylulykken, 700 manSv fra nedfald fra atmosfæriske atomprøvesprængninger og 200 manSv fra europæiske oparbejdningsanlæg. Andre kilder, så som nukleare anlæg, bidrager langt mindre (1 manSv). Den radioaktive forurening bidrager dog langt mindre til den samlede kollektivdosis end naturligt forekommende radioaktivitet, hvor ^{210}Po i samme periode giver en kollektiv dosis på 20000 manSv.

De sundhedsmæssige konsekvenser af udslip af radioaktivitet fra ubådsreaktorerne, der blev dumpet af Sovjetunionen i Karahavet, er blevet vurderet ved modelberegninger. Kollektivdoserne til verdens befolkning blev omkring 1 manSv fremskrevet over 10000 år.

Endvidere blev de sundhedsmæssige konsekvenser af hypotetiske udslip af plutonium fra den sovjetiske Konsomolets ubåd, der sank på 1600 m dybde i 1989 ved Bjørnøen undersøgt ved modelberegninger. Kollektivdosen til verdens befolkning blev beregnet til 8 manSv ved en fremskrivning på 1000 år.

Pålideligheden af modelberegningerne er blevet undersøgt ved sammenligning mellem beregnede og målte værdier, sensitivitetsanalyser og parameter-usikkerhedsanalyser. Usikkerhedsanalysen af kollektivdoserne fra dumpningerne af ubådsreaktorerne i Karahavet har vist sig at give en variation på to størrelsesordener. Dette betyder, at kollektivdosen på 1 manSv vurderes til ikke at overstige 100 manSv.

Laboratorieundersøgelser

Laboratorieundersøgelser har bidraget til at forbedre forståelsen af vekselvirkningen mellem vand og sediment. Sediment-fordelingskoefficienten K_d har vist sig at variere med hensyn til både sediment type og salinitet. Variationen med sedimenttyper blev fundet til en størrelsesorden, og samme sedimenttyper viste K_d-værdier, der var op til to størrelsesordener højere for ferskvand end for havvand. Dette medfører, at oversvømmelser ved floder hvor kontaminerede sedimenter transporteres til havet, kan forårsage frigørelser af radioaktiv forurening fra sedimenterne. Resultaterne af laboratorieundersøgelserne er også vigtige for modelarbejdet, hvor K_d er en vigtig parameter. Undersøgelserne giver en forbedret forståelse og kvantificering af den variation, man ser for denne parameter.

Feltstudier

Feltstudier har fundet sted inden for to hovedområder:

1. Miljøer med særlige fysiske og kemiske forhold (vægt på processer)
2. Miljøer typiske for norden (vægt på regionale forhold)

Miljøer med særlige fysiske og kemiske forhold (processtudier)

Undersøgelserne af processer har koncentreret sig om, hvorledes plutonium opfører sig i sedimenter og vekselvirker med sedimenter. Man har længe spekuleret over muligheden af remobilisering af plutonium fra sedimenter, men indtil nu har det ikke været bevist. I særlig grad har man undersøgt muligheden for remobilisering under anoxiske (iltfattige) forhold. Ringe tilførsel af iltet vand og tilstedeværelse af organisk materiale forårsager anoxiske forhold i betragtelige områder ved bunden i Østersøen (omkring 19% af det samlede område). Denne situation kan også opstå i bugte og fjorde i Skagerrak og Kattegat. En undersøgelse i Framvaren Fjord i Norge er den første, hvor man har fundet bevis på remobilisering, og hvor men har udviklet en model til at beskrive processen. Det fremgår, at remobiliseringen af aktinider fra sedimentet kræver, at de anoxiske forhold har været gældende i en længere årrække. Undersøgelser af plutonium under anoxiske betingelser er også blevet foretaget i Hestholmsfjärden Bugt i Finland. Radioaktive stoffer fra Loviisa atomkraftværket udledes til bugten, der bliver anoxisk i visse perioder af året. Her var det imidlertid ikke muligt at påvise remobilisering af plutonium eller cæsium fra sedimenterne. Dette er imidlertid ikke i uoverensstemmelse med resultaterne fra Framvaren Fjord, som viste at de anoxiske forhold skal være til stede i lang tid, før remobiliseringen finder sted. Plutonium kan transporteres med floder til marine økosystemer. En undersøgelse af de processer, der finder sted i et område med opblanding af ferskvand og havvand blev foretaget i udløbet af Kalix elven i Nordsverige, hvor elven løber ud i Østersøen. Prøver af lagdelte sedimentkerner og opslemmede sedimentpartikler indsamlet i mundingen af Kalix elven viser, at tilførslen af plutonium til Østersøen med floder på nuværende tidspunkt er af mindre betydning, men at tilførslen på længere sigt kan blive en vigtig kilde, da opholdstiden af plutonium i Østersøens vand er meget kortere end opholdstiden i flodernes afvandingsområder. Afvandingsområder, der består af moser og andre vådområder, vil i denne sammenhæng være af særlig betydning.

Miljøer typiske for norden (regionale forhold)

Der er foretaget feltundersøgelser i en række områder af betydning for de nordiske lande. Områderne har strakt sig fra Thule på vestkysten af Grønland til de Arktiske have nord for Sibirien. Desuden har Østersøen og dele af Atlanterhavet været inddraget. For det meste er dybdefordelingen af radioaktive stoffer i sedimenterne bestemt tillige med sedimentationshastigheder og sediment fordelingskoefficienter K_d. Ved undersøgelserne er parameterværdier blevet bestemt for forskellige lokaliteter. Undersøgelserne viste endvidere, at sedimentationshastigheder i visse tilfælde ikke kunne bestemmes korrekt ved kun at benytte en enkelt metode. I en undersøgelse i Østersøen blev foretaget en omfattende sammenligning af forskellige metoder. Der blev ikke fundet systematiske forskelle mellem metoderne, men ingen af metoderne blev fundet velegnet til rutinemæssig anvendelse. Det blev konkluderet, at man så vidt muligt bør benytte mere end en enkelt metode til at bestemme sedimentationshastigheder.

En undersøgelse i de arktiske have viser ingen tegn på større forureningskilder for radioaktivitet, når man ser bort fra de velkendte bidrag fra nedfaldet fra stormagternes atmosfæriske atomvåbenforsøg, udslip fra europæiske oparbejdningsanlæg og Tjernobylulykken. Undersøgelsen udelukker dog ikke muligheden af mindre udslip fra dumpet radioaktivt affald i Karahavet og udslip med de sibirske floder fra rusiske nukleare anlæg.

Kvalitetssikring

Kvalitetssikring har været et væsentligt element i arbejdet med EKO-1 projektet. Der er især blevet lagt vægt på følgende to forhold:

a) Prøveindsamling

b) Radiometrisk analyse

Udformningen af et apparat til prøvetagning af sedimenter kan være afgørende for resultaterne. En prøvetager, der er velegnet til visse forhold eller undersøgelser kan være mindre egnet til andre forhold. I forbindelse med projektet blev foretaget en opgørelse af de prøvetagere, der benyttes af projektdeltagerne. Opgørelsen er afrapporteret sammen med en gennemgang af fordele og ulemper ved de forskellige prøvetagere.

Blandt deltagerne i NKS EKO-projekter blev desuden foretaget en sammenligning af analyseresultater for radioaktivitet i sedimentprøver. Enkelte andre laboratorier blev inviteret til at deltage, nogle fra de Baltiske stater. To typer sedimenter blev sendt til deltagerne for bestemmelse af radioaktivitetsindhold, og resultaterne blev derefter sammenlignet og sammenstillet i en rapport. Sammenligningen viser, at mange laboratorier ikke opnåede tilfredsstillende resultater i forhold til de anvendte kriterier. Dette var især tilfældet for beta-emittere (f.eks. ^{90}Sr). Undersøgelsen viste også, at kvaliteten af analyserne af gamma-emittere (f.eks. ^{137}Cs), kan forbedres betydeligt for flere laboratorier. Arbejdet med at forbedre kvaliteten ved gammaspektrometriske analyser er efterfølgende blevet taget op i EKO-3.2 projektet.

EKO-1 seminarer

EKO-1 seminarerne har været endnu en faktor, der har bidraget til at forøge den nordiske kompetence indenfor området. Radioøkologi rummer mange discipliner og anvender mange metoder, der også anvendes til at bestemme stabile stoffer. EKO-1 seminarerne er derfor blevet benyttet til at samle såvel deltagere i EKO-1 projektet og andre nordiske eksperter som nordiske eksperter indenfor beslægtede områder og andre internationale eksperter. I løbet af projektperioden er der blevet afholdt to seminarer med temaer af betydning for projektarbejdet:

- Kristineberg, 20-21 september 1995, *Sedimentationsprocesser*
- Helsinki, 2-3 April 1997, *Datering af sedimenter og bestemmelse af sedimentationshastighed*

Flere deltagere i EKO-1 projektet har givet udtryk for, at disse seminarer har givet dem stort fagligt udbytte.

Brug af internet teknologi til informationsformål

I løbet af projektperioden 1994-1997 er der sket en eksplosiv vækst i brugen af internet teknologi. EKO-1 projektet har benyttet sig af de muligheder internettet giver både med hensyn til elektronisk post og information via World Wide Web. Megen kommunikation indenfor projektgruppen mellem projektlederen og deltagerne er foregået via elektronisk post, og adskillige tekster og dokumenter er blevet fordelt til projektdeltagerne og andre via EKO-1 hjemmesider.

Kontakt til de Baltiske stater

Institutioner i de baltiske stater er blevet inviteret til at deltage i EKO-1 sammenligningen af analyseresultater for radioaktivitet i sedimentprøver. Efterfølgende deltog flere af disse laboratorier i EKO-3.2 arbejdet med kvalitetssikring i radioanalytisk arbejde med gammaspektrometri. De deltagende baltiske laboratorier har givet udtryk for stor tilfredshed ved at deltage i NKS arbejdet og udtrykt ønske om at fortsætte. Deltagelsen af de baltiske laboratorier i EKO-1 sammenligningen fandt sted uden betaling fra NKS.

Anbefalinger

Det økonomiske bidrag, som NKS har stillet til rådighed for EKO-1 projektet har været af afgørende betydning for arbejdet. Projektet har dog ikke kunne gennemføres med de opnåede resultater uden de nationale økonomiske bidrag, der har været betydelige. NKS's bidrag har imidlertid været afgørende for, at samle aktiviteterne fra deltagerne til en fælles-nordisk aktivitet. EKO-1 projektet har medført, at de nordiske lande nu er langt bedre i stand til at vurdere kort- og langtidskonsekvenserne af udslip af radioaktivitet til det marine miljø.

Radioaktiv havforurening er fortsat et emne, der undertiden skaber bekymring i de nordiske lande. Østersøen er et vigtigt spisekammer for mange af de nordiske lande. Selv om strålingsdoser til mennesker fra konsum af fisk fra Østersøen er relativt små, kan de være betragtelige i forhold til strålingsdoser fra andre typer af radioaktiv forurening. I Danmark skyldes hovedparten af strålingsdoserne til befolkningen fra radioaktiv forurening konsum af fisk fra Østersøen.

Et NKS projekt med fokus på Østersøen vil være af stor betydning for de nordiske lande. Projektet kan kombineres med undersøgelser af kort- og langtidsvirkninger af transport af radioaktivitet med floder fra landområder til Østersøen.

Der har for nylig været offentlig opmærksomhed i de nordiske lande på de mulige virkninger af forøgede udslip at radioaktivitet (^{99}Tc) fra Sellafield i England. Det er af betydning for de nordiske myndigheder for strålingsbeskyttelse at kunne vurdere betydningen af disse udslip. I forbindelse med et nyt NKS projekt kan udslippet af ^{99}Tc endvidere benyttes som sporstof for vandstrømme i Atlanterhavet, det Arktiske Ocean og Østersøen.

NKS giver et vigtigt bidrag til at vedligeholde kompetencen inden for radioaktivitet i miljøet i de nordiske lande. Men NKS bidrager også til at opbygge ny viden. I forbindelse med det tidligere NKS program (1990-1993) blev et kursus i radioøkologi afholdt i Lund i 1991. Mange deltagere fra dette kursus har været aktive i EKO-1 og EKO-2 projekter under den nuværende programperiode. Et nyt kursus i radioøkologi er blevet planlagt til foråret 1998. Bidrag fra NKS til støtte for kurset og andre aktiviteter vil forstærke opbygningen af kompetence.

De nordiske lande har indenfor NKS etableret en tæt og uformel måde at samarbejde på. Gennem EKO-1 projektet har NKS skabt et netværk af sagkyndige i de nordiske lande inden for marin radioøkologi. Andre former for internationalt samarbejde og projekter kan ikke erstatte dette netværk. Det er vigtigt, at NKS fortsat støtter forskning indenfor radioaktivitet i det marine miljø. I modsat fald kan det på længere sigt blive vanskeligt at håndtere problemer om radioaktiv havforurening, der vækker bekymring i de nordiske lande, især i tilfælde af et pludseligt udslip.

Table of contents

Introduction

The marine environment contributes generally a relatively small part of the doses to the population from man made sources. This contribution can however be very significant, and in some cases even a relatively large part of the dose from man made radionuclides. Currently the major part of the dose to the Danish population from man made radionuclides comes from sea food caught in the Baltic Sea. Releases of radionucldies to the marine environment can also have a considerable economic effect even though the resulting doses may be relatively small. It is therefore important for the authorities to be able to assess as accurately as possible the effects of releases (real or possible) of radionuclides to the marine environment.

Objective of the EKO-1 project

> *The main aim of the EKO-1 project was to enable better and faster assessments to be made of the effects of releases of radionuclides to the marine environment, taking health and economic factors into account.*

To achieve this aim one needs accurate and reliable radioecological models. An assessment of the effects of releases of radionuclides to the marine environment must be based on radioecological modelling. All of the knowledge about the marine ecosystem cannot however be quantified. A sound understanding of the marine ecosystem is essential for an efficient use of radioecological models and for interpreting the results.

Project outline

The EKO-1 project concentrated on improving the usefulness of available models. Emphasis was also put on supporting research and other activities (quality assurance, strengthening competence, dissemination of information) which are an essential basis for radioecological modelling. This will contribute to:

- improved input for decision makers, e.g., by means of environmental impact calculations and dose and risk assessment.
- better and quicker information to mass media, the general public and other interested parties.

The work in the proposed project therefore concentrated on:

a) Model work, identifying parameters of main importance where data is needed for parameter determination or validation.

b) Research with the aim of providing the data needed in (a)

Some of the factors that determine the dose to humans after a release of radionuclides to the marine environment are:

a) Behaviour of radionuclides in sea water after release, e.g. the interaction of radionuclides with particles and sediments
b) Uptake of radionuclides by biota and fish from sea water
c) Transfer of radionuclides from sea food to humans by consumption

The interaction of radionuclides with sediments has not been studied to the same degree as (b) and (c). This interaction is complex and is furthermore affected by:

- site specific factors
- special physical and chemical factors
- anoxic conditions (lack of oxygen) in the sea can under certain conditions make Pu much more soluble in sea water and cause it to be reintroduced from sediments.

Factors (b) and (c) have been studied for a number of years. Less emphasis has been put on factor (a) since it does not require thorough studies, as long as the emphasis is put on short term assessments of releases of soluble radionuclides (e.g. Cs and Sr, which interact little with particles and sediments).

In short term assessments of soluble radionuclides (e.g. Cs and Sr) the interaction with sediments is of relatively minor importance. For long term assessments however the interaction with particles and sediments must be taken into account. This interaction can lead to the removal and reintroduction of radionuclides to the ecosystem.

Insoluble radionuclides (e.g. Pu) can be very reactive towards sediments. These processes can be of major importance for the behaviour of such nuclides right from the beginning of the release. Not even short term consequences can be adequately described without taking these processes into account, let alone long term consequences.

Organisation of work

The EKO-1 project work was planned as follows:

1. *Model work* - Identifying, estimating and validating parameters of main interest
2. *Research*
    - 2a) *Field studies:*
        - 2a1) Environments typical for various Nordic regions
        - 2a2) Environments with special physical or chemical characteristics
    - 2b) *Laboratory studies*
3. *Dissemination of information* - Seminars, reports, articles

In the project work emphasis was also put on other aspects viewed to be important for the aim of the project:

- Quality assurance
- Use of internet technology for more efficient dissemination of information
- Maintaining a link with the related work done within NKS/EKO-2.3 on fresh-water ecosystems
- Following what was being done internationally in a similar field and avoiding duplicate work
- Supporting developments of plans for a Nordic course on radioecology.

Division into sub-projects organised on a national basis

Sediment sampling from the marine environment can be very expensive. Field work had therefore to be co-ordinated with work done in each country in order to make the best use of the available resources. This meant that the work in the project had to be divided into sub-projects, at least one in each participating country. The reports from the individual sub-projects can be found in a separate EKO-1 technical report, NKS/EKO-1(98)TR-1 *Final reports from sub-projects within the Nordic Nuclear Safety Research Project EKO-1*. In the text of this report references are made to the work in the subprojects by using a two letter ISO country code (DK, FI, IS, NO and SE) and a project number (1 - 3). Co-ordination of the work in the project was achieved by holding work group meetings twice per year, where the participants described the status of their work, results obtained and future plans. Internet technology (e-mail and web pages) was also used for exchange of information within the group.

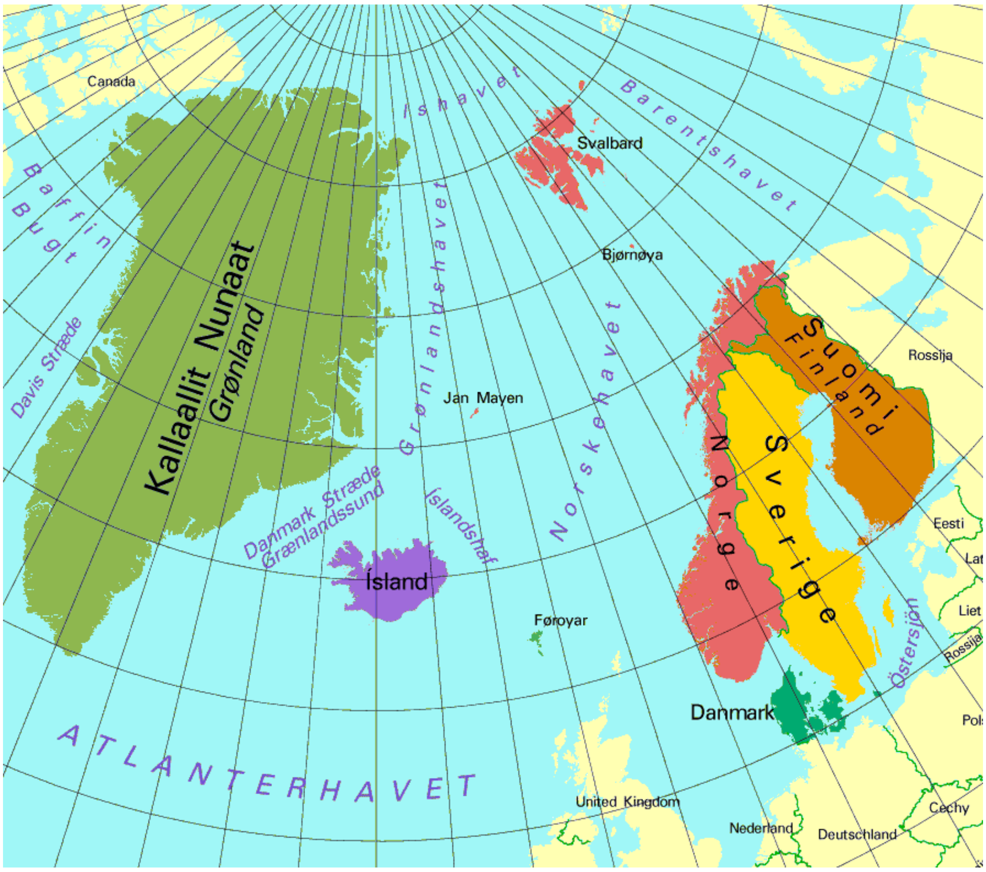
Structure of report

The main text of the report is in the following two chapters "Background" and "Results and discussion". The former discusses some relevant background information while the latter describes results obtained within the project.

The text of the report was generally written by the project leader or compiled by him from material contributed by the national project groups. Some sections, however, were written by other members of the project group. The name of the author is then given at the beginning of the section.

Background

Maps



Field studies were carried out in various areas of concern for the Nordic countries. The study area spanned from Thule on the west coast of Greenland to the Arctic Seas north of Siberia. The Baltic Sea was included and parts of the Atlantic Ocean. The study areas are shown in more detail in each national country report, which can be found in the EKO-1 technical report NKS/EKO-1(98)TR-1.

Radionuclide Source Terms to the Nordic Seas

Elis Holm, Lund University

There major sources regarding the presence of radioactive elements in the Nordic Seas can be distinguished, fallout from nuclear tests, releases from European reprocessing facilities and fallout from the Chernobyl accident. In addition there are local releases from nuclear power plants but such are dominated by activation products. We will here only deal with the selected radionuclides ^{137}Cs, $^{239+240}$Pu, ^{90}Sr and ^{99}Tc.

The Baltic Sea

The Baltic Sea is a semi-enclosed, shallow sea in which dissolved substances will remain for a relatively long period compared to many other areas.
The inventory of ^{137}Cs following the Chernobyl accident was estimated to 5 200 TBq in 1986 (1) compared to 600 TBq(2) from nuclear test fallout. Considering decay and outflow (50-60 TBq per year) and inflow (9 TBq per year) the total inventory can today (1997) be estimated to about 4 000 TBq. About 40% of radiocaesium is associated with the sediments.
The inventory of $^{239+240}$Pu was 16.5 TBq from nuclear test fallout and 1.5 TBq from the Chernobyl accident of which the major part is bound to the sediments(3).
The activity ratio ^{137}Cs/^{90}Sr is 1.6 in nuclear test fallout. In 1981 Salo *et al*.(2) estimated the inventory of ^{90}Sr in the Baltic Sea to 552 TBq and only 2.2% was associated with the sediments. Roughly, a total inventory of 400 TBq of ^{90}Sr can be estimated today of which 30 TBq originates from the Chernobyl accident.
Aarkrog et al.(4) investigated the inventory of ^{99}Tc in the Baltic Sea which was estimated to 1.3 TBq of which 1.05 TBq originates from European reprocessing facilities and the remaining mainly from nuclear test fallout.

Temperate and Arctic waters of the Nordic Seas

Fallout from nuclear tests in the Atlantic Ocean can for ^{137}Cs, ^{90}Sr, $^{239+240}$Pu and ^{99}Tc be estimated to 150 000 TBq, 94 000 TBq, 1 800 TBq and 22 TBq respectively north of the 30^0 latitude band excluding the Chernobyl contribution and eventual local fallout. About 57% of the caesium and strontium has decayed until today. About 21% of the surface can be estimated to represent the Nordic Seas i.e. the inventories in temperate and Arctic waters of the Nordic Seas correspond to 13 400 TBq, 8 300 TBq, 380 TBq and 4.6 TBq respectively for the four radionuclides. These are very rough figures since we have not fully considered the latitudinal variation in deposition.
The annual outflow of radiocaesium following the Chernobyl accident from the Baltic Sea into the North Sea is about 60 TBq(3) which means that this source has contributed with about 660 TBq.

Releases of ^{99}Tc from the Sellafield reprocessing plant can be estimated to 760 TBq from 1952 to 1983 compared to 200 TBq from the nuclear fuel reprocessing plant at la Hague in France(5). The releases from Sellafield peaked in 1978 (178 TBq) and were lower thereafter until 1994 and 1995 when about 70 TBq and 190 TBq respectively were released.
Regarding $^{239+240}Pu$ releases from Cap de la Hague can be estimated to 4.5 TBq compared to about 700 TBq from the Sellafield reprocessing plant.
An undecayed amount of 1 200 TBq of ^{137}Cs has been released from Cap de la Hague compared to 40 000 TBq from the Sellafield plant. The corresponding figures for ^{90}Sr are 1 300 TBq and 7 200 TBq respectively. The major releases of radiocaesium and radiostrontium from the Sellafield plant occurred during 1974 - 1982 and consequently about 62 % of the activity remain today
A major fraction of the caesium, strontium and technetium activities have been transported into the North and Norwegian Seas and further into the Arctic Ocean. Plutonium from the European reprocessing facilities has mainly been deposited in sediments in the vicinity of the release source and only about 4%(soluble fraction) has been transported along with caesium and technetium. Fallout plutonium is mainly present in the higher oxidation states (+V +VI) and behaves relatively more conservative with an effective ecological residence time of 8 years in open Atlantic waters(6).

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Collective doses to man from radioactive pollution in the sea

Sven P. Nielsen
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Table 1 summarises collective doses calculated in various projects from a range of actual and predicted releases of radioactivity to the marine environment. The marine environments cover mainly the Arctic Ocean, the Atlantic Ocean, the Baltic Sea and the Mediterranean Sea. The results are not all directly comparable, since the range of radionuclides vary significantly between the different sources and also the time over which doses are integrated. But nevertheless the results roughly illustrate the range of collective doses to man from marine exposure pathways and provide a ranking of the importance due to this index.

Table 1 Collective doses from marine exposure pathways from a range of actual and predicted releases of radioactivity to the marine environment.

Sources	Release (PBq)	Collective Doses (manSv)
Fallout from weapons tests	500	9000
Sellafield, La Hague etc.	150	5200
Chernobyl fallout in Baltic	5	1400
Dumping in NE Atlantic	26	1200
Weapons fallout in Baltic	2	700
Nuclear plants in Europe	1	90
Komsomolets submarine	0.02	8
Marcoule plant	0.5	2
Dumping in Arctic waters	4	1

Processes of relevance for predicting radiological consequences

Sven P. Nielsen
Risø National Laboratory

Important hydrodynamic processes that determine the dispersion of dissolved pollutants (e.g. caesium and technetium) in the seas are advection due to sea currents, and mixing due to tidal and wind energy. Pollutants that associate readily to particulates in the water column (e.g. cobalt and transuranics) are transferred to surface sediments due to sedimentation processes and are not dispersed by hydrodynamic processes to the same extent as dissolved substances. From the surface sediment pollutants may be transferred back into the water column by resuspension processes or further isolated from the water column by continuing burial in the sediment by the sedimentation. Mixing processes occur within the surface sediment due to diffusion and bioturbation and thus counteract the sediment burial process. Pollutants in the water column and sediment are transferred to biota in the marine environment and further to man by ingestion of seafood. Some elements are transferred mainly to fish (e.g. caesium), others mainly to crustaceans (e.g. technetium), and others to molluscs (e.g. transuranics). Other marine exposure pathways than that of ingestion may be considered for individuals who spend extended time on beaches (e.g. fishermen), particularly inhalation of resuspended sea spray and coastal sediment particulates, and external exposure.

Methods used in estimation of the sedimentation rates

Jukka Mattila and Erkki Ilus, STUK

Pb-210 based methods

CF:CS-model (Constant Flux Constant Sedimentation rate-model)

Basic assumptions (Robbins, 1978;):

- the sedimentation rate is constant
- the Pb-210 activity flux to the sediment is constant

The model in practice:

- When the unsupported Pb-210 concentration is plotted on a logarithmic scale against the cumulative dry mass (massdepth), the resulting Pb-210 profile will be close to linear according to the assumptions. The mass flux can be determined from the mean slope of the profile using the least-squares fit procedure (e.g. Oldfield and Appleby, 1984).
- To find the most optimal mean slope and to estimate the error for this slope, the tools of the regression analysis can be effectively used. It is also possible to calculate the coefficient of determination for this linear regression model.
- Calculations are principally based on the equation (Robbins, 1978; Oldfield and Appleby, 1984)

(1) $$C(x) = C(o)e^{\frac{-l_{210} \times m}{r}}$$, where

C(o) = the unsupported ^{210}Pb concentration at the sediment/water interface [Bq kg^{-1}],
C(x)= the unsupported ^{210}Pb concentration in the sediment at the depth x [Bq kg^{-1}],
l_{210} = ^{210}Pb decay constant,
r = the sedimentation rate [g cm^{-2} y^{-1}] and
m = the massdepth [g cm^{-2}] of the observed depth x [cm].

The slope b of the least-squares fit is then

(2) $$b = \frac{l_{210}}{r}$$,

wherefrom sedimentation rate r can be resolved. The error for sedimentation rate is possible to estimate by using the standard error of the slope b.

CRS-model (Constant Rate of Supply-model)

Basic assumptions (Appleby and Oldfield, 1978; Oldfield and Appleby, 1984):

- the sedimentation rate can be variable
- the ^{210}Pb activity flux to the surface of the water column is constant **and** the transfer of ^{210}Pb activity from water to sediment is also constant.

This means that the unsupported ^{210}Pb consentration in the sinking material is variying if the amount of sinking material is variying. Therefore it is assumed that the measured activity profile of unsupported ^{210}Pb in sediment is due to changes in the sedimentation rate and to the radioactive decay of ^{210}Pb.

The CRS-model in practice:

- The concentration of unsupported Pb-210 in sediment has to be measured at each layer of the sediment down to the depth, where the concentration is near to zero. Age estimation of a sediment layer is based on the total activity of unsupported Pb-210 in the sediment profile and the activity of unsupported Pb-210 below the sediment layer to be dated.
- CRS-model calculations are based on the equation (Appleby and Oldfield, 1978)

(3) $$A(x) = A(o)e^{-l_{210} \times t}$$, where

A(o) = total residual activity of unsupported ^{210}Pb in the sediment column [Bq cm^{-2}],
A(x)= the cumulative residual activity of unsupported Pb-210 in sediments below the depth x [Bq cm^{-2}]. The age t [y] of the sediment layer at the depth x is then

(4) $$t = \frac{1}{l_{210}} \times \ln\left[\frac{A(o)}{A(x)}\right]$$

and the sedimentation rate r [g cm^{-2} y^{-1}] is possible to calculate from (Robbins, 1978)

(5) $$r = \frac{m}{t}$$, where

m = massdepth [g cm^{-2}] of the dated sediment layer. It is possible to count an error for the age t (Binford, 1990), which can be used when counting error for the sedimentation rate r.

CIC-model (Constant Initial Concentration-model)

Basic assumptions (e.g. Oldfield and Appleby, 1984):

- the sedimentation rate can be variable
- the Pb-210 activity flux to the surface of the water column is constant **but** the transfer of Pb-210 activity from water to sediment is increasing when the amount of sinking particles is increasing.

According to this assumptions the sinking matter should always have the same initial concentration and therefore the measured activity of unsupported Pb-210 should decline unambiquously with the depth in sediments.

The CIC-model in practice (e.g. Oldfield and Appleby, 1984):

- The concentration of unsupported Pb-210 in sediment has to be measured at each layer of the sediment
- CIC-model calculations are based on the equation

(6) $$C(x) = C(o)e^{-\lambda_{210} t}\text{ , where}$$

C(o) = the unsupported ^{210}Pb concentration at the sediment/water interface [Bq kg^{-1}],
C(x)= the unsupported ^{210}Pb concentration in the sediment at the depth x [Bq kg^{-1}],
λ_{210} = Pb-210 decay constant,
t = the age [y] of the sediment layer at the depth x, which can be calculated with the equation

(7) $$t = \frac{1}{\lambda_{210}} \times \ln\left[\frac{C(o)}{C(x)}\right].$$

In CIC-model it is also possible to count an error for the age t (Binford, 1990), which can be used when counting error for the sedimentation rate r.

Cs-137 based method

Basic assumptions:

- the observed activity peak of Cs-137 at some depth of the sediment profile corresponds the year 1986 of the Chernobyl accident.
- the sedimentation rate has been constant between the year 1986 and the sampling year.

The Cs-137 method in practice:

- Cs-137 concentrations are measured in a sediment profile. The massdepth of highest Cs-137 concentration in the sediment profile corresponds the year 1986.
- Calculation of sedimentation rate r [g cm^{-2} y^{-1}] is based on the equation

(8) $$r = \frac{m}{t}, \text{ where}$$

m = massdepth [g cm^{-2}] of the Cs-137 peak and
t = time in [y] between sampling and the year 1986.

The minimum and maximum sedimentation rates r in [g cm^{-2} y^{-1}] have been calculated using the massdepths of the upper and lower edge of the sediment slice, or a broader sediment layer, where the peak concentration exists (Ilus et al., 1997).

Pu-239,240 based method

Basic assumptions:

- the observed Pu-239,240 activity peak at some depth of the sediment/profile, correspods the year 1963, when the maximum global fallout occured.
- the sedimentation rate has been constant between the year 1963 and the sampling year.

The Pu-239,240 method in practice

- Pu-239,240 concentrations are measured in a sediment profile. The massdepth of highest Pu-239,240 concentration in the sediment profile corresponds the year 1963.
- Calculation of the sedimentation rate r [g cm^{-2} y^{-1}] and its error is similar to that of the Cs-137 method (Ilus et al., 1997).

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Results and discussion

This chapter summarises the work done within the EKO-1 project. More information on the work can be found in the EKO-1 technical report, NKS/EKO-1(98)TR-1 *Final reports from sub-projects within the Nordic Nuclear Safety Research Project EKO-1* and the references given there.

Laboratory Studies

Deborah Oughton
NLH

Introduction

Sediments can act as both sinks and sources of radionuclides in the marine environment. The radionuclide speciation and interaction with sediment components determines the degree to which contaminants will be transported to other parts of the ecosystem (Salbu and Oughton, 1995). Hence variations in K_d can have a profound influence on the fate of radionuclides in marine ecosystems. With this in mind, the aim of laboratory studies was to provide some information on water-sediment interaction processes. In particular, ^{134}Cs and ^{85}Sr tracer experiments were used to investigate the kinetics of radionuclide water-sediment interactions, and to determine which parameters influence sorption processes, K_ds and kinetics. The main variables studied were: time, sediment type and salinity. Experiments included water-sediment interactions in both marine and freshwater systems.

Sediment Characteristics

Tracer studies were carried out on marine, estuarine and dirty ice sediments collected during a number of different research expeditions, and included samples from the open Kara Sea, Stepovogo Fjords, Ob Estuary, Yenisey Estuary, Norwegian Coast, Irish Sea, the "Mix" area North of the Ob-Yenisey estuary mouths and sediments collected from Dirty Ice north of the Ob-Yenisey estuary mouths. Dry weight (d.w., 105°C), organic content by loss on ignition (LOI, 550°C), pH, cation exchange capacity (CEC), trace element concentrations, surface area, and mineral type were determined on sediment sub-samples. The sediments varied in terms of organic content, CEC and stable element concentration, but pH was similar: between 7 and 8. Apart from the Irish Sea and Stepovogo sediments, radionuclide concentrations were low.

Tracer Studies

All tracer experiments were carried out under aerobic, dark conditions at 4 °C using wet sediments. Seawater was collected from the Oslo Fjord (pH 8.2, salinity 34

‰); freshwater was from Maridalsvann (pH 5.8). Tracer solutions (^{134}Cs and ^{85}Sr) were added to static tanks containing wet sediment (equivalent to ca. 10 g dry weight) and water (2 l) that had been allowed to equilibrate for two weeks. The sorption of tracers to sediments and rates of change in K_d (ml g^{-1}) for ^{134}Cs and ^{85}Sr were measured as a function of time (Oughton et al. 1997). Variations in K_d according to sediment type were documented. Alternatively, batch studies were carried out wherein labelled seawater or freshwater (20 ml) was added to sediment aliquots (equivalent to 2 g d.w.) and the subsequent remobilization of tracers from the labelled sediments studied as a function of tracer contact time and salinity. Where possible, laboratory-derived K_ds and remobilization of ionic ^{134}Cs and ^{85}Sr tracers were compared to environmental K_ds and remobilization of ^{137}Cs and ^{90}Sr.

Results and Conclusions: Relevance for risk assessment

Distribution coefficients (K_d ml g^{-1}) for ^{134}Cs in seawater-sediment systems varied with contact time, mixing conditions, sediment type, and salinity, and showed a rather good correlation with cation exchange capacity (Fig. 1, Table 1). After 1 year contact time of labelled seawater with different sediments under static conditions, K_d for ^{134}Cs varied by an order of magnitude, ranging from 150 to 1680 ml g^{-1}. As expected, the K_d increased as a function of time, but only relatively small increases were observed after 150-200 days contact time. Thus, for Cs-isotopes, the variation in K_d with time should not result in significant deviations for dispersal box models, which have integration steps of more than 200 days.

Table 1 Distribution coefficients (K_d) for ^{134}Cs and ^{85}Sr tracer in seawater-sediment and freshwater-sediment systems. Average ± SD (n = 2-3).

	^{134}Cs K_d (ml g^{-1})		^{85}Sr K_d (ml g^{-1})		CEC[b]
	1 week	1 year	1 month	1 year	(meq /100g)
Seawater-sediment (pH 7.2, salinity 33‰)					
Mix	430 ± 30	1680 ± 30	11 ± 3	na	84
Ob-I Ob-II	380 ± 40 140 ± 10	1520 ± 80 na[c]			74 na
Kara	300 ± 10	1480 ± 20			68
Irish Sea	190 ± 10	740 ±10			25
Stepovogo	160 ± 10	980 ± 40			46
Yenisey	260 ± 10	900 ± 70			24
Dirty Ice	390 ± 20	na			120
Norway-16	120 ± 10	460 ± 20			29
Norway-26	70 ± 10	150 ± 10			32
Norway-34	100 ± 10	280 ± 20			23
Norway-39	80 ± 10	170 ± 10			27
Freshwater-sediment (pH 5.8)					
Ob-II	5900 ± 400[a]	na	1600 ± 300[a]	na	na
Dirty Ice	24700 ± 2900[a]	na	2300 ± 60[a]	na	120
Norway-26	8 ± 2	2500 ± 300	50 ± 3	160 ± 10	29
Norway-39	8 ± 2	2700 ± 400	50 ± 4	170 ± 10	32
Heimdalen	100 ± 5	12000 ± 2000	110 ± 20	250 ± 10	na
Lösjöen	30 ± 4	7200 ± 800	60 ± 9	170 ± 10	na

a- batch study, shaken;

b- Correlation of K_d with CEC for ^{134}Cs in sediment-seawater system after one year contact time: K_d = 19.8 CEC (R2 = 0.77).

c - na -not analysed

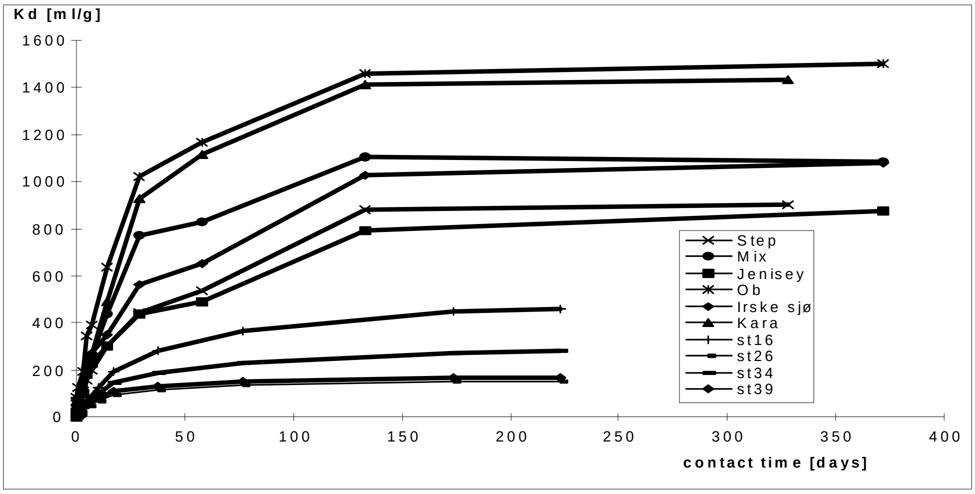

Figure 1 Variation in K_d for Cs-134 as a function of contact time.

Extraction studies showed that the majority of ^{134}Cs was rapidly strongly bound to sediment components. After only 1 hr contact time, more than 70% of the ^{134}Cs was found in the HNO_3 and residual fractions. A small fraction (1-10%) was easily displaced (NH_4Ac extractable) and remained in dynamic equilibrium with soluble species. Hence, physical mobilisation of sediments (e.g. resuspension), rather than chemical remobilization, can represent a major transport mechanism of Cs-isotopes in the environment. Comparison with results from extraction studies on ^{137}Cs in Irish Sea and Stepovogo sediments gave similar distributions for the two Cs isotopes.

Ionic ^{85}Sr was rather conservative in seawater, showing little transfer to sediments: K_d was 11 ± 3 ml g^{-1}. Furthermore, more than 99% of the adsorbed ^{85}Sr was easily extractable with NH_4Ac. Sequential extraction of Irish Sea and Stepovogo sediments indicated, however, that ^{85}Sr was rather less mobile than the ionic tracer (Oughton et al 1997; Børretzen et al.,1995). This could reflect a time-dependent effect, different ^{90}Sr species in the discharge (i.e. non-ionic), or the existence of other water-to-sediment removal mechanisms besides ion-exchange and surface sorption in the natural environment (i.e. biogeochemical cycles). Nevertheless, chemical mobilisation from contaminated sediments, rather than physical mobilisation of sediments, will be more significant for Sr-isotopes than for Cs-isotopes.

The kinetics of the water-sediment water interactions of the ionic ^{134}Cs tracer have been modelled using a simple box model (Fig.2). The rate of decrease of ^{134}Cs activity levels in seawater could be resolved into a two component function, probably represented by relatively fast interaction with sediments by surface sorption or ion exchange followed by slow removal through "less reversibly" (i.e. slower exchange rates) to "irreversibly" or "fixed" sites. Although the model cannot give absolute rate constants for the chemical processes, it is a useful tool in parameterising the sorption rates so that the different sediments can be compared. Optimisation of the model gave similar rate constants for exchange between water and the "reversibly bound" pool , k_1 0.17 ± 0.03 d^{-1} and k_2 0.09 ± 0.05 d^{-1}, Rate constants for transfer to the "fixed"pool varied between the different sediments: k_3 from 0.0003 to 0.001 d^{-1}. Rate constants for remobilization from the fixed pool, k_4 showed large uncertainties, but half-lives seemed to be in the order of decades. Fixation of radionuclides in sediments (in kinetic terms, removal to binding sites in sediments wherein release rates are greater than the half-life of the radionuclide) can in effect represent a mechanism by which the contaminant is removed from the system. Other removal mechanisms include physical decay and burial in the sediment profile. Slow transfer to the "fixed" compartment means that contaminated sediments could act as a secondary source of radionuclides to marine systems (Hunt and Kershaw, 1990). This is particularly important for "pulse" or episodic inputs of radionuclides to marine systems, or for sites with low sedimentation rates.

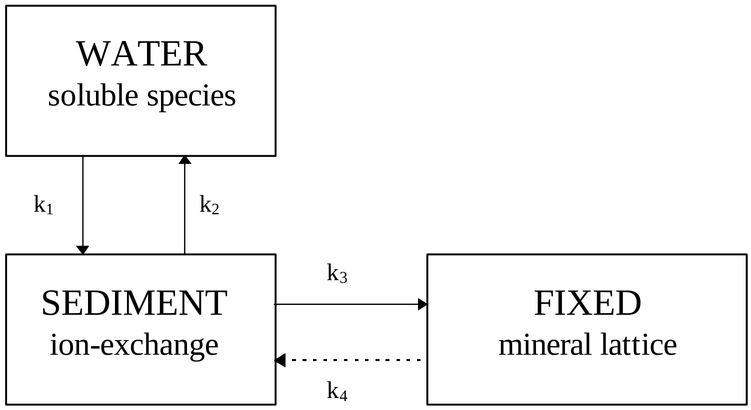

Figure 2 Simple box model describing sorption of Cs-134 tracer to marine sediments

Salinity had a significant effect on K_d, for both ^{134}Cs and ^{85}Sr. For the same sediments, K_ds were up to two orders of magnitude higher in freshwater as compared to seawater (Table 1). This implies a potential for remobilization of radionuclides if contaminated sediments are transferred from freshwater to marine environments, for example, during flooding. The influence of sediment type and salinity on K_ds observed in laboratory experiments, suggest that for ^{137}Cs and ^{90}Sr a K_d range of two orders of magnitude would not be inappropriate for testing the sensitivity of box models.

Freshwater-sediment static tank studies were also carried out on samples collected from Heimdalen (Norway) and Loppesjö (Sweden) lakes (studied in EKO-2 projects), as well as two of the Norwegian coast marine sediments. Results confirmed that after 1 year contact time K_ds for both ^{134}Cs and ^{85}Sr were higher in freshwater-sediment than in seawater-sediment systems (Table 1). However, under static conditions, sorption process were significantly slower in the freshwater as compared to the seawater studies. This could reflect the importance of increased colloid aggregation and particle precipitation in high ionic strength seawater systems. When sediments were shaken with labelled waters and then separated by high speed centrifugation (batch studies), sorption was rapid. In the static system, Cs sorption kinetics were best described by a 3 box model. Sr sorption kinetics were well described by a 2-box model representing water and reversibly-bound components.

The data reported here represents laboratory studies, thus results will always be open to questions regarding the application "real-life" situations. However, the intention of tracer experiments is to investigate some of the processes influencing water-sediment interactions and not to attempt to mimic natural conditions. Differences between K_ds observed in laboratory studies and those derived from environmental measurements could reflect many factors. As well as the obvious variations in physical and chemical parameters, these factors might also include contact time (i.e. non steady-state conditions in the environment), variations in activity concentrations with depth (both for water and sediments) or different source terms. Nevertheless, laboratory studies can offer some important insights into processes influencing the transport of contaminants in the environment. For example, knowledge on the types of parameters which can result in variations in K_ds are useful for assessment of box model predictions and sensitivity.

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Field studies

The field studies were divided into two main categories:

1. Environments with special physical or chemical characteristics (emphasis on processes)
2. Environments typical for various Nordic regions (emphasis on regional characteristics)

Each study is identified with a two letter ISO code for the country responsible for the study followed by a serial number (1-3). A detailed description of each study can be found in the appendix.

Environments with special physical or chemical characteristics (process studies)

The process studies focused mainly on the behaviour of plutonium in sediments and its interaction with water. For 20 years there have been speculations about the possible remobilisation of plutonium from sediments, but until now it had not been proven that it takes place. Special attention has been paid to possible releases under anoxic (oxygen deficient) conditions. This is a situation that can occur in near bottom water in the Baltic Sea and also in bays and fjords of Skagerak and Kattegat. The following studies are described in this section:

1. Anoxic conditions, areas of study: Framvaren, Gullmaren and Byfjorden (SE-1)
2. Anoxic conditions, area of study: Hästholmsfjärden Bay (FI-2)
3. Mixing zone between freshwater and seawater (SE-2)

Anoxic conditions, areas of study: Framvaren, Gullmaren and Byfjorden (SE-1)

The sediments in the anoxic Framvaren fjord acts as a source for actinides to the overlaying water column. The remobilisation process is most likely linked to early diagenetic alteration of the marine organic material in the sediments. This is indicated by the close correlation between Pu, Am and dissolved organic carbon depth profiles in the water column. Speciation studies of the plutonium and americium in the water column shows that both to a large degree are associated to colloidal material in the size range 0.01-0.45 µm. Less than 2% is retained by a 0.45 µm filter which is reflected in the low K_D-values obtained of about 20 000, which is at least a factor of 10 lower than in typical coastal waters. It is also proven that the plutonium exist almost entirely in the trivalent state in the anoxic water column.

This study is the first ever to show extensive remobilisation of plutonium and americium from sediments in anoxic marine basins. Similar remobilisation from sediments most likely occur in other anoxic marine waters where early diagenesis results in humic and fulvic acid production.

Although the remobilised actinides in the Framvaren fjord at present don't pose any radiological hazard due to the lack of fish in anoxic waters it is of great concern to identify processes involved in the remobilisation of actinides from anoxic sediments as such sediments likely will be a major source for actinides in the Baltic Sea and other oxygen sensitive basins in the long term perspective. In such basins the remobilised plutonium may reach oxygenated and biological productive waters by convection.

Results from the temporarily oxygen deficient Gullmarn fjord on the Swedish west coast shows that remobilisation from sediments can not be identified during short (a few months) periods of oxygen deficient water. The rapid bioturbation (quantified by tracer studies) in this fjord results in that sedimenting organic material rapidly is burried and distributed within the upper 10-15 cm of sediments. This means that the organic material is diluted with large amounts of inorganic material which may act as traps for the mobilised actinides.

It is likely that anoxic conditions must persist for a long enough time to allow organic rich sediment to accumulate in sufficient amounts before substantial amount of humics are produced that may mobilise the actinides. This time depends on the organic matter flux to the sediments.

Anoxic conditions, area of study: Hästholmsfjärden Bay (FI-2)

Varying redox conditions may affect the occurrence and concentrations of certain radionuclides in the surface layers of sediments and in near-bottom waters by causing remobilization of radionuclides from surface sediments to the overlying water and their settling back into the sediment. In recent decades about 70.000 km^2 of the sea bottom in the deepest part of the Baltic Sea (about 19% of its total area) have withstood almost continuous anoxic conditions; thus, it is important to know to what extent depletion of oxygen can affect the behaviour of these radionuclides in near-bottom waters. The aim of the project was to resolve the above question in a coastal basin periodically undergoing anoxic conditions.

Radioecological processes in sediments and in near-bottom water under varying redox-conditions were studied in the deep area of the Hästholmsfjärden Bay in Loviisa (eastern Gulf of Finland) in 1995-1996. The Hästholmsfjärden Bay is a semienclosed basin between the mainland and the archipelago and is connected with the open Gulf of Finland only through narrow, shallow sounds.

In 1995, total depletion of oxygen occurred in the hypolimnion of Hästholmsfjärden Bay during 2 periods in late summer and autumn. In 1996, oxygen conditions were the worst ever observed in the Hästholmsfjärden deep. During early autumn anoxic conditions prevailed for more than 1 month in the near-bottom water. The highest total phosphorus and total nitrogen concentrations in the near-bottom water during these periods were 20- and 4- fold compared with the corresponding values in surface water.

According to the results obtained in this project, remobilization of Cs-137 and Pu-239,240 from sediments to near-bottom water is negligible or non-existent in the Hästholmsfjärden deep. If it does occur, however, it may be so slight that it is not possible to observe with the methods used in this study. Although the anoxic periods are quite short in the Hästholmsfjärden deep, they are of sufficient length for strong remobilization of nutrients. If the initiation of remobilization processes in the case of caesium or plutonium were retarded, then the situation may be different in the deep areas of the Baltic Sea where depletion of oxygen has been almost continuous.

Mixing zone between freshwater and seawater (SE-2)

Laminated sediments and suspended matter collected in the estuary of the Kalix river shows that the input of plutonium to the Baltic Sea from river run-off at present is of rather small importance but may in the long term perspective become one of the more important sources as the residence time of plutonium in the Baltic Sea water column is much shorter than the residence time in the river drainage basins. Drainage basins containing large percentage of mire and wetland will be of particular importance.

Environments typical for various Nordic regions (regional characteristics)

Field studies were carried out in various areas of concern for the Nordic countries. The spanned from Thule on the west coast of Greenland to the Arctic Seas north of Siberia. The Baltic Sea was included and parts of the Atlantic Ocean. In most cases the distribution in sediments of various radionuclides was determined as well as sedimentation rate and distribution coefficient (K_d). The studies helped to determine site specific parameters for the different areas. They also showed that the sedimentation rate could not in some cases be correctly determined by using just one method. In one study there was an extensive comparison of the different methods in the Baltic Sea. None of the methods was judged suitable for routine use and no systemic difference between methods could be seen. More than one method should be used whenever possible. The following study areas are described in this section:

1. The Baltic Sea (FI-1)
2. Continental shelf along the European-Siberian Tundra Coast (SE-3)
3. Russian Arctic Seas (FI-3)
4. Various sites from the North Sea to the Arctic (NO-1)
5. Icelandic waters (IS-1)
6. The Thule area, Greenland (DK-1)

The Baltic Sea (FI-1)

Determination of sedimentation rates plays an important role in material balance and model calculations of seas and other bodies of water. The Baltic Sea offers an exceptionally good opportunity to study processes in sediments and sedimentation rates with radioecological methods, because the concentration peaks of Cs-137 and Pu-239,240 are easily detectable in its sediments. In 1995-1996 sediment profiles were taken at 51 sampling stations situated in the Baltic Proper, Bothnian Bay, Bothnian Sea and Gulf of Finland. The aim was to estimate sedimentation rates in different parts of the Baltic Sea by using alternative methods and to consider reasons for eventual differences in results. The Pb-210, Cs-137, Pu-239,240 and the sediment trap methods were used in estimations.

The results show that the accumulation rates of dry matter may vary between 0.006 and 0.90 g $cm^{-2}y^{-1}$ at different sampling stations of the Baltic Sea and the sedimentation rates between 0.2 and 29 mm y^{-1} depending on the sedimentation itself and the method used in calculation. This is a considerable range in results, considering that all of the sampling stations were located in areas of soft sediment bottoms. In general, the sedimentation rates were highest at the Bothnian Sea sampling stations. In the Gulf of Finland the sedimentation rates were highest in the eastern part, while in the Bothnian Bay and in the Baltic Proper the rates were in general lower than in the 2 areas first mentioned.

The differences among the results obtained with various methods varied unsystematically; thus it was not possible to predict that anyone of the methods would always give higher results than any of the others or *vice versa.* The results show that in the Baltic Sea the use of more than 1 parallel methods in estimation of sedimentation rate is highly recommended. None of the methods is necessarily suitable for routine use in the Baltic Sea. In those cases where the Cs-137 or Pu-239,240 peak is distinct, sharply defined and at sufficient depth, these methods may result in the best estimates. On the other hand, at those stations where the peaks are spread out over a broad range of depths, the methods based on Pb-210 may give more accurate results.

One of the advantages of the Pb-210-based methods is that they give information on sedimentation rate over longer period of time than the Cs-137 and Pu-239,240 methods. In contrast to the Pu-239,240 method, Pb-210 can be analysed gammaspectrometrically; however the gammaspectrometric method for Pb-210 is very demanding both for the equipment and the staff. At least in the case of our material, the practice used in the CF:CS-model appeared to be most useable Pb-210-method for the Baltic Sea sediments.

In comparing the methods the sedimentation rates should be presented as accumulation rates of dry matter [g cm^{-2} y^{-1}] to avoid the error caused by compaction of the sediment and its impact on the results. The time scale of the estimation should always be considered because of the changes in the Baltic Sea environment affecting the sedimentation rate. The importance of undisturbed and high-quality samples is especially pronounced in the sedimentation rate studies, because the loss of soft surface sediments during sampling may significantly affect the results when using the Cs-137 and Pu-239,240 methods.

The project has markedly increased our knowledge of sedimentation rates in various subregions of the Baltic Sea, and in addition it has provided further data on vertical distribution of Cs-137 and Pu-239,240, as well as on the total inventory of Cs-137 in Baltic Sea sediments.

Continental shelf along the European-Siberian Tundra Coast (SE-3)

The contribution of ^{137}Cs, ^{134}Cs and ^{90}Sr from the European sources to the arctic seas have decreased in the first half of the 1990's. This is reflected in the measured activities in the different arctic seas which all show lower concentrations compared to earlier measurements. In the Eurasian shelf seas a continuos decrease towards east is recorded and in the central Arctic Ocean comparisons with earlier measurements show lower activities in 1996. The influence from the Chernobyl accident were about one third of the total activity of ^{137}Cs at the Eurasian continental shelf in 1994 and between 10-30 % in the surface water in the central Arctic Ocean in 1996. The distribution pattern of the activity suggests a transport pathway for the European derived activity along the Norwegian coast, passing Barents and Kara Sea to the Laptev Sea. From there the transport is northward and follows the so called transpolar drift across the north pole and onwards to the north Atlantic.

Measurements on ^{137}Cs in bottom sediment on the Eurasian continental shelf point at extensive sediment mixing and high deposition, 500-1000 Bq/m^2. Due to the low sedimentation rates the deposition of ^{137}Cs in the central arctic is much lower, 10-100 Bq/m^2.

The obtained results give no indication of any large extra sources for anthropogenic activity besides the well known fallout from atmospheric nuclear bombs test, discharges from European reprocessing plants and the Chernobyl accident releases. However smaller or local contributions from e.g. the dumped nuclear material in the Kara Sea and releases by the Siberian rivers from Russian nuclear facilities are not possible to exclude in this investigation.

Russian Arctic Seas (FI-3)

The levels of the anthropogenic radionuclides in the Russian Arctic Seas are low compared to the potential sources of pollution and originate meanly from the global fallout, Chernobyl fallout and from the western nuclear fuel reprocessing plants. Fresh release of radioactivity was noticed in this study only in the Kola Bay and in two sampling location in the White Sea. The increased ^{137}Cs concentrations measured in the estuaries of River Dvina and River Yenisey are caused by the riverine transport from the large catchment area.

The sediments of the Russian Arctic Seas are hard. Good and enough long cores for sedimentation rate determination were obtained only in two locations in the White Sea. All the cores from river estuaries were badly mixed.

Various sites from the North Sea to the Arctic (NO-1)

The NO-1 part of EKO-1 involved both laboratory and field studies. The laboratory studies have been described earlier in this report. The following is a summary of the field studies.

At station 26 (Norwegian Sea) the sediments seem to be influenced by radiocesium from the Chernobyl accident. This may be due to direct fallout deposition to the sea surface and followed by a rapid sinking and sedimentation.

At station 16 (North Sea) some influence from Sellafield plutonium is suggested, as the plutonium ratio is significantly higher (0.07-0.09) than would be expected from global fallout (0.03).

Sedimentation rates based on analysis of ^{210}Pb or ^{210}Po varied between 0.03 cm/year - 0.25 cm/year. A suprisingly low sedimentation rate was found in the Yenisey Bay (0.05 cm/year).
It is possible that the dating method is less suited in this area, due to the long winter ice cover.

In general, the rough estimates on K_d values for ^{137}Cs obtained empirically are higher than K_d values obtained from the laboratory studies. This may be due to the fact that the 2 cm surface sediment in most cases has accumulated over many years, carrying contamination from the early eighties when levels of ^{137}Cs in the sea water were higher. The ^{137}Cs in the sediment is now fixed, or being remobilized only very slowly. Burial of the contamination by sedimentation may also make it unavailable for exchange with free water masses.

Icelandic waters (IS-1)

The sea bottom around Iceland has a very uneven topology. This is understandable with Iceland being a part of the Mid-Atlantic Ridge. Iceland is also at the junction of different ocean currents. These factors contribute to create rich fishing grounds around Iceland. Over the years there has been considerable oceanographic research in Iceland and there has been some research concerning sediments. But there had been no studies of radionuclides in sediments prior to the start of the EKO-1 project.

Sampling of sediment cores was attempted at 35 stations around Iceland, repeatedly at most of them. The conditions proved however to be very difficult and only 5 good quality sediment cores were obtained. These cores were analysed for ^{137}Cs, ^{210}Pb and ^{226}Ra. The sedimentation rates were determined using regression analysis of the unsupported ^{210}Pb profile. The obtained fit was very good in all cases (R^2 ranging from 0.94 to 0.99) which indicates that the results are of good quality. Interpretation of the ^{137}Cs profiles was also consistent with the interpretation based on the ^{210}Pb data. The shallower depths (106 m – 215 m) gave sedimentation rates of 0.1 – 0.3 g cm^{-2} y^{-1} (3 – 4 mm y^{-1}) while the cores from 256 m and 972 m gave considerably lower rates (0.06 g cm^{-2} y^{-1} and 0.03 g cm^{-2} y^{-1} respectively).

Two of the sediment cores were taken in the same area, but at different times. These are the cores sampled at depths 210 m and 215 m. The difference in the exact position of these two sites is approximately 100 m. The difference in the obtained results illustrates clearly the variability that can be expected within an area.

The project has been important for research in this field in Iceland. Not only has data for the area been obtained, but also valuable experience gained in sampling under these difficult conditions and in the subsequent interpretation of results.

The Thule area, Greenland (DK-1)

Analyses of radionuclide profiles in sediment cores collected in 1991 at Thule in Greenland have provided information on processes that occur in the sediments. The radionuclides include the naturally occurring ^{210}Pb, $^{239+240}$Pu originating from the aircraft accident in 1968, and ^{137}Cs originating from nuclear weapons testing. The processes include mixing of the surface sediments mainly from biological activity and burial of sediments due to particle scavenging. These processes influence the time scale and the extent to which the plutonium contamination is in contact with seawater and thus available for further dispersion. The quantified description of these processes is necessary for numerical modelling of the impact of radioactive contamination of the marine environment. These processes are of particular importance for the transuranic elements due to the relatively high radiotoxicity, the long physical half lives and the sediment-reactive properties of these elements.

The radionuclide profiles have been analysed with a numerical model to identify values of parameters describing the sediment processes. The average parameter values for the Thule area are: 0.3 cm y^{-1} for the sedimentation rate, 5 cm for the mixing depth and 1 cm^2 y^{-1} for the mixing rate. It was found that the mixing parameters were not correctly identified from the interpretation of the unsupported

^{210}Pb profile alone, but that information from the $^{239+240}Pu$ profile was necessary. This stresses the need for caution when interpreting ^{210}Pb profiles where no other information is available.

Summary of results

The experimental results are not always comparable and therefore it is best to go to each of the national reports in the appendix for details. The following compilation gives some indication of the results obtained.

Area	**Depth** (m)	**Mass accumulation rate** ($g\ cm^{-2}\ y^{-1}$)	**Sedimentation rate** ($mm\ y^{-1}$)
The Baltic Sea (FI-1)		0.006 - 0.90	0.2 - 29
The Kara Sea (NO-1)			0.6
The Barents Sea (NO-1)	290	0.12	1.2
Yenisey Bay (NO-1)			0.5
West of Svalbard (NO-1)	3200		1
Near “Komsomolets” (NO-1)	1680	0.11	1.2
Norwegian Sea (NO-1)	270	0.09	0.4
Skagerak (NO-1)	535	0.15	2.7
The North Sea (NO-1)	280	0.04	0.3
NE of Iceland (NO-1)	420	0.11	2.6
NW of Iceland (IS-1)	250-1000	0.03-0.06	0.6-1.1
NW of Iceland (IS-1)	100-220	0.1-0.3	3-4
Thule (DK-1)			3

In some cases the values just represent one sediment core, in other cases the values represent values from comprehensive surveys (especially the Baltic Sea survey in FI-1)

Model work

This section summarises work done during the project period on models for radiological assessment covering marine pathways. Due to recent concern about the risks from dumping of nuclear waste in the Kara Sea by the former Soviet Union, the work has focused mainly on Arctic waters and the potential contamination of seafood. However, work has also been carried out on the Baltic Sea.

Model description

The marine assessment models simulate the dispersion of radionuclides in the marine environment, the transfer to biota and further to man where doses are estimated to individuals of critical groups and populations. The dispersion of radionuclides in the marine environment is simulated with mathematical models. The models simulate the dispersion of radioactivity in the water due to advective transport including mixing from wind and tidal forces. Association of radionuclides to suspended sediment material is considered along with subsequent transfers to sediments through particle scavenging. Further transfer of radionuclides between the water column and the sediments includes diffusion, bioturbation and resuspension. The models calculate time-dependent concentrations in seawater and sediments from specified inputs of radioactivity to the marine environment.

For box models, the area of interest is divided into a number of boxes which determine the spatial resolution. Box models can provide estimates of radiological consequences considering short-range and short-term as well as long-range and long-term dispersion of radionuclides in the marine environment. This is in contrast to the more realistic hydrodynamic models that provide results with high resolution in time and space, but with a projective time scale of a few tens of years.

Box-model analysis assumes instantaneous uniform mixing within each box with rates of transfer being proportional to the inventories of material in the source boxes. Box models have been used previously in connection with studies of the dispersion of discharges of radioactivity from European civil nuclear installations[i,ii,iii,iv]. The present model for Arctic waters is a combination of two models: 1) one is an adjusted version[v] of a regional box model used for radiological assessments in north-west European coastal areas[vi], and 2) the other is a larger box model covering the Arctic Ocean and the North Atlantic[vii]. The larger box model is derived from a World Ocean general circulation model from which the results have been used for the design of a box structure in the Arctic Ocean and surrounding waters. The resulting structure and water fluxes were selected taking into account expert information based on experimental data from the Barents Sea.

The box-model analysis uses first order differential equations to describe the transfer of pollutants between the boxes. The equations are of the form:

$$\frac{dA_i}{dt} = \sum_{j=1}^{n} k_{ji} A_j - \sum_{j=1}^{n} k_{ij} A_i - k_i A_i + Q_i$$

where $k_{ii}=0$ for all i, A_i and A_j are activities (Bq) at time t in boxes i and j, k_{ij} and k_{ji} are rates of transfer (y^{-1}) between boxes i and j, k_i is an effective rate of transfer of activity (y^{-1}) from box i taking into account loss of material from the compartment without transfer to another, for example radioactive decay, Q is a continuous source of input into box i (Bq y^{-1}) and n is the number of boxes in the system.

The rates of transfer between the aquatic boxes, k_{ij} (y^{-1}) are related to the volume exchanges, R_{ij} ($km^3\ y^{-1}$) according to:

$$R_{ij} = k_{ij} V_i ,$$

where V_i is the volume of water represented by box i.

Figures 1 and 2 show the regions used in the marine box model and Figure 3 shows the structure of the water boxes and their interconnections. Each of the water compartments has associated suspended sediment and the water compartments in contact with the seabed have underlying seabed sediment compartments. The latter are not shown in Figure 3.

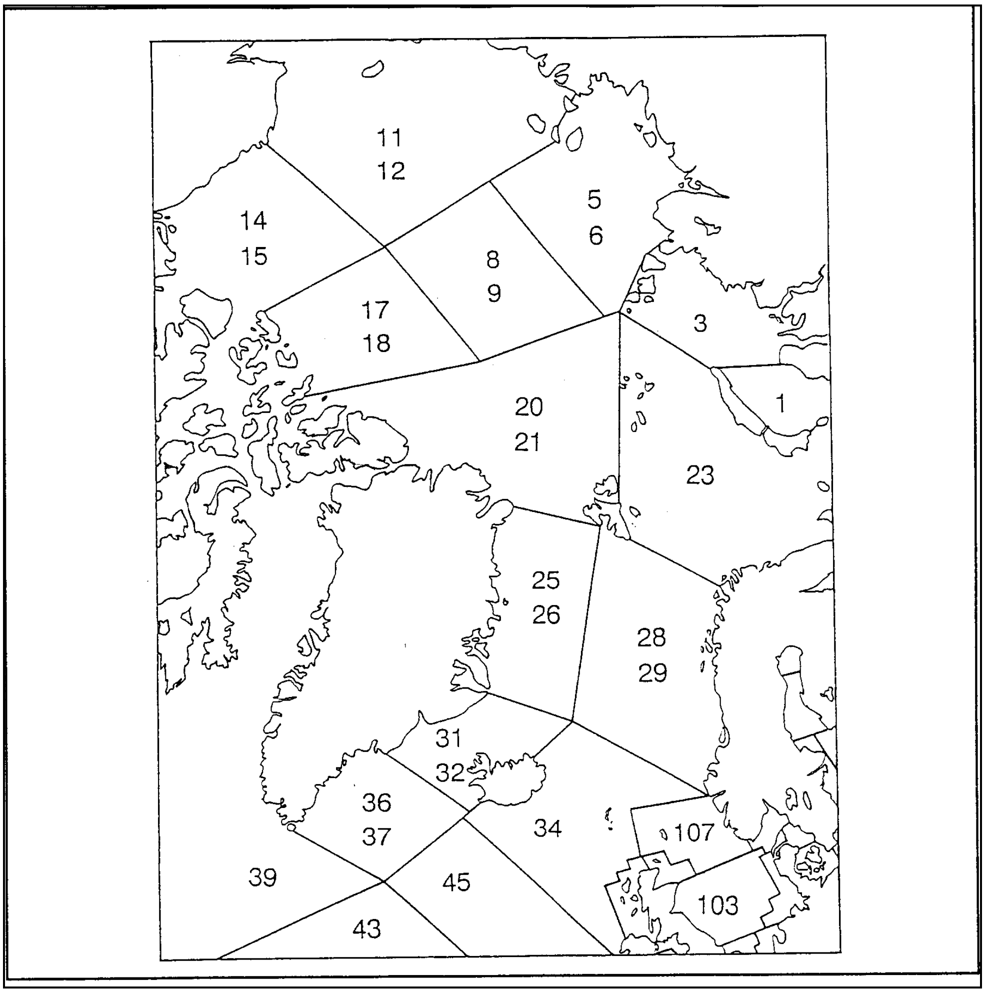

Figure 1 Regions in the Arctic Ocean covered by the box model. The numbers refer to water boxes.

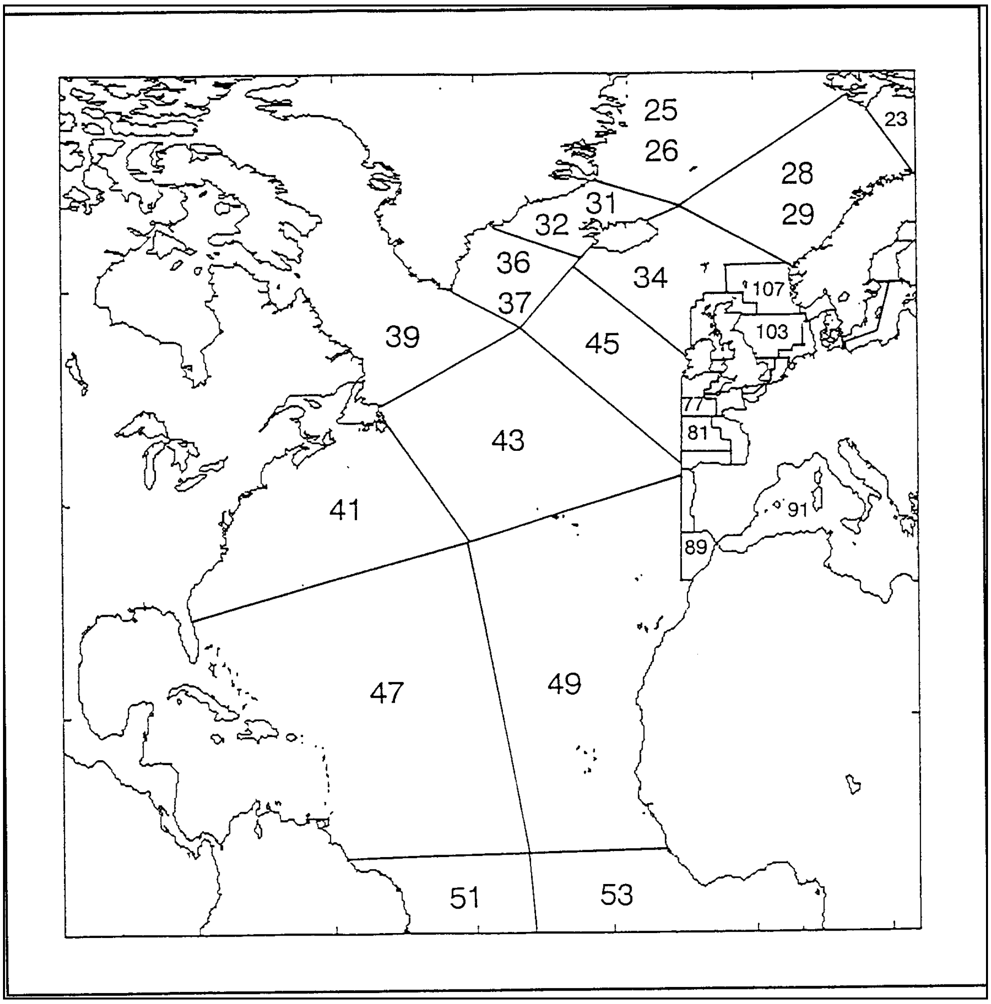

Figure 2 Regions in the North Atlantic covered by the box model. The numbers refer to water boxes.

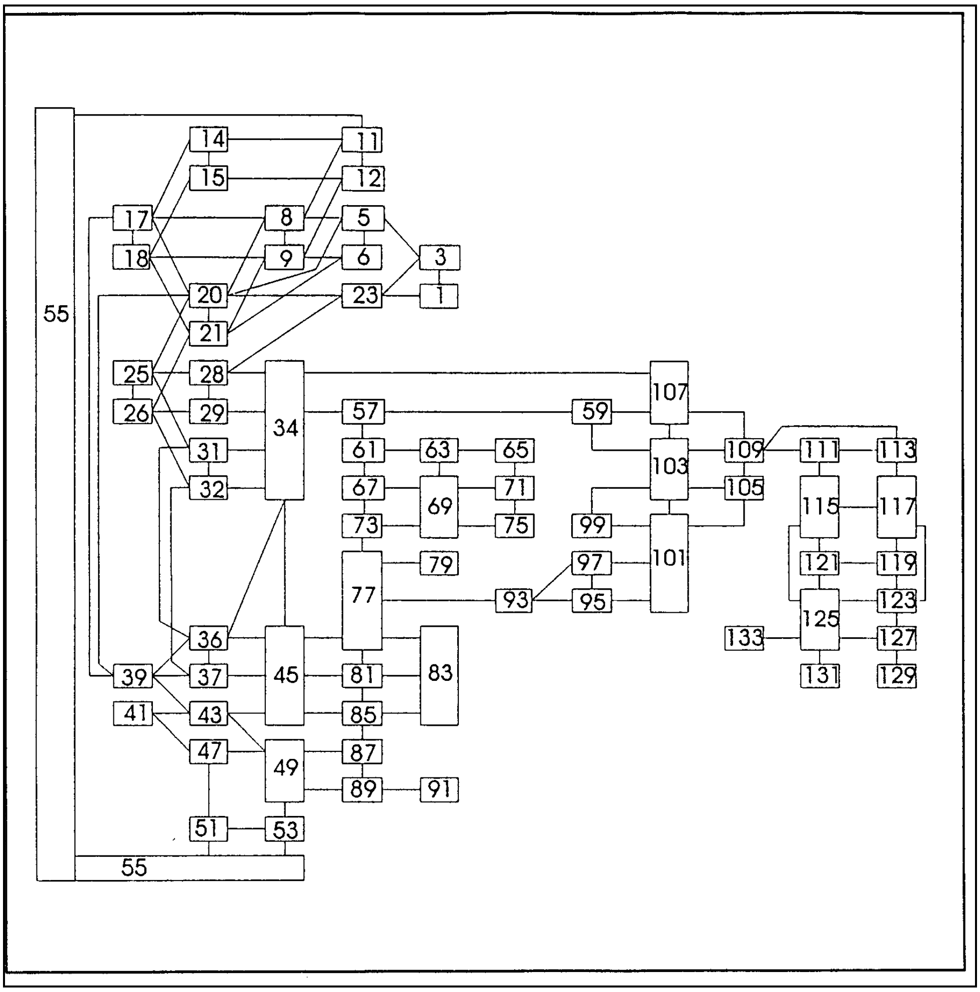

Figure 3 Schematic box structure of the model showing the water boxes only. The lines connecting the boxes indicate the water fluxes between adjacent boxes.

At any given time, the activity in the water column is partitioned between the water phase and the suspended sediment material. The fraction of the activity (F_W) in the water column which is in solution is given by:

$$F_W = \frac{1}{1 + K_d \ SSL},$$

where K_d is the sediment concentration factor ($m^3 \ t^{-1}$) and SSL the suspended sediment load ($t \ m^{-3}$). Activity on suspended sediments is lost to the underlying boxes when particulates settle out. The fractional transfer from a water column (box i) to the sediments (box j) due to sedimentation is given by:

$$k_{ij} = \frac{K_d \ SR_i}{d_i \ (1 + K_d \ SSL_i)},$$

where d_i (m) is the mean water depth of the water column and SR (t m^{-2} y^{-1}) the mass sedimentation rate.

The model also includes the transfer of radioactivity between the surface sediment layer and the bottom boundary layer comprising diffusivity through the pore water and mixing due to bioturbation modelled as a diffusive process. Furthermore, removal of activity from the top surface sediment to lower sediment layers is taken into account by assuming that the burial rate is equal to the flux of particles which settle from the overlying waters. Radioactive decay is included in all the boxes.

The dispersion of radioactive material in the marine environment is modelled from the description given above. The contamination of fish, crustaceans and molluscs is further calculated from the radionuclide concentrations in filtered seawater in the different water regions. For this purpose, concentration factors for biological material are used. Dose factors for inhalation and ingestion are for members of the public and based on the recent International Basic Safety Standards issued by the IAEA[viii].

Calculated Doses

The models have been used for assessments of radioactivity in the Baltic Sea[ix] and in Arctic waters[x]. The assessments have included doses to human individuals and populations from marine pathways.

Radioactivity in the Baltic Sea

The assessment for the Baltic Sea has included the radionuclides Cs-137 and Sr-90 from nuclear weapons testing, fallout from the Chernobyl accident, discharges to sea from Sellafield and La Hague and discharges to sea from nuclear installations bordering the Baltic Sea. The source terms are shown in Table 1.

Table 1 Time-integrated input of Cs-137 and Sr-90 to the Baltic Sea from1950 to 1994.

Source	Cs-137 (TBq)	Sr-90 (TBq)
Weapons fallout, direct deposition to sea	1780	1130
Weapons fallout, river runoff	70	410
Chernobyl fallout	4500	90
European reprocessing	380	70
Nuclear facilities	1.7	0.014
Total	6732	1700

Collective dose rates and doses to members of the public were calculated based on fishery statistics and on predicted concentrations of Cs-137 and Sr-90 in biota. The collective dose rate peaks in 1986 at a level of 160 manSv y^{-1} due to fallout from the Chernobyl accident. The total collective dose from Cs-137 and Sr-90 in the Baltic Sea is estimated at 2300 manSv of which about 60% originates from Chernobyl fall-out, about 30% from fallout from nuclear weapons testing, about 10% from European reprocessing facilities, and about 0.03% from nuclear installations bordering the Baltic Sea area. Doses from Cs-137 dominate (99%) over those from Sr-90 (1%). Doses from naturally-occurring radioactivity in seafood (polonium-210) were calculated on a similar basis and compared with the doses from Cs-137 and Sr-90. The results of this comparison are shown in the Table 2 which summarises the calculated doses.

Table 2 Collective exposure of the population around the Baltic Sea for the period from 1950 to 2050 from Cs-137, Sr-90 and Po-210 in Baltic seawater.

Sources	Collective dose to year 2050 (manSv)
Chernobyl fallout	1400
Nuclear weapons fallout	700
European reprocessing	200
Nuclear installations	1
Natural radioactivity (Po-210)	20000

Dumped nuclear waste in the Kara Sea

An assessment commissioned by the European Commission was carried out to evaluate the radiological consequences of the marine reactors that were dumped in the Kara Sea by the former Soviet Union[xi]. Radionuclide inventories of fission products, actinides and corrosion products were calculated and release rates to the marine environment estimated based on rates of corrosion in seawater of steels, nuclear fuel and other materials. Table 3 gives an overview of the time-integrated releases from the waste sites for the best-estimate scenario.

Table 3 Time-integrated release of radionuclides to the Kara Sea.

Nuclide	Release (TBq)
Pu-238	1.1E-03
Pu-239	8.3E+00
Pu-240	2.3E+00
Pu-241	1.1E-11
Am-241	1.3E+00
Sr-90	3.8E-05
Cs-137	6.6E-04
Sm-151	1.0E-02
Tc-99	1.1E-01
I-129	3.8E-04
Fe-55	2.5E+01
Co-60	3.5E-01
Ni-59	4.9E+00
Ni-63	4.7E+00

The box model for Arctic waters mentioned above was used for the calculation of doses to man from these source terms. For the collective dose truncated at 10000 years to the world population, a total dose of about 1 manSv was obtained. The dose rate to individuals from a critical group located in the western Kara Sea is shown in Fig. 4. The break down of the maximum dose rates in year 1970 and 3700 by nuclide and exposure pathway are shown in Figs. 5 and 6 which show the percentage contributions to the maximum dose rates.

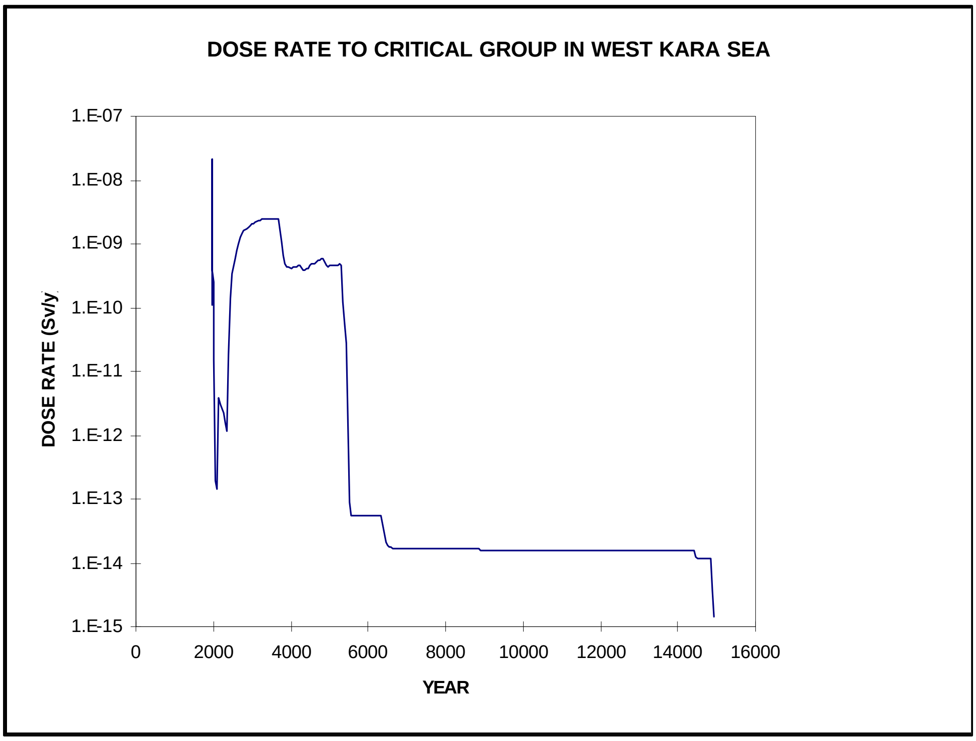

Figure 4 Annual dose rate (Sv y^{-1}) to the critical group on the Yamal peninsula in the West Kara Sea.

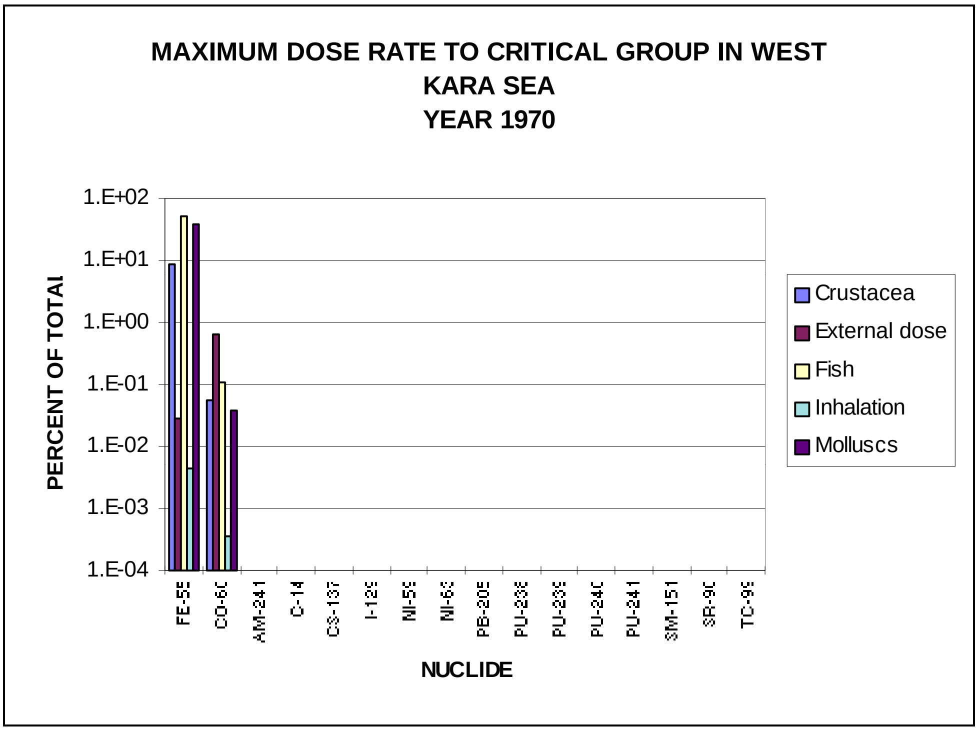

Figure 5 Relative contributions (per cent) by nuclide and exposure pathway to the peak annual dose in year 1970 to the critical group on the Yamal peninsula.

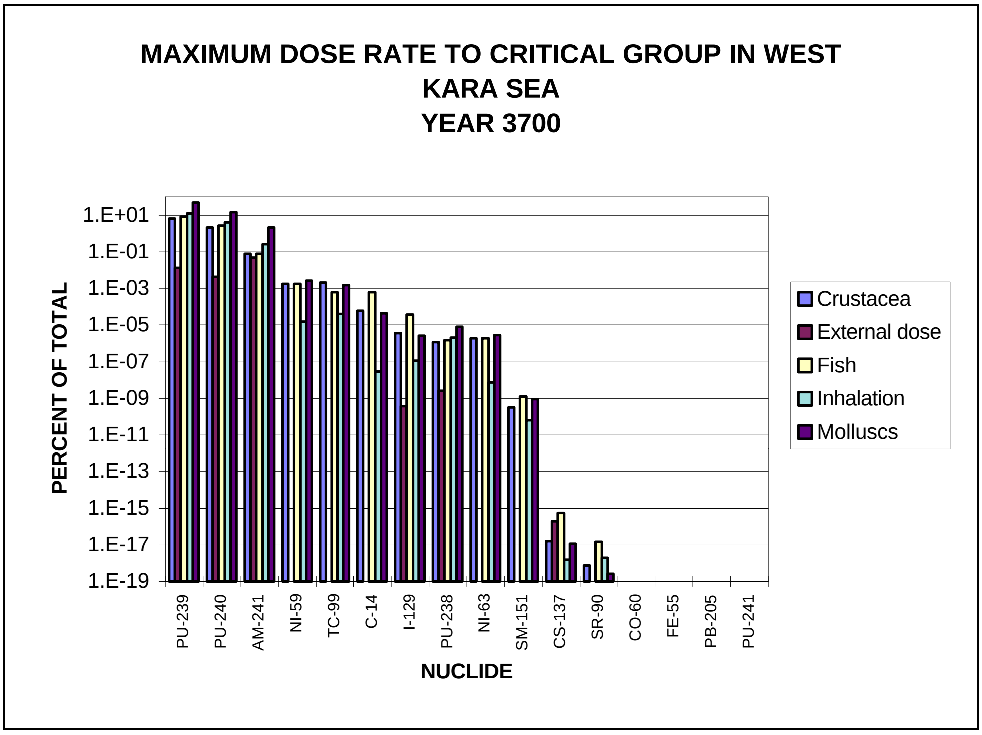

Figure 6 Relative contributions (per cent) by nuclide and exposure pathway to the peak annual dose in year 3700 to the critical group on the Yamal peninsula.

The maximum annual dose occurs in year 1970 at a value of 2E-8 Sv y^{-1} with contributions from Fe-55 and Co-60 at 99.2% and 0.8%, respectively. Figure 5 shows that the dominating exposure pathways are ingestion of fish for Fe-55 and external exposure for Co-60. The overall pathway contributions to the maximum annual dose are ingestion of fish 52%, ingestion of molluscs 38%, ingestion of crustaceans 9%, external exposure 0.7% and inhalation 0.005%. The peak annual dose in year 3700 has a value of 2E-9 Sv/y and is dominated by the Pu-isotopes as shown in Fig. 6. The pathway contributions to the latter peak annual dose are ingestion of molluscs 64%, inhalation 16%, ingestion of fish 11%, ingestion of crustaceans 9%, and external exposure 0.07%.

The Komsomolets Submarine

An assessment was made with the Arctic model of the potential radiological consequences from the release of Pu-239 from the submarine Komsomolets which suffered an accident in the Norwegian Sea in 1989 and sank about 200 km southwest of Bear Island at a depth of 1600 m.

The source term of radioactivity was assumed to comprise 8 kg of Pu-239 of which the 6 kg originates from the nuclear warheads and the rest from the nuclear reactor. The specific activity of Pu-239 is 2.3 TBq per kg, so the source term is assumed at a rounded number of 20 TBq. The rate of release has been assumed to be rather fast (one year) based on information from Russian sources concerning accelerated galvanic corrosion due to the titanium hull of the submarine. The source was assumed situated in a small local box (of 1 km^3 volume) on the seabed connected to the deep waters of the Norwegian Sea.

The dispersion of plutonium has been simulated with the box model covering the Arctic Ocean and the North Atlantic providing the basis for calculating the transfer of plutonium to marine biota. Global fisheries statistics have been used to estimate the transfer of plutonium to man and the corresponding collective dose to the world population. The calculation of the dose was truncated at 1000 y. The total collective dose from ingestion of seafood was estimated at a value of 8 manSv.

Model testing

The reliability of the predictions made from box models has been tested for the Baltic Sea covering a comparison between calculated and observed annual mean concentrations of the radionuclides Cs-137 and Sr-90 for the time period 1970-1991. The sources of radioactivity considered include fallout from nuclear weapons testing, fallout from the Chernobyl accident, discharges to sea from Sellafield and La Hague and discharges to sea from nuclear facilities bordering the Baltic Sea.

The comparison is illustrated in Fig. 7 showing a scatterplot between observed and predicted annual average concentrations of Cs-137 in seawater. The full line gives the ideal 1:1 relationship. The geometric mean of the predicted-to-observed (P/O) ratio is 1.1 with a geometric standard deviation of 1.4. Fig. 8 shows a scatterplot of the observed and predicted annual average levels of Sr-90 in seawater. The geometric mean of the P/O values is 0.8 with a geometric standard deviation of 1.4.

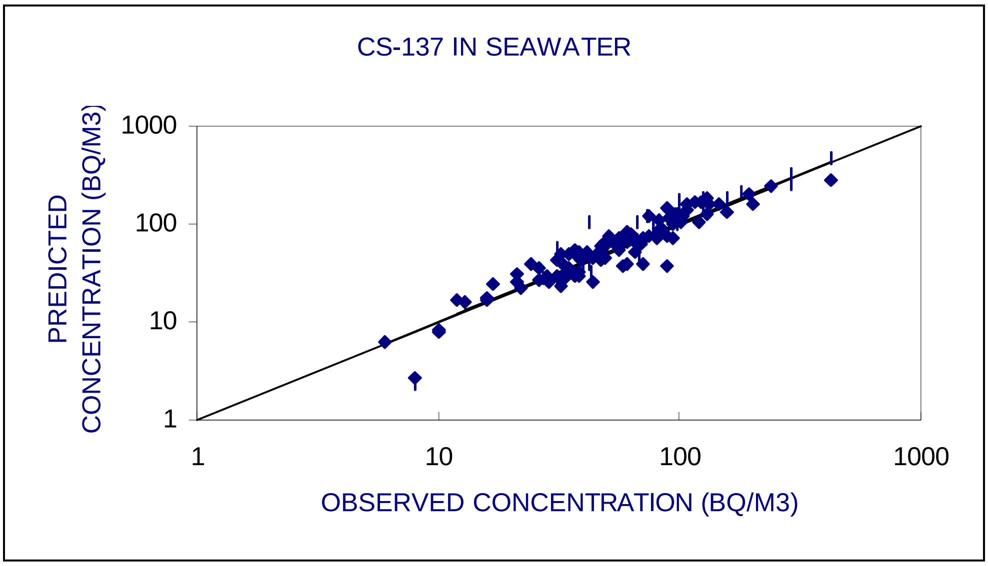

Figure 7 Scatterplot of predicted and observed annual mean concentrations of Cs-137 in Baltic seawater. The line indicates the 1:1 relationship.

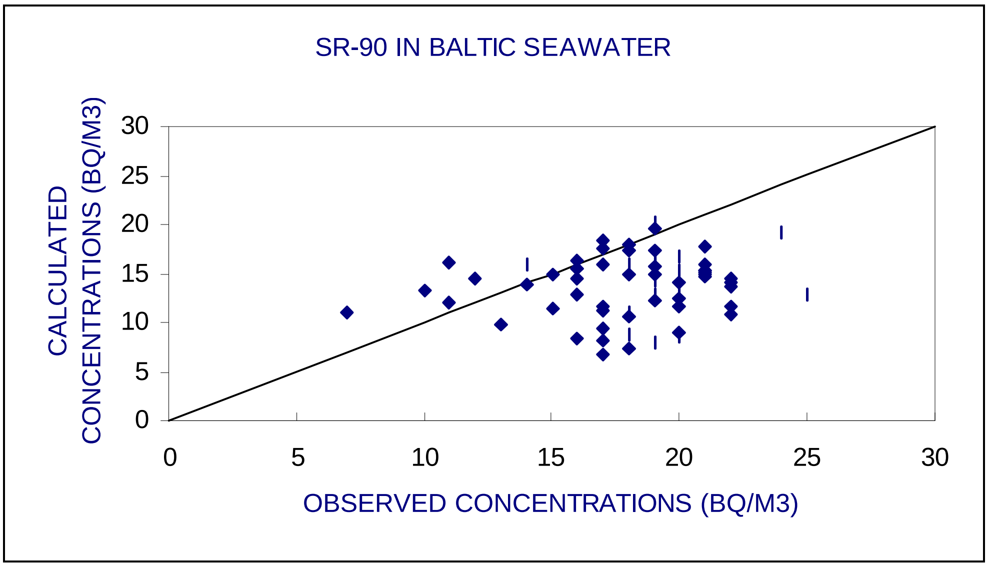

Figure 8 Scatterplot of predicted and observed annual mean concentrations of Sr-90 in Baltic seawater. The line indicates the 1:1 relationship.

Sensitivity analysis

A sensitivity analysis was carried out to identify components of the Arctic model that are potentially important contributors to the predictive accuracy. The components investigated include features associated with water transport and mixing, particle scavenging, water-sediment interaction and biological uptake. The source terms used for the calculations were obtained from the IAEA's International Arctic Seas Assessment Project (IASAP). The best-estimate release scenario was selected comprising release rates of radionuclides from shallow bays on the east coast of Novaya Zemlya (Abrosimov Bay and Tsivolki Bay) and from the deeper waters of the Novaya Zemlya Trough. The radionuclide-specific releases considered are shown in Table 4, and Fig. 9 shows the release rate assumed for the Tsivolki Bay where the reactor compartment and spent nuclear fuel from the Lenin icebreaker was dumped in 1967. The graph shows that the best-estimate scenario assumes release of the activation products only (Co-60, Ni-59, Ni-63) until the year 2300 due to corrosion of the protective barriers. At that time contact is assumed between the seawater and the spent nuclear fuel, and a peaked release of fission products (Cs-137 and Sr-90) and transuranics (Pu-239, Pu-240, Am-241) is assumed followed by a steady release due to corrosion of the nuclear fuel.

Table 4 Integrated release of radionuclides (TBq) dumped in the Kara Sea, IASAP best-estimate scenario.

	Tsivolki Bay	Abrosimov Bay	Novaya Zemlya Trough
Pu-239	4.9	0.73	0.09
Pu-240	2.1	0.32	0.04
Am-241	0.41	0.12	0.01
Co-60	0.03	2.9	0.08
Ni-63	370	110	1.9
Ni-59	40	110	2.3
Cs-137	0.10	41	8.7
Sr-90	0.07	34	7.4

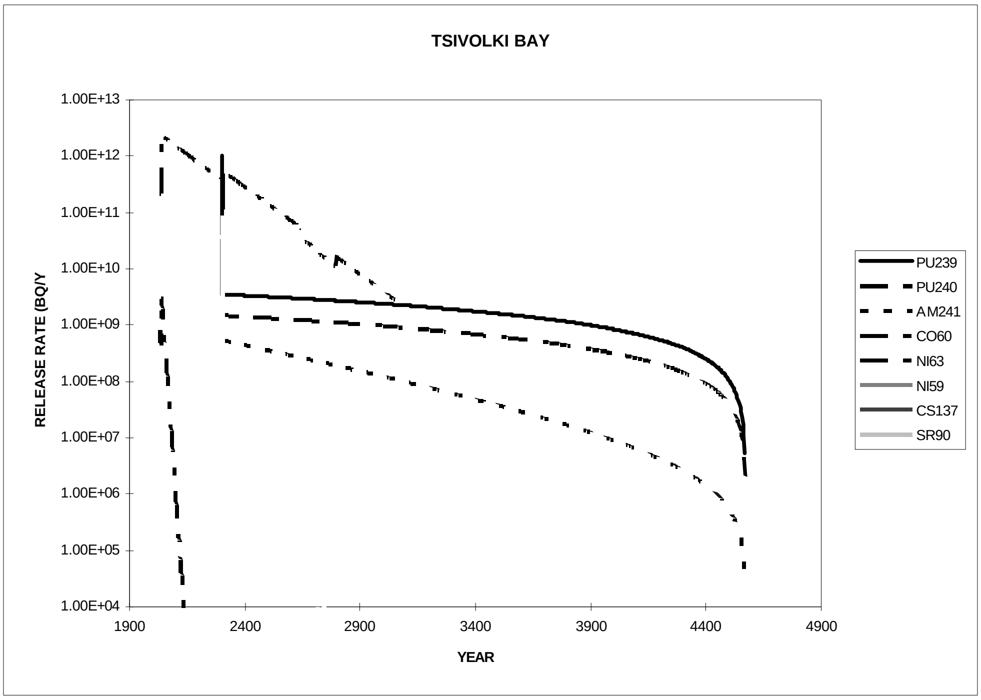

Figure 9 Radionuclide release rates (TBq y^{-1}) for Tsivolki Bay estimated by the IASAP Source Term Group.

The parameter sensitivity of the doses to individuals and populations has been investigated concerning three main processes: water movement and mixing, sediment-water interaction and biological transfer. For the hydrodynamical processes, four parameters were investigated: the advection and mixing between the Kara Sea and the Laptev Sea to the east, the advection and mixing between the Kara Sea and the Barents Sea to the west, the vertical mixing between surface and deeper waters over the Novaya Zemlya Trough, and the rates of exchange of water between the bays containing the dumped waste and the open Kara Sea. The sediment-related parameters were the following: sedimentation rates, sediment distribution coefficients (K_d), suspended sediment loads, depth of the mixed surface sediment layer, and the mixing rates in the surface sediments. For the biological transfer processes, the three parameters representing biological concentration factors for fish, crustaceans and molluscs were included.

The parameter sensitivity analysis was carried out by assigning identical variabilities of 10% to all the above mentioned parameters and running the model repeatedly (about 500 times). Correlation coefficients were calculated between parameter values and dose values, and the square of the correlation coefficients (R^2) were interpreted as how much of the variation of the doses is explained by the linear relationship to the parameters. The parameter sensitivities are thus expressed in percent of the total variability.

The results of the parameter sensitivity analysis for the collective doses are shown in Table 5 which summarises the integrated releases, the collective doses and the main parameter sensitivities (> 1%). The total collective dose is calculated to 0.4 manSv (truncated at 1000 y) with dominating contributions from plutonium isotopes of 57% and from Cs-137 of 37%. The parameter sensitivities are seen to vary across the radionuclides from Sr-90 which has a low K_d to the transuranics and activation products which have high K_d's. For the total collective dose the main parameter sensitivities are due to sedimentation processes (sedimentation rate, K_d, suspended sediment load) and biological transfer processes (concentration factor for fish).

Table 5 Integrated releases (TBq), collective doses (manSv) and main parameter sensitivities (%).

Nuclide	^{137}Cs	^{90}Sr	^{239}Pu	^{240}Pu	^{60}Co	^{241}Am	^{63}Ni	^{59}Ni	Total
Total release (TBq)	49	42	6	2.4	3	0.5	480	150	
Coll. dose (manSv)	0.14	0.02	0.16	0.06	7E-05	0.001	0.002	0.001	0.4
Coll. dose (%)	37	5	41	16	0.02	0.4	0.6	0.3	100
Sedimentation rate, (%)	19		66	66	54	61	53	53	45
Conc. factor, fish (%)	65	90			13		4	4	28
K_d (%)	11		13	13	25	7	24	24	12
Susp. sediment load, (%)			16	16	4	14	1	1	9
Conc. factor, molluscs (%)			9	9		5	1	1	5
Adv., Kara Sea, west (%)	4	1					4	4	2

Parameter uncertainty analysis

Variabilities were assigned to the most sensitive parameters identified above. The variabilities take into account current observed variability and estimated future variability. The variabilities are expressed as ranges around the central values (c), so that for each parameter the same factor (f) determines the high value (c·f) and the low value (c/f). The factors of variability for the sediment related parameters were 2 for the sedimentation rate, 2 for the suspended sediment load and 10 for the sediment distribution coefficient, K_d. A factor of 3 was used for the variability of each of the three biological concentration factors for fish, crustaceans and molluscs. A factor of 2 was used for the variability of the rate of exchange of the water between the Novaya Zemlya bays and the Kara Sea. Finally, a factor of 2 was used for variability of the depth of the surface sediment layer in the Novaya Zemlya bays. There was no specific information available on suitable probability distributions for the different parameter values for which reason it was decided to sample (Monte Carlo technique) the values from log-uniform distributions with upper and lower values as specified above. The factors of variability were applied globally for each parameter across the model for each simulation. One hundred simulations were carried out for each model endpoint involving collective doses and doses to critical groups from the dominating nuclides (Fe-55, Co-60, Cs-137 and Pu-239) and source locations (bay and Novaya Zemlya depression) to determine the ranges on the model output values. This number of simulations was considered adequate for the purpose as confirmed from carrying out 500 simulations for one of the above combinations.

Uncertainties of maximum annual doses to critical groups

The results of the parameter uncertainty calculations on the peak annual doses to the critical groups are shown in graphical form in Figure 10.

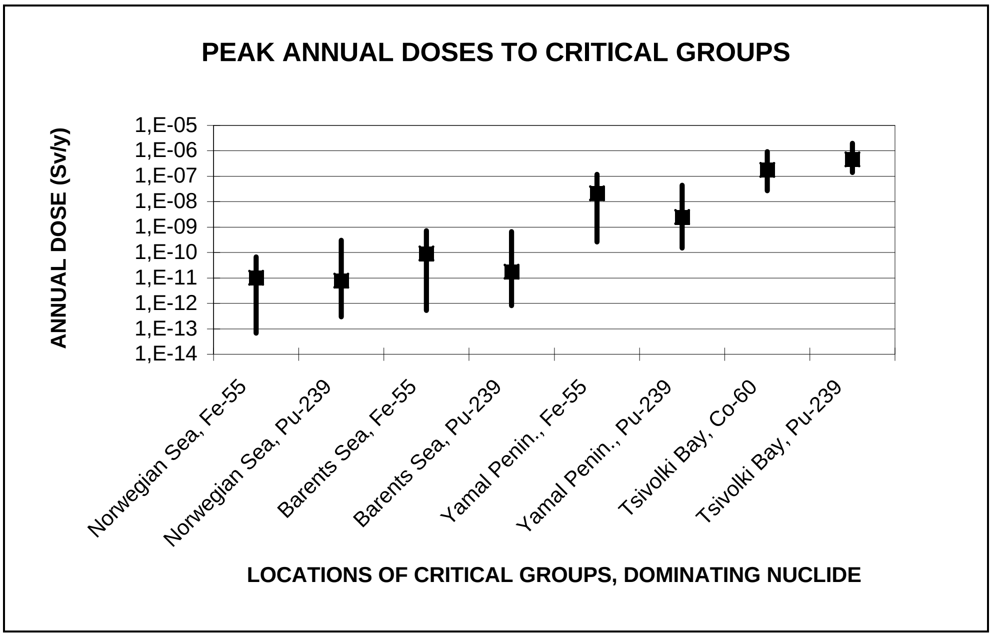

Figure 10 Peak annual doses to critical groups (Sv y^{-1}) with ranges of variability estimated from parameter uncertainty analysis.

Correlation analysis was carried out using rank correlation coefficients to identify and quantify the main components of variability. Only correlation coefficients numerically greater than 0.5 are mentioned. The peak annual doses to the critical groups show negative correlation coefficients with the sediment distribution coefficients, K_d's, of -0.8 for Fe-55 and -0.6 for Pu-239 while the sedimentation rates and the peak annual doses give negative correlation coefficients of -0.5 for both radionuclides. This means that high doses are associated with low values of the two parameters. Both radionuclides have high K_d values and are therefore particularly sensitive to sedimentation processes that transfer the radionuclides from the water column to the sediments and reduce the wider dispersion of the nuclides in the marine environment. For the critical group located in Tsivolki Bay on Novaya Zemlya, the analysis shows other results due to the different exposure pathways that apply here. The peak annual dose from Co-60 shows a negative correlation coefficient of -0.8 with the depth of the surface sediment layer (mixing layer). This is due to the dominating exposure pathway which in this case is external exposure from Co-60 in coastal sediments in which the concentration is inversely related to the depth of the mixing layer. The peak annual dose from Pu-239 shows a negative correlation coefficient of -0.7 with the bay flushing rate. In this case inhalation is the dominating exposure pathway, which is controlled by the concentration of Pu-239 in the water of the bay, and the peak concentration is inversely related to the bay flushing rate.

The predicted variabilities of the peak annual doses to the critical groups around the mean values increase with distance from the Kara Sea. For the doses dominated by long-lived plutonium isotopes, the variability is nearly symmetrical around the mean value on a log-scale corresponding to the factors of variability applied to the parameters. The predicted variabilities on the doses from Pu-isotopes range from a factor of about 4 in Tsivolki Bay to a factor of about 40 in Norway. For the doses dominated by the short-lived Fe-55, the variability is non-symmetrical around the mean value on a log-scale due to the low values being associated with delayed transfer involving further physical decay. The variabilities for Fe-55 of the maximum values relative to the mean values range from a factor of 6 in the Kara Sea to a factor of 10 in Norway, and the variabilities of the minimum values relative to the mean values range from a factor of about 80 in the Kara Sea to a factor of about 250 in Norway.

Uncertainties of collective doses

The predicted variability of the collective dose to the world population is based on the variability predicted from the plutonium isotopes. Correlation analysis shows that the collective dose is negatively correlated with the sediment related parameters yielding a correlation coefficient of -0.7 for the sediment distribution coefficient and a correlation coefficient of -0.5 for the sedimentation rate.

The predicted variability of the collective dose is about two orders of magnitude up and down around the mean value of about one man sievert. The collective dose is thus predicted at a level of about one man sievert and not to exceed one hundred man sieverts.

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Quality assurance

Quality assurance has been an important factor in the EKO-1 project work. Emphasis was put on the following two factors:

a) Sampling
b) Analysis

The EKO-1 study on quality assurance in sediment sampling

Bottom sediments play an important role in aquatic ecosystems. In general, they act as a final sink for most of the organic material produced in the water phase, as well as for other particles transported by water currents and/or from adjacent terrestrial areas. During their slow sinking the particles tend to bind foreign substances from the water and sweep them to the bottom (e.g. radionuclides and other contaminants).

In favourable sedimentation conditions suspended particles form undisturbed laminae in a historical order, and the bottom sediments create an archive, from which historical features of a lake or a sea area can be read. Various particle-bound substances can be used as markers of certain historical events or periods. With the aid of these markers the laminae can be dated.

The radioactive fallouts caused by nuclear weapons tests in the 1960s and the accident at the Chernobyl NPP in 1986 have created useful markers in the sediments of many Nordic waters. These have been successfully used not only in radioecological studies but also in limnological and marine research dealing with sedimentation processes and rates.

To be able to study sedimentation and processes in sediments, it is essential to obtain reliable samples from sediments. False conclusions are an obvious risk if the studies are based on biased field samples. More strictly, it is unreasonable to perform exacting and expensive analyses if the samples themselves are unreliable or of poor quality.

Sampling of bottom sediments is difficult and requires, in addition to properly designed devices, standard and precise working methods, as well as experience and competence on the part of the crew, including knowledge of sources of error. Sediment sampling is beyond doubt one of the most exacting sampling tasks in the aquatic environment. The prime reason is that the upper, most recent layers of sediment are often very soft and susceptible to resuspension, because the interface between water and sediment usually resembles a line drawn in water. That the crew normally cannot see what happens when the sampling device penetrates into the sediment makes the work particularly difficult.

In the planning phase of the EKO-1 Project the need for intercalibration of sediment sampling methods and equipment among the participating laboratories was discussed. The idea was dismissed due to the high costs involved, and because many of the devices had been earlier used in similar intercomparisons arranged by NKA and the HELCOM/MORS Expert Group. It was instead decided to arrange this inquiry and evaluation.

On the basis of experience from earlier intercalibration exercises it was evident that differences in the types of sediment and sedimentation processes are the main reasons for using different sampling equipment and philosophies. It was also obvious that no universal sampler exists for meeting all the requirements of sediment sampling. Consequently, intercomparison of the results has by no means constituted a competition between devices. The goal has been to obtain proper samples for interpreting the results and processes in a most reliable manner.

The instruments best suited for quantitative sampling of softbottom sediments appear to be those based on the coring principle. Box corers can be reliably used mainly for bulk sampling of coherent sediments and some silty and sandy sediments. Nevertheless, it should be borne in mind that the same instrument may not be the best alternative for all types of bottom, but various circumstances require different types of instrumentation.

Many factors speak in favour of relatively large diameters/areas of the corer orifices. It is not possible however, to increase the tube diameter endlessly without negative impact to the corer's handiness and increasing difficulty in handling and slicing of the cores.

Despite the large variety of sampling instruments and many sources of error involved in the use of different instruments, it is most important to know and account for the disadvantages and to work as carefully as possible towards minimising errors and obtaining undisturbed, reliable samples.

A fuller description of this study was published in an EKO-1 report by Erkki Ilus *"Evaluation of sediment sampling devices and methods used in the EKO-1 project"* (NKS/EKO-1(96)TR-1).

The EKO-1 laboratory intercomparison exercise

This intercomparison exercise dealt with the analysis of artificial and natural radionuclides in two sediment samples, one from the Baltic Sea and one from the Kattegat. The measurement techniques have included direct gamma-ray spectrometry with Ge and NaI detectors, and radiochemical procedures followed by beta counting and alpha spectrometry. The participants have comprised 21 laboratories of which 14 are from the Nordic countries, 6 from the Baltic republics and one from Japan.

Results were submitted for a large number of radionuclides comprising Cs-137, Cs-134, Co-60, Sb-125, Pu-239,240, Pu-238, Am-241, Sr-90, Ra-226, Th-232 and K-40, Pb-210, Po-210 and U-235. The analytical performance of the participants has been evaluated for those radionuclides where six or more data sets were received. Statistical tests were made to see if individual results agreed with overall average radionuclide concentrations in the two sediment materials within target standard deviations. The results of these tests are summarised in Table 1 which shows the radionuclides, the selected target standard deviations, the number of data sets submitted and the number of data sets which disagree significantly with the overall mean values.

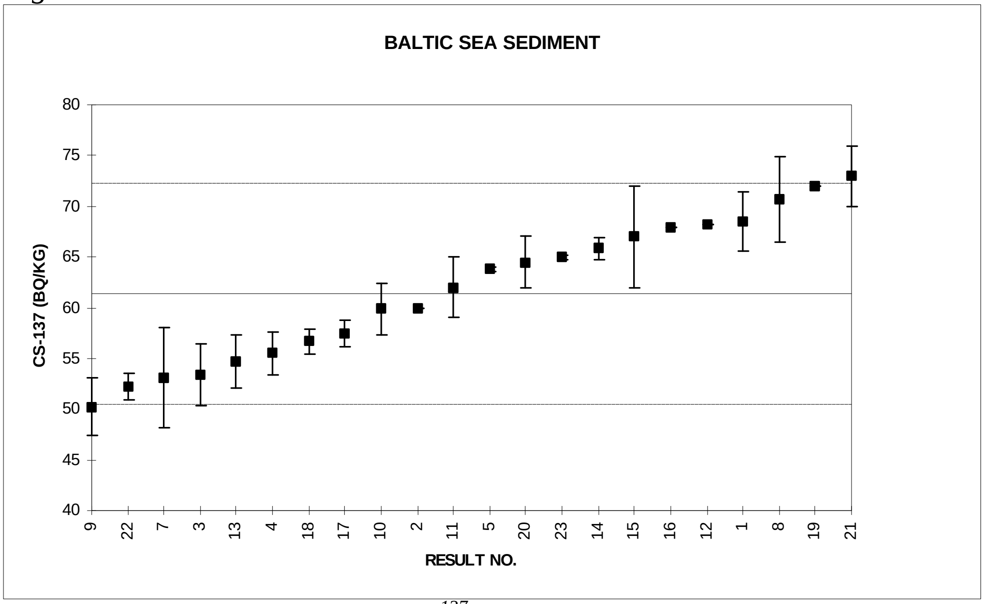
It is noteworthy that with the exception of Co-60 the performance criteria are not met in about 20-40% of the data sets for the relatively straight forward gamma-spectrometric analyses of Cs-137, Cs-134, Ra-226, Th-232 and K-40. For the more complicated radiochemical analyses of plutonium isotopes only 1 in 8 of the data sets do not meet the performance criteria. The test of the overall analytical performance shows that 60% of the data sets do not meet the combined performance criteria. This shows that there is room for considerable improvement of analytical quality for most of the laboratories that have participated in this intercomparison.

The intercomparison exercise has furthermore demonstrated several elementary problems in analytical work such as interchange of samples, mistakes in calculations and correction of background levels. High-quality analytical performance requires careful and dedicated staff using well-established laboratory procedures and frequent participation in international intercomparison exercises.

Table 1. Summary of analytical performance across all data sets.

Nuclide	Target standard deviation (%)	Number of data sets	Number of data sets that disagree significantly with performance criteria
Cs-137	5	23	9 (39%)
Cs-134	20	6	2 (33%)
Co-60	15	6	0 (0%)
Pu-239,240	10	8	1 (13%)
Pu-238	20	8	1 (13%)
Ra-226	10	12	5 (42%)
Th-232	10	12	5 (42%)
K-40	10	20	4 (20%)
Nuclides combined		23	14 (61%)

Figure 1.



Distribution of analytical results for ^{137}Cs in Baltic Sea sediment

A fuller description of the results can be found in Sven P. Nielsen's report *"An Intercomparison Exercise on Radionuclides in Sediment Samples"*, published as Risø-R-914(EN) report in July 1996.

The EKO-1 seminars

Another important factor in increasing the Nordic competence in this field have been the EKO-1 seminars. Radioecology is a multi-disciplinary field. Much of the work done is related to work done on stable nuclides. The EKO-1 seminars have therefore been used to bring together on one hand participants in the EKO-1 project and other Nordic experts in radioecology and on the other hand Nordic experts in related fields and other leading international experts. During the project period two seminars have been held dealing with issues which were considered to be of major relevance for the project work:

- Kristineberg September 20th - 21st, 1995, *Sedimentation processes*
- Helsinki April 2-3, 1997, *Dating of sediments and determination of sedimentation rate*

Many of those who have participated in the EKO-1 project work have commented that for them personally these seminars were the most important aspects of the EKO-1 work.

The Kristineberg Seminar on Sedimentation Processes

The first seminar was held September 20-21, 1995 at Kristinebergs Marine Research Station, Sweden and was organised by the Department of Radiation Physics, Lund University, Sweden. The lectures covered not only radionuclides and sedimentation processes, but also heavy metals and organic compounds. This seminar gave, in a very friendly atmosphere, a very useful external input to both the EKO-1 as well as the EKO-2.3 project (on freshwater ecosystems). The seminar was attended by most participants in the EKO-1 and EKO-2.3 projects.
The following presentations were given and the lecturers also provided copies of overheads and a brief text which was put together in a booklet.

- **Rob Comans**, The Netherlands: Ion-exchange and surface complexation processes controlling the partitioning of radiocaesium and heavy metals between sediments and pore waters.
- **John Hilton**, UK: Modelling pollutant loss from the water column: sedimentation, resedimentation and direct diffusion losses.
- **Jack Cornett**, Canada: radionuclide Residence Time Models: The next generation.
- **Mats Jansson**, Sweden: The role of sedimentation and sedimentological processes for retention and downstream transport of inorganic and organic material in lake Öträsket, a humic lake in northern Sweden.
- **Jurg Bloesch**, Switzerland: past, present and future aspects of sediment trap methodology and settling flux measurements.
- **Scott Fowler**, Monaco: Scavenging and vertical transport of radionuclides and trace elements by sinking particulate matter.
- **Per Hall**, Sweden: Input, recycling and burial of organic matter in marine sediments.
- **Jens Skei**, Norway: Environmental implications of contaminated sediments.
- **Johan Ingri**, Sweden: Chemical composition of suspended and deposited sediments in the Kalix River and its estuary.
- **Lauri Niemistö**, Finland: Sediment and sedimentation in the Baltic Sea.

The Helsinki seminar on dating of sediments and determination of sedimentation rates

The second NKS/EKO-1 Seminar ‘Dating of sediments and determination of sedimentation rate’ was held at the Finnish Centre for Radiation and Nuclear Safety (STUK) on April 2-3, 1997. The programme of the Seminar was compiled from 8 invited lecturers and 6 other scientific presentations. The lectures were given by renowned European experts, such as Dr. Peter G. Appleby and Dr. Neil L. Rose from the UK. The lectures and presentations of the Seminar will be published in the Report Series STUK-A in early 1998. The Seminar was attended by 48 scientists from 9 countries.

Dating of sediments and determination of sedimentation rate are of crucial importance in all types of sedimentological study and model calculations of fluxes of substances in the aquatic environment. In many cases these tasks have been closely related to radioecological studies undertaken in marine or freshwater environments, because they are often based on measured depth profiles of certain natural and artificial radionuclides present in sediments. During recent decades Pb-210 has proved to be very useful in dating of sediments, but other radionuclides have also been successfully used, e.g. some transuranic elements and Cs-137 (the last-mentioned especially after the Chernobyl accident in 1986).

Despite its wide use, ‘‘it is unlikely that dating by Pb-210 will ever be a totally routine procedure...The situation is considerably more difficult in marine environments where there is no *a priori* reason to suppose that Pb-210 supply rates are governed by any simple relationship’’ (cit. from P.G. Appleby’s lecture). Examples of existing limitations, difficulties and problems in the use of the Pb-210 dating methods were discussed in the Seminar, as well as solutions for solving these problems. Mixing models in the interpretation of Pb-210 concentration profiles in sediments were introduced in one of the lectures.

The usability of several artificial radionuclides in estimation of sediment accumulation rate (e.g. Tc-99, Cs-137, Np-237, Pu-238, Pu-239,240 and Am-241), as well as their mobility in sediments were discussed in many lectures. The use of radiocesium in dating of sediments is based on the premise that it is highly and selectively bound to illitic clay minerals. Some findings suggest that the long-term exchangeability of radiocesium in sediments may be higher than the low percentage that is generally assumed. It was concluded, however, that weapons-test Cs-137 will remain as an important marker for dating. It is likely that its future use will be limited by radioactive decay rather than diffusive mobility, while the mobility of the Chernobyl cesium may be still greater.

This type of seminar in which renowned outside experts and participants in the project can meet and exchange their knowledge and experience on the topic of the ongoing NKS project has proved to be very useful and to form an essential part of the project.

Use of internet technology for the dissemination of information

The project period 1994-1997 has seen an explosive growth in the use of internet technology. The EKO-1 project has tried to make use of the increasing opportunities the Internet offers. Texts have been distributed within the EKO-1 work group, and World Wide Web (WWW) pages have been set up.

Contacts with the Baltic States

The Baltic States were invited to participate in the EKO-1 intercomparison for laboratory measurements. Subsequently most of the laboratories also participated in EKO-3.2 work on quality assurance in laboratory measurements. The participating laboratories have expressed their pleasure with being able to participate in this NKS work and expressed a wish for some form of continuation. The participation of the Baltic States in the EKO-1 intercomparison did not require any NKS funding.

Concluding remarks

The funding that the NKS provided for work in the current period was very valuable, even though it only covered part of the project cost. It provided the necessary stimulus to turn work in each of the participating countries into a joint Nordic undertaking. The EKO-1 project has resulted in that the Nordic countries are now in a much better position than before to assess the short and long term effects of radionuclides that have been (or might be) released and incorporated into sediments. The individual countries can now continue their own work within this field.

Marine radioactivity continues to provide topics of concern for the Nordic countries. The Baltic Sea is an important source of food for many of the Nordic countries. Even though the individual doses resulting from the consumption of sea food from the Baltic Sea are relatively low, they can nevertheless be considerable compared to other doses caused by man-made radionuclides. In Denmark the major part of doses to humans from man-made radionuclides comes from sea food caught in the Baltic Sea.

A project focusing on the Baltic Sea would be of considerable relevance for the Nordic countries. The project could also be linked with studies concerned with the short and long term effect of runoff from the catchment area and into the Baltic Sea.

Recently there has been considerable public concern in the Nordic countries over the possible effects of the increased release of ^{99}Tc from Sellafield. It is important for authorities in the Nordic countries to be able to make an assessment of the effect of this release. Furthermore the ^{99}Tc pulse can be a very useful tracer for studying processes in the Atlantic Ocean, the Arctic seas and the Baltic Sea.

The NKS has made a major contribution to the maintenance of competence in the field of environmental radioactivity in the Nordic countries. But it has also been an important factor in the build-up of competence. As a part of the previous project period (1990-1993) a course on radioecology was held in 1991 at Lund, Sweden. Many of the students from that course have been active participants in the work within EKO-1 and EKO-2 in the present period. Now a new course has been scheduled for the spring of 1998. Support from the NKS for this course and other activities strengthening the build-up of competence would be valuable.

The Nordic countries have established a culture of co-operating together in a very close and informal manner. The NKS has created a network of competent people in the field of marine radioactivity in the Nordic countries. Other forms of international co-operation and projects cannot replace this network. It is important that work on marine radioactivity will continue to be supported by the NKS. Otherwise it would be much more difficult to deal with many issues of concern for the Nordic population in the marine environment, especially in case of a sudden release of radionuclides to the environment.

Acknowledgements

The work described in this report was supported by the NKS and by the participating institutes. The project group wants also to express gratitude to all those who contributed in various ways to the implementation of the EKO-1 and making the described work possible, whether by participation or in other ways.

Appendix 1

List of project participants:

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4: Hafrannsóknastofnunin / Marine Research Institute (IS)
5: NRPA, Norwegian Radiation Protection Authority (NO)
6: NLH, Agricultural University of Norway (NO)
7: IFE, Institute of Energy Technology (NO)
8: Lund University (SE)

Appendix 2

Publications

The work in the individual EKO-1 sub-projects is described in more details in the EKO-1 technical report:

> Pálsson S E (ed.) (1998). *Final reports from sub-projects within the Nordic Nuclear Safety Research Project EKO-1.* NKS, NKS/EKO-1(98)TR-1.

Apart from annual reports etc. the following publications can be regarded as EKO-1 reports, since they are the outcome of work that the EKO-1 work group as a whole decided that should be undertaken and they are not the result of a nationally run project.

> Nielsen S P (1996). *An Intercomparison Exercise on Radionuclides in Sediment Samples.* Risø National Laboratory, Risø-R-914(EN).
>
> Ilus E (1996). *Evaluation of Sediment Sampling Devices and Methods used in the EKO-1 Project.* NKS, NKS/EKO-1(96)TR-1.
>
> Ilus E (ed.) (1998). *Proceedings of a Seminar on Dating of Sediments and Determination of Sedimentation Rate, Helsinki, April 2-3, 1997.* STUK, (Radiation and Nuclear Safety Authority, Finland), STUK-A145.

Only the report by Erkki Ilus on sediment samplers was published formally as a NKS/EKO-1 technical report, the two other reports were published by the authors' institutes.

Most of the EKO-1 work has been reported in various scientific articles by the participants. References can be found in individual sections of the main report as well as in the different national reports, which can be found in technical report NKS/EKO-1(98)TR-1 referred to above.

Appendix 3

Abbreviations

Explanation of some of the abbreviations used in this report

CEC	Cation Exchange Capacity
K_d	Distribution Coefficient, ratio of concentration of a radionuclide in sediment compared to concentration in water
CIC	Constant Initial Concentration
CRS	Constant Rate of Supply
CF:CS	Constant Flux Constant Sedimentation rate
LOI	Loss on Ignition

Appendix 4

International project work with links to EKO-1

There are various international projects, which deal with the same areas as the EKO-1 projects. The EKO-1 project participants have participated actively in some of these projects and had some connections with others. This helps to make the work within EKO-1 and the other project complementary to each other and decreases the risk of parallel work.

- **HELCOM / MORS** involves monitoring of radioactive substances in the Baltic Sea, compilation of results and advise to the authorities. The HELCOM / MORS does not however have any funds to support research in this field. Some EKO-1 project participants are also working within HELCOM / MORS.
- **AMAP (Arctic Monitoring and Assessment Programme)**. A programme that collects and evaluates data from the Arctic region, but does not give financial support to research projects. EKO-1 project participants were involved in AMAP work.
- **ANWAP - Arctic Nuclear Waste Assessment Program**. This program is run by the US Department of Defence / Office of Naval Research and its aim is to assess the disposition of radioactive waste deposited in the Arctic region by the former Soviet Union. The program is comprised of approximately 70 different projects and the total funding has been $30 million over 3 years. Some contacts have been made with the project management by EKO-1 project participants.
- **NATO / CCMS *“Cross-Border Environmental Problems Emanating from Defence-related Installations and Activities”***. This work is not producing new data but rather compiling information from current data. Project members have been associated with work in this project.
- **European Union project works**. Many EKO-1 project participants are also participating in European Union project work. Most of the participants in the EKO-1 project group are also participating in the EU ARMARA project.
- **IAEA’s International Arctic Seas Assessment Project (IASAP)**. This project was established in 1993 and it is run by the IAEA in co-operation with the Norwegian and Russian Governments. It also involves experts from relevant IAEA member states. The project will last for four years and the objectives are to assess the risks associated with the radioactive wastes dumped in the Kara and Barents Seas and to examine possible remedial actions. Project members were associated with work within this project.