

BALTIC SEA ENVIRONMENT PROCEEDINGS

No. 85

RADIOACTIVITY IN THE BALTIC SEA 1992-1998



HELSINKI COMMISSION
Baltic Marine Environment Protection Commission

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RADIOACTIVITY IN THE BALTIC SEA 1992-1998

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PREFACE

Monitoring data on radioactivity in the Baltic Sea has been collected since 1984 within the framework of the Helsinki Commission – the Baltic Marine Environment Protection Commission. This monitoring has been carried out and supervised by a group of experts known as the MORS group (Monitoring of Radioactive Substances in the Baltic Sea), whose members represent the Contracting Parties of the Helsinki Convention. Until 1996 the group was a permanent expert group responsible for advising and reporting to the HELCOM Environment Committee. Following organisational adjustments at HELCOM in 1996, the group's work was redefined as a 5-year project, MORS-PRO 1996-2000. The MORS group subsequently reported to the Monitoring and Assessment Group – HELCOM MONAS, although their basic activities remained unchanged.

The first report of the MORS group “Three Years Observations of the Levels of Some Radionuclides in the Baltic Sea after the Chernobyl Accident” was published in 1989 as the Baltic Sea Environment Proceedings, No. 31. A second report “Radioactivity in the Baltic Sea 1984-1991” was published in 1995 as Baltic Sea Environment Proceedings, No. 61.

The present report provides an overview of the work carried out by the MORS Group during the period 1992-1998. This work was facilitated by close co-operation from all HELCOM Contracting Parties (Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Poland, Russia, and Sweden), as well as the IAEA, who carried out sampling and analyses, and reported data and findings at the annual MORS meetings, and to the data consultants.

Project Manager
Sven P. Nielsen

EXECUTIVE SUMMARY

Since 1984 the contracting parties to the Helsinki Convention have collected monitoring data on radionuclides in the Baltic Sea. The data covers radioactivity in the Baltic marine environment and in discharges from nuclear installations (nuclear power plants and nuclear research facilities) within the catchment area of the Baltic Sea.

Due to the relatively slow exchange of water between the Baltic Sea and the North Sea, contaminants such as anthropogenic radionuclides have a prolonged residence time in the Baltic Sea. Levels of ^{90}Sr and ^{137}Cs are consequently still high in the Baltic Sea compared with other water bodies around the world. Strontium-90 contamination originates from atmospheric nuclear weapons testing, which peaked in the 1960's and led to direct input to the Baltic Sea in the shape of atmospheric fallout, and to delayed input via rivers. Caesium-137 was also released during atmospheric nuclear weapons tests, but the related input was small compared to the direct input in fallout from the Chernobyl accident in 1986. The delayed input of ^{137}Cs to the Baltic Sea from rivers is smaller than the direct atmospheric fallout of ^{137}Cs , and also much smaller than delayed inputs of ^{90}Sr .

Since the Chernobyl accident in 1986, levels of anthropogenic radionuclides in the Baltic Sea have generally declined, mainly due to radioactive decay and the gradual outflow of water through the Belt Sea and the Kattegat. However, increased discharges of radionuclides into the Irish Sea from Sellafield since 1994 have affected European coastal waters including the Baltic Sea, which receives small but measurable amounts of technetium-99 in inflow from the North Sea.

Baltic Sea biota received the most important contribution to their radionuclide levels from the Chernobyl accident in 1986, predominantly in the form of ^{137}Cs and ^{134}Cs . Time trends of ^{137}Cs in fish did not always closely follow the corresponding trends in seawater. The high trophic level species, including predators such as cod and pike, showed the highest ^{137}Cs levels, but maximum values after 1986 occurred considerably later than in seawater. In the long-term, however, ^{137}Cs time trends in biota evidently do follow trends in seawater.

Levels of ^{137}Cs in fish in the Åland Sea and the Bothnian Sea, where initial concentrations in seawater were highest after Chernobyl, have decreased since the end of the 1980's. Slightly lower values were observed in the Gulf of Finland. In the Baltic Proper, the area with the highest production of fish for human consumption, levels of ^{137}Cs in fish increased until the beginning of the 1990's and then only gradually decreased. Fish in the Belt Sea showed lower values. The Kattegat area showed the lowest ^{137}Cs levels in fish from about 1990 onwards, with no obvious impact from Chernobyl.

Marine algae from nuclear power plant (NPP)

monitoring stations in the Baltic Sea, used as bio-indicators of radionuclides in the Baltic Sea, show low activities for radionuclides representing NPP discharges. Some of these radionuclides were also found in samples of benthic fauna. Caesium-137 levels in seaweed from various parts of the Baltic Sea regions closely followed the ^{137}Cs time trend for surface waters.

Bottom sediments play an important role in radiological processes in the marine environment, because they usually act as a final sink for radionuclides in the sea. Large amounts of radioactive substances entering the sea are adsorbed in the course of time on suspended particulate matter, and subsequently deposited in bottom sediments.

The most important event with significant impact on the inventories of artificial radionuclides in the Baltic Sea sediments was the accident at the Chernobyl nuclear power plant in 1986. Following this accident, the amounts of ^{137}Cs and ^{134}Cs increased considerably, especially in the seabed of the Bothnian Sea and the eastern Gulf of Finland. During the 1990s the deposition of long-lived fallout nuclides continued from the water phase into the sediments, but at a decreasing rate. The highest amount of ^{137}Cs detected in the Baltic Sea sediments was 125 kBq m^{-2} in the northern Bothnian Sea. The total inventory of ^{137}Cs in the seabed of the Baltic Sea was estimated at 1,940 – 2,210 TBq in 1998. Considerable differences were still observed between the different sub-basins, and even between sampling stations situated very close to each other.

The radiological consequences of radioactivity in the Baltic Sea have been assessed based on information on input and observed levels of radioactivity in the Baltic Sea for 1950-1996. Doses to humans were calculated using a model designed to cover the waters in the North Atlantic region, including the Baltic Sea. The calculations cover the time period from 1950 to 2000, and include source contributions from nuclear weapons testing, the Chernobyl accident, European reprocessing plants, and nuclear installations bordering on the Baltic Sea area.

Dose rates from anthropogenic radioactivity to individual members of the public were calculated based on rates of annual intake and beach occupancy time. The dose rates to individuals living around the Bothnian Sea and the Gulf of Finland are predicted to be larger than for any other area around the Baltic Sea, due to the pattern of fallout from the Chernobyl accident. Dose rates are predicted to have peaked in 1986 at a value of 0.2 mSv y^{-1} .

An assessment of the dumping of low-level radioactive waste in the Baltic Sea in the 1960's by Sweden and the Soviet Union indicated that related doses to humans are negligible.

Doses from naturally occurring radioactivity in seafood were also calculated and compared with the doses from anthropogenic radioactivity ob-

tained through marine pathways. This comparison shows that dose rates and doses from natural radioactivity generally dominate, except in 1986, when individual dose rates from Chernobyl fallout in some regions of the Baltic Sea approached those from natural radioactivity.

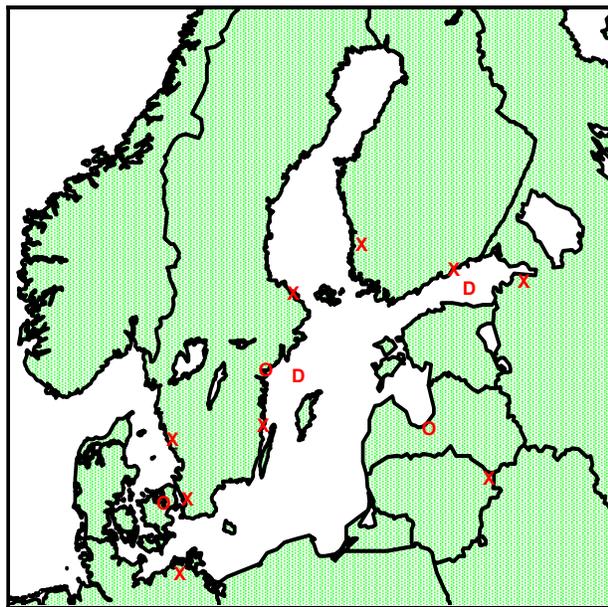
The maximum annual dose during the period 1950-2000 to individuals from any critical group in the Baltic Sea area is estimated at 0.2 mSv y^{-1} , which is below the dose limit of 1 mSv y^{-1} for the exposure of the general public set out in the Basic Safety Standards of the IAEA and the EU. Even considering the uncertainties involved in this assessment, it is unlikely that any individual has been exposed through marine pathways to doses at levels above this dose limit. Doses due to liquid

discharges from nuclear power plants in the Baltic Sea area are estimated to be at or below the levels specified in the Basic Safety Standards as being of no regulatory concern.

1 INTRODUCTION

Since 1984, the Contracting Parties to the Helsinki Convention have collected monitoring data on radionuclides in the Baltic Sea. The data covers concentrations of radioactivity in the Baltic marine environment and discharges from nuclear installations (nuclear power plants and nuclear research facilities) in the catchment area of the Baltic Sea. Additionally, important sources of anthropogenic radionuclides in the Baltic Sea have been considered – namely atmospheric fallout from nuclear weapons testing and the Chernobyl accident, and discharges into sea from European reprocessing facilities at Sellafield in the UK and La Hague in France.

Figure 1.1 Nuclear power plants (X), nuclear research facilities (O) and sites for dumping of radioactive waste (D) in the Baltic Sea area.



Due to the relatively slow exchange of water between the Baltic Sea and the North Sea, contaminants such as anthropogenic radionuclides have a prolonged residence time in the Baltic Sea. Levels of ^{90}Sr and ^{137}Cs are consequently still high in the Baltic Sea compared with other water bodies around the world. Strontium-90 contamination originates from atmospheric nuclear weapons testing, which peaked in the 1960's and led to direct input to the Baltic Sea from atmospheric fallout, and to delayed input via rivers. Caesium-137 was also released during atmospheric nuclear weapons tests, but the related input was small compared to the direct input from the Chernobyl accident in 1986. The delayed input of ^{137}Cs to the Baltic Sea from rivers is smaller than the direct atmospheric fallout of ^{137}Cs , and also much smaller than delayed inputs of ^{90}Sr , since due to its chemical properties, caesium is less mobile in

the environment than strontium.

The anthropogenic radionuclides present in the Baltic Sea originate from several sources and modes of input. Direct atmospheric fallout has accounted for the main inputs, from atmospheric nuclear weapons tests and from the Chernobyl accident. Run-off from the land into sea via rivers has also contributed significantly for ^{90}Sr , and to a lesser extent for ^{137}Cs . Hydrodynamic transport has been significant for radionuclides from European nuclear reprocessing facilities, even though these are located beyond the North Sea, and far away from the Baltic Sea. Only a small proportion of the total radioactive discharges into the sea from these facilities is estimated to reach the Baltic Sea, but on a relative scale this input is significant for the Baltic Sea. Finally, direct discharges into the Baltic Sea occur in coastal waters from the routine operations of nuclear facilities along the shores of the Baltic Sea itself. These discharges are very low, however, and are authorised by national regulatory authorities. Other anthropogenic radionuclides are present in the Baltic Sea, but ^{137}Cs is by far the most important radionuclide with respect to radiation doses to humans.

Since the Chernobyl accident in 1986, the levels of anthropogenic radionuclides in the Baltic Sea have generally declined mainly due to radioactive decay and the gradual outflow of water through the Belt Sea and the Kattegat. However, increased discharges of radionuclides into the Irish sea from Sellafield since 1994 have affected European coastal waters including the Baltic Sea, which receives small but measurable amounts of technetium-99 in inflow from the North Sea.

This report describes the work carried out by the MORS group during the period 1992-1998 concerning radioactive substances in the Baltic Sea. Chapter 2 lists the sources of anthropogenic radionuclides in the Baltic Sea. Chapter 3 describes the monitoring network which forms the backbone of the HELCOM Recommendation 18/1 on monitoring activities. Chapter 4 describes the work carried out on the analytical quality of the monitoring data. Chapters 5, 6 and 7 describe the levels of radionuclides in seawater, sediments and biota, respectively. Chapter 8 describes the modelling of radioactivity in the Baltic Sea, and Chapter 9 covers the assessment of radiation doses to humans from radioactivity in the Baltic Sea.

References

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2 SOURCES OF RADIOACTIVITY IN THE BALTIC SEA

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In the previous Joint Report of the HELCOM/MORS Group and in the Final Report of the Marina Balt Project (Nies et al., 1995, Ilus & Ilus, 2000) various sources of radioactive substances in the Baltic Sea were discerned, with some more significant than the others. In this chapter these data are updated. The sources are classified as follows:

- I. **Aquatic discharges into the Baltic Sea**, including all nuclear facilities (power reactors, research reactors, waste handling facilities, fuel production etc) located in the catchment area of the Baltic Sea and discharging directly or indirectly into the sea.
- II. **Aquatic discharges from facilities located outside the Baltic Sea catchment area**, including inputs from facilities (mainly nuclear reprocessing plants) releasing discharges outside the area which are nevertheless detectable in the Baltic Sea.
- III. **The Chernobyl accident** (Current impact)
- IV. **Atmospheric nuclear weapons tests**
- V. **Other significant sources**, including hospitals, research institutes and conventional industrial plants using radioactive substances.

2.1 Discharges into the Baltic Sea

Nuclear facilities around the Baltic Sea are divided into different types: nuclear power reactors, research reactors, waste handling facilities and a fuel fabrication plant. The main characteristics of these facilities are given below and summarised in Table 2.1. The locations of the facilities are shown in Figure 2.1.

2.1.1 Nuclear power plants (NPPs)

The discharge patterns for most of the NPPs are similar, and the most abundant nuclides in the discharges are shown in Table 2.1. The amounts of the most significant radionuclides discharged (³H, ⁶⁰Co, ¹³⁷Cs and ⁹⁰Sr) are shown in Figures 2.2-2.10 (Ilus & Ilus, 2000).

Finland

Loviisa

The Loviisa NPP is located on the north coast of the Gulf of Finland, on Hästholmen Island, about 12 km south-east of the town of Loviisa. The plant consists of two 488 MW_e pressurised water reactors (VVER-440). The first unit started commercial operation in 1977 and the second in 1980. Both units have subsequently been in continuous use, except during brief annual outages. The discharge area of the NPP, Hästholmsfjärden Bay, is a semi-enclosed basin between the mainland and the outer archipelago, where many islands, narrow and shallow straits and underwater sills limit the exchange of water with the open Gulf of Finland (Ilus & Ilus, 2000).

Olkiluoto

The Olkiluoto NPP is located on the east coast of the southern Bothnian Sea, about 12 km north of the town of Rauma. The plant consists of two 840 MW_e boiling-water reactors (ASEA). The first unit started commercial operation in 1978 and the second in 1980. Both units have subsequently been in continuous use, except during brief annual outages. Liquid discharges are conducted through a narrow channel into the Bothnian Sea. Although there are many skerries off Olkiluoto Island, the discharge area is quite open and the exchange of water with the open Bothnian Sea is relatively free (Ilus & Ilus, 2000).

Germany

Greifswald

The Greifswald NPP is located on the south coast of the southern Baltic Proper (Arkona Sea), about 22 km north-east of the town of Greifswald. The plant was operated by the German Democratic Republic, and shut down in 1990. The plant consisted of five 408 MW_e pressurised water reactors (VVER-440) (Fujii & Morishima, 1994) which were started up in 1973, 1974, 1977, 1979 and 1990. The discharge area of the Greifswald NPP was the open but shallow coastal waters at the mouth of Greifswalder Bodden Bay. Discharge data are available for the period from 1983 to 1998, including the period after shutdown (Ilus & Ilus, 2000).

Lithuania

Ignalina

The Ignalina NPP is located about 230 km inland in the catchment area of the Baltic Sea, but the distance to the sea along water courses is about 600 km. The plant consists of two 1450 MW_e light-water-cooled, graphite-moderated reactors (RBMK-1500) (Fujii & Morishima, 1994), which started commercial operation in 1984 and 1987. Liquid discharges from the Ignalina NPP are led into Lake Druksiai, which is drained by the River Prorva a tributary of the River Daugava. A small proportion of these discharges could theoretically reach the Gulf of Riga via the River Daugava (Ilus & Ilus, 2000).

Russia

Leningrad

The Leningrad NPP is located on the southern coast of the eastern Gulf of Finland near the town of Sosnovy Bor. The plant consists of four 960 MW_e light-water-cooled, graphite-moderated reactors (RBMK-1000) (Fujii & Morishima, 1994), which started commercial operation in 1973, 1975, 1979 and 1981. The discharge area of the Leningrad NPP, Koporian Bay, is a wide, open bay with an effective exchange of water with the Gulf of Finland (Ilus & Ilus, 2000).

Sweden

Barsebäck

The Barsebäck NPP is located on the south-west coast of Sweden, i.e. in the Sound directly opposite the Danish capital, Copenhagen, 20 km away. The plant consists of two 600 MW_e boiling water reactors (ASEA), which started commercial operation in 1975 and 1977 (Fujii & Morishima, 1994). Liquid discharges are released directly into the open Sound, where the exchange of water is effective. The prevailing direction of sea currents in the Sound is northwards (Ilus & Ilus, 2000). The first reactor at Barsebäck NPP was shut down in 1999 .

Forsmark

The Forsmark NPP is located on the south-west coast of the Bothnian Sea, about 70 km north-east of the town of Uppsala. The plant consists of 3 boiling-water reactors (ASEA) with rated powers of 970 MW_e (2 units) and 1060 MW_e (1 unit) (Fujii & Morishima, 1994), which have been operating since 1980, 1981 and 1985. Discharges are conducted either directly (F3) or through a bio-test basin (F1, F2) into the sea. This bio-test basin is an artificially enclosed body of water (0.9 km²) in the archipelago set up for ecological and radio-ecological studies. The coast by the bio-test basin is open and the exchange of water with the Bothnian Sea is free (Ilus & Ilus, 2000).

Oskarshamn

The Oskarshamn NPP is located on the west coast of the central Baltic Proper, about 20 km north of the town of Oskarshamn, at approximately the same latitude as the northernmost point of Öland. The plant consists of 3 boiling-water reactors (ASEA) with rated powers of 440, 595 and 1060 MW_e (Fujii & Morishima, 1994), taken into operation in 1972, 1974 and 1985. Liquid discharges are conducted into Hamnefjärden, a small bay connected by a narrow, shallow sound to the Baltic Proper. The coast is open off Hamnefjärden (Ilus & Ilus, 2000).

Ringhals

The Ringhals NPP is located on the east coast of the Kattegat, about 50 km south of Gothenburg. The plant consists of one 750 MW_e boiling-water reactor (ASEA) and 3 pressurised water reactors (WEST/STAL-LAVAL) with rated powers of 800 (1 unit) and 915 MW_e (2 units) (Fujii & Morishima, 1994). The units have been in commercial operation since 1975, 1976, 1981 and 1983, respectively. The discharge area of the Ringhals NPP is a stretch of open coast on the Kattegat with an effective exchange of water (Ilus & Ilus, 2000).

2.1.2 Research reactors

Risø (Denmark)

Risø National Laboratory is located on the Roskilde Fjord about 6 km north of the town of Roskilde, on the island of Zealand about 40 km west of Copenhagen. Risø operates one heavy-water-moderated research reactor (DR3, 10 MW) which releases discharges of ³H. Liquid discharges from the reactor's secondary cooling system and from Risø's waste-treatment station enter the fjord, which is connected by narrow straits via Isefjord to the Kattegat. The ³H discharges from the waste-treatment station predominate (about 90%) due to the processing of wastewater collected from the reactor (Ilus & Ilus, 2000).

Salaspils (Latvia)

The Salaspils Research Centre (Nuclear Research Centre of the Latvian Academy of Sciences) is located near Salaspils, about 25 km east of Riga. The Research Reactor (IRT) was a 5 MW pool reactor using 90% enriched ²³⁵U. The reactor was started up in 1961, reconstructed in 1979, and shut down in 1998. The reactor was located inland at a distance of 2.5 km from the River Daugava. Liquid wastes from the closed

waste-collection system of the Salaspils reactor were irregularly discharged into a municipal wastewater system. Discharges could conceivably reach the Gulf of Riga along this route via the Daugava River (Ilus & Ilus, 2000).

Studsvik (Sweden)

The Studsvik facilities are located on the west coast of the northern Baltic Proper, about 22 km east of the town of Nyköping. Discharges are conducted into a narrow strait that connects Tvären, a large semi-enclosed bay in the archipelago, to the Baltic Proper. The facility includes 2 small research reactors of 1 MW and 50 MW which started operating in 1960. These reactors are used for tests of material and nuclear fuel and the production of isotopes for medicinal and scientific purposes. The site also includes laboratories, an incineration facility and a melting facility.

2.1.3 Waste handling and other facilities

Waste handling facilities differ considerably in their operation, producing various discharge patterns.

Paldiski (Estonia)

Paldiski is located on the south coast of the western Gulf of Finland. During the Soviet era, Paldiski was a naval base, where crews of nuclear-powered submarines were trained. Two nuclear reactors (70 and 90 MW) operated at the base until 1989, when on the initiative of the Estonian government the reactors were decommissioned and moved to Russia. A Finnish company (IVO International) purified the residual liquid waste in 1995, and the treated water was discharged into the Gulf of Finland. Since 1995 no discharges have been reported from Paldiski into the sea. No data is available on possible previous discharges from the naval training centre. The sea off Paldiski consists of an open bay with a free exchange of water with the Gulf of Finland (Ilus & Ilus, 2000).

Sillamäe (Estonia)

In 1948 a chemical metallurgy plant was established at Sillamäe on the south coast of the Gulf of Finland, for processing uranium originally from alum-shale, and later from uranium ore. In 1970 the plant was switched to process loparite, a mineral which is rich in niobium and other rare metals but also contains uranium and in particular thorium (Ehdwall et al., 1993, 1994, Putnik et al., 1994).

Table 2.1 Nuclear facilities in the catchment area of the Baltic Sea (locations shown in Figure 2.1).

Facility	Country	Type of facility; number of units	Main radionuclides discharged before 1998	Remarks
Loviisa	Finland	Power Plant; 2 PWR	^3H , ^{124}Sb , ^{137}Cs , ^{134}Cs , ^{60}Co , $^{110\text{m}}\text{Ag}$	
Olkiluoto	Finland	Power Plant; 2 BWR	^3H , ^{60}Co , ^{51}Cr , ^{54}Mn , ^{58}Co	
Greifswald	Germany	Power plant; 5 PWR	^3H , $^{110\text{m}}\text{Ag}$, ^{58}Co , ^{58}Co , ^{54}Mn , ^{60}Co , ^{137}Cs , ^{134}Cs	Shut down in 1990
Ignalina	Lithuania	Power Plant; 2 RBMK	^3H , ^{60}Co , ^{137}Cs , ^{54}Mn	
Leningrad	Russia	Power Plant; 4 RBMK	^{60}Co , ^{137}Cs , ^{134}Cs	^3H not reported
Barsebäck	Sweden	Power plant; 2 BWR	^3H , ^{60}Co , ^{51}Cr , ^{58}Co	First reactor shut down in 1999
Forsmark	Sweden	Power Plant; 3 BWR	^3H , ^{60}Co , ^{51}Cr , ^{141}Ce	
Oskarshamn	Sweden	Power Plant; 3 BWR	^3H , ^{60}Co , ^{65}Zn , ^{137}Cs , ^{51}Cr	
Ringhals	Sweden	Power Plant; 3 PWR, 1 BWR	^3H , ^{58}Co , ^{60}Co , ^{51}Cr	
Risø	Denmark	Research reactor	^3H	
Salaspils	Latvia	Research reactor	^3H , ^{137}Cs , ^{90}Sr , ^{134}Cs	Shut down in 1998
Studsvik	Sweden	Research reactor, Waste handling facility	^3H , ^{137}Cs , ^{90}Sr , ^{60}Co , ^{134}Cs	
Paldiski	Estonia	Training centre	^3H , ^{90}Sr , ^{137}Cs , ^{60}Co	Shut down in 1989. No discharges after 1995
Sillamäe	Estonia	Chemical metallurgy plant, waste depository	^{238}U , ^{234}U , ^{226}Ra , ^{210}Pb , ^{232}Th	
ABB Atom	Sweden	Fuel fabrication plant	^{234}U , ^{234}Th , ^{238}U , ^{235}U , ^{231}Th , ^{60}Co	

Between 1948 and 1959, waste from uranium processing was transported and heaped on the first coastal terrace of the Gulf of Finland outside the plant.

In 1959, a waste depository was established on the same site, and during 1969-1970 it was expanded to its present size. Today the depository is an oval retention impoundment on the shore of Narva Bay with a total area of about 330 000 m², the top of the dam being about 25 m above sea level. The sea in front of the depository is open, and the exchange of water with the Gulf of Finland is effective (Ilus & Ilus, 2000). Consequently, the impact of the depository on the Gulf of Finland has been estimated to be small. Risk assessment has shown that in a normal situation leakage of the depository causes an individual committed dose of 1 µSv over 50 years from Narva Bay. The collective committed dose over 50 years was estimated to be 1 manSv (Ehdwall et al., 1994, Bergström et al., 1994).

Studsvik (Sweden)

In addition to operating a research reactor, the facilities at Studsvik also handle radioactive waste. There is a facility for melting scrap metal and an incinerator for handling combustible waste on the site. The melting facility is licensed for the production of 900 tonnes and the incinerator 600 tonnes per year (Studsviks årsredovisning 1998). The plant's location is described above.

2.1.4 Fuel fabrication plants

ABB Atom (Sweden)

ABB Atom is located 100 km west of Stockholm near the city of Västerås. The plant releases discharges into a small river that runs into a bay of Lake Mälaren, which is connected to the Baltic Sea. The plant produces fuel for nuclear reactors and has a licence to produce fuel using the equivalent of 600 tonnes of converted uranium dioxide per year. These discharges could theoretically reach the Baltic Sea.

Figures 2.7-2.10 show that the cumulative amounts of locally discharged, long-lived nuclides, such as ³H, ⁹⁰Sr and ¹³⁷Cs, are still slowly increasing in the Baltic Sea, although the discharges of ³H and ¹³⁷Cs have decreased during recent years. In contrast, the cumulative amount of ⁶⁰Co (with a shorter half-life) has begun to decrease, due to lower discharges in recent years (Ilus & Ilus, 2000).

2.2 Discharges from facilities located outside the Baltic Sea catchment area

2.2.1 Nuclear reprocessing plants

Small proportions of the discharges from Sellafield, which discharges into the Irish Sea from the north-west coast of England, and La Hague, which discharges from the north-west coast of France into the English Channel, are transported by the inflow of saline water through the Danish Straits into the Baltic Sea. The transport time for radionuclides is about 4-5 years after discharge into the Irish Sea (Sellafield) or about 2 years after discharge into the English Channel (La Hague) (Nies et al., 1995). Models indicate that only about 4% of discharges from Sellafield and about 8% of the discharges from La Hague reach the Skagerrak. Due to the efficient mixing of water layers in the Kattegat and the Belt Sea, most of the radioactivity returns to Skagerrak, and only about 1% enters the Baltic Sea (Nielsen et al., 1995).

2.2.2 Fuel fabrication plant (Springfields, UK)

The Springfields fuel fabrication plant is located south of Sellafield. Discharges into the Irish Sea are lower than those from Sellafield, but it can be assumed that the same transportation patterns apply for discharges from Springfields. The plant manufactures fuel elements and uranium hexafluoride. Liquid radioactive waste consists mainly of thorium and uranium and their decay products, and is released from a pipeline into the estuary of the River Ribble. Beta-emitting radionuclides make up the main contribution to the radiological impact from this facility (MAFF, 1999).

Discharges from these 3 facilities are given in Table 2.2.

2.2.3 Other nuclear facilities (NPPs and research reactors)

Discharges from other nuclear facilities are of at least one magnitude lower than those from the reprocessing plants mentioned above, so they are of minor significance, and therefore not addressed here.

Table 2.2 Discharges from Sellafield, La Hague and Springfields 1992 – 1998 (OSPAR, 1996, 1997, 1998).

Sellafield

Nuclide TBq y ⁻¹	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
Tritium	1,724	2,144	1,699	1,803	1,199	2,310	1,680	2,700	3,000	2,600	2,130
Total-α	2.1	2.7	2.2	2.1	1.6	2.6	1.0	0.4	0.27	0.18	0.17
Total-β	81	101	71	62	57.2	97	125	190	140	140	85.5
⁹⁹ Tc	4.2	6.1	3.8	3.9	3.2	6.1	72	190	150	84	52.7
¹³⁷ Cs	13.3	28.6	23.5	15.6	15.3	21.9	13.8	12	10	7.9	7.5
⁹⁰ Sr	10.1	9.2	4.2	4.1	4.2	17.1	28.9	28	16	37	17.7
¹⁴ C	3.0	2.0	2.0	2.4	0.8	2	8.2	12	11	4.4	3.75

La Hague

Nuclide TBq y ⁻¹	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
Tritium	2,540	3,720	3,260	4,710	3,770	5,150	8,090	9,610	10,500	11,900	10,500
Total-α	0.36	0.37	0.37	0.15	0.11	0.1	0.097	0.0701	0.046	0.048	0.047
Total-β	575	588	314	116	76.5	73.2	70.2	52.9	29.4	26.6	26.5
⁹⁹ Tc	10.1	7.1	5.7	0.9	0.47	0.64	0.37	0.10	0.12	0.13	0.219
¹³⁷ Cs	8.5	12.6	12.5	5.6	3.0	4.4	10.5	4.62	2.41	2.46	2.51
⁹⁰ Sr	39.5	28.5	15.8	29.8	18	25	16	15	5.30	3.73	2.53
¹⁴ C	1.7	2.6	2.9	3.9	3.60	4.90	7.40	9.20	9.94	9.65	9.76

Springfields

Nuclide TBq y ⁻¹	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
Total-α	NI	0.4	0.2	0.14	0.1	0.08	0.16	0.12	0.12	0.12	0.195
Total-β	NI	114	92	38.9	120	63	114	112	150	140	150
⁹⁹ Tc	NI	NI	NI	NI	0.1	0.1	0.016	0.03	0.033	0.033	0.0027
²³⁰ Th	NI	NI	NI	NI	0.03	0.02	0.09	0.057	0.048	0.052	0.085

NI = No Information

2.3 The Chernobyl accident

The accident in Chernobyl which occurred in 1986 has subsequently been the main source of radioactivity in the Baltic Sea.

2.3.1 The current situation

The total input of ¹³⁷Cs from the Chernobyl accident into the Baltic Sea was estimated to be 4.5 PBq by the CEC (1991). This estimate was later adjusted to 4.7 PBq (Nielsen et al., 1999). The HELCOM/MORS Expert Group estimated in its previous Joint Report that 4.1-5.1 PBq of ¹³⁷Cs and 0.08 PBq of ⁹⁰Sr (decay-corrected to 1991)

entered the Baltic Sea as a consequence of the Chernobyl accident (Nies et al., 1995). Fallout from Chernobyl was very unevenly distributed over the catchment area of the Baltic Sea; the most contaminated land areas were around the Bothnian Sea and the eastern Gulf of Finland.

2.3.2 River discharges

The amount of Chernobyl-derived ¹³⁷Cs carried into the Baltic Sea by river runoff has been evaluated for all Finnish rivers discharging into the Baltic Sea; for 5 rivers discharging from the former Soviet Union; and for the River Vistula in Poland.

Thorough calculations showed that a total of 65 TBq of ^{137}Cs was discharged by Finnish rivers into the Baltic Sea during the period 1986-1996 (Saxén & Ilus, 2000). Corresponding calculations performed in Russia showed that 14 TBq was discharged by five large rivers from the territory of the former Soviet Union during 1986-1988 (Gavrilov et al., 1990). A Polish estimate made for the River Vistula showed that a total of 18 TBq of ^{137}Cs was discharged during the period 1986-1996 (Ilus & Ilus, 2000). The total river input of ^{137}Cs may have been roughly 300 TBq during the period 1986-1996 (Ilus & Ilus, 2000).

2.4 Atmospheric nuclear weapons tests

The impact of global fallout as a source of radioactivity in the Baltic Sea was assessed in detail in the previous Joint Evaluation Report of the HELCOM/MORS Group (Nies et al., 1995). According to recent calculations (Nielsen, pers. comm.), the total inputs of ^{90}Sr and ^{137}Cs from weapons tests into the Baltic Sea were 0.5 and 0.8 PBq ($5.0\text{E}+14$ and $8.0\text{E}+14$ Bq), respectively (decay-corrected to 1998). Inventories based on measured concentrations of these nuclides in water and sea-floor sediments resulted in quite similar values: $4.9\text{E}+14$ for ^{90}Sr and $6.2\text{E}+14$ for ^{137}Cs (calculated to 1981) (Salo et al., 1986).

2.5 Other significant sources

2.5.1 Dumping of radioactive waste

Five officially confirmed dumpings of radioactive waste have occurred in the Baltic Sea region at three different sites. All these small-scale dumpings were performed in the late 1950s or early 1960s. A radiological assessment of these dumpings has shown that doses to man from these activities were negligible (Nielsen et al., 1999).

2.5.2 Other types of facilities (e.g. hospitals, research institutes etc)

Radionuclides are used for various purposes in industry, medicine and research, and their use is increasing. It was generally not possible to obtain reliable information about discharges from these sources. However, according to reports from UN-SCEAR, their contribution to overall anthropogenic exposures is relatively insignificant. Most radionuclides used in hospitals are short-lived, while discharges are also small, so their impact

on the radioactivity of the Baltic Sea is negligible and very local (Ilus & Ilus, 2000).

2.6 Conclusions

The most significant source of radioactivity with respect to the total inventory of artificial radionuclides in the Baltic Sea is the fallout from the accident at the Chernobyl NPP in 1986. The most important radionuclides present in this deposition were ^{137}Cs and ^{134}Cs . The total input of ^{137}Cs from Chernobyl to the Baltic Sea has been estimated at 4,700 TBq. Post-Chernobyl river discharges of ^{137}Cs have been calculated in the Marina Balt Study as amounting to some 300 TBq, comprising 6-7% of the total input (Table 2.3).

The second most important source is global fallout from atmospheric nuclear weapons tests carried out during the late 1950s and early 1960s. The predominant radionuclides in this global fallout were ^{137}Cs and ^{90}Sr , in an activity ratio of about 1.6. During the late 1990s the decay-corrected amounts of weapons-test ^{137}Cs and ^{90}Sr in the Baltic Sea have been evaluated at 800 and 500 TBq, respectively (Table 2.3).

The corresponding decay-corrected total injections of ^{137}Cs and ^{90}Sr originating from nuclear reprocessing plants in Western Europe (Sellafield and La Hague) have been estimated to 250 and 40 TBq, respectively (Table 2.3). At present this source has become of minor importance, due to significant reductions in discharges from Sellafield in recent years.

The predominant radionuclide in the discharges from nuclear power plants and research reactors in the Baltic Sea region is ^3H . Total discharges of ^3H from these local sources have amounted to 2,400 TBq and those of other beta-gamma nuclides to about 20 TBq. The total discharges of alpha nuclides have been 0.0001 TBq (Ilus&Ilus, 2000). During the period 1992-1998, the total discharges of ^3H were 700 TBq (decay-corrected to 1998), while discharges of ^{137}Cs were 0.07 TBq (Table 2.4).

For ^{137}Cs , the main sources were fallout from Chernobyl (82%) and nuclear weapons tests (14%). For ^{90}Sr , the main source of contamination was the fallout from nuclear weapons testing (81%), while the proportion from the Chernobyl fallout was smaller (13%).

Table 2.3 Total inputs of ^{137}Cs and ^{90}Sr into the Baltic Sea from different sources (decay-corrected to 1998).

Source	^{137}Cs TBq	Percentage of total	^{90}Sr TBq	Per- centag e of total
Chernobyl accident ^{1,4)} incl. river discharges ⁵⁾	4,700 300	82	80	13
Nuclear weapons tests ⁶⁾	800	14	500	81
Discharges from sources outside the Baltic region ²⁾ ³⁾	250	4	40	6
Discharges into the Bal- tic ¹⁾ , Cumulative amount up to 1998	1.66	0.03	0.76	0.1

1) based on measurements
2) estimated
3) according to the Joint Report of HELCOM/MORS, 1995
4) according to Nielsen et al., 1999
5) according to Ilus & Ilus, 2000
6) according to Nielsen (unpublished)

Table 2.4 Annual and cumulative discharges of certain radionuclides into the Baltic Sea from all nuclear facilities during the period 1992-1998.

Annual discharges Decay corrected to 1998 Bq								
Nuclide / half-life (years)	1992	1993	1994	1995	1996	1997	1998	Total
^3H (12.3)	1.45E+14 1.03E+14	1.07E+14 8.07E+13	9.10E+13 7.26E+13	9.99E+13 8.44E+13	8.30E+13 7.41E+13	1.03E+14 9.76E+13	1.10E+14 1.10E+14	7.00E+14
^{54}Mn (0.85)	3.30E+10 2.59E+08	2.16E+10 3.79E+08	1.77E+10 6.98E+08	2.04E+10 1.81E+09	7.33E+10 1.46E+10	2.16E+10 9.63E+09	8.34E+09 8.34E+09	3.57E+10
^{58}Co (0.19)	1.34E+11 6.54E+01	8.91E+10 1.55E+03	9.04E+10 5.60E+04	6.91E+10 1.53E+06	1.04E+11 8.19E+07	4.73E+10 1.33E+09	3.22E+10 3.22E+10	3.36E+10
^{60}Co (5.27)	2.46E+11 1.12E+11	2.28E+11 1.18E+11	1.95E+11 1.15E+11	1.66E+11 1.12E+11	1.89E+11 1.46E+11	2.10E+11 1.85E+11	7.33E+10 7.33E+10	9.56E+11
^{125}Sb (2.77)	1.04E+10 2.31E+09	1.12E+10 3.19E+09	8.32E+09 3.06E+09	7.64E+09 3.61E+09	7.65E+09 4.64E+09	7.86E+09 6.12E+09	4.74E+09 4.74E+09	2.94E+10
^{134}Cs (2.06)	2.31E+10 3.06E+09	5.09E+10 9.46E+09	4.14E+10 1.08E+10	2.09E+10 7.63E+09	1.95E+10 9.93E+09	2.45E+10 1.75E+10	1.14E+10 1.14E+10	7.10E+10
^{137}Cs (30.2)	6.29E+10 5.48E+10	8.65E+10 7.71E+10	7.46E+10 6.81E+10	5.37E+10 5.01E+10	4.50E+10 4.30E+10	5.02E+10 4.90E+10	3.24E+10 3.24E+10	4.26E+11
^{238}Pu (87.8)				7.40E+04 7.23E+04	9.98E+05 9.82E+05	3.35E+05 3.32E+05	2.60E+05 2.60E+05	1.65E+06
^{239}Pu (24100)				1.80E+04 1.80E+04	8.50E+03 8.50E+03	4.70E+04 4.70E+04	1.10E+04 1.10E+04	8.45E+04
^{241}Am (432)				2.70E+04 2.69E+04	1.50E+06 1.50E+06	7.65E+05 7.63E+05	1.71E+08 1.71E+08	1.73E+08

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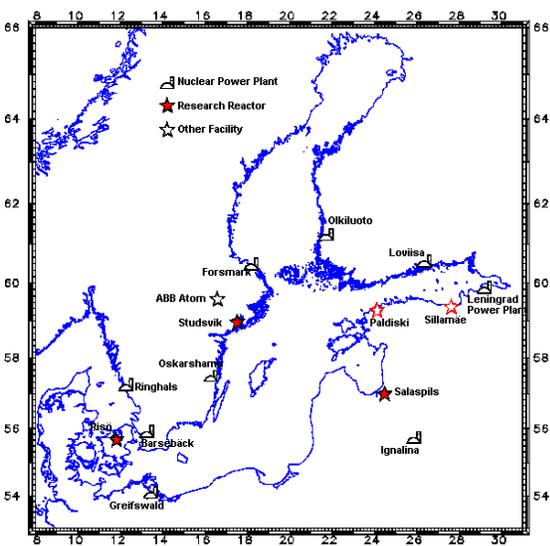


Figure 2.1 Locations of nuclear facilities in the Baltic Sea region.

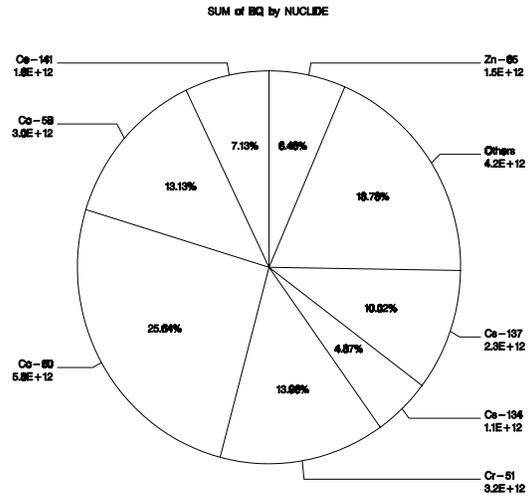


Figure 2.2 Total aquatic discharges into the Baltic Sea until the end of 1998, excluding ^3H .

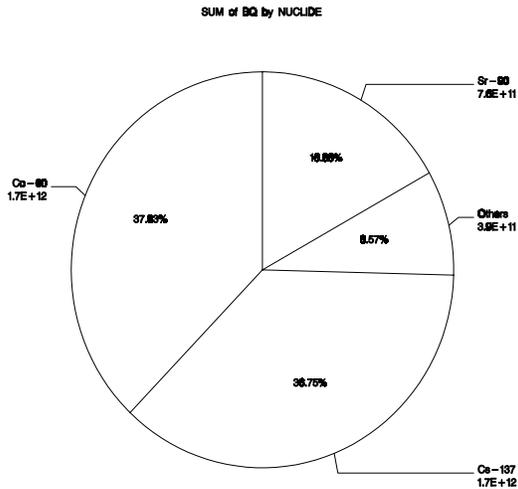


Figure 2.3 Cumulative aquatic discharges into the Baltic Sea decay corrected to the end of 1998, excluding ^3H .

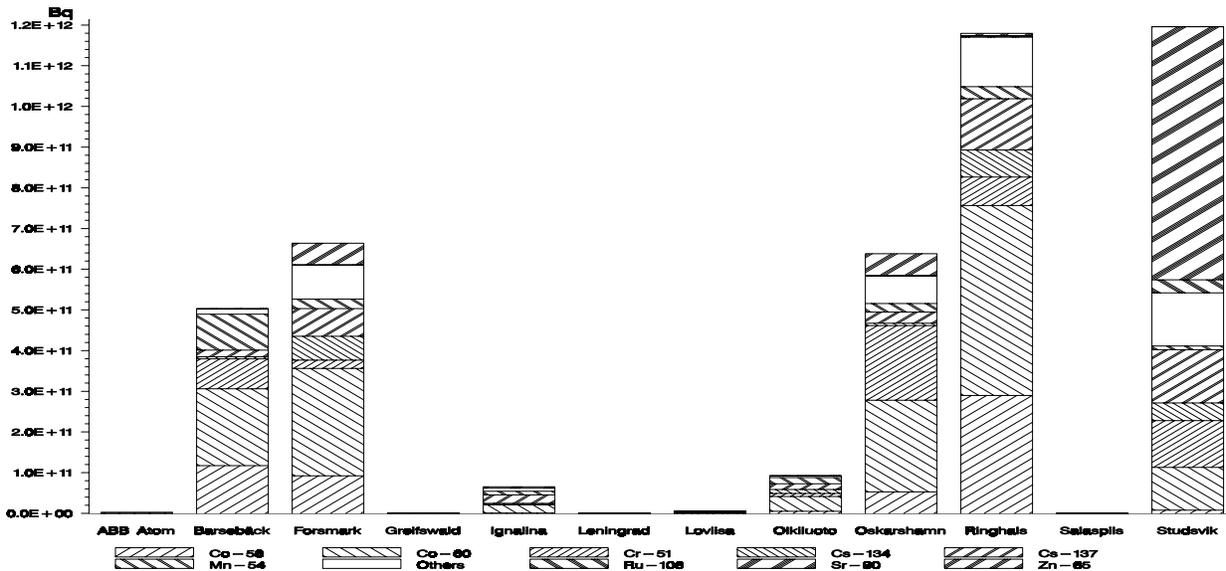


Figure 2.4 Total aquatic discharges into the Baltic Sea 1992-1998, excluding ^3H .

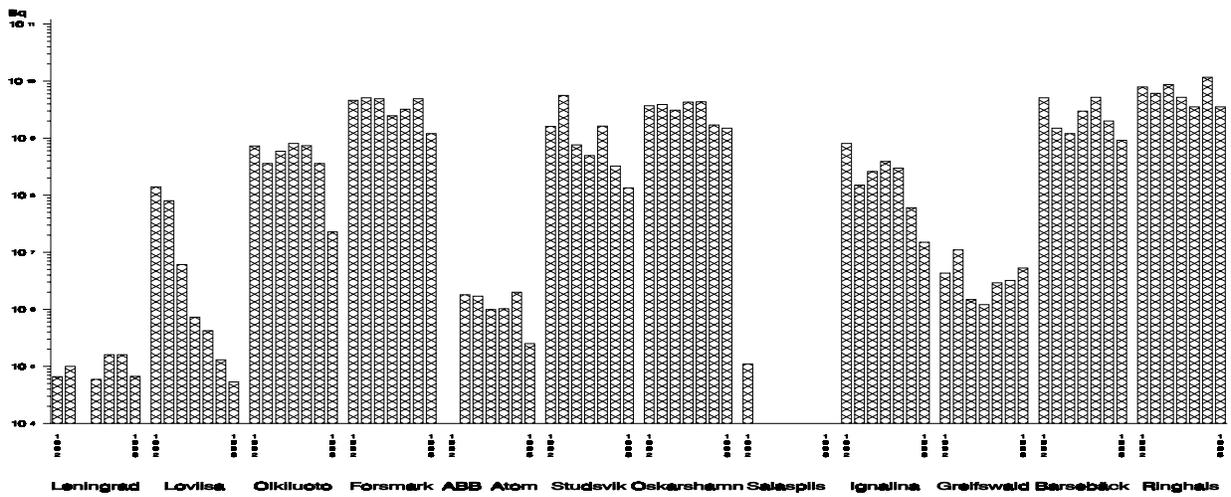


Figure 2.5 Annual ^{60}Co discharges from nuclear facilities into the Baltic Sea 1992-1998.

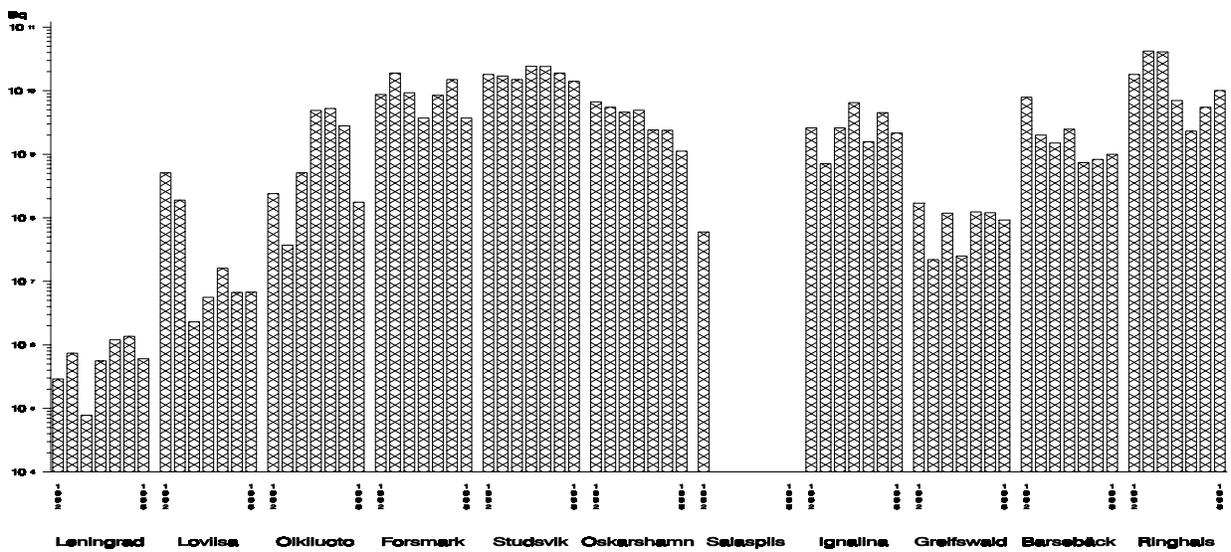


Figure 2.6 Annual ^{137}Cs discharges from nuclear facilities into the Baltic Sea 1992-1998.

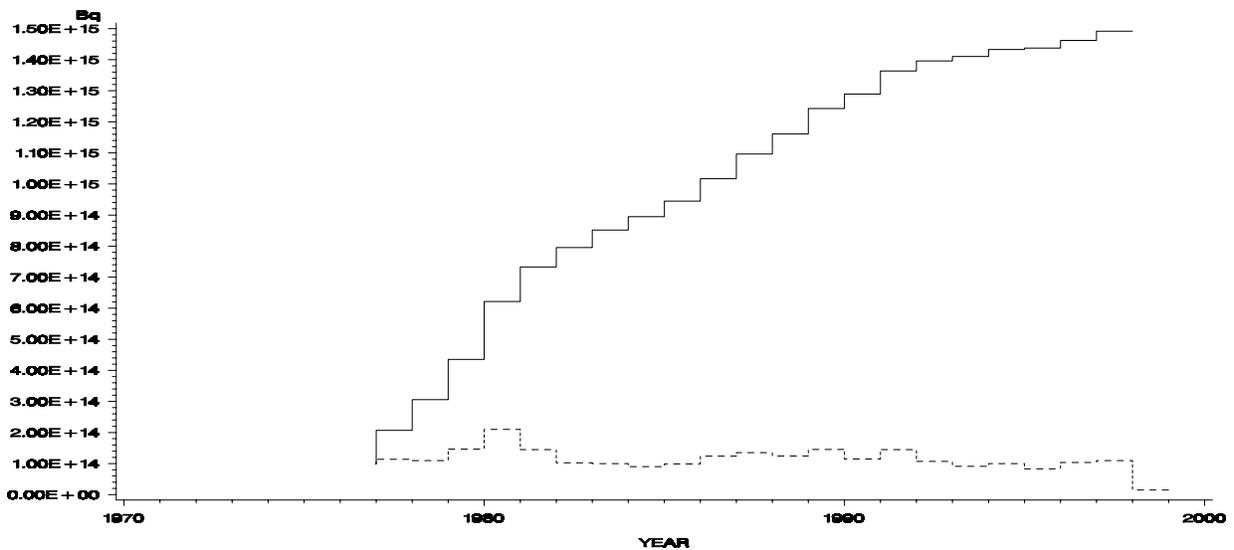


Figure 2.7 Cumulative aquatic ^3H discharges into the Baltic Sea 1977-1998.

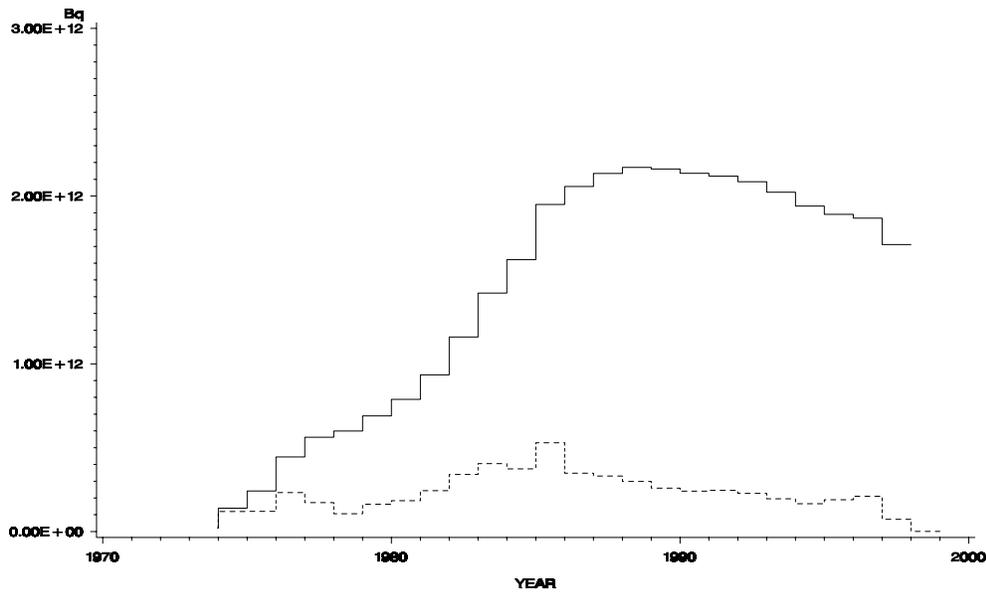


Figure 2.8 Cumulative aquatic ^{60}Co discharges into the Baltic Sea 1974-1998.

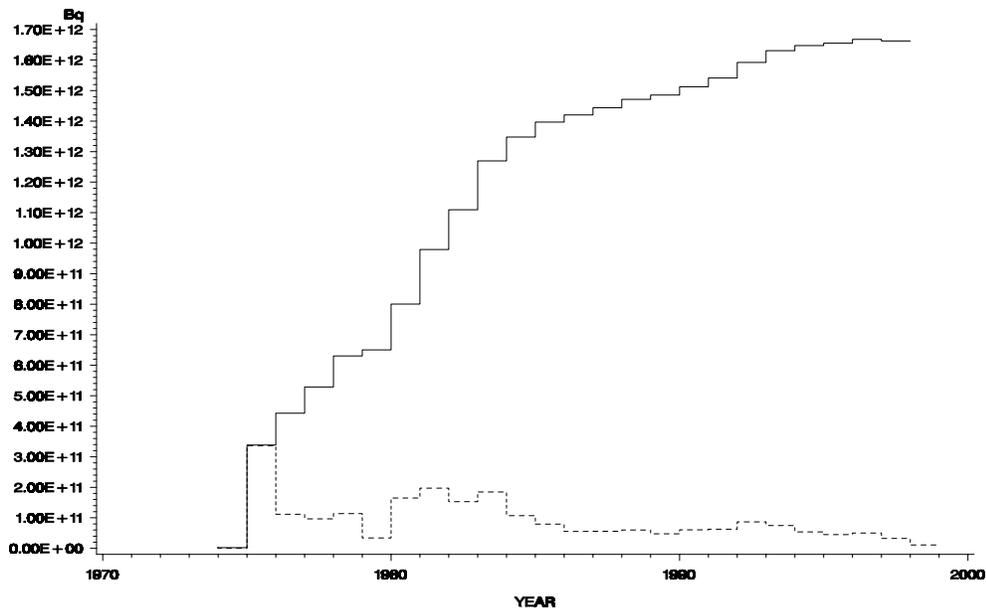


Figure 2.9 Cumulative aquatic ^{137}Cs discharges into the Baltic Sea 1974-1998.

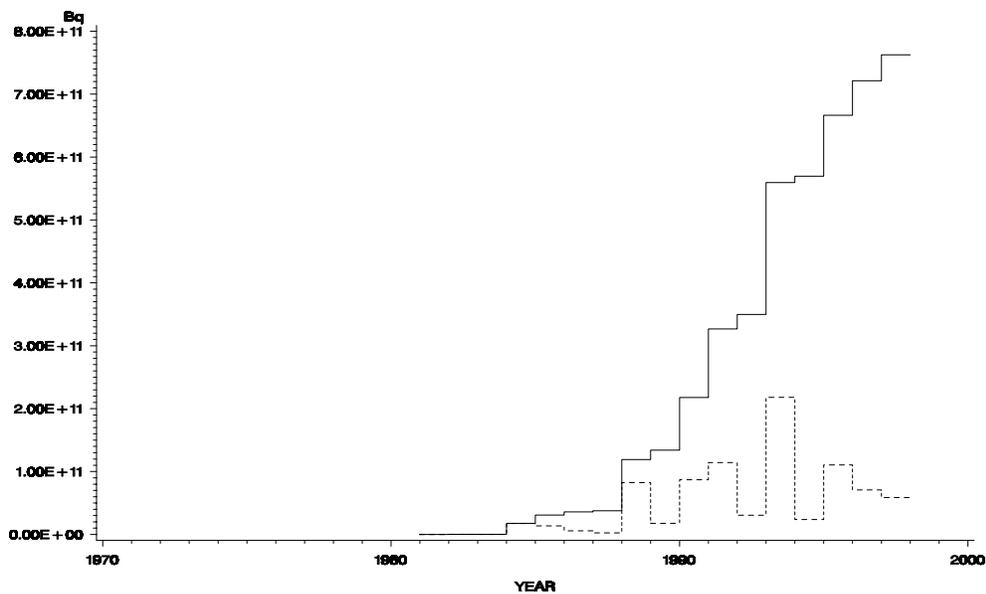


Figure 2.10 Cumulative aquatic ^{90}Sr discharges into the Baltic Sea 1981-1998.

3 THE MONITORING NETWORK

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3.1 Stations

To fulfil the objectives set out in the Project Plan of the Project Group for Monitoring of Radioactive Substances in the Baltic Sea (MORS) the Contracting Parties have agreed on a network of sampling stations covering all the sub-areas of the Baltic Sea. The radioactive substances monitoring programme is defined in HELCOM Recommendation 18/1, which was adopted in 1997. The programme includes regular sampling of sea water, sediments, fish, aquatic plants and benthic animals at permanent sites, with analysis results

reported annually for the HELCOM Data Base.

According to the HELCOM Recommendation 18/1 each Baltic Sea coastal state should establish environmental stations at sea or on the coast, and inform the Commission of their locations. The recommendation also stipulates that the list of stations contained in the Guidelines should be used as a basis for the environmental monitoring programme in each country, bearing in mind that the list consists of minimum requirements, and that additional voluntary measurements should be taken to provide complementary information. The routine station network for the regular monitoring programme is shown in Figures 3.1.1 - 3.1.5.

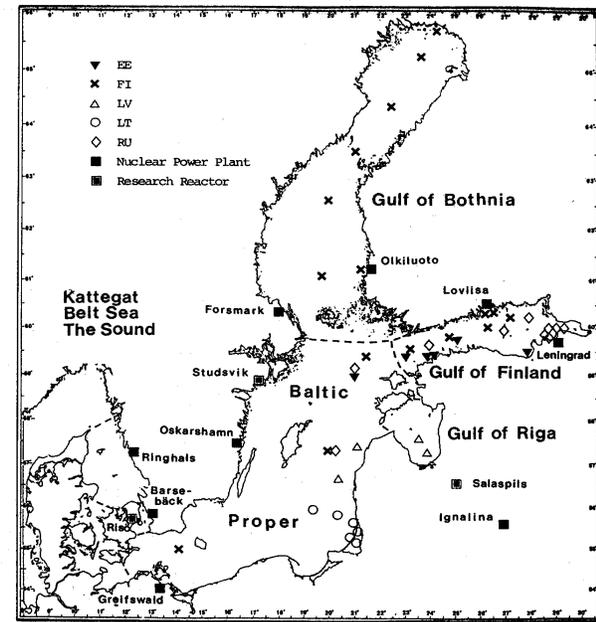


Figure 3.1.1 Sampling stations set up by Estonia, Finland, Latvia, Lithuania and Russia for monitoring sea water; and the locations of nuclear power plants and research reactors around the Baltic Sea.



Figure 3.1.2 Sampling stations set up by Denmark, Germany and Poland for monitoring sea water.

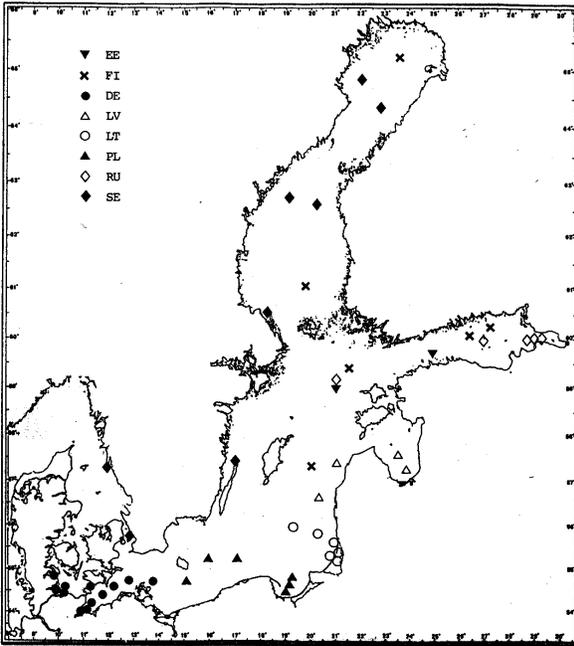


Figure 3.1.3 Sediment sampling stations

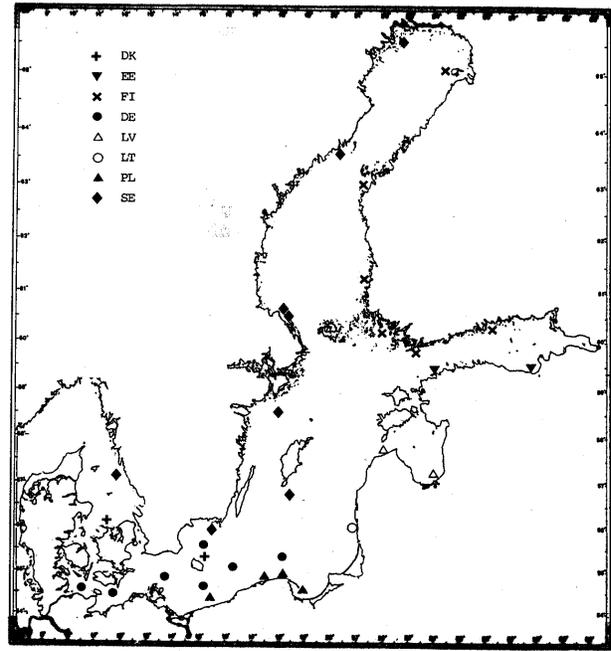


Figure 3.1.4 Fish sampling areas

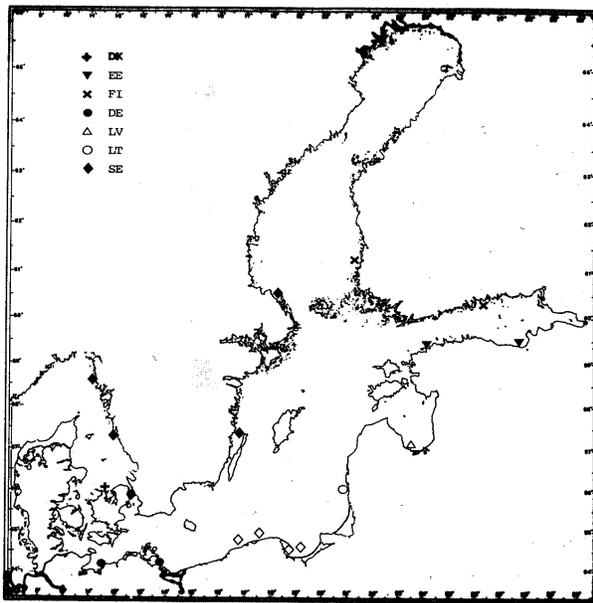


Figure 3.1.5 Sampling sites for aquatic plants and benthic animals.

The monitoring of radionuclides in the Baltic Sea is carried out by individual countries according to their technical capabilities and the equipment available. The routine monitoring network consists of varying numbers of sampling stations set up for the monitoring of sea water, sediment and biota in different sub-areas of the Baltic Sea as follows:

Sub-area	Sea water	Sedi-ment	Fish	Aquatic plants	Benthic animals
Gulf of Bothnia	7	7	8	3	3
Gulf of Finland	20	7	4	4	1
Gulf of Riga	2	2	1	1	0
Baltic Proper	43	22	15	4	6
Belt Sea, Kattegat and the Sound	37	11	4	4	5
Total	109	49	32	16	13

The following laboratories were involved in the work of the MORS Project during the period 1992-1998:

- Denmark: - Risø National Laboratory, Roskilde
- Estonia: - Estonian Meteorological and Hydrological Institute, Tallinn, 1993-1994
- Estonian Radiation Protection Centre, Tallinn, 1997-1998
- Finland - STUK - Radiation and Nuclear Safety Authority, Helsinki (formerly the Finnish Centre for Radiation and Nuclear Safety, until 1997)
- Germany - Federal Maritime and Hydrographic Agency, Hamburg
- Federal Fisheries Research Centre, Hamburg
- Latvia - Ministry of Environmental Protection and Regional Development, Liel-Riga
- Regional Environmental Board, Riga, 1995-1998
- Lithuania - Central Environmental Research Laboratory, Vilnius, 1993-1994
- Environmental Protection Ministry, Joint Research Centre, Vilnius, 1995-1998
- Poland - Central Laboratory for Radiological Protection, Warsaw
- Russia - V.G. Khlopin Radium Institute, St. Petersburg
- Sweden - Swedish Radiation Protection Institute, Stockholm

In addition to the environmental data on radionuclides, data on radioactive discharges from nuclear power plants and research reactors operating in the Baltic Sea catchment area is also collected for the HELCOM MORS discharge database. According to HELCOM Recommendation 18/1 Contracting Parties should report the relevant discharge data to the Commission annually. Reporting of liquid discharges into the aquatic environment is compulsory, while discharges into air are reported on a voluntary basis. Only nuclides with a half-life longer than one week need to be reported, and data should be nuclide specific. The locations of the nuclear power plants and research reactors are shown in Figure 3.1.1.

3.2 Radionuclides monitored in the environment

Lists of the obligatory and voluntary analyses for the 5 sample types are included in Recommendation 18/1, and reproduced here in Table 3.2.1. In addition to data on radionuclides, information on other factors such as water temperature, salinity,

sediment type, and the numbers and sizes of animal specimens in samples are also reported for the database.

Table 3.2.1 Monitoring of radionuclides in the Baltic Sea.

Sample		Obligatory	Voluntary
A.	Sea water (results in Bq m ⁻³)	Radiocaesium *) ⁹⁰ Sr**)	³ H; ⁹⁹ Tc; ^{239,240} Pu; ²⁴¹ Am; γ-emitters
B.	Sediments (results in Bq kg ⁻¹ dry wt. and Bq m ⁻²)	γ-emitters ***)	⁹⁰ Sr; ^{239,240} Pu; ²⁴¹ Am; natural radionuclides (e.g. ²¹⁰ Po)
C.	Fish (results in Bq kg ⁻¹ fresh wt.)	γ-emitters ***)	⁹⁰ Sr; natural radionuclides (e.g. ²¹⁰ Po)
D.	Aquatic plants (results in Bq kg ⁻¹ dry wt.)	γ-emitters ***)	⁹⁰ Sr, ⁹⁹ Tc; ^{239,240} Pu; ²⁴¹ Am; natural radionuclides
E.	Benthic animals (results in Bq kg ⁻¹ dry wt.)	γ-emitters ***)	⁹⁰ Sr, ⁹⁹ Tc; natural radionuclides (e.g. ²¹⁰ Po); ^{239,240} Pu; ²⁴¹ Am

*) ¹³⁷Cs and ¹³⁴Cs, if possible

**) regularly, on a carefully selected number of samples

***) ⁴⁰K, ¹³⁷Cs and other γ-emitters identified in the γ-spectrum

Table 3.2.2 shows the amounts of data reported for the environmental database during the period 1992-1998. The numbers of sea water and sediment samples analysed (which increased substantially following the Chernobyl accident in 1986) decreased to some extent towards the end of the reporting period. No data on ⁹⁹Tc in benthic animals was submitted. One set of sediment data from January 1999 was included, because sampling planned for the end of 1998 was delayed due to bad weather conditions.

Table 3.2.2 Amounts of data on different constituents reported for the HELCOM data bank 1992-1998.

Constituents	Number of analyses						
	1992	1993	1994	1995	1996	1997	1998
A. WATER							
Obligatory							
¹³⁷ Cs	287	286	312	211	332	258	249
¹³⁴ Cs	256	195	222	157	231	126	102
⁹⁰ Sr	167	132	96	89	97	122	88
Voluntary							
³ H	18	20	19	4	4	19	6
⁹⁹ Tc	-	-	-	-	-	-	8
^{239,240} Pu	22	9	40	9	47	5	-
²⁴¹ Am	16	8	40	4	40	2	-
other gamma emitters (excluding Cs)	43	36	66	32	66	27	26
B. SEDIMENT							
Obligatory							
Gamma emitters	1532	508	1006	662	478	571	744
Voluntary							
⁹⁰ Sr	-	-	-	-	-	-	7
^{239,240} Pu	46	21	18	32	59	74	41
²⁴¹ Am	40	16	6	17	5	8	-
Natural radionuclides	756	202	578	374	211	253	387
C. FISH							
Obligatory							
Gamma emitters	106	293	200	311	303	217	235
Voluntary							
⁹⁰ Sr	15	21	16	20	12	8	16
Natural radionuclides	33	79	56	86	90	64	70
D. AQUATIC PLANTS							
Obligatory							
Gamma emitters	61	58	70	62	61	74	71
Voluntary							
⁹⁰ Sr	1	2	1	1	2	6	2
⁹⁹ Tc	12	-	-	-	-	-	3
^{239,240} Pu	-	2	-	3	4	5	-
²⁴¹ Am	-	-	-	-	2	4	4
Natural radionuclides	5	9	9	13	12	18	17

Table 3.2.2 The amounts of data on different constituents reported for the HELCOM data bank 1992-1998 (continued).

Constituents	Number of analyses						
	1992	1993	1994	1995	1996	1997	1998
BENTHIC ANIMALS							
Obligatory							
Gamma emitters	30	43	34	65	62	45	45
Voluntary							
⁹⁰ Sr	2	2	2	6	6	4	2
⁹⁹ Tc	-	-	-	-	-	-	-
^{239,240} Pu	-	-	-	4	6	1	1
²⁴¹ Am	-	-	-	4	4	1	-
Natural radionuclides	7	14	9	17	13	13	13

The sources of environmental data on radionuclides in sea water and sediments submitted for the HELCOM database are quite evenly distributed throughout the Baltic Sea. The number of sampling locations for sediments is much smaller than that for water,

however (Figs. 3.2.1 and 3.2.2). The majority of the sampling locations for fish, aquatic plants and benthic animals are concentrated in the southern waters of the Baltic Sea (Figs. 3.2.3 - 3.2.5).

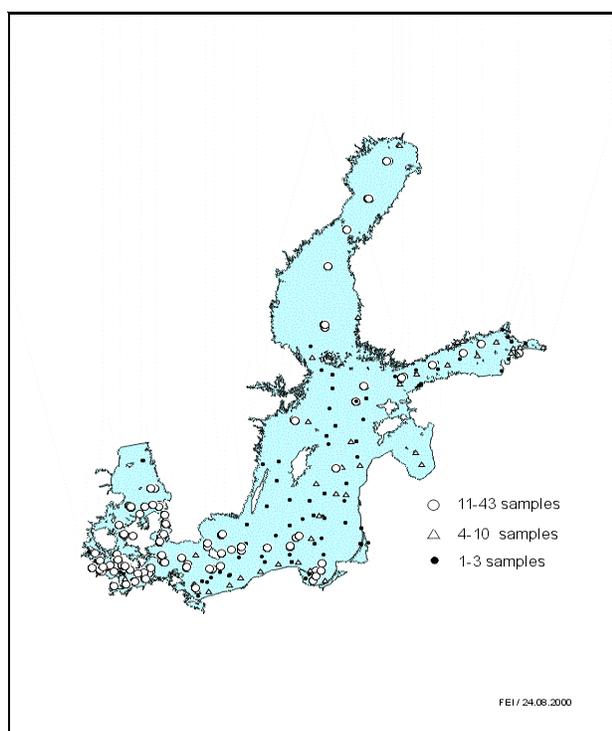


Figure 3.2.1 Sea water sampling locations for measurements of radioactive substances 1992-1998.

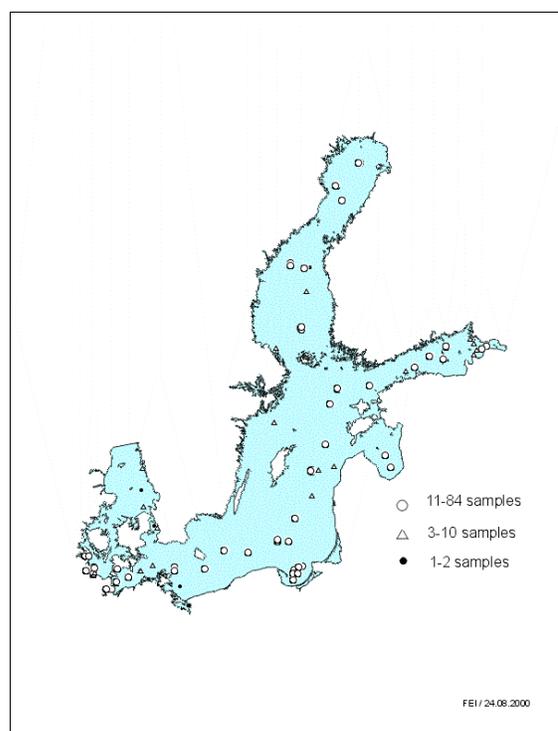


Figure 3.2.2 Sediment sampling locations for measurements of radioactive substances 1992-1998.

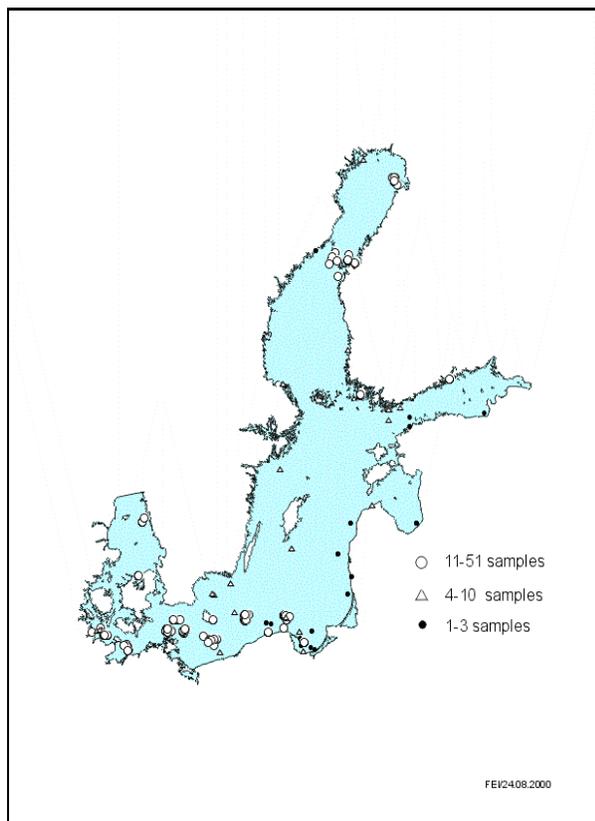


Figure 3.2.3 Fish sampling locations for measurements of radioactive substances 1992-1998.

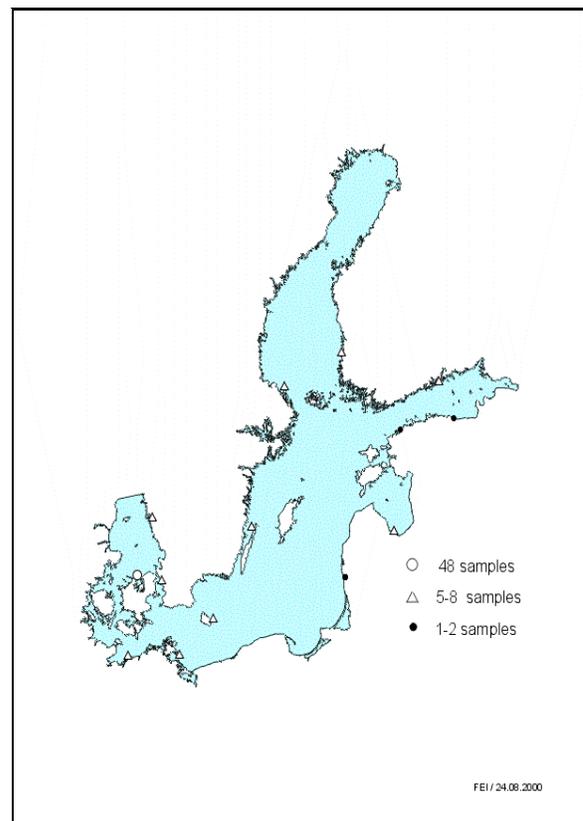


Figure 3.2.4 *Fucus vesiculosus* sampling locations for measurements of radioactive substances 1992-1998.

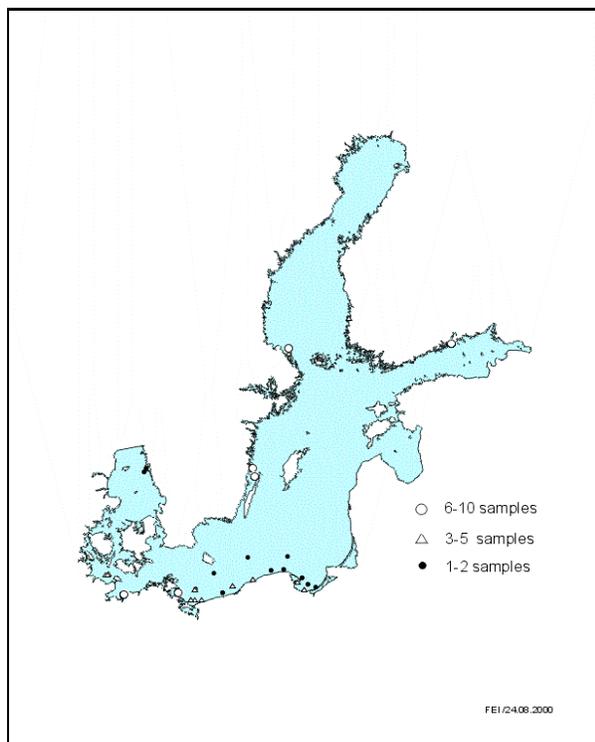


Figure 3.2.5 Benthic animal sampling locations for measurements of radioactive substances 1992-1998.

3.3 Description of the HELCOM Databases

The results of the monitoring programme and the discharge data are submitted for the HELCOM databases, which are managed on consultant basis. The environmental data is managed by the Finnish Environment Institute (FEI) while the discharge data is managed by STUK -- the Radiation and Nuclear Safety Authority, Finland.

3.3.1 HELCOM MORS environmental database

Technical environment

The environmental radioactivity database was originally developed using the tools of the Ingres Program, and applications were run on VMS operating system computers. During the reporting period the original environmental database was converted to MS ACCESS97 software, which can be run in a PC with no network connection needed. Only minor changes needed to be made to the structure of the database and its files during this conversion, since which all data has been submitted as Excel tables. After verification in Excel, data is converted to ACCESS97 for manipulation.

The database has been linked with ODBC (Open DataBase Connectivity) to GIS (Geographic Information System) software, and ArcView for map presentations. The maps have been finalised using CorelDraw software.

Logical structure

The design of the logical structure of the MORS database follows the structure used in the reporting guidelines. Data definitions are not as strict as before, due to the more flexible data entry parameters of MS Excel 97. Some additional modifications were carried out in order to avoid 'the year 2000 problem'. Some new fields have been added, and some old fields modified.

For the present reporting period three mediums were used: sea water, sediment and biota. Data on each medium was arranged in two tables: one consisting of information on sampling and the sample itself (sample table), and the other consisting of the analysis results (nuclide table). Some descriptions of the codes used are also added.

Coding

The codes and variables used in the environmental database are as follows:

Sea water (sample):

Key code, Country code, Laboratory code, Sequence number, Date (Year, Month, Day), Station, Latitude (dd.mmmm), Longitude (dd.mmmm), Latitude (dd.dddd), Longitude (dd.dddd), Total depth, Sampling depth, Salinity, Water temperature, Filtration code and Basin code.

Sea water (nuclides):

Key code, Nuclide, Method code, < (less than, if needed), Value in Bq m⁻³ and Error %

Sediment (sample):

Key code, Country code, Laboratory code, Sequence number, Date (Year, Month, Day), Station, Latitude (dd.mmmm), Longitude (dd.mmmm), Latitude (dd.dddd), Longitude (dd.dddd), Device code, Total depth, Slice depth (upper and lower horizon), Sampled area, Sediment type code, State of oxidation, Dry weight (%), Ignition loss (%) and Basin code.

Sediment (nuclides):

Key code, Nuclide, Method code, < (less than, if needed), Value in Bq kg⁻¹ dry weight, Error % and Value in Bq m⁻²

Biota (sample):

Key code, Country code, Laboratory code, Sequence number, Date (Year, Month, Day), Station, Latitude (dd.mmmm), Longitude (dd.mmmm), Latitude (dd.dddd), Longitude (dd.dddd),

Sampling depth, Species code (Rubin), Tissue code, Number of specimen, Average size of specimen, Dry weight (%), Ignition loss (%) and Basin code.

Biota (nuclides):

Key code, Nuclide, Method code, < (less than, if needed), Value in Bq kg⁻¹ wet weight for fish and in Bq kg⁻¹ dry weight for other biota, Basis (dry or wet) code and Error %.

The database and its contents

There are six main data tables, two for each reporting medium: sea water, sediment and biota. The number of data rows covering the years 1992-1998 is:

	sea water	sediment	biota
sample records	1,985	1,787	855
analyse records	4,662	6,097	2,853

3.3.2 HELCOM MORS discharge database

The HELCOM/MORS discharge register for radioactive discharges from nuclear power plants and other nuclear facilities in the Baltic Sea catchment area is managed by STUK. Aquatic discharges from 13 facilities operating in the area have been reported for the database since they started operating. Six nuclear power plant units and one research reactor were shut down during the 1990s. Although airborne discharges are reported on a voluntary basis, reporting is almost as completely implemented as for aquatic discharges.

The discharge database has been built up using tools available in the SAS product family under Microsoft Windows NT. Incoming data is in ASCII-format or Excel-files. All output routines are made using SAS.

The database consists of a single table with six variables: the name of the power plant or facility, type of discharge (aquatic/airborne), year, radionuclide discharged, annual discharge in Bq, and notes. Only nuclides with a half-life longer than one week are covered in the database. A total of 2,478 sets of data were reported during the period 1992-1998.

4 DATA QUALITY

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- 2) Marine Environment Laboratory (MEL), International Atomic Energy Agency (IAEA), Monaco

4.1 Introduction

Over ten laboratories from the nine Baltic Sea coastal countries have performed gamma-spectrometrical and radiochemical analyses on marine samples within the Baltic Sea monitoring programmes. New information from Estonian, Latvian and Lithuanian laboratories has been incorporated for the first time into this joint report. The IAEA/Monaco Laboratory has arranged and participated in intercomparisons. Data from the Monaco Laboratory is also presented here, although it does not directly concern the Baltic Sea monitoring.

In general, the methods for measuring radionuclides have not changed much since earlier reports were made, but certain developments have led to improvements in measuring techniques and quality control. These trends are discussed below. Detailed data on sampling, the pre-treatment of samples, the analysis of radionuclides and quality control in the Baltic Sea marine environment are presented in the tables below.

Seven intercomparison exercises have been performed during the project period 1992 -1998. The results of these exercises are also presented here.

4.2 Analytical procedures used by MORS laboratories

4.2.1 Sampling and pre-treatment of marine samples

Sampling data is reported for the first time in this report. Information on sampling methods, related know-how and those responsible for sampling is also given. Typically, a respected institute is responsible for sampling, but representatives of commercial organisations and fishermen may be involved in fish sampling. Nowadays it is very well realised that sampling procedures can greatly affect the accuracy of the results. However, quality control of sampling is still relatively undeveloped. One reason for this may be that it is quite difficult to perform. A few comparisons have been done on the HELCOM/MORS sampling of sediments.

Those results are presented in "Intercomparison of sediment sampling devices by means of artificial radionuclides in the Baltic Sea sediments", Baltic Sea Environment Proceedings No. 80 (2000).

Pre-treatment procedures for samples have not changed much during the period in question. Sampling and pre-treatment procedures are described in detail in Table 4.1.

4.2.2 Gamma-spectrometry

Most detectors used in gamma-spectrometrical analyses today are high-purity Ge detectors. Operating environments in various laboratories have become more uniform: personal communicators, more tool programmes for nuclide data (libraries), and high resolution graphics can all be used. Gamma measurement procedures are described in detail in Table 4.2.

4.2.3 Radiochemical determination of strontium-90

Classic nitric acid precipitation method and extraction methods have generally been used for the separation of strontium, although one laboratory used a new ion chromatography method (Sr-specific resin). In measuring systems, liquid scintillation equipment has been increasingly used, since modern liquid scintillators now have low background count rates and can also handle spectra easier and better than earlier. Procedures for the determination of strontium are described in detail in Table 4.3.

4.2.4 Radiochemical determination of transuranic elements

Likewise, methods for measuring plutonium and americium have not changed much in recent years. Anion exchange and extraction methods have been used separately or together to separate transuranic elements. In spectral analysis more commercial programmes have come into use. Alpha detectors have also improved to give better energy resolution and counting efficiency, while certain detectors are now cleanable. Procedures for the determination of transuranic elements are described in detail in Table 4.4.

4.3 Quality assurance, internal and external checking

Interest in quality has generally increased considerably during the period in question. Quality sys-

tems are more consistent and precise than previously. Accreditation has become an important issue, which is at least under discussion in all the Baltic laboratories. Some laboratories or methods have already been accredited, and in other laboratories the accreditation procedure is in progress. These discussions and procedures have increased the level of quality, transparency and traceability in general. Accreditation will remain an important issue in the future. It could also be

useful to focus more on sampling and the effects of sampling on repeatability and errors. Intercomparisons are discussed in the next part of this chapter.

Quality control procedures in various countries are described in detail in Table 4.5.

Table 4.1 Sampling and pre-treatment

	Sea water			Biota		Sediment	
	Sampling (performed by)	Pre-treatment for		Sampling (performed by)	Pre-treatment	Sampling device (performed by)	Pre-treatment
		Cs	Sr				
Denmark	With hose and pump (Danish Navy)	Adsorption on AMP in laboratory	Analysed after AMP adsorption from the same sample	Fish: Bought at fish markets or from fishermen. Seaweed: by hand from shore (Riso)	Drying or freeze drying, ashing at 450°C or wet ashing	-	-
Estonia	From surface with a pail (the same person every time)	Evaporation to 2.5 litres, taking 1 litre sub-samples	-	Fish: trawls (commercial fish catches) Seaweed: from the surface with a scraper (the same person every time)	Fish: cleaning to eatable parts, drying at 105°C, ashing at 450°C Other biota: drying at 105°C, ashing at 450°C	Kajak type corer with 10 cm diameter (the same person every time)	Slicing into 2 cm slices, drying at 105°C, homogenising
Finland	Submerged pump-hose system or a large water sampler (Finnish Institute of Marine Research)	Acidifying, adding of carriers, evaporation to 0.5 litres	Analysed after gamma measurement from the same sample	Fish: Fishnets (Research centres or local fishermen) Seaweed: by diving (STUK) Other biota: bait-nets, Ekman-Birge crab or by diving (STUK)	Fish: cleaning to eatable parts, drying at 105°C, ashing at 450°C Other biota: drying at 105°C, ashing at 450°C (Sr) or wet ashing (TU)	92-94: Aquarius Box-corer, 1haul, 95-98: Gemini Twin corer, 1 tube (Finnish Institute of Marine Research)	Slicing into 5 cm slices, freeze drying, homogenising
Germany	Surface water with fire pump from 4 m, 30 litre Rosette, 50 litre stainless steel (BSH)	Pre-filtration (5 µm), acidifying, adsorption to potassium-cobalt-ferrocyanate, drying at 60°C	Acidifying	Fish: bottom trawl (BFFG) Other biota: bottom trawls or with a dredge, Fucus from fisher boats (Federal State Institute in Rostock)	Fish: cleaning to fillets, also liver (cod), drying at 110°C, ashing at 420°C Other biota: Drying at 110°C, ashing at 420°C	Small box corer, Gemini - corer (BSH)	Slicing into 2 cm slices, freeze drying, homogenizing
IAEA/MEL	30 litre Rosette, 270 litre Gerard surface pump from 5 – 5000m (IAEA)	Pre-filtration 0.45 µm, acidifying, carriers, AMP.	Acidifying, carriers, oxalate precipitation	Fish trawling, plankton net. freezing (IAEA)	Lyophilization. homogenisation	Large box corer 750. 112mm plexiglas liners	X-Ray, slicing at mm -scale, lyophilization (IAEA)

Table 4.1 Sampling and pre-treatment (continued)

	Sea water			Biota		Sediment	
	Sampling (performed by)	Pre-treatment for		Sampling (performed by)	Pre-treatment	Sampling device (performed by)	Pre-treatment
		Cs	Sr				
Latvia	5 l van Dorn sampler (Hydroecological Institute of Latvian University)	Acidifying, adding carriers, evaporation to 1 litre	Acidifying, adding carriers, evaporation to 6 litres, oxalate precipitation	Fish: fishnets (Latvian Fisheries Research Institute) Seaweed: loose drifting plants by rake (Lielrīga Reg. Env. Board, Ecol. lab.)	Fish: cleaning to eatable parts, ashing at 420-450°C Other biota: air drying, drying at 105°C, homogenising	Kajak corer (Hydroecol. Institute of Latvian University)	Slicing into 2 cm slices, drying at 105°C, homogenising
Lithuania	Surface water with fire pump, 60 litres	Adding Cs and Sr carriers, co-precipitation of carbonate and $K_4Fe(CN)_6$	Co-precipitation of carbonate and $K_4Fe(CN)_6$	Fish: commercial catches Seaweed: by hand from shore	Fish: - homogenisation of fresh fish for gamma spectrum analysis - drying at 105-110°C, homogenising, ashing at 610°C for Sr separation Other biota: drying at 105-110°C, homogenising, ashing at 610°C	Grab sampler	Drying at 105°C and homogenising
Poland	Rosette (IMGW)	Adsorption on AMP on board	Precipitation with oxalic acid on board	Fish: research catches (Sea Fisheries Institute), and commercial catches Other biota: small bottom trawl (CLOR and IMGW)	Fish: cleaning to fillets, drying at 105°C, ashing at 450°C Other biota: drying at 105°C, ashing at 450°C	92-95: Sprut corer of diam. 8.8 cm; 96-98: Niemistö-corer (CLOR and IMGW)	Slicing into 1cm slices (0-5 cm) and 2 cm slices (5-19 cm), drying at 105°C, homogenising
Russia	Surface water by plastic vessel, large water sampler 30 litres (Khlopin)	Acidifying, adding of carriers on board, precipitation in lab.	Acidifying, adding of carriers on board	Fish: bought from a fisherman; Other biota: occasional sampling - some types of algae from Kopskaya Bay	Fish: cleaning to eatable parts, ashing at 450°C Other biota: air drying, drying at 105°C, ashing at 450°C, homogenising	SPRUT corer, (Khlopin)	Slicing into 2-5 cm slices in lab., drying at 100°C, homogenising
Sweden	No sampling	-	-	Fish: fishing nets (several staff and other external consultants) Other biota: fish trap, trawling, rake diving and hand-picking (several staff and other external consultants)	Fish: cleaning, drying at 80°C and ashing at 450°C Other biota: freeze drying or drying at 80°C, homogenising	Willner-corer (6.4 cm diam.), Willner-corer (7.0 cm), Niemistö-corer (5.0 cm), Kajak-corer (8.0 cm) (Umeå Marine Fisk. centre, Fiskeriverkets and other external consultants)	Slicing into 2 cm or 5 cm slices, drying at 80°C or freeze drying, ashing at 450°C, homogenising

Table 4.2 Gamma Spectrometry

	Equipment	Geometry	Analysis of spectra	Calibrations
Denmark	Ge-detectors, efficiencies 15-40%, resolution 2-3 keV, 4000 channels, energy range 0-3 MeV	Cylinder 10-200 ml, Marinelli 0.8-1 l, well detector 0-2 ml	PC, Riso software: - correction for densities and coincidence losses - calculation of analytical error - nuclide library (typically 10 nuclides used defined) - graphics	With mixed liquids for all geometries, low frequency, calibration error less than 5%, weekly energy control
Estonia	HP Ge-detector, efficiency 20 %, resolution 1.82 keV, 8000 channels, energy range 0.05-2.7 MeV	Cylinder 35 and 60 ml, Marinelli 0.5 and 1 l	Tennelec OXFORD soft.: - calc. anal. error - nuclide library (20 nucl.) -graphics	With mixed standard solutions, once a year, calibration error approx. 5%, energy control daily
Finland	HP Ge-detectors, efficiencies 20-100%, resolution 1.71-2.0 keV, (4000) 8000 channels, energy range (0.01) 0.04-2.7 MeV	Cylinder 0- 110 ml, Marinelli 0.5 l	PC, STUK software: - correction for height and density, and coincidence losses - nuclide library (103 nucl.) - calculation of analytical error and calibration of uncertainty - high resolution graphics	With mixed standard solutions, every 2-3 years, calibration error 4%, calculation of efficiency, transfer energy control/every measurement
Germany BFAFi	Ge and Ge(Li)-detectors, efficiency 15-41%, resolution 1.8-2.3 keV, 4000 channels, energy range 0.03-1.9 MeV	cylinders 0-50 ml and 0-200 ml	BFAFi (based on SAMPO80) software: - correction for self absorption and coincidence losses for some nuclides - nuclide library (55 nucl) - calculation of analytical error and calibration of uncertainty - high resolution graphics	92-94: with ash and sediment of diff. densities and several nuclides. 94-98: with mixed standard solutions, less than once a year, calibr. error 5 %, energy control every 1 or 2 weeks
BHS	Ge detectors, efficiency 18.5-44 %, resolution 1.78-1.88 keV, 8000 channels, energy range 0.045-2 MeV	Cylinder 0- 200 ml, Marinelli 1 l	Gammavision/Ortec soft: - correction for self adsorption - nuclide library (91 nucl) - calculation of analytical error - high resolution graphics	New detectors with mixed radionuclide solution, calibr. error 5 %, energy control weekly
IAEA/MEL	HP Ge-detectors, efficiencies 20-150 %, resolution 1.71-2.0 keV, (4000) 8000 channels, energy range (0.01) 0.04-2.7 MeV	Cylinder 10-200 ml, Marinelli 0.8-1 l, well detector 0.5 to 8 ml	Gammavision/Ortec soft: - corr. self adsorption - nuclide library (91 nucl) - calc. anal. error - high resolution graphics	With mixed (standards) liquids for all geometries, low frequency, calibration error less than 5%, monthly energy control

Table 4.2 Gamma Spectrometry (continued)

	Equipment	Geometry	Analysis of spectra	Calibrations
Latvia	Ge detectors, efficiency 12-25 %, resolution 1.7-2.7 keV, 4000/8000 channels, energy range 0.03-2.5 MeV	Cylinder 35 ml, Marinelli 0.5 and 1 l	Gammavision/Ortec or Canberra ASAP softw.: - corr. densities and coincidence losses - nuclide library (80 nucl) - calculation of analytical error	With mixed nuclide source and separate nuclides, once a year, calibration error 5%, energy control monthly
Lithuania	HP Ge detectors, efficiency 25 %, resolution 1.8 keV, 8000 channels, energy range 0.10-2.2 MeV	Marinelli 0.5 l Cylinders 10-50 ml	Tennelec Oxford software: - nuclide identification (20 nuclide library) - calculation of activities - analytical error estimation - high resolution graphics	- Efficiency calibration: with mixed standard solution, activity uncertainties 3.5% twice a year, calibration error 6%, - Energy control: every measurement - Energy calibration: irregularly (in cases of spectrum shift)
Poland	HP Ge detectors, efficiency 30 %, resolution 1.8 keV, 4000 channels, energy range 0.08-2.2 MeV	Cylinders 15-75 ml and 220 ml	Canberra 90+ software: Spectran AT with correction of height	With mixed standard solutions, 3 times/year, calibration error 5%, energy control 1/month
Russia	Ge(Li) detectors, efficiency 20-30 %, resolution 1.8-2.8 keV, 4000-8000 channels energy range 50-2500 MeV	Cylinder 0- 1 l	PC, RI dev. software: - correction for density, height and coincidence losses - nuclide library (100 n.) - high resolution graphics	With mixed standard solutions, twice a year, calibration error 2%, energy control/every measurement
Sweden	HP Ge detectors, efficiency 20-40 %, resolution 1.9 keV, 4000 channels energy range 50-2600 MeV	Marinelli 0.5 - 1 l, cylinder 35 - 100 ml	PC Genie-2000 software: - nuclide libraries (50 n.) calculation of analytical error and calibration uncertainty - high resolution graphics	With mixed nuclide source, low frequency, calibration error 5% energy and BG checking every day

Table 4.3 Determination of Strontium-90

	Separation method	Yield measurement	Equipment	Calibration
Denmark	Classic nitric acid method	⁹⁰ Sr: with ⁸⁵ Sr gamma ⁹⁰ Y: gravimetry	Low level beta counter DL: 0.004_Bq/sample	Occasionally with ⁹⁰ Sr standard solution, stability checked weekly with ref. source
Estonia	-	-	-	-
Finland	Ion chromatography method with Sr-specific resin	⁹⁰ Sr: stable carrier with AAS ⁹⁰ Y: by titration	Low level beta counter or low level liquid scintillation equip.(Wallac) DL: 0.002Bq/sample (beta counter) or 0.005 Bq/sample (liquid)	With ⁹⁰ Sr standard solution, 4 times/year, several times with each detector, stability checked with ref. source/each measurement
Germany BFAFi	Classic nitric acid or extraction method	⁹⁰ Sr: with ⁸⁵ Sr gamma ⁹⁰ Y: by titration	Low level beta counter DL: 0.008-0.012 Bq/sample	With ⁹⁰ Sr standard solution, once/year
BSH	Extraction method	⁹⁰ Sr: gravimetry ⁹⁰ Y: gravimetry	Low level beta counter DL: 0.015 Bq/sample	With ¹³⁷ Cs source, once a year
IAEA/MEL	Classic nitric acid method	⁹⁰ Sr:with ⁸⁵ Sr gamma ⁹⁰ Y: gravimetry	Low level beta counter DL: 0.002 Bq/sample	with ⁹⁰ Sr standard solution, stability checked weekly with ref. source
Latvia	Extraction method	⁹⁰ Y: by titration	Low level liquid scintillation equip. DL: 0.02 Bq/sample	With ⁹⁰ Sr standard solution, 2 times /year
Lithuania	Extraction of Y	⁹⁰ Y: gravimetry	Low level beta counter	With ⁹⁰ Sr standard solution once/year stability checking every mount with ref. source
Poland	Classic nitric acid method	⁹⁰ Sr: gravimetry ⁹⁰ Y: gravimetry	Low level beta counter DL: 0.008 Bq/sample	With ⁹⁰ Sr standard solution, once/year
Russia	Co-precipitation method (Ferrocyanide-carbonate precipitations)	⁹⁰ Sr: AAS ⁹⁰ Y: gravimetry	Low level liquid scintillation equipment (Wallac), Cherenkov-measurement. DL: 0.02 Bq/sample	With ⁹⁰ Sr standard solution, 4 times/year
Sweden	HDEHP-extraction method	⁹⁰ Y: titration	Low level liquid scintillation equipment (Wallac), DL: 0.001 Bq/sample	Occasionally with ⁹⁰ Sr standard solution, stability checked weekly with ref. source

Table 4.4 Determination of Transuranic Elements

	Separation of Pu	Separation of Am	Yield measurement	Equipment	Calibration
Denmark	Anion exchange method, electrode position	Co-precipitation with HDEHP, ion exchanges, electrode position	Internal tracers: ^{242}Pu and ^{243}Am	PIPS no-alpha detectors, multichannel analyser, spread sheet calculation DL: 0.2 mBq/sample	With standard solution sources occasionally
Estonia	–	–	–	–	–
Finland	Anion exchange method, electrode position	Co-precipitation with HDEHD, ion exchanges, electrode position	Internal tracers: ^{242}Pu , ^{243}Am	PIPS no-alpha detectors, Canberra AlphaAnalyser system DL: 0.2 mBq/sample	With mixed alpha nuclide source and ^{241}Am standard, once a year, pulser checking once a month
Germany BFAFi and BSH	Combined ion exchange/extraction method, electrode position	Combined ion exchange/extraction method, electrode position	Internal tracers: ^{242}Pu , ^{243}Am	Ruggedised surface barrier and PIPS detectors, multichannel analyser, self-made programme DL: 0.1-0.2 mBq/sample	With mixed alpha nuclide source and ^{241}Am standard, once a year, automatic energy calibration/every measurement
IAEA/MEL	Combined ion exchange/extraction method and electrode position	Co-precipitation with HDEHP, ion exchanges, electrode position	Internal tracers: ^{242}Pu , ^{243}Am	Surface barrier, multichannel Analyser. AlphaVision	Mixed alpha nuclide source and ^{241}Am standard twice a year. Energy calibration
Latvia	-	–	–	–	-
Lithuania	Not measured	–	–	–	–
Poland	Anion exchange method, electrode position	–	Internal tracer: ^{242}Pu	PIPS detectors, multichannel analyser, Canberra 90+ software, DL: 0.1 mBq/sample	With mixed alpha nuclide source, 6 times/year
Russia	Anion exchange method, electrode position	-	Internal tracer: ^{236}Pu or ^{242}Pu	Si surface barrier detectors, multichannel analyser, Canberra software, DL: 0.1 mBq/sample	With Pu standard isotopes, 5 times/year
Sweden	Anion exchange method, electrode position	–	Internal tracer: ^{242}Pu	IPC detector, multichannel analyser, Tennelec (DMR software), DL: 0.1 mBq/sample	With mixed alpha nuclide source, occasionally

Table 4.5 Quality control (QC)

	QC for sampling and pre-treatment	QC for gamma spectrometry	QC for strontium analysis	QC for transuranic analysis
Denmark	Participation in inter-comparisons	Participation in inter-comparisons and proficiency tests; long-term BG measurements: 2-3/year	Participation in inter-comparisons and proficiency tests; BG measurements with every sample measurement.	Participation in inter-comparisons and proficiency tests; long-term BG measurements: several times/year
Estonia	-	Participation in inter-comparisons and proficiency tests	-	-
Finland	Pre-treatment accredited, parallel samples occasionally for both sampling and pre-treatment, participation in inter-comparisons, when arranged	Accredited method, analysis of parallel and reference samples, participation in national and international Intercomparisons; long-term BG measurements: 2-3/ year	Accredited method, analysis of parallel and reference samples, participation in national and international intercomparisons and proficiency tests; analysis of reagent blanks; BG measurements: at least once a month	Accredited method, analysis of parallel and reference samples, participation in national and international intercomparisons and proficiency tests; analysis of reagent blanks; long-term BG measurements: 1-2/year
Germany BFAFi	Control of sample weight reducing factors, reduction factors for ashing in the analysis of inter-comparison samples by radiochemical methods	Control of each detector every two weeks; one reference sample/year for each detector; participation in national and international Intercomparisons; long-term BG measurements: once/ year	Control of each detector every week; participation in national and international Intercomparisons; BG measurements: once a month	Control of detection efficiencies; control of peak positions/each measurement, analysis of reagent blanks for approx 10% of measurements; participation in national and international Intercomparisons; long-term BG measurements: 1-2/year
BSH	Parallel samples occasionally for both sampling and pre-treatment; participation in inter-comparisons, when arranged	Accredited method; analysis of parallel and reference samples; participation in national and international Intercomparisons; long-term BG measurements: 2-3/ year	Analysis of parallel and reference samples; participation in national and international intercomparisons and proficiency tests; analysis of reagent blanks; BG measurements: twice a month, efficiency check once/ week	Analysis of parallel and reference samples, participation in national and international intercomparisons and proficiency tests; analysis of reagent blanks; long-term BG measurements: 1-2/year
IAEA/MEL				
Latvia	Water sampling accredited; laboratories accredited	Accredited methods; periodical analysis of reference and parallel samples; participation in national and international Intercomparisons; long-term Bg measurements: 4 times/year	Accredited method; analysis of parallel and reference samples; participation in international inter-comparisons and proficiency tests; BG measurements: 2/ year	-

Table 4.5 Quality control (QC) (continued)

	QC for sampling and pre-treatment	QC for gamma spectrometry	QC for strontium analysis	QC for transuranic analysis
Lithuania	–	Participation in national and international intercomparisons	Participation in national and international intercomparisons	–
Poland	Parallel samples for pre-treatment occasionally	Analysis of parallel and reference samples; participation in intercomparisons; long-term BG measurements: 3 times/year	Analysis of parallel and reference samples; participation in intercomparisons; BG measurements with every sample measurement	Analysis of parallel and reference samples; analysis of reagent blanks; participation in intercomparisons; long-term BG measurements: 3 times/year
Russia	Accredited laboratory; certified pre-treatment methods; parallel samples	Certified gamma measurement method, analysis of reference samples 3-4 times/year; analysis of parallel samples; participation in intercomparisons; BG checking: monthly	Certified method, analysis of reagent blanks and reference samples; participation in intercomparisons and proficiency tests; BG measurements monthly	Certified method; analysis of reagent blanks and reference samples; participation in intercomparisons; BG measurements monthly
Sweden	-	Participation in national and international intercomparisons; long-term BG measurements: 2-3/year; QA-control every day	Participation in national and international intercomparisons; BG measurements every week	Participation in national and international intercomparisons; calibration occasionally

4.4 Intercalibration exercises (1992-1998)

The International Atomic Energy Agency (IAEA) has conducted seven intercomparison exercises involving the MORS group under its continuing analytical quality control programme. Most of these exercises have been especially designed for the Baltic Sea Members working in the MORS Program under the umbrella of the Helsinki Commission. These samples consisted of sediment and sea water collected in the Baltic Sea. Irish Sea fish flesh was used for a biota sample, and one exercise was carried out on standard solutions of ^{137}Cs and ^{90}Sr with a known activity.

Twelve laboratories from the 9 Baltic Sea coastal countries and the IAEA's Monaco Laboratory participated in at least some of these intercalibration exercises. The results of these intercomparisons were very satisfactory.

These exercises were designed to offer the laboratories the chance to check their analytical results, to discover and eliminate possible systematic errors, and to give the data a common basis in order to ensure overall comparability. It was agreed from the outset that analytical quality control would be absolutely necessary for these studies in order to take full advantage of the data accumulated through the concerted efforts of all participants over the period 1992-1998.

4.4.1 List of exercises

During the period 1992-1998, 7 intercomparison exercises were organised by the IAEA. The characteristics of the different exercises are presented in Table 4.6. With the exception of IAEA 381 (on water from the Irish Sea) and IAEA 382 (on the flesh of fish from the Irish Sea), all the exercises were designed for the Baltic Sea. There were three intercomparisons on Baltic Marine Sediment (IAEA 300, 378 and 379) and two on Baltic Sea water samples (IAEA 337 and 299). It was agreed that participants would measure ^{90}Sr and ^{134}Cs and ^{137}Cs isotopes over a period of 5 years (IAEA 299), with each laboratory requested to analyse the same sample every year. The participant laboratories were also provided each year with a set of standard solutions of ^{90}Sr and ^{137}Cs in order to check their equipment and chemical procedures.

A complete list of the laboratories that participated in one or more of the IAEA-MEL intercomparison exercises organised during this period is given in Table 4.7.

4.4.2 Data treatment

All the data submitted by the Baltic Sea coastal countries over the period 1992-1998 has been incorporated in the data compilation of the various exercises.

Data was selected from individual reports with minor alterations, mainly involving the removal of excessive digits, the standardising of activity units and the matching of activities to reference dates. Laboratory averages were calculated as necessary from individual results and then given as arithmetical means, with margins of error. The principles and application of the statistical programme were discussed in a previous report.

Identification of outlying data

One of the main objectives of the intercalibration was to check if sets of data produced within the Baltic group are mutually consistent, and can thus be regarded as belonging to the same population. Several criteria have been proposed for the rejection of suspected values. However, none of these criteria are fully satisfactory, because of several assumptions that must be taken "a priori". A non-parametric procedure developed by Veglia (1981) and modified by Pszonicki et al. (1983) has been in use for some years. But box and whisker plots with the option for determining outliers have recently been adopted. Although the latter is not a statistical test, it does however define items of data as outliers based on the upper and lower range of the mean + 1 standard error or the 75th percentile times an outlier coefficient, normally set up to 1.5. Outliers have also been checked according to Cochran's test (ISO5735-2:1994). This statistical test depends on the ratio of variance of a submitted value, and the total variance of the results reported. This latter method has corroborated what the whisker plots recognised as outliers in only 25% of all cases. The 95% Confidence Interval was calculated with 1 sigma of the normally distributed set of data. (Sokal and Rohlf, 1969)

Some of the participating laboratories had submitted more radionuclides than requested for the exercise. Although this extra data is presented here (Tables 4.14 - 4.20), only the data on ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ (Tables 4.8 - 4.13 and 4.22 - 4.28) will be fully discussed. These isotopes represent alpha, beta and gamma emitting radionuclides, and are also considered as the most relevant isotopes in terms of doses to man.

Evaluation of the data

A total of 12 laboratories participated in these exercises. Data was reported on 9 artificial and 16 natural radionuclides. Data on ^{90}Sr , ^{137}Cs and

$^{239,240}\text{Pu}$, as well as less frequent radionuclides measured reported by the laboratories of the MORS Group during the period of 1992-1998, is presented in Tables 4.8 to 4.30.

The analytical methods used by the participants are:

Alpha counting

Code

- A Unspecified
- A1 Leaching, spontaneous deposition on Ag, Cu, Ni, Au discs
- A2 Ashing, leaching, precipitation, ion exchange, electrode position
- A5 Ashing, leaching, precipitation, ion exchange, liquid-liquid extraction and electrode position

Beta Counting

Code

B Unspecified

- B7 Oxalate precipitation, carbonate, separation with fuming or conc. nitric acid, scavenging of Ra and Fe, 2 weeks ingrowth period, precipitation (hydroxide, oxalate, carbonate), beta counting of ^{90}Y as Y_2O_3 or liquid scintillation

Gamma-spectrometry

Code

G Unspecified

- G1 High resolution (Ge detectors), direct
- G2 High resolution (Ge detectors), indirect from daughters in equilibrium

Other methods

Code

- Em Radon Emanation
- ICPMS Inductively Coupled Plasma Mass Spectrometry

For data sets comprising more than 4 accepted values, medians, ranges and 95% Confidence Intervals (CI) were calculated as estimations of true activity concentrations.

A great variety of radio-isotopes have been measured and reported for IAEA 300, IAEA 378 and IAEA 379. The artificial radionuclides reported were: ^{54}Mn ^{125}Sb ^{90}Sr ^{137}Cs ^{134}Cs ^{238}Pu $^{239,240}\text{Pu}$ and ^{241}Am .

^{90}Sr

Seven laboratories submitted data for ^{90}Sr (Tables 4.8, 4.10 and 4.12). 90% of the laborato-

ries used method code B7. In spite of the large scatter in IAEA 300, (1.7 to 32.7 Bq kg^{-1}) the median value obtained was 10.85 Bq kg^{-1} . This value is very similar to the median value obtained in the world wide intercomparison exercise of this sample, that is, 10.8 Bq kg^{-1} with a range of accepted values of 1.7 to 16 Bq kg^{-1} , inhomogeneity may be the cause of this wide scattering.

In the data sets for IAEA 378 and IAEA 379, only one result was rejected as being one order of magnitude higher than any other result reported for this radio-isotope in these samples.

The strontium values reported for IAEA 299 all statistically belonged to a single population of data points (Table 4.22 and Figure 4.1) ANOVA was applied to test the hypothesis that the mean values generated from 1992-1996 by each laboratory were statistically the same. The 5% significance level could not reject this hypothesis, so the mean values were all defined as statistically not different from each other.

For the exercises IAEA 381 and IAEA 337, data was reported by 7 and 6 laboratories, respectively (Table 4.26). No outliers were found, and most of the values fall within the 95% confidence interval.

^{137}Cs and ^{134}Cs

All the results reported were measured by gamma-spectrometry. Only two values were rejected (Table 4.10 and 4.12). The lowest values were found in IAEA 378 (5.7 to 6.2 Bq kg^{-1}), and the highest values in IAEA 300, reaching up to 1081 Bq kg^{-1} .

In IAEA 299 most of the laboratories were very consistent over the whole period, where ^{137}Cs measurements were concerned (Table 4.23). One outlier was detected in 1995, and two in 1996. ANOVA testing found differences among the mean values reported by the laboratories. The HDS-Tukey *a posteriori* test found that Khlopin's mean value was statistically different from the means reported by STUK and BSH.

In conjunction with the IAEA 299 exercise, a set of ampoules was distributed to each participant laboratory with a known concentration of ^{137}Cs . The results for the 1992-1996 period are shown in Table 4.29 and Figure 4.3.

For exercises IAEA 381 and IAEA 337, data was reported by 8 and 9 laboratories, respectively (Table 4.27). Only one outlier was found in IAEA 337, but certain laboratories consistently showed higher values than others.

^{238}Pu and $^{239,240}\text{Pu}$

Only two data points were rejected as outliers. One of these values (for ^{238}Pu) falls well within the accepted range for this isotope in the world-wide intercomparison exercise. It can therefore be concluded that all the plutonium data reported by the MORS group is comparable to data reported in the world-wide exercise IAEA 300.

In IAEA 381, (6 laboratories; Table 4.28) the reported data is generally highly homogeneous; although one laboratory had consistently higher values for both ^{238}Pu and $^{239,240}\text{Pu}$ than the other laboratories.

^{241}Am

The data sets reported on Americium show a similar pattern to the data on plutonium isotopes. The values considered as outliers fall within the accepted range for this isotope in the world-wide intercomparison exercise; thus the values reported by MORS group can be considered as satisfactory.

In IAEA 381, only six results are reported here. In general, the values are in good agreement and no outliers were found (Table 4.28).

Natural radio-isotopes

The natural radionuclides reported were: ^{40}K , ^{210}Po , ^{210}Pb , ^{214}Pb , ^{223}Ra , ^{224}Ra , ^{226}Ra , ^{227}Th , ^{228}Ac , ^{227}Th , ^{228}Th , ^{232}Th , ^{234}Th , ^{234}U , ^{235}U and ^{238}U . The most frequent radio-isotopes reported are shown in Tables 4.15-4.17 and the less frequent ones are shown in Tables 4.18-4.21.

^{40}K

All the reported results were obtained by gamma-spectrometry. Only three outliers were found – two in IAEA 300 and one in IAEA 378. The outliers found in IAEA 300 fall within the accepted range for this radio-isotope in the world-wide intercomparison exercise. Here too, the values reported by the MORS group are in good agreement with those reported in the world-wide exercise.

^{210}Pb

All the reported results were determined by gamma-spectrometry with the exception of one determined by ingrowth of ^{210}Po by alpha-spectrometry. The limited quantity of data obtained did not allow any statistical evaluation.

Radium isotopes

Seven laboratories reported data on radium, and in most cases the isotope was measured by gamma-spectrometry. Only one data point was

measured by the ^{222}Rn emanation technique. Both methods are in good agreement. Only one outlier was found in the data set for IAEA 378.

Less frequently reported isotopes

From the limited amount of data reported on ^{210}Po and ^{210}Pb for IAEA 300, IAEA 378 and IAEA 379, it can be observed that these radionuclides are in secular equilibrium. The other radionuclides are scarce, and no conclusions could be drawn on them.

Only one exercise has been carried out on a biota sample, namely IAEA 382 on Irish Sea fish flesh. Too little data was reported for any serious conclusions to be drawn. The data presented here (Tables 4.28 and 4.29) nevertheless shows some scattering of values, particularly for ^{137}Cs and $^{239,240}\text{Pu}$ isotopes. This may be the result of heterogeneity within the sample. The IAEA-MEL has run several homogeneity tests with a wide range of values for $^{239,240}\text{Pu}$. This exercise may be run in the future again.

^{54}Mn , ^{60}Co and ^{125}Sb

Results were reported only in IAEA 300. All measurements were obtained by gamma-spectrometry. In general the values submitted are within the accepted ranges found in the world-wide exercise for this sample (Table 4.14) with the exception of the reported values for ^{125}Sb (world-wide range: 5.2 to 16.5 Bq kg⁻¹). In this last case, two values fall outside the 95% CI calculated from the data presented here. The ranges found in the world-wide intercomparison for Mn and Co were 1 to 3.2 Bq kg⁻¹ for ^{54}Mn , and 0.6 to 2.6 Bq kg⁻¹ for ^{60}Co .

4.4.3 Discussion

Intercomparison exercises are among the most effective elements of quality assurance programmes. The median concentrations for each set of data - after the rejection of outliers - were chosen as the most reliable estimates for the true values. A summary of the recommended values is reported together with ranges of accepted laboratory values for IAEA 300, IAEA 378 and IAEA 379 in Table 4.21.

The methods used to analyse the different radionuclides by the various members of the MORS group are quite comparable, based on the statistical evaluation of the results obtained and reported here. It is also important to note that the results reported for IAEA 300 by the MORS group are in very good agreement with the statistical evaluation in the world-wide intercomparison exercise on this particular sample.

Table 4.6 Intercomparison exercises on marine environmental samples carried out by the MORS Group during the period 1992-1998.

Sample code	Sample type	Source	Year of collection	Year of exercise	No. of Baltic participants
IAEA 300	Sediment	Baltic Sea	1992	1993	12
IAEA 378	Sediment	Baltic Sea	1992	1993	12
IAEA 379	Sediment	Baltic Sea	1991	1993	12
IAEA 299	Sea water	Baltic Sea	1991	1992-1996	9
IAEA 337	Sea water	Baltic Sea	1996	1997-present	9
IAEA-381	Sea water	Irish Sea	1986	1998	9
IAEA-382	fish flesh	Irish Sea	1996	1997	5

Table 4.7 List of laboratories participating in the IAEA-MEL Intercomparison Programme during the period 1992-1998.

Main Investigator	Laboratory
Nielsen (DK)	RISØ Risø National Laboratory
Ikäheimonen/Klemola/Ilus (FIN)	STUK Radiation and Nuclear Safety Authority
Kanisch (DE)	BFAFi Federal Fisheries Research Institute
Herrmann (DE)	BSH Federal Maritime and Hydrographic Agency
Bojanowski (PL)	PAN, Sopot Institute of Oceanology
Tomczak(PL)	IMWM Institute for Meteorology and Water Management
Grzybowska/Suplinska (PL)	CLRP Central Laboratory for Radiological Protection
Panteleev (RU)	Khlopin V.G. Khlopin Radium Institute
Melin (S)	NIRP National Institute of Radiation Protection
Neumann (S)	NSEPB National Swedish Environmental Protection Board
Sandell (S)	Studsvik Nuclear AB
Skujina(LV)	LREB Liel-Riga Regional Environmental Board
Motiejunas (LT)	EPDRL, Mins.Envirion.
Jakobson (EE)	ERPC Estonian Radiation Protection Agency
Putnik(EE)	EMHI Estonian Meteorological and Hydrological Institute
Suomela (S)	SSI Swedish Radiation Protection Institute
IAEA	International Atomic Energy Agency, Monaco

Table 4.8 Measurements of ^{90}Sr , ^{134}Cs and ^{137}Cs in sediments, taken by the Baltic MORS group 1992-1998; intercomparison exercises IAEA 300 (reference date: January 1st 1993; unit: Bq kg^{-1} dry weight)

Laboratory	Country	Samples	Method	^{90}Sr	^{134}Cs	^{137}Cs
RISØ	DK	4	B7	5.9 ± 0.7		
RISØ	DK	6	G1		63 ± 4	940 ± 56
SUAS	S	1	G1		74 ± 1	1081 ± 3
EMTI	EE	1	G1		57 ± 3.6	1075 ± 43
STUK	FIN	2	G1		69.0 ± 3.5	1000 ± 50
BFAFi	DE	4	B7	10.3 ± 2.4		
BFAFi	DE	5	G1		73 ± 2	1053 ± 53
BSH	DE	2	G1		69.1 ± 1.8	1101 ± 32
PAN	PL	3	B7	27.3 ± 6.7		
PAN	PL	1	G1		69 ± 3	1080 ± 32
CLRP	PL	2	G1		73.3 ± 10.9	1097 ± 142
IMWM	PL	4	G1		63 ± 10	1034 ± 23
Khlopin	RU	10	G1		66.7 ± 6.1	1076 ± 48
Lund	S	1	B	1.7 ± 0.2		
Lund	S	1	G1		60 ± 3	970 ± 40
Envt. Min.	LT	1	B	32.7		
Envt. Min.	LT		G1			1013
IAEA-MEL	MC	11	B7	11.4 ± 3.2		
IAEA-MEL	MC	7	G1		68.9 ± 1.4	1048 ± 50
Median				10.85	69	1053
AQCS values				1.7 – 32.7	57 - 94	940 - 1101
Confidence Interval				1.7 – 32.7	63 - 73	1000 - 1081

Laboratory	Country	Samples	Method	²³⁸ Pu	^{239,240} Pu	²⁴¹ Am
RISØ	DK	2	A2	0.14 ± 0.03	3.09 ± 0.19	
RISØ	DK	2	A5			1.14 ± 0.07
STUK	FIN	4	A2	0.15 ± 0.04	3.5 ± 0.35	1.2 ± 0.18
BSH	DE	4	A5	0.15 ± 0.03	3.55 ± 0.1	1.4 ± 0.03
BFAFi	DE	4	A5	0.14 ± 0.005	3.44 ± 0.12	
CLRP	PL	3	A2		3.2 ± 0.4	
Khlopin	RU	4	A2	<0.2	3.9 ± 0.5	
Lund	SW	1	A2	0.17 ± 0.02*	3.1 ± 0.2	
Lund	SW	1	A2			0.9 ± 0.1
IAEA-MEL	MC	14	A2	0.137 ± 0.02	3.41 ± 0.15	
IAEA-MEL	MC	13	A5		0.8 ± 0.4	1.27 ± 0.08
Median				0.14	3.43	1.23
Range accepted				0.137 – 0.15	3.09 – 3.9	1.14 – 1.4
Confidence Interval						

Table 4.9
Measurements of ²³⁸Pu, ^{239,240}Pu and ²⁴¹Am in sediments, taken by the Baltic MORS group 1992-1998; intercomparison exercises IAEA 300; unit: Bq kg⁻¹

*result rejected

Laboratory	Country	Samples	Method	⁹⁰ Sr	¹³⁴ Cs	¹³⁷ Cs
RISØ	DK	2	B7	0.42 ± 0.05		
SUAS	S					4.6 ± 0.2
EMHI	EE					6.1 ± 1.0
RISØ	DK	6	G1		<0.5	5.9 ± 0.4
STUK	FIN	2	B7	0.47 ± 0.1		
STUK	FIN	2	G1			6.2 ± 0.6
BSH	DE	2	G1			6.5 ± 0.2
PAN	PL	1	B7	0.6 ± 0.2		
PAN	PL	1	G1			3.2 ± 0.3
CLRP	PL	4	G1		<0.4	5.4 ± 0.5
IMWM	PL	2	G1		2.2 ± 4.0	5.7 ± 6.0
Khlopin	RU	6	G1			4.3 ± 0.8
Lund	S	1	G1			4.6 ± 0.4
Min. Environ	LT	1	B	5.3*		
Min. Environ	LT	1	G			9.7*
IAEA-MEL	MC	4	B7	0.26 ± 0.05		
IAEA-MEL	MC	3	G1		<0.5	5.7 ± 0.5
Median				0.45		5.7
Range accepted				0.26 – 0.6		3.2 – 6.5
Conf. Interval						4.3 – 6.2

Table 4.10
Measurements of ⁹⁰Sr, ¹³⁴Cs and ¹³⁷Cs in sediments, taken by the Baltic MORS group 1992-1998; intercomparison exercises IAEA 378; unit: Bq kg⁻¹

*result rejected

Table 4.11
Measurements
of ^{238}Pu , $^{239,240}\text{Pu}$
and ^{241}Am in
sediments, taken
by the Baltic
MORS group;
intercomparison
exercise IAEA
378; unit: Bq kg^{-1} .

Laboratory	Country	Samples	Method	^{238}Pu	$^{239,240}\text{Pu}$	^{241}Am
RISØ	DK	4	A2	<0.1	0.11 ± 0.01	0.037 ± 0.007
STUK	FIN	2	A2	<0.03	0.13 ± 0.01	
BSH	DE	4	A5	0.013 ± 0.004	0.15 ± 0.01	0.09 ± 0.02
CLRP	PL	3	A2		0.12 ± 0.02	
Khlopin	RU	2	A2	<0.2	$0.21 \pm 0.06^*$	
Lund	S	2	A2		0.14 ± 0.04	
Lund	S	2	A2			0.04 ± 0.01
IAEA-MEL	MC	14	A2	<0.03	0.14 ± 0.04	
IAEA-MEL	MC	13	A5			0.033 ± 0.02
Median					0.135	0.039
Range accepted					0.11 – 0.15	0.033 – 0.09
Confidence Interval					0.11 – 0.15	

*result rejected

Table 4.12
Measurements of
 ^{90}Sr , ^{134}Cs and
 ^{137}Cs in sedi-
ments, taken by
the Baltic MORS
group 1992-1998;
intercomparison
exercises IAEA
379; unit: Bq kg^{-1}

Laboratory	Country	Samples	Method	^{90}Sr	^{134}Cs	^{137}Cs
RISØ	DK	2	B7	0.5 ± 0.04		
RISØ	DK	4	G1		<0.5	39 ± 2
SUAS	S	1	G1		1.6 ± 1	45 ± 1
EMHI	EE	1	G1			44 ± 1.8
STUK	FIN	2	B7	1.2 ± 0.24		
STUK	FIN	2	G1			38 ± 3
BSH	DE	4	G1			44 ± 0.8
PAN	PL	1	B7	1.85 ± 0.05		
PAN	PL	1	G1		0.45 ± 0.12	39 ± 2.5
CLRP	PL	4	G1		<0.4	37.9 ± 3.0
IMWM	PL	2	G1		1.7 ± 4.0	38 ± 8.0
Khlopin	RU	6	G1		$3.8 \pm 0.7^*$	43.5 ± 1.6
Lund	S	1	G1			39 ± 2.0
Min. Environ	LT	1	B	12.7*		
Min. Environ	LT	1	G			40
IAEA-MEL	MC	4	B7	2.0 ± 0.2		
IAEA-MEL	MC	3	G1		0.8 ± 0.4	41.4 ± 3.0
Median				1.53	1.2	39.5
Range accepted				0.5 - 2	0.45 – 1.7	37.9 - 45
Confidence Interval						38 - 44

*result rejected

Laboratory	Country	Samples	Method	²³⁸ Pu	^{239,240} Pu	²⁴¹ Am
RISØ	DK	4	A2	0.1 ± 0.01	3.2 ± 0.2	
RISØ	DK	5	A5			1.17 ± 0.07
STUK	FI	2	A2	0.15 ± 0.04	3.7 ± 0.36	
BSH	DE	4	A5	0.12 ± 0.01	3.8 ± 0.2	1.6 ± 0.12
CLRP	PL	3	A2		3.6 ± 0.5	
Khlopin	RU	2	A2	<0.2	4.1 ± 0.4	
Lund	S	2	A2	0.11 ± 0.02	3.8 ± 0.2	
Lund	S	2	A2			1.9 ± 0.1
IAEA-MEL	MC	8	A2	0.12 ± 0.04	4 ± 0.3	
AQCS values						
Median				0.12	3.8	1.6
Range accepted				0.1 – 0.15	3.1 – 4.1	1.17 – 1.9
Confidence Interval				0.1 - 0.15	3.1 – 4.1	

Table 4.13 Measurements of ²³⁸Pu, ^{239,240}Pu and ²⁴¹Am in sediments, taken by the Baltic MORS group; inter-comparison exercise IAEA 379; unit: Bq kg⁻¹.

Laboratory	Country	Samples	Method	⁵⁴ Mn	⁶⁰ Co	¹²⁵ Sb
STUK	FI	2	G1			8.9 ± 2.4
BSH	DE	4	G1		1.5 ± 0.3	15.2 ± 2.8
BFAFi	DE	5	G1		0.74 ± 0.09	10.1 ± 0.5
PAN	PL	1	G1			15.7 ± 0.9
Lund	SW	2	G1	1.0 ± 0.3	0.8 ± 0.3	
IAEA-MEL	MC	8	G1	1.5 ± 0.5	1.3 ± 0.3	18 ± 6
Median					1.05	15.2
Range accepted					0.74 – 1.5	10.1 - 18
Confidence Interval					0.72 – 1.45	10.7 – 16.5

Table 4.14 Measurements of ⁵⁴Mn, ⁶⁰Co and ¹²⁵Sb in sediments, taken by the Baltic MORS group; intercomparison exercises IAEA 300 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Table 4.15
Measurements of ^{226}Ra , ^{210}Pb and ^{40}K in sediments, taken by the Baltic MORS group; intercomparison exercises IAEA 300 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Laboratory	Country	Samples	Method	^{226}Ra	^{210}Pb	^{40}K
RISØ	DK	6	G	53 ± 3	350 ± 21	960 ± 58
STUK	FI	2	G1			990 ± 50
EMHI	EE	1	G1			973 ± 58
SUAS	S	1	G1			1040 ± 12
BSH	DE	7	G2	60.0 ± 4.7		
BSH	DE	1	G1		352 ± 52	1032 ± 35
BFAFi	DE	5	G1	86.8 ± 17.4		1005 ± 51
PAN	PL	1	A1		400 ± 18	
PAN	PL	1	G1			1150 ± 51*
CLRP	PL	1	Em	95.7 ± 12		
CLRP	PL	2	G1			1064 ± 159
CLRP	PL	4	G	43 ± 16		1007 ± 158
Lund	S	1	G1			985 ± 45
Khlopin	RU	2	A2	41.3 ± 2.5		820 ± 190*
IAEA-MEL	MC	8	G2	58.8 ± 2.9		
IAEA-MEL	MC	3-8	G2		397 ± 45	1043 ± 9
AQCS values						
Median				58.8	375	1006
Range accepted				41.3 – 95.7	350 – 400	960 - 1064
Confidence Interval				47.1 – 78.2	347.8 – 401.7	987 - 1034

*result rejected

Table 4.16
Measurements of ^{226}Ra , ^{210}Pb and ^{40}K in sediments, taken by the Baltic MORS group; intercomparison exercises IAEA 378 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Laboratory	Country	Samples	Method	^{226}Ra	^{210}Pb	^{40}K
RISØ	DK	6	G	120 ± 6	185 ± 13	1040 ± 52
SUAS	S	1	G1			981 ± 10
EMHI	EE	1	G1			959 ± 48
STUK	FI	2	G1			890 ± 50
BSH	DE	4	G2	125.5 ± 7.3		
BSH	DE	2	G1		153 ± 17	980 ± 33
PAN	PL	1	A1		190 ± 10	
PAN	PL	1	G1			586 ± 22*
CLRP	PL	1	G	114 ± 14		
CLRP	PL	2	Em	132 ± 15		962 ± 66
CLRP	PL	4	G			855 ± 140
Lund	SW	1	G1			938 ± 62
Khlopin	RU	2	A2	103.2 ± 3.3		894 ± 36
IAEA-MEL	MC	3	G1		191 ± 38	961 ± 16
IAEA-MEL	MC	6	G2	116 8		
AQCS values						
Median				118	187.5	937
Range accepted				103.2 – 132	153 – 191	855 - 1040
Confidence Interval				111 – 125	166 – 193	893 – 981

Laboratory	Country	Samples	Method	²²⁶ Ra	²¹⁰ Pb	⁴⁰ K
RISØ	DK	6	G	26 ± 1	228 ± 16	790 ± 40
SUAS	S	1	G1			790 ± 17
EMTI	EE	1	G1			727 ± 43
STUK	FIN	2	G1			727 ± 43
BSH	DE	5	G2	29 ± 2.6		
BSH	DE	2	G1		203.9 ± 3.7	757 ± 28
PAN	PL	1	A1		240 ± 20	
PAN	PL	1	G1			692 ± 43
CLRP	PL	1	G	40.3 ± 4.8		
CLRP	PL	2	Em	40.6 ± 5.6		749 ± 53
IMWH	PL	4	G	48 ± 20		795 ± 80
Lund	S	1	G1			713 ± 47
Khlopin	RU	2	A2	22.1 ± 1.6		636 ± 45
EMHI	EE					727 ± 43
SUAS	S					790 ± 17
IAEA-MEL	MC	3	G1	28.1 ± 3		
IAEA-MEL	MC	6	G2		230 ± 12	751 ± 40
AQCS values						
Median				29	229	751
Range accepted				22.1 - 48	203.9 - 240	690 - 795
Confidence Interval				26.4 – 40.5	214.1 – 236.8	697 - 819

Table 4.17 Measurements of ²²⁶Ra, ²¹⁰Pb and ⁴⁰K in sediments, taken by the Baltic MORS group. Intercomparison exercises IAEA 379 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Table 4.18 Measurements of less frequent radionuclides reported by the MORS group; intercomparison exercise IAEA 300 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Laboratory	Country	Samples	Method	Isotope	Activity
RISØ	DK	3	A1	²¹⁰ Po	350 ± 21
IAEA-MEL	MC	5	A1	²¹⁰ Po	361 ± 11
BFAFi	DE	5	G1	²¹⁴ Pb	58.5 ± 0.6
PAN	PL	1	G2	²²⁴ Ra	66.0 ± 3.7
BFAFi	DE	5	G	²²⁷ Th	4.9 ± 0.7
BSH	DE	7	G	²²⁸ Ac	66.9 ± 3.3
BSH	DE	7	G2	²²⁸ Ac	64.0 ± 4.3
BFAFi	DE	5	G2	²²⁸ Ac	59.5 ± 3.0
RISØ	DK	6	G	²²⁸ Ra	64 ± 4
Khlopin	RU	10	G	²²⁸ Ra	56 ± 12
IAEA-MEL	MC	3	G2	²²⁸ Ra	62.3 ± 15
PAN	PL	1	G2	²²⁸ Th	64 ± 4.1
Khlopin	RU	10	G	²²⁸ Th	51.3 ± 5.2
BFAFi	DE	5	G2	²²⁸ Th	64.3 ± 3.0
IAEA-MEL	MC	3	ICPMS	²³² Th	68.9 ± 2.6
BFAFi	DE	5	G2	²³² Th	64.3 ± 3.0
BSH	DE	6	G	²³⁴ Th	65.2 ± 4.7
IAEA-MEL	MC	7	A2	²³⁴ U	61.7 5.3
BSH	DE	7	G	²³⁵ U	4.7 1.2
IAEA-MEL	MC	7	A2	²³⁵ U	2.4 0.2
PAN	PL	1	G2	²³⁸ U	99 4.9
IAEA-MEL	MC	3	ICPMS	²³⁸ U	68.7 0.9
IAEA-MEL	MC	7	A2	²³⁸ U	58.5 5.7
BFAFi	DE	5	G2	²³⁸ U	66.1 13.2

Table 4.19 Measurements of less frequent radionuclides reported by the MORS group for the intercomparison exercise IAEA 378 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

Laboratory	Country	Samples	Method	Isotope	Activity
RISØ	DK	2	A1	²¹⁰ Po	188 ± 13
PAN	PL	1	G	²²⁴ Ra	33 ± 2.7
BSH	DE	4	G	²²⁸ Ac	55.7 ± 1.8
BSH	DE	4	G2	²²⁸ Ac	55 ± 1.9
RISØ	DK	6	G	²²⁸ Ra	54 ± 3
Khlopin	RU	6	G	²²⁸ Ra	43.5 ± 4.1
IAEA-MEL	MC	6	G2	²²⁸ Ra	55.2 ± 2.2
PAN	PL	1	G	²²⁸ Th	31 ± 2
Khlopin	RU	6	G	²²⁸ Th	52.1 ± 3.7
IAEA-MEL	MC	6	G2	²²⁸ Th	55.7 ± 15.9
BSH	DE	4	G	²³⁴ Th	75.2 ± 6.2
BSH	DE	3	G	²³⁵ U	5.3 ± 1.6
PAN	PL	1	G	²³⁵ U	3.6 ± 1.2
PAN	PL	1	G	²³⁸ U	72 ± 2.2

Laboratory	Country	Samples	Method	Isotope	Activity
RISØ	DK	2	A1	²¹⁰ Po	218 ± 15
Lund	S	1	G1	⁵⁴ Mn	0.7 ± 0.1
PAN	PL	1	G	²²³ Ra	2.3 ± 0.4
PAN	PL	1	G	²²⁴ Ra	39 ± 3.3
BSH	DE	4	G	²²⁸ Ac	46.1 ± 3.8
BSH	DE	4	G2	²²⁸ Ac	44 ± 1.9
RISØ	DK	6	G	²²⁸ Ra	42 ± 2
Khlopin	RU	6	G	²²⁸ Ra	47 ± 7
IAEA-MEL	MC	6	G2	²²⁸ Ra	41 ± 4
PAN	PL	1	G	²²⁸ Th	39 ± 2.5
Khlopin	RU	6	G	²²⁸ Th	43 ± 1.8
IAEA-MEL	MC	6	G2	²²⁸ Th	43.6 ± 3.6
BSH	DE	4	G	²³⁴ Th	47.9 ± 3.4
BSH	DE	3	G	²³⁵ U	3.4 ± 1
PAN	PL	1	G	²³⁵ U	1.6 ± 1.1
PAN	PL	1	G	²³⁸ U	64 ± 3.6

Table 4.20 Measurements of less frequent radionuclides reported by the MORS group for the intercomparison exercise IAEA 379 (reference date: January 1st 1993; unit: Bq kg⁻¹ dry weight).

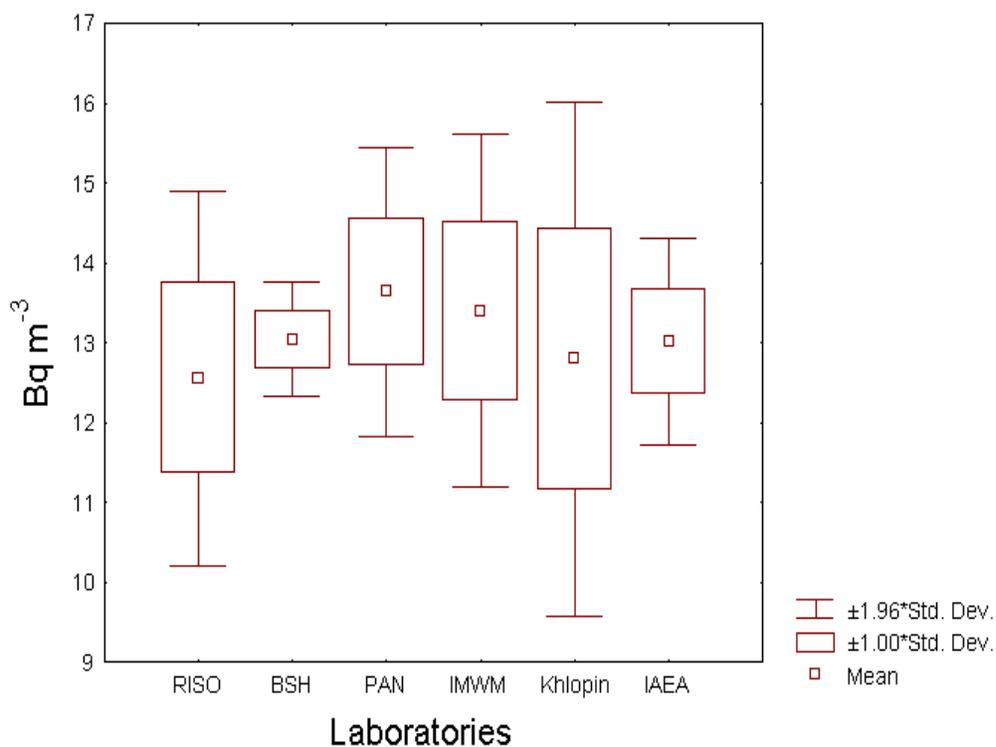
Radionuclide	IAEA 300	IAEA 378	IAEA 379
⁹⁰ Sr		0.45 [0.26 - 0.6]]	1.53 [0.5 – 2.0]
¹³⁴ Cs	0.69 [54-74]		1.2 [0.45 –1.7]
¹³⁷ Cs	1053 [940 – 1101]	5.7 [3.2 – 6.5]	39.5 [37.9 – 45]
⁶⁰ Co	0.77 [0.74 – 1.5]		
¹²⁵ Sb	15.2 [8.9 – 18]		
²³⁸ Pu	0.14 [0.14 – 0.15]		0.12 [0.1 – 0.15]
^{239,240} Pu	3.43 [3.1 – 3.9]	0.135 [0.11 – 0.15]	3.8 [3.2 – 4.1]
²⁴¹ Am	1.23 [1.14 – 1.4]	0.039 [0.033 – 0.09]	1.6 [1.17 – 1.9]
⁴⁰ K	1006 [960 – 1064]	960 [855 – 1040]	749 [636 – 795]
²¹⁰ Pb	375 [350 – 400]	188 [153 – 191]	229 [203.9 – 240]
²²⁶ Ra	58.8 [41.3 – 95.7]	118 [103.2 – 132]	29 [22.1 – 48]

Table 4.21 Summary of radionuclide concentrations measured in Baltic Sea Marine Sediment for intercomparison exercises IAEA 300, 378 and 379; median and range of accepted values (in parentheses); unit: Bq kg⁻¹ dry weight.

Table 4.22 ^{90}Sr measurements reported by the MORS group for intercomparison exercise IAEA 299, 1992-1996 (reference date: June 27th 1991; unit: Bq m^{-3}).

Laboratory	1992	1993	1994	1995	1996	Mean
RISØ (DK)	13.3 ± 0.3	13.1 ± 0.7	11.0 ± 0.9	13.8 ± 0.7	11.6 ± 0.5	12.6 ± 1.2
STUK (FIN)					11.8 ± 0.8	
BSH (DE)	12.9 ± 0.2	13.2 ± 0.4	13.6 ± 0.5	12.8 ± 0.3	12.7 ± 0.9	13.0 ± 0.4
PAN (PL)	14.4 ± 0.2	13.5 ± 0.3	14.2 ± 0.5	14.0 ± 0.5	12.1 ± 1.1	13.6 ± 0.9
CLRP (PL)						
IMWM (PL)	12.8 ± 0.5	12.7 ± 0.2	14.7 ± 1.8			13.4 ± 1.1
Khlopin (RU)	12 ± 2	11 ± 2	15 ± 1	12 ± 2	14 ± 2	13 ± 2
IAEA (MC)	13.9 ± 1.5	13.0 ± 0.3	12.8 ± 0.7	12.1 ± 0.7	13.3 ± 0.5	13.0 ± 0.7
Median	13.1	13.05	13.9	12.8	12.4	13
Range	12 – 14.4	11 – 13.5	11 – 15	12 - 14	11.6 – 14.0	12.6 – 13.4
95% CI	12.6 – 13.8	12 – 13.4	12.5 – 14.6	12.2 – 13.6	11.9 – 13.4	12.8 – 13.4

Figure 4.1 ^{90}Sr values grouped by laboratory for data reported for IAEA 299 during the period 1992-1996. No significant differences were detected among the mean values reported by the participants (ANOVA, $p > 0.05$).



Laboratory	1992	1993	1994	1995	1996	mean
RISØ (DK)	72.3 ± 1.2	69.7 ± 0.7	70.1 ± 3.5	70.6 ± 0.7	72.1 ± 0.7	70.96 ± 1.17
STUK (FIN)	68 ± 5	69 ± 3	68 ± 5	69 ± 2	66.6 ± 8	68.12 ± 0.98
BSH (DE)	69.9 ± 1.3	71.8 ± 2.7	65.2 ± 2.0	69.5 ± 1.5	69.3 ± 6.8	69.14 ± 2.41
PAN (PL)	72 ± 1	68.7 ± 0.6	72.3 ± 2.5	73 ± 2	71.9 ± 12	71.58 ± 1.66
Studsvik (S)	72.1 ± 1.9	70.8 ± 3.1	71.1 ± 2.9	75.1 ± 2.2	77.6 ± 3.6*	72.27 ± 1.96
NSEP B	72.3 ± 4.0	70.1 ± 3	72 ± 1.5	69.8 ± 1.9	70.2 ± 1.5	70.88 ± 1.17
IMWM (PL)	69.7 ± 5.4	68.6 ± 1.4	72.5 ± 4.6	72 ± 1.3	67.4 ± 0.8	70.04 ± 2.18
Khlopin (RU)	76 ± 8	73 ± 5	74 ± 6	73 ± 6	80 ± 7*	74 ± 1.412
IAEA (MC)	67.7 ± 3.0	73.1 ± 3.5	74.5 ± 2.8	63.6 ± 1.4*	67.3 ± 2.9	70.65 ± 3.68
Median	72	70.1	72	71.3	69.3	
Range	67.7 – 76	68.6 – 73	68 – 74.5	63.6 – 75.1	67.3 – 77.6	
95% CI	69.2 – 73.0	68.9 – 73	68.9 – 73.3	69.9 – 73	67.6 – 70.9	

Table 4.23 ¹³⁷Cs measurements reported by the MORS group for intercomparison exercise IAEA 299, 1992-1996 (reference date June 27th 1991; unit: mBq m⁻³).

*rejected values

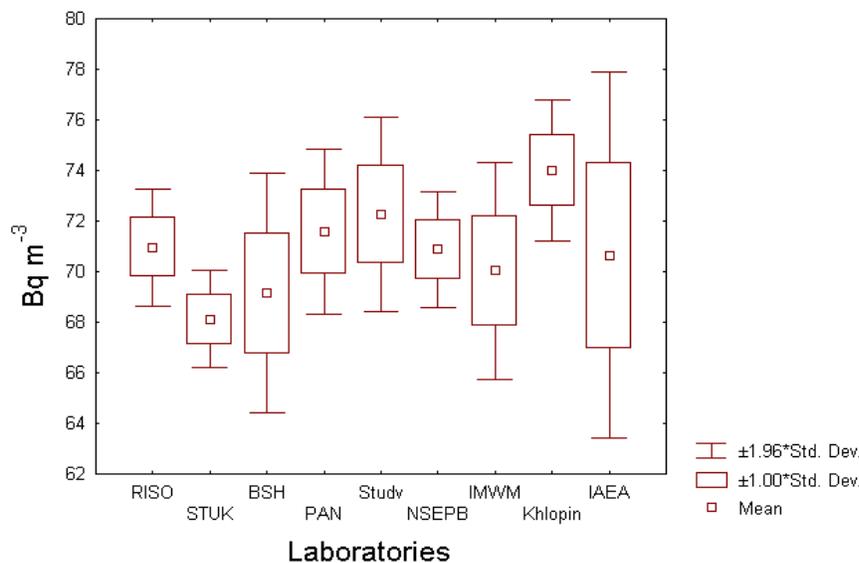


Figure 4.2 ¹³⁷Cs values grouped by laboratory for data reported for IAEA 299, 1992-1996. Khlopin's mean value was statistically different (ANOVA, $p < 0.05$ and HDS Tukey) from those reported by STUK and BSH.

Laboratory	1992	1993	1994	1995	1996
RISØ (DK)	50.5	48.3	48.9	48.7	nr
STUK (FIN)	49 ± 2.9	49.7 ± 1.5	48 ± 1.4	47.7 ± 1.4	44.9 ± 1.3
BSH (DE)	42.3	48.1 ± 0.4	49.5 ± 0.55	45.3	47.9 ± 0.1
PAN (PL)	49.8 ± 1.5	52 ± 3	51 ± 3	Nr	49.6 ± 0.3
Studsvik (S)	50.2 ± 0.5	50.4 ± 0.6	49.1 ± 0.6	49.0 ± 0.5	50.6 ± 0.7
IMWM (PL)	nr	48.3 ± 1.8	48.5 ± 3	49.3 ± 0.5	49.9 ± 0.6
IAEA (MC)	46.8 ± 0.5	50.9 ± 1.5	51 ± 3	Nr	nr
Std. Value	49.125	49.125	49.125	49.125	49.125
Median	49.4	49.7	49.1	48.7	49.6
Range	42 – 50.2	48.1 – 52	48 – 51	47.7 – 49.3	44.9 – 50.6
95% CI	45.7 – 50.4	48.6 – 50.8	48.6 – 50.3	46.8 – 49.2	46.9 – 50.3

Table 4.24 ¹³⁷Cs measurements reported by the MORS group for intercomparison exercise IAEA 299 on standard ampoules, 1992-1996 (reference date: June 27th 1991; unit: Bq ampoule⁻¹).

Figure 4.3 ^{137}Cs values grouped by laboratory for data reported for IAEA 299 on standard – ampoules, 1992-1996. PAN's mean value was statistically different (ANOVA, $p < 0.05$ and HDS Tukey) from that reported by BSH.

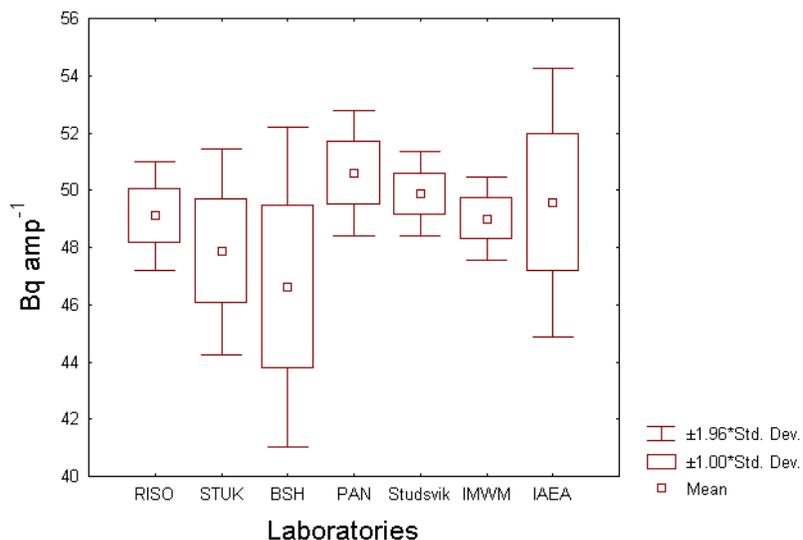
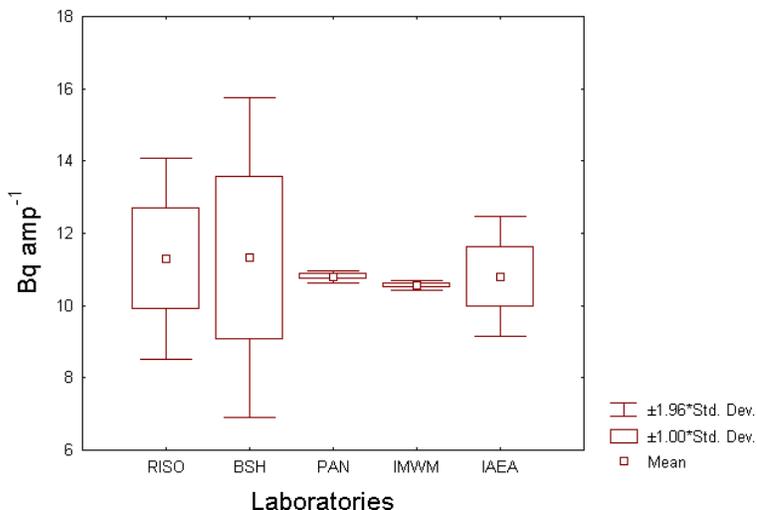


Table 4.25 ^{90}Sr measurements reported by the MORS group for the intercomparison exercise on standard ampoules 1992-1996 (reference date: June 27th 1991; unit: Bq ampoule⁻¹).

Laboratory	1992	1993	1994	1995	1996
RISØ (DK)		10.95	13.3 ± 0.7	11	9.96
BSH (DE)	14.7	10.3 ± 0.04	10.3 ± 0.21		9.9 ± 0.05
PAN (PL)	10.9 ± 0.3	10.8 ± 0.5	10.8 ± 0.5		10.7 ± 0.3
IMWM (PL)		10.6 ± 0.2	10.5 ± 0.2		
IAEA (MC)	10.2 ± 0.3	11.4 ± 0.5			
Median	10.9	10.8	10.65		9.96
AQCS value	10.6	10.6	10.6	10.6	10.6
Range	10.2 – 14.7	10.3 – 11.4	10.3 – 13.3		9.96 – 10.7
95% CI	10.1 – 13.7	10.5 – 11.1	10.1 – 12.3		9.87 – 10.5

Figure 4.4 ^{90}Sr values grouped by laboratory for data reported for IAEA 299 on standard ampoules, 1992-1996. None of the mean values reported are statistically different.



Laboratory	Country	² IAEA 337 (Bq m ⁻³)	³ IAEA 381 (Bq l ⁻¹)
RISØ	DK	13.3 ± 0.4	0.142 ± 0.011
STUK	FIN	13.6 ± 1.1	0.14 ± 0.001
BSH	S	13.5 ± 0.8	0.149 ± 0.005
PAN	PL		0.158 ± 0.013
Khlopin	RU	17.3 ± 1.7	
LREB	PL	18.4 ± 1.2	0.14 ± 0.01
Min. Environ	LT	7.7 ± 1.3	0.1
ERPC	EE		0.12 ± 0.06
IAEA-MEL	MC		0.16 ± 0.008
Median		13.6	
AQCS values			0.14
Range accepted		7.7 – 18.4	
95% CI		11.3 – 15.8	0.13 – 0.15

Table 4.26 ⁹⁰Sr measurements reported by the Baltic Sea coastal countries for sea water samples for intercomparison exercises IAEA 337 and IAEA 381.

² Reference date: July 1st, 1996

³ Reference date: September 7th, 1993

Laboratory	Country	² IAEA 337 (Bq m ⁻³)	³ IAEA 381 (Bq l ⁻¹)
RISØ	DK	75.65 ± 1.19*	0.425 ± 0.02
STUK	FIN	85.75 ± 3.35*	0.52 ± 0.09
BSH	S	72.4 ± 2.9	0.495 ± 0.02
PAN	PL		0.475 ± 0.03
CLRP	PL		0.496 ± 0.048
IMWMI	PL	73.4 ± 1.44	
Khlopin	RU	96.3 ± 8	
LREB	PL	91.85 ± 6.86	0.53 ± 0.04
Min. Environ	LT	64.8 ± 3	.1
ERPC	EE	82 ± 1.42*	0.483 ± 0.035
IAEA-MEL	MC	75 ± 0.14*	0.523 ± 0.037
Median		77.01	
AQCS values			0.482
Range accepted		64.8 – 96.7	
95% Conf. Interval		73.1 – 80.8	0.48 – 0.50

Table 4.27 ¹³⁷Cs measurements reported by the Baltic Sea coastal countries for sea water samples for intercomparison exercises IAEA 337 and IAEA 381.

* > 2 samples

² Reference date: July 1st, 1996

³ Reference date: September 7th, 1993

Laboratory	Country	²³⁸ Pu	^{239,240} Pu	²⁴¹ Am
RISØ	DK	2.8 ± 0.2	13.1 ± 0.7	16.0 ± 0.8
STUK	FI	3.5 ± 0.4	14 ± 1	13 ± 1
BSH	DE	5.63 ± 0.42	22.96 ± 1.21	18.93 ± 0.91
CLRP	PL	3.1 ± 0.4	13.2 ± 1.5	
IAEA-MEL	MC	3.95 ± 0.31	15.7 ± 0.5	17.7 ± 1.4
AQCS value		3.17	13.2	17.1
Conf. Interval		3.1 – 3.5	13.0 – 14.0	13.3 – 17.6

Table 4.28 ²³⁸Pu, ^{239,240}Pu, ²⁴¹Am measurements for sea water samples for IAEA 381 (reference date: September 7, 1993; unit: mBq kg⁻¹).

Laboratory	Country	⁹⁰ Sr (Bq kg ⁻¹)	¹³⁷ Cs (Bq kg ⁻¹)	¹³⁴ Cs (Bq kg ⁻¹)	⁴⁰ K (Bq kg ⁻¹)
STUK	FIN	1.04 ± 0.156	43.1 ± 0.47		434 ± 138
CLRP	PL		40.4 ± 4.4		455.8 ± 36.5
LREB	LV	1.3 ± 0.3	45.2 ± 4.8	7.9 ± 2.3	
Min. Environ	LT	3.5 ± 0.5	49.6 ± 4		463 ± 30
ERPC	EE		49.3 ± 3.5		

Table 4.29 ⁹⁰Sr, ¹³⁷Cs and ¹³⁴Cs measurements reported by the Baltic Sea coastal countries for fish samples for intercomparison exercise IAEA 382 (reference date: January 1st 1997).

Laboratory	Country	²³⁸ Pu (Bq kg ⁻¹)	^{239, 240} Pu (Bq kg ⁻¹)	²⁴¹ Am (Bq kg ⁻¹)	²³⁸ Pu ^{239, 240} Pu
STUK	FIN	0.11 ± 0.017	0.66 ± 0.05	0.82 ± 0.08	0.167
CLRP	PL	0.131 ± 0.02	0.72 ± 0.06		0.181
ERPC	EE	0.15 ± 0.02	0.87 ± 0.05		0.172
IAEA-MEL	MC	0.20 ± 0.06	1.07 ± 0.3		0.186

Table 4.30 ²³⁸Pu, ²³⁹, ²⁴⁰Pu and ²⁴¹Am measurements reported by the Baltic countries for 382 fish samples for intercomparison exercise IAEA 382 (reference date: January 1st 1997).

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5 RADIONUCLIDES IN SEA WATER

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5.1 Introduction

This chapter contains reports on the current status of the concentrations of radioactive substances in the waters of the Baltic Sea as determined between 1984 and 1998 at selected stations, as shown in Figure 5.1. The data presented here is restricted to the most commonly measured artificial radionuclides in the Baltic Sea, namely ¹³⁷Cs, ⁹⁰Sr and ^{239,240}Pu, with some ⁹⁹Tc data also included.

Between 1984 and 1998 nine countries have contributed data on a total of 4,935 samples covering all the 14 distinct basins of the Baltic Sea. A detailed description of procedures followed in the early years, until 1992, was given in the two preceding Joint Reports (IAEA 1986a, Pantelev 1995).

5.2 Distribution and temporal evolution of radionuclides

The temporal trends in ¹³⁷Cs, ⁹⁰Sr and ^{239,240}Pu concentrations in surface and near-bottom water during the period 1984 - 1998 are shown in Figures 5.2 - 5.5. Concentrations have been summarised as yearly averages for each of the 14 basins defined in Figure 5.1. This study focuses on the behaviour of ¹³⁷Cs. Concentrations of ¹³⁴Cs had decreased to almost undetectable levels by 1998, due to radioactive decay.

For more detailed examinations, 6 stations around the Baltic Sea were selected to represent different sea areas. Observations were based on the regular sampling and measurements of two radionuclides, ¹³⁷Cs and ⁹⁰Sr. The stations are: 1) LL3a, Gulf of Finland, 2) BY15, Baltic Proper, 3) EB1, Bothnian Sea, 4) C VI, Bothnian Bay, 5) P5, Bornholm Sea and 6) Schlei, Western Baltic (Figs. 5.2 - 5.4). The locations of stations and the basins are mapped in Figure 5.1.

¹³⁷ Cs

During the period 1992 -1998 concentrations of

¹³⁷Cs in the surface waters decreased in all parts of the Baltic Sea (Fig. 5.7). However, there were still clear differences between the different basins. Concentrations in the Bothnian Sea decreased from about 170 Bq m⁻³ to 95 Bq m⁻³, but remained the highest of all the basins. The decrease in the Baltic Proper was less pronounced, from 110 to 80 Bq m⁻³. This was due to the inflow of more contaminated water from the northern part of the Baltic Sea. Reductions in the Bothnian Bay (from 120 to 65 Bq m⁻³) were also significant. Concentrations in the Gulf of Finland were already lower than in the Gulf of Bothnia by 1992. At the end of 1998 the average concentration of ¹³⁷Cs in the Gulf of Finland was 60 Bq m⁻³. In the Bornholm Sea, concentrations did not decrease evenly; and were at an exceptionally high level in 1996 (120 Bq m⁻³) for reasons that remain unclear. Nevertheless, the level had fallen to about 70 Bq m⁻³ in 1998. In the Western Baltic Sea ¹³⁷Cs concentrations had been relatively low for almost all the time since the Chernobyl accident, but a decrease is also discernible here during the period 1992 -1998; from 65 to 45 Bq m⁻³.

According to these observations, ¹³⁷Cs concentrations in the Bothnian Sea remained twice as high as those in the southernmost parts of the Baltic Sea. One reason for this is the slow exchange of water between this basin and the Baltic Proper. The most contaminated catchment areas in the Baltic Sea region were around the Bothnian Sea. River inflow from these areas can also affect the higher concentrations in this basin (Saxén and Ilus 2001).

Differences between concentrations in surface and near-bottom waters over the years 1992 - 1998 in different basins can clearly be seen in Figures 5.7 and 5.8. In the Baltic Proper and the Bornholm Sea the ratio of concentrations in surface water to those in near-bottom water has constantly been over one. No clear mixing between surface and near-bottom waters can be seen due, to the strong halocline. In the Bothnian Sea and the Western Baltic Sea the concentrations in surface water were higher than those in near-bottom water in the beginning of the period, but in recent years they have been more or less equal. In contrast, in the Gulf of Finland and the Bothnian Bay ¹³⁷Cs concentrations in near-bottom water have been higher than those in surface waters during the whole period. This is due to the sinking of the small particles containing most of ¹³⁷Cs to the bottom, and the mixing of surface and near-bottom waters. Re-suspension from bottom sediment could also contribute to the higher amounts of caesium in near-bottom water, although this phenomenon has not yet been verified.

⁹⁰Sr

The levels of ⁹⁰Sr concentration were roughly be-

tween 12 and 20 Bq m⁻³ during the period 1992 - 1998 (Fig. 5.4). In surface waters ⁹⁰Sr is evenly distributed throughout the Baltic Sea, and concentrations are gradually decreasing over time (Fig. 5.9). The rate of decrease for ⁹⁰Sr is clearly slower than that for ¹³⁷Cs, reflecting differences in their behaviour in the aquatic environment. In near-bottom waters ⁹⁰Sr showed very similar patterns to those observed in surface waters (Fig. 5.10).

^{239, 240}Pu

Concentrations of ^{239, 240}Pu were generally very low in both surface and near-bottom waters, usually only a few mBq per cubic metre, in the whole of the Baltic Sea. There were some exceptions, however, with concentrations six to ten times higher than elsewhere observed in the Baltic Proper in 1994 and in the Bothnian Bay in 1997 for reasons unknown (Fig. 5.5), although the uncertainties in plutonium results are considerable. It is difficult to draw any clear overall temporal trends for the behaviour of ^{239, 240}Pu. This is due to the lack of data and very low concentrations of ^{239, 240}Pu, rather than the presence of sources of plutonium in the Baltic Sea area.

⁹⁹Tc

The ⁹⁹Tc values given in this report are mainly from the Arkona Sea, the Bornholm Sea, the Sound and the Kattegat. Figure 5.6 shows that the highest concentrations were found in the Kattegat, and that concentrations declined towards the Bornholm Sea. Two small patches of higher values were found in the Sound (Nielsen 2001). This data suggest that ⁹⁹Tc enters the Baltic Sea from the North Sea. No ⁹⁹Tc data is yet available for the other basins of the Baltic Sea, however.

5.3 Sea water inventories

No detailed calculations of inventories of ¹³⁷Cs in water from the Baltic Sea have been done since the previous Joint Report. In that report the total inventory of ¹³⁷Cs was evaluated to be 2,330 TBq (Panteleev 1995). Since concentrations are

clearly decreasing in the different parts of the Baltic Sea, however, it can be very roughly estimated that the total amount of ¹³⁷Cs i has decreased about 30% in the water phase across the whole sea. Consequently, an approximate estimate for the inventory of ¹³⁷Cs in the water of the Baltic Sea would be of the order of 1,630 TBq in 1998.

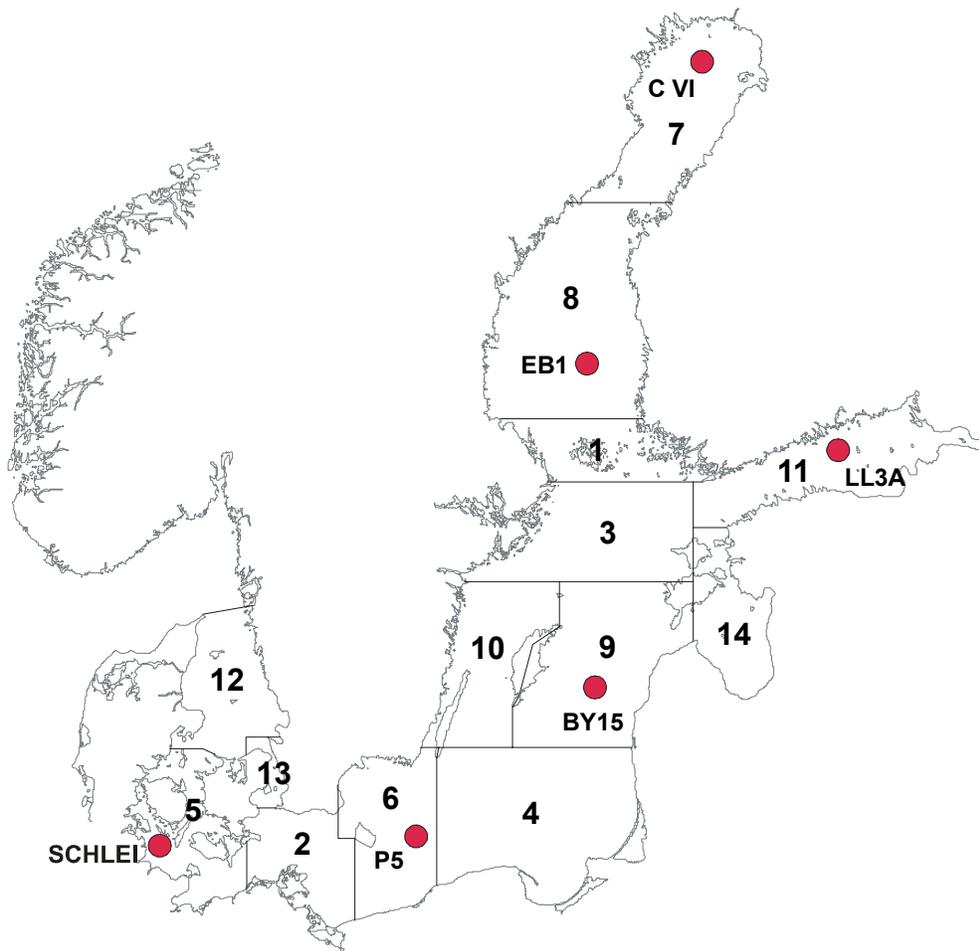
5.4 Conclusions

In conclusion, it can be inferred that the origin of most of the ¹³⁷Cs in the Baltic Sea is from Chernobyl fallout, whereas the source for ⁹⁰Sr and ^{239, 240}Pu is the earlier global fallout from nuclear weapons tests.

¹³⁷Cs concentrations are one of the main indicators of the radioactive status of the waters of the Baltic Sea. The temporal trends observed so far indicate that ¹³⁷Cs concentrations will continue to decrease in the future.

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- | | |
|-----------------------------|--------------------|
| 1 Archipelago and Åland Sea | 8 Bothnian Sea |
| 2 Arkona Sea | 9 Gotland East |
| 3 Northern Baltic Proper | 10 Gotland West |
| 4 Southern Baltic Proper | 11 Gulf of Finland |
| 5 Western Baltic | 12 Kattegat |
| 6 Bornholm Sea | 13 The Sound |
| 7 Bay of Bothnia | 14 Gulf of Riga |

Figure 5.1 Basins (numbered) and seawater sampling points (dots).

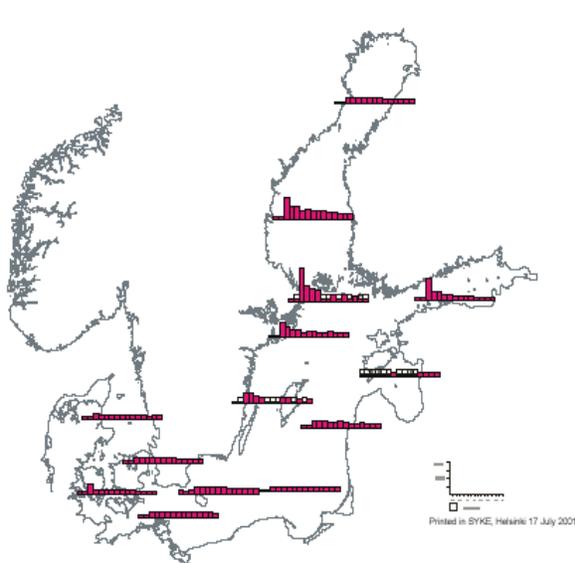


Figure 5.2a ^{137}Cs concentrations (Bq m^{-3}) in surface water 1984-1998.

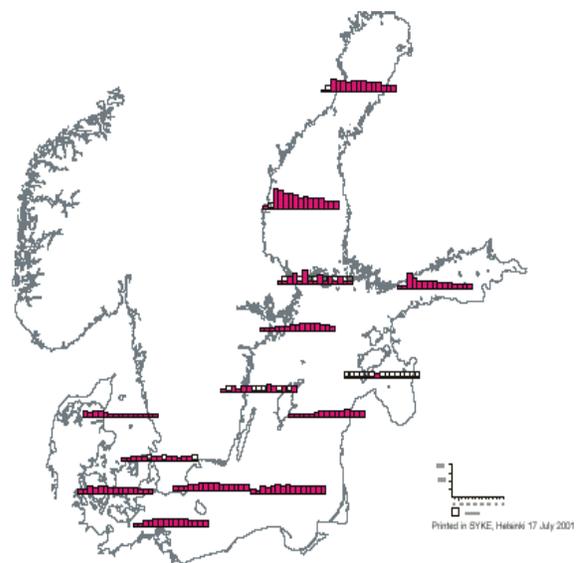


Figure 5.2b ^{137}Cs concentrations (Bq m^{-3}) in bottom water 1984-1998.

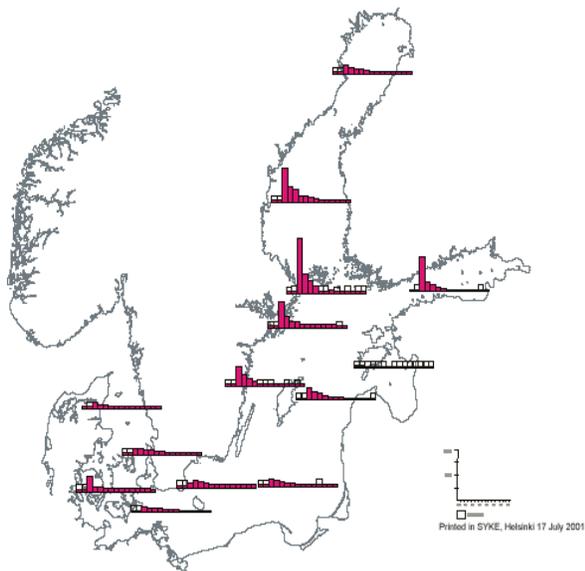


Figure 5.3a ^{134}Cs concentrations (Bq m^{-3}) in surface water 1984-1998.

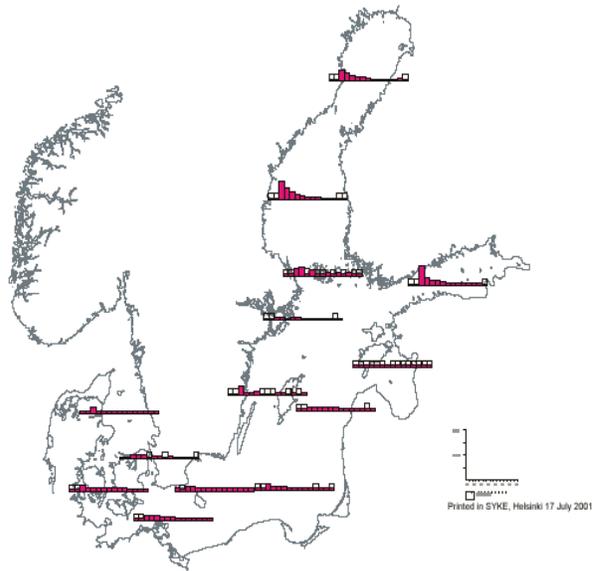


Figure 5.3b ^{134}Cs concentrations (Bq m^{-3}) in bottom water 1984-1998.

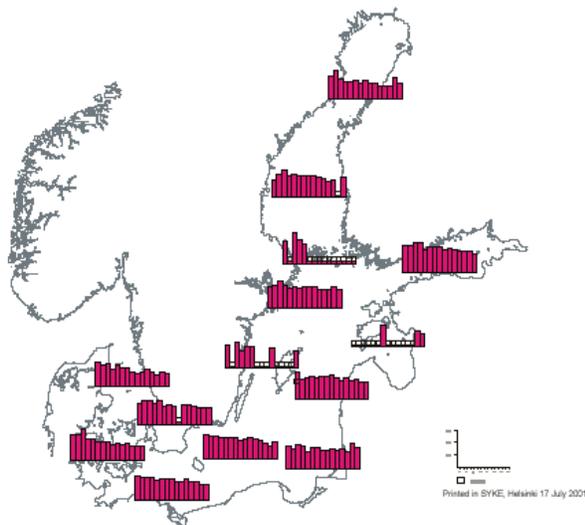


Figure 5.4a ^{90}Sr concentrations (Bq m^{-3}) in surface water 1984-1998.

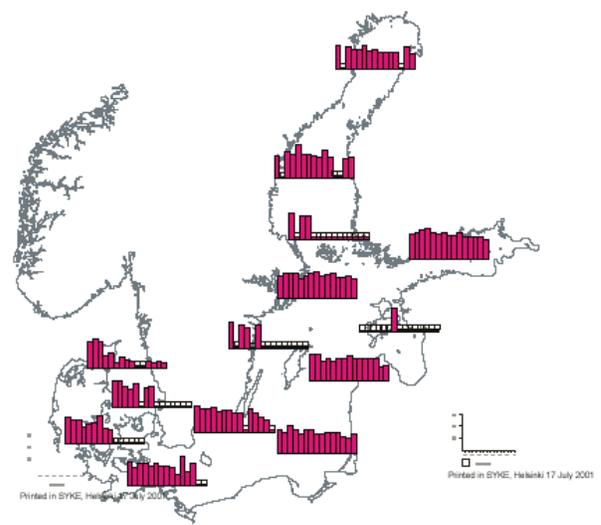


Figure 5.4b ^{90}Sr concentrations (Bq m^{-3}) in bottom water 1984-1998.

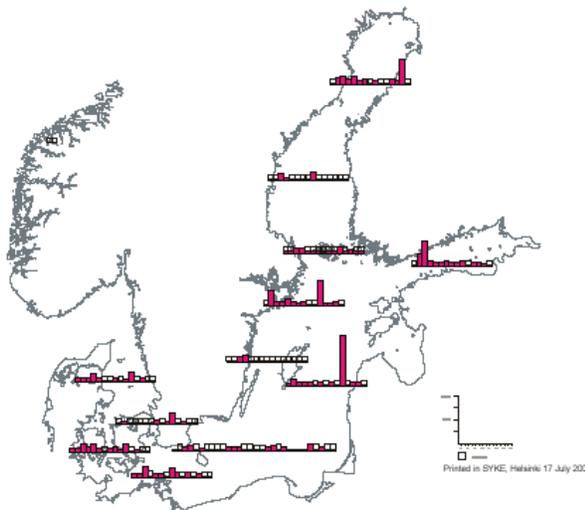


Figure 5.5a $^{239,240}\text{Pu}$ concentrations (Bq m^{-3}) in surface water 1984-1998.

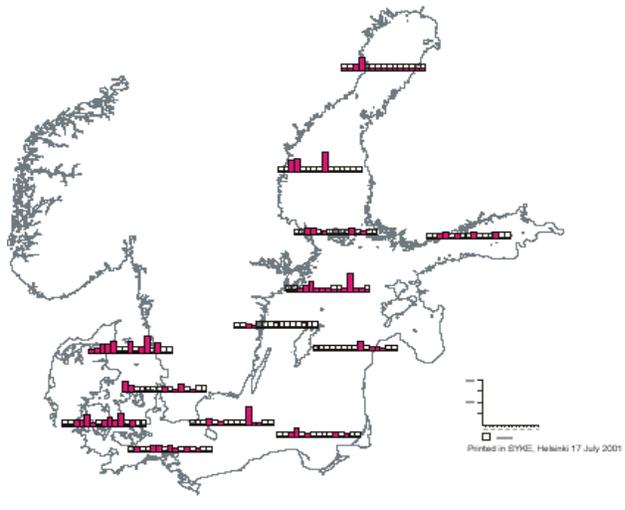
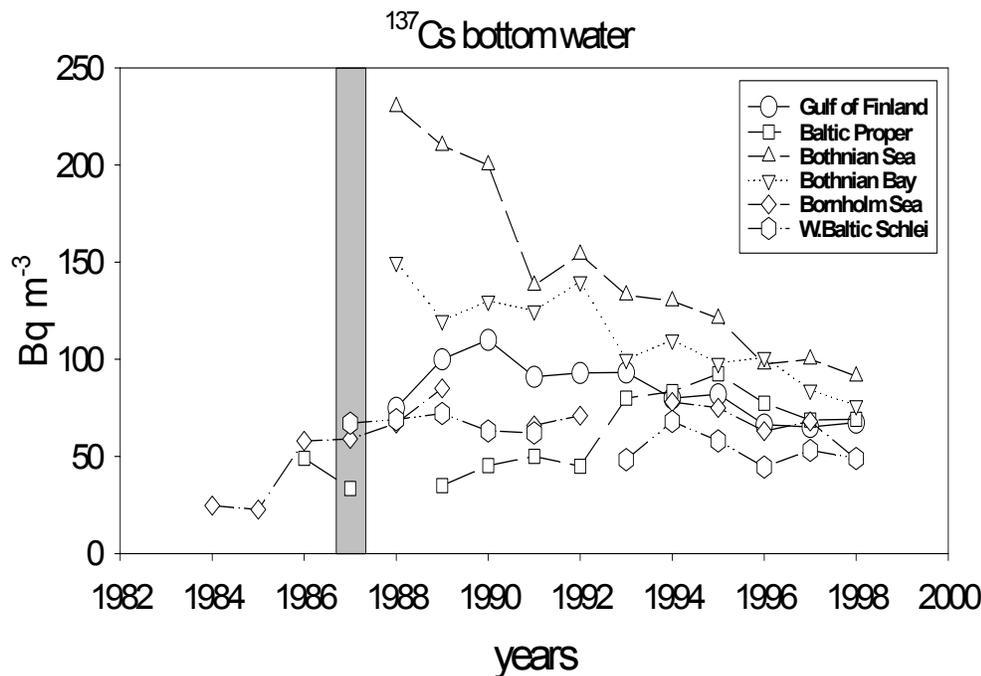
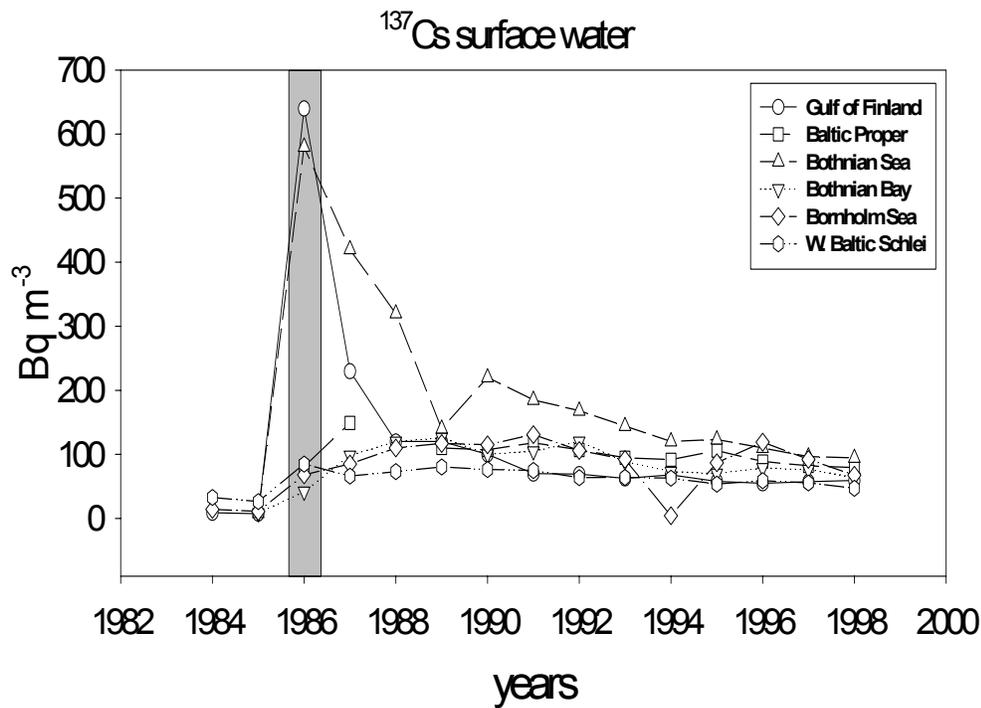
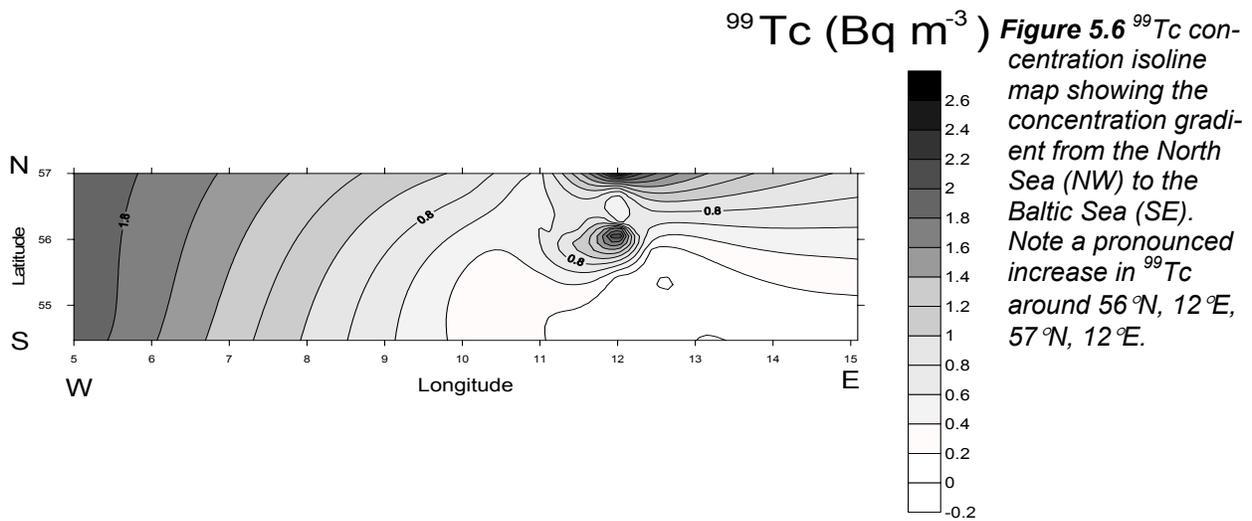


Figure 5.5b $^{239,240}\text{Pu}$ concentrations (Bq m^{-3}) in bottom water 1984-1998.



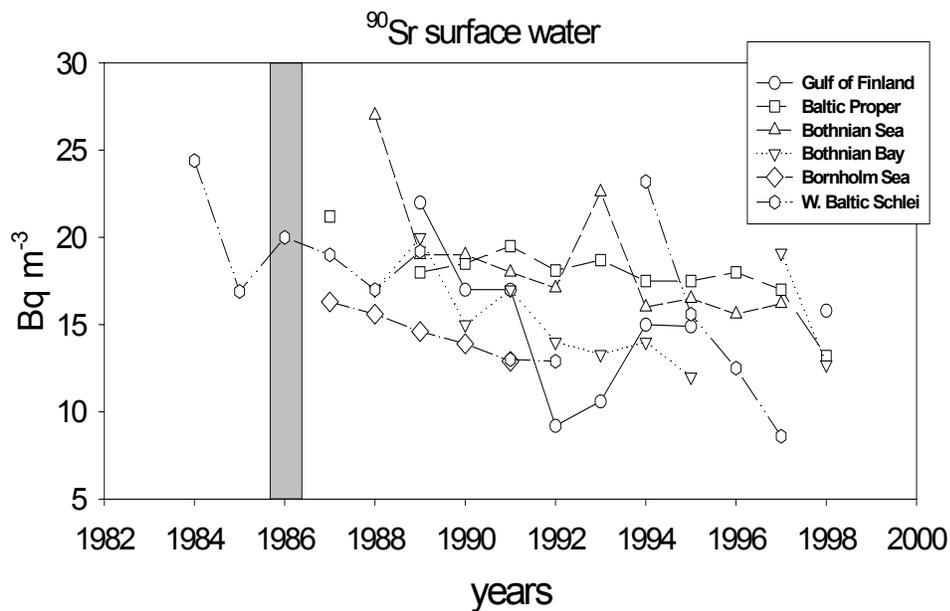


Figure 5.9 Temporal variations in ⁹⁰Sr concentrations measured at 6 stations in various parts of the Baltic Sea. The values represent concentrations in surface water in Bq m⁻³. The shaded area marks the year of the Chernobyl Accident.

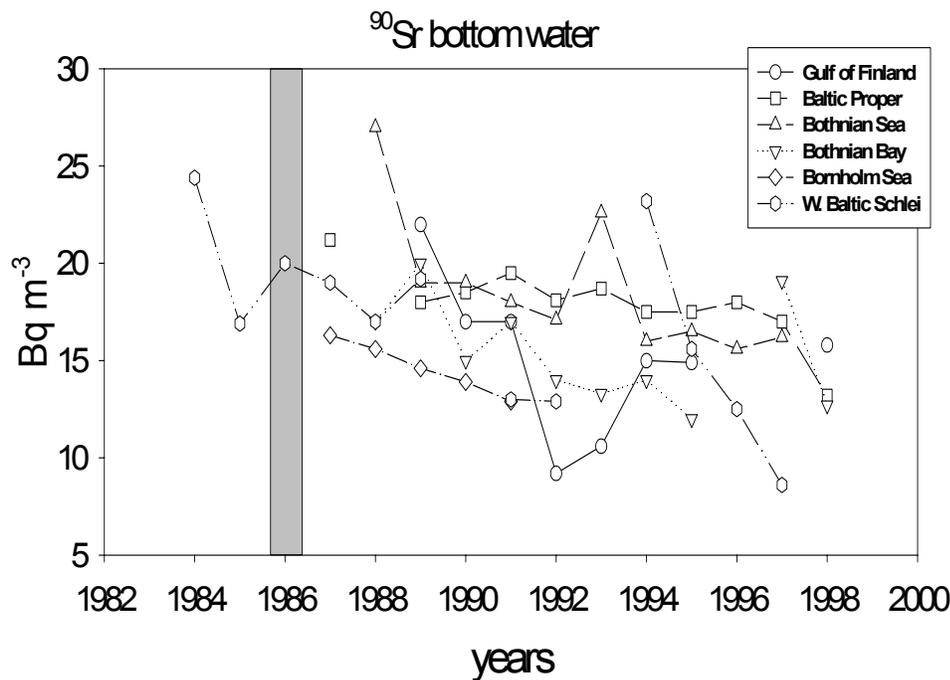


Figure 5.10 Temporal variations in ⁹⁰Sr concentrations measured at 6 stations located in various parts of the Baltic Sea. The values represent concentrations in bottom water in Bq m⁻³. The shaded area marks the year of the Chernobyl Accident.

6 RADIONUCLIDES IN SEDIMENTS

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6.1 Introduction

In radio-ecological studies of the marine environment, bottom sediments play an important role. This is because large proportions of the radioactive substances entering the sea are adsorbed over time onto suspended particulate matter, and subsequently deposited in sediments. Sediments are generally a final sink for most organic material produced in the water phase, and for other particles transported by water currents from other parts of the sea or from adjacent terrestrial sources. During their slow settling these particles tend to bind radionuclides from the water phase and drag them to the bottom.

In favourable conditions, such deposited particles form undisturbed laminae in a stratigraphic sequence on the sea-bed, creating an archive recording the history of the area. Various particle-bound substances can be identified as markers of specific historical events or periods, and laminae can be dated with the aid of such marker horizons. For example, the radioactive fallout from atmospheric nuclear weapon tests in the 1950s and 1960s and the accident at the Chernobyl Nuclear Power Plant in April 1986 have created useful markers in marine sediments in many seas, and especially in the Baltic Sea.

The Baltic Sea offers exceptional opportunities for sedimentological studies, since average rates of sedimentation are much higher here than in the deeper oceans or most other coastal seas. The anoxic conditions in the near-bottom water of the Baltic Proper mean that extensive areas of the sea-bed have few or no benthic animals, thus preventing bioturbation (disturbance by animals, e.g. worm burrows), so sediment laminae can be sampled in these areas in an undisturbed stratigraphic sequence. However, it must always be remembered that sediment sampling is extremely sensitive to errors, which can cause considerable differences in results.

6.2 Sampling techniques

To get valid results, it is essential to obtain reliable samples from sediments. False conclusions are an obvious risk if studies are based on biased samples. Sampling of the least consolidated top sedi-

ments is difficult and requires standardised and precise working methods, as well as properly designed devices. An experienced and competent team, aware of all possible sources of error, is always needed in sediment sampling. The main problem is that the topmost sediment layers are usually very soft and susceptible to re-suspension, because the interface between water and sediment resembles "a line drawn in water" (Ilus et al., 2000).

The main errors involved in sediment sampling are as follows (Ilus et al., 2000):

- 1) The loss of soft, unconsolidated sediments as a result of scavenging (blow-away), caused by the pressure wave built up in front of the descending sampler.
- 2) Mixing and redistribution of sampled sediment layers.
- 3) Cores may be shortened during sampling.
- 4) Deeper sediment layers may be smeared with particles from upper layers.
- 5) The loss of material from the margins of the sediment core (edge effect).
- 6) Brimming of the corer may occur.
- 7) Tilting of the corer away from the vertical.
- 8) The loss of enclosed material as a result of brimming or tilting.
- 9) Sediment from outside the true sample may enter the sample due to brimming, tilting or re-suspension.

Sediment sampling techniques and sampling devices differ widely, also within the HELCOM/MORS Group. This variation most probably causes differences in results.

Study material and description of sampling devices and methods used by the MORS Group

Estonia

The Estonian Radiation Protection Centre only started sediment studies quite recently. During the period 1992-1998 results for only one sediment core (taken in 1997) were reported for the HELCOM/MORS database.

The sediment samples were taken with a Kajak Corer, with a 10 cm inner diameter coring tube. Usually one core is taken at each station and sliced into 2-cm-thick sub-samples.

Finland

In accordance with the HELCOM/MORS monitoring programme, STUK makes annual reports for the database with sediment results from six sampling stations situated in the Baltic Proper, the Gulf of Finland, the Bothnian Sea and the Bothnian Bay (Fig. 3.1.3). All the samples were analysed for artificial gamma nuclides; a selected group of samples was also analysed for ²³⁸Pu, ^{239,240}Pu, and in

one case ^{241}Pu .

The sediment samples were taken by the Finnish Research Vessel Aranda using two types of sampling device. In 1992-1994 samples were mainly taken with an Aquarius Box Corer (in 3 cases with a Gemini Twin Corer) and in 1995-1998 only the Gemini Twin Corer was used. These corers are described in Ilus et al. (2000).

The Aquarius Corer is a frame-supported box corer with an inner coring "aquarium" made of Plexiglas. The total height of the corer is 85 cm and the total weight 26 kg, with optional additional weights up to 12.5 kg. The height of the "aquarium" is 30 cm and the inner square cross-section measures 18.2 x 18.3 cm. The "aquarium" is removed from the corer for sectioning the core. The sectioning device consists of a plastic square piston. A Plexiglas slicer base and slice holders with thin sliding cover-plates are used to make sections of the sub-samples. One core was taken at each station, and sectioned into 5-cm-thick sub-samples.

The Gemini Twin Corer is a gravity corer with two parallel Plexiglas coring tubes in stainless steel core barrels. The corer is 132 cm long and weighs 33 kg, with optional additional weights up to 32 kg. The inner diameter of the coring tubes is 8.0 cm, and the length 79 cm. The sectioning device is identical with that of the traditional Niemistö Corer (Niemistö, 1974) consisting of a screw-operated extruder-piston, a Plexiglas slicer base and Plexiglas slicing rings with centimetre scales. Normally only one of the parallel sediment cores was sectioned into 5-cm-thick sub-samples for radionuclide analysis.

After slicing, the samples were put into plastic bags or boxes, and stored frozen, then freeze-dried and homogenised before analysis.

Germany

The Federal Maritime and Hydrographic Agency (BSH) reported sediment data from 22 sampling stations in 1992, from 10-12 stations in 1993-1997, and from 20 stations in 1998. The 12 German sediment sampling stations in the HELCOM/MORS monitoring programme are located in the south-western part of the Baltic Sea, in the Arkona Sea and the Belt Sea (Fig. 3.1.3). The BSH data for 1992 and 1998 also includes results from the Baltic Proper, the Gulf of Finland and the Bothnian Sea. All samples were analysed for artificial gamma nuclides, and selected samples were also analysed for ^{238}Pu , $^{239,240}\text{Pu}$ and ^{241}Am .

In 1992, most of the samples were taken with a Small Box Corer (SBC), and some with a Large Box Corer (LBC) or a German Niemistö Corer. In 1993, only the Small Box Corer was used, and

since 1994 the samples have been taken in equal numbers with the Small Box Corer and the Gemini Twin Corer. The Gemini Corer is normally used on soft bottoms, whereas the Small Box Corer is used for sediments with high sand content. All these corers are described in Ilus et al. (2000).

The SBC and LBC are frame-supported box corers with inner stainless steel boxes. The weight of the SBC is from 20 to 80 kg (adjusted with lead weights) and the LBC weighs 800 kg. The corer heights are 160 cm and 255 cm, respectively. The inner dimensions of the boxes are 15 x 15 cm (SBC) and 50 x 50 cm (LBC), with heights of 20 cm and 50 cm, respectively. After SBC and LBC cores are brought on board, they are sampled with hand-operated 9.4 cm inner diameter acrylic glass coring tubes. The sectioning device consists of a plastic piston with aluminium rings used in a special cutting head. Two parallel tube cores are normally taken from the LBC, and one from the SBC.

The German Niemistö Corer is a gravity corer with an inner Plexiglas coring tube (a copy of the original Niemistö Corer [Niemistö, 1974] with a larger diameter). The total corer length is 140 cm and it weighs 13 kg with additional weights of 32 kg. The coring tube inner diameter is 5.4 cm and its length is 88 cm. The sectioning apparatus consists of a screw-operated core-extrusion piston, with a Plexiglas slicer base and slice holders. 3-4 parallel samples were taken with the German Niemistö Corer at each station, and the parallel slices were combined for analysis.

The Gemini Twin Corer and its sectioning device are the same as the Finnish instruments described above, with some small modifications. The standard sectioning interval in all the German sediment studies is 2 cm. After sampling, the samples are stored in a deep freezer and freeze-dried before analysis.

Latvia

The Regional Environmental Board of Latvia started sediment studies quite recently. In 1996-1998 sediment samples were taken at two sampling stations in the Gulf of Riga, with samples also taken at three stations in the Baltic Proper in 1997. The results from the Gulf of Riga provide the only information in the database from this sea area. Samples were analysed for ^{137}Cs , and in 1998 also for ^{90}Sr . These are the only ^{90}Sr results reported from Baltic Sea sediments for the database during the period 1992-1998.

Sediment samples were taken with a Kajak Corer with an inner diameter of 8 cm, measuring 55 cm in length and weighing 10 kg. Two cores were

usually taken at each station, and then sliced into 2-cm-thick sub-samples with a Plexiglas slicer unit.

Poland

The Polish contribution to the HELCOM/MORS monitoring programme for sediments consisted of sample data from three open sea stations in the southern Baltic Proper and three coastal stations in the Gulf of Gdansk (Fig. 3.1.3). During the period 1992-1998 results from 2-8 stations were reported annually for the database. All samples were analysed for artificial gamma nuclides. Most samples were additionally analysed for ^{226}Ra , and many also for ^{238}Pu and $^{239,240}\text{Pu}$.

The sediment samples from the Central Laboratory for Radiological Protection were taken by the Polish Research Vessels Hydromet, Baltica and Orp Arctowsky, using a Sprut Corer in 1992-1995 and a Niemistö Corer from 1996 onwards. The Sprut Corer is described below in the Russian section and the Niemistö Corer in Niemistö (1974) (cf. also Ilus et al., 2000). In general, two parallel cores were taken with the Sprut Corer and sectioned into 1 or 2 cm-thick sub-samples using an extruder piston, a slicing plate and a slicing ring.

The Niemistö Corer is a gravity corer with an inner Plexiglas coring tube. The total length of the corer is 124 cm, and it weighs 38.5 kg, with additional weights always used. The inner diameter of the 80-cm-long coring tube is 5.0 cm. Five parallel cores were taken with the Niemistö Corer and sectioned in the same manner as those taken with the Sprut Corer.

Sub-samples were put in plastic boxes, stored frozen, and then air-dried and homogenised before analysis. Parallel sub-samples were combined for analysis.

Russia

The Russian contribution to the HELCOM/MORS monitoring programme for sediments consisted of sample data from four stations in the eastern Gulf of Finland and one in the northern Baltic Proper (Fig. 3.1.3). Results were reported for the database from 8 stations in 1992, 9 stations in 1994 and 5 stations in 1995. Samples were analysed for artificial gamma nuclides and regularly also for ^{226}Ra , ^{228}Ra and ^{228}Th .

The samples were taken aboard the yacht Boyan of the V.G. Khlopin Radium Institute using a Sprut Corer described e.g. in Ilus et al. (2000). This is a gravity corer with an inner Plexiglas coring tube surrounded by a stainless steel framework. The corer is 88 cm long and weight 20 kg (+8 additional weights of 1.25 kg). The inner diameter of

the 59-cm-long coring tube is 8.6 cm. In general, one core was taken at each station and sectioned into 2-cm-thick or 5-cm-thick sub-samples, either manually, or using an extruder piston, a slicing plate and a slicing ring.

Sweden

The Swedish contribution to the HELCOM/MORS monitoring programme for sediments consisted of sample data from four coastal stations situated close to the Swedish nuclear power plants, and additionally since 1995 from four open sea stations situated in the Bothnian Sea and the Bothnian Bay (Fig. 3.1.3). Results from the 3 coastal stations were reported for the database for the whole period 1992-1998, and from the 4 open sea stations for 1995-1998. The last sampling at three open sea stations was postponed from late 1998 to January 1999 due to bad weather conditions, but the results have been considered here as 1998 results. Samples were analysed for artificial gamma nuclides, and in some cases for ^{226}Ra .

Sediment samples from the vicinities of the nuclear power plants were taken with Willner Corers. At Forsmark, the inner diameter of the corer was 6.5 cm; at Barsebäck and Ringhals 7.0 cm. Five parallel cores were taken at each station. At Forsmark and Ringhals the uppermost 2 centimetres and at Barsebäck the uppermost 3 centimetres from each core were pooled for analysis.

Until 1999 the sediment samples at the open sea stations were taken with a Kajak Corer (diameter 8.0 cm); in January 1999 a Gemax Corer with an inner diameter of 9.0 cm was used. Three parallel cores were taken at each station and treated as follows: two cores were sectioned into 0-5cm and 5-10 cm slices and the parallel slices were pooled for analysis. The third core was sectioned into 5-cm-slices from 0 to 25 cm and then analysed. The Gemax Corer is very similar to the Gemini Twin Corer, with some improvements. The main difference is a peeling slicer, which peels the core during the slicing so that the diameter of the sample itself is the same as in the Gemini Corer (8.0 cm). Winterhalter (1998) has described the Gemax Corer.

Results of the MOSSIE exercise

In 1992, the HELCOM/MORS Group arranged a comparison exercise which became known as "MOSSIE" on sediment sampling devices and methods. The following corers (and laboratories) as described above were involved in the exercise: 1) Aquarius Box Corer (STUK), 2) Gemini Twin Corer (STUK), 3) German Niemistö Corer (BSH), 4) Large Box Corer (BSH), 5) Niemistö Corer (STUK and Swedish University of Agricultural Sci-

ences), 6) Small Box Corer (BSH) and 7) Sprut Corer (CLOR and KRIL). The results of the comparison were published by Ilus et al. (2000), but a short summary is given below.

The instruments best suited to quantitative sampling of soft sediments such as those typically found in the Baltic Sea appear to be those based on the coring principle. Many factors work in favour of corer orifices with relatively large diameters or areas. However, it is not possible to increase the tube diameter endlessly without having a negative impact on usability in terms of the handling and slicing of cores. However, it should be remembered that the same instrument may not be the best alternative for all types of bottom, and that different circumstances require different types of instrumentation. Box corers can be reliably used for bulk sampling of coherent sediments and some silt and sandy sediments.

The comparison was based on five criteria: 1) Lowest dry weight [g cm^{-3}] in surface layer. 2) Highest ^{137}Cs peak [Bq kg^{-1} dry wt.] in any of the layers. 3) Highest total amount of ^{137}Cs [Bq m^{-2}] in the whole sediment profile. 4) Proximity of total amount of ^{137}Cs to the median test result. 5) Proximity of total amount of ^{137}Cs to the amount found with the original Niemistö Corer.

One corer at a time was chosen as a reference corer, and the results from the other corers were compared with those from the reference corers. The average relative differences were as follows (the lowest values differ least from the reference):

Gemini Twin Corer	0.29
German Niemistö Corer	0.30
Niemistö Corer	0.31
Aquarius Box Corer	0.37
Sprut Corer	0.38
Small Box Corer	0.70
Large Box Corer	1.02

The values show that the results of the Gemini and the Niemistö corers were quite close to each other. The Aquarius and Sprut corers differed slightly more from the references, but the Small Box Corer and especially the Large Box Corer differed significantly from the others.

Typical sources of error involved in sampling with the different corers were registered as follows:

Aquarius Box Corer: There is a potential risk of scavenging or tilting. Slicing is not as precise as in tube corers with smaller diameters. However, very fine samples were obtained with this corer.

Gemini Twin Corer: There is a small risk of smearing, tilting or core shortening. In general this corer yields undisturbed, good-looking samples.

Large Box Corer: There is a significant risk of scavenging and filling up to the brim where the

very soft bottoms of the Baltic Sea are involved. This corer is probably more useful for hard clay and silt bottoms.

Niemistö Corer: The inherent problems of small-diameter corers (smearing, core shortening, edge effect) and the risk of tilting are the main disadvantages. But the original slicing device is still one of the most sophisticated available. These problems are probably a little less significant in the larger diameter **German Niemistö Corer**.

Small Box Corer: There is an obvious risk of scavenging and filling up to the brim where the very soft bottoms of the Baltic Sea are involved. This corer is probably more useful for hard clay and silt bottoms.

Sprut Corer: The main weakness of this corer is its closing mechanism, which may cause "bubbling" and mixing of the core.

Kajak Corer: This model was not tested in the "MOSSIE" exercise, but its main weakness is the vacuum-based closing system, since the closing valve is situated at the top of the coring tube. Especially when working aboard large research vessels with high rails the sample is often lost when the corer comes through the water surface (Ilus, 1996).

In general, the results showed marked differences in the total amounts of ^{137}Cs per square metre, even when the ^{137}Cs concentrations per kg of dry weight were equal. The results calculated per m^2 showed a significant direct correlation with the area of the corer, being lowest in the corers with smallest diameter; whereas no correlation was found between the ^{137}Cs value per kg of dry weight in the surface layer, and the area of the corer. This is due to an "edge effect", in which the inner walls of the coring tubes or barrels cause a loss of material from the margin of the core. This loss is greater in corers with a small diameter, and this error is multiplied when several parallel samples taken with small-diameter corers are used for analyses (Ilus et al., 2000). However, it should be remembered that despite the disadvantages associated with the small-diameter corers, their slicing systems are generally most accurate.

The "MOSSIE" exercise showed that the results obtained with different sampling devices may differ significantly. As well as documenting the variability of results, this work proved valuable in that it showed how much emphasis must be put on sampling procedures in sediment studies. In this sense the exercise was valuable not only for scientists studying radioactive substances, but for all scientists working on sediments.

Experiences from this and earlier exercises indicate that differences in sediment types and sedimentation processes should be the main consideration when selecting the sampling equipment and method. It is also clear that no universal sampler exists that would meet all the requirements

for sediment sampling.

6.3 Sediment types and geomorphology of the Baltic Sea floor

The Baltic Sea is a shallow brackish sea with only a limited water exchange with the open seas through the Danish Straits. Tidal sea level fluctuations are hardly noticeable, but long-term sea level changes have been considerable, mainly due to the regional isostatic uplift of the earth's crust following the melting of the continental ice sheet (Winterhalter et al., 1981).

Although the Baltic Sea is a shallow sea, the morphology of the sea floor is as diverse as that of its shores. The main features are pre-glacial in origin. Glacial erosion and deposition, and later erosion and deposition by waves and currents, have played a rather limited role. The differences between the pre-glacial and the present-day sea floor relief range from a few meters to some tens of meters (Winterhalter et al., 1981).

The majority of the more prominent morphological features in the Baltic Sea consist of various types of deeps – depressions and troughs – in most cases partly filled with Quaternary sediments. However, the depths of the depressions are rather modest: the depth of the Gotland Deep east of Gotland is 245 m. If Quaternary deposits were removed, its depth would be about 280 m. The Landsort Deep, north of Gotland, is the deepest part of the Baltic Sea at 459 meters deep, with an estimated 150 m of Quaternary sediments (Winterhalter et al., 1981).

The Baltic Proper is connected to the Gulf of Bothnia by a number of deep channels running through the Archipelago Sea between the Åland Islands and the mainland of SW Finland. A set of very prominent channels links the Åland Deep with the Bothnian Sea. The trough forming the deepest part of the Åland Sea attains a maximum depth of nearly 300 m (Winterhalter et al., 1981).

In the southern part of the Bothnia Sea, the sea floor is very rugged, but further north Palaeozoic sedimentary rocks cover the crystalline basement rocks, giving the bottom more gently undulating forms with thick post-glacial sediments filling the Eastern Basin. Further north the sea floor is again more rugged. There is a sharp contrast between the morphology of the sea floor off the Finnish coast and off the Swedish coast. Nearer Finland, the sea floor is largely fairly flat, whereas nearer Sweden it is characterised by a series of faults and fractures which make the bottom morphology highly irregular, even near the coast. Large shallow-water areas are also found in the central and southern parts of the Bothnian Sea (Winterhalter et al., 1981).

In the north, the Bothnian Sea is linked by a shallow strait known as the Quark to the Bothnian Bay. The same asymmetric morphology, rugged on the Swedish side and gentle on the Finnish side, is present in the Bothnian Bay. The Härnösand Deep in the northern part of the Bothnian Sea is 230 m deep, while the deepest part of the Bothnian Bay is a mere 147 m (Winterhalter et al., 1981).

Despite the presence of several considerable deeps, mean depths across much of the Baltic Sea are rather modest: for the Baltic Proper 65 m, for the Bothnian Sea 68 m, and for the Bothnian Bay 43 m (Winterhalter et al., 1981).

The Gulf of Finland shows a general decrease in depth eastwards. The south-eastern part of the Gulf is characterised by large-scale forms running in a NNW-SSE direction rising some 25-50 m above the sea floor. Off the Finnish coast, bottom morphology is governed by the bedrock surface, and is rugged in the shallow zone and successively levelled off by late-glacial and post-glacial sediments deeper waters (Winterhalter et al., 1981).

The areas around the Hiiumaa and Saaremaa Islands and the adjacent Gulf of Riga are characterised by rather flat sea floors, rarely deeper than 25 m. However, in the central part of the Gulf of Riga, the bottom slopes gently to a depth of 50 m (Winterhalter et al., 1981).

The southern part of the Baltic Sea is rather shallow, with depths rarely exceeding 50 m. Depths greater than 100 m occur only in two areas: the Gdansk depression and a basin NE of Bornholm. The Gdansk Depression in the south-eastern Baltic Proper is only 116 m deep. The Gotland Deep and the Gdansk Depression are bounded to the west by shoals forming a broad ridge extending from Gotland to the Polish coast in the south. The connection between the North Sea and the Baltic Sea has its deepest passage, 18 m, through the Danish Straits. The Sound, between Denmark and Sweden, is flat and shallow, with a sill depth of only 8 m (Winterhalter et al., 1981).

The uneven distribution of different sediments in the Baltic Sea reveals the dynamic nature of the sedimentation processes involved. Current sedimentation processes include a wide range of processes, from erosion and transport to accumulation. The Baltic Sea is characterised by rapid land upheaval in the North, and by considerable differences in seabed geology, ranging from the rugged forms of crystalline bedrock in the Gulfs of Bothnia and Finland to the smoother sedimentary formations in the southern Baltic Sea.

In the south-western part of the Baltic Sea, around the Danish Straits and the Sound, the bot-

tom mainly consists of sand, except in deeper waters where soft bottoms dominate. The south-western Baltic Sea is characterised by vast areas with sandy bottoms. Within the southern and central parts of the Baltic Sea, each of the three main bottom types cover large areas. Soft bottoms are generally dominant in deeper waters, where the sediment thickness amounts to several tens of meters. Sandy bottoms are present along the southern and eastern coastal zones. Hard bottoms can be found north of Poland and off the south-eastern coast of Sweden, for example (Winterhalter et al., 1981). Southern and central regions have large accumulative basins with fine-grained, soft silt and muddy sediments.

The northern part of the Baltic Proper is characterised by irregular topography with depressions, fairly flat areas and shoals. Around Gotland and Öland, in the Gulf of Riga, and also further north, there are sand and till deposits of considerable dimensions. Soft bottoms occur mainly where the water exceeds 80 m depth. The northern part of the Baltic Proper is largely composed of alternating small areas with hard or soft bottoms (Winterhalter et al., 1981). The irregular topography is dominated in certain areas by a mosaic of small-scale sedimentation basins, but there are also a few larger basins, including the Gotland Basin – the largest in the Baltic Sea.

The Gulf of Finland is characterised by a great variations in bottom type. Rugged bedrock forms create a series of very small, irregular separate basins. The largest sedimentation basins are situated in the southern Gulf, while the estuary of the Neva River dominates the eastern Gulf. Sedimentation in the eastern region is high, as a result of river transport of particulate matter. The northern part of the Gulf consists of rocky areas alternating with clay sediments filling the deeper parts. Large deposits of glacial drift are typical of the south-eastern part, where deeps between these deposits are filled with soft sediments (Winterhalter et al., 1981).

In the Åland Sea, soft bottoms dominate in the Åland Depression. Bottom currents erode the deepest and narrowest parts, so the top surface consists of coarse-grained lag sediments. The area north-east of the depression is characterised by rough bedrock topography with a consequent alternation of hard and soft bottoms (Winterhalter et al., 1981).

Sediments in the Gulf of Bothnia are characterised by the "basin-filling-nature" of sedimentation in the deepest areas. Rapid land uplift continuously exposes sediments to erosion by waves and currents. As a result, the sedimentary material is often a mixture of very "old" and very "young" material. A dense population of amphipods continuously mixes the sediment surface.

In the Bothnian Sea, hard bottoms dominate large areas. Along the coasts of Sweden and Finland, hard bottoms are separated by smaller areas with soft bottoms. Very extensive deposits of glacial drift occur in the north-western part of the Bothnian Sea just off the coast of Sweden. Further to the east there is a large drumlin field. All of these drift forms are more or less exposed, with soft sediments between and sometimes alongside them (Winterhalter et al., 1981).

In the Bothnian Bay, the northernmost part of the Baltic Sea, conditions are dominated by the re-sedimentation of material derived from shallower areas. This is due to the very rapid land uplift in the region, and the vast shallow areas consequently exposed to wave erosion. Sandy bottoms are very common in the north-eastern part of the Bay (Winterhalter et al., 1981).

6.4 Changes in the occurrence of radionuclides in sediments during the period 1992-1998

This Evaluation report is based on data reported by the Contracting Parties for the HELCOM/MORS database during the period 1992-1998. Sediment analysis data was reported by seven countries as follows: Estonia 1 sediment profile, Finland 43, Germany 98, Latvia 9, Poland 37, Russia 25 and Sweden 37, a total of 250 sediment profiles. If all slices are counted the material consists of 1,743 samples. The data submissions included results for 17 radionuclides: ^7Be , ^{40}K , ^{54}Mn , ^{60}Co , ^{90}Sr , ^{106}Ru , ^{125}Sb , ^{134}Cs , ^{137}Cs , ^{210}Pb , ^{226}Ra , ^{228}Ra , ^{228}Th , ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Pu and ^{241}Am . Of these ^7Be , ^{40}K , ^{210}Pb , ^{226}Ra , ^{228}Ra and ^{228}Th are classified as naturally occurring radionuclides, and some (e.g. ^{90}Sr and ^{241}Pu) have been reported only in a few cases. Most data refers to ^{137}Cs .

To date only the accident at the Chernobyl Nuclear Power Plant and atmospheric nuclear weapons tests have significantly affected the amounts of artificial radionuclides in Baltic Sea sediments. During the reporting period 1992-1998, there were no such events with notable impact on the total inventories of radionuclides. Thus the main emphasis was to follow up the occurrence and behaviour of the fallout nuclides originating from the above-mentioned events. Monitoring has duly focused on sedimentation processes and the sedimentation rates of radionuclides, as well as on the accumulation of the fallout-nuclides in sediments. Over the course of time, the water in the Baltic Sea has been purified of the fallout nuclides; and the ongoing deposition of particulate matter containing ever-decreasing quantities of these nuclides means that high-concentration peaks are being buried in ever-deeper sediment layers.

6.4.1 Sedimentation rates and the burial of fallout peaks in deeper sediment layers

During the last four decades Baltic Sea sediments have offered an extraordinary research opportunity for radioecologists and other scientists to study sedimentation and other processes in sediments by using radionuclides deposited in a historical sequence in sediment laminae on the seabed. This is due to two major events which caused significant radioactive fallout in the Baltic Sea region, creating distinct markers in the corresponding sediment layers. One was the global fallout from atmospheric nuclear weapons tests in the late 1950s and early 1960s, which resulted in clear peaks of long-lived radionuclides such as ^{137}Cs , ^{90}Sr and certain transuranic elements (notably $^{239,240}\text{Pu}$) in sediments. The other event was the fallout from the accident at Chernobyl Nuclear Power Plant, then in the USSR, in April 1986. The first radioactive clouds from Chernobyl initially travelled northwards, causing high deposition in the Baltic Sea region, and the Baltic Sea was the marine area most affected by Chernobyl accident anywhere in the world (Povinec et al., 1996). The most significant long-lived constituents of this fallout were ^{137}Cs and ^{134}Cs . These radioactive markers have been widely utilised in the dating of sediments and determination of sedimentation rates (e.g. Kankaanpää et al., 1997, Ilus, 1998).

In 1986, soon after the initial deposition of Chernobyl fallout on the surface of the Baltic Sea, the sinking of fresh fallout through the water column was observed to be quite rapid. The sinking of fallout-nuclides was most probably accelerated by phytoplankton, which were at the end phase of their spring maximum during the main fallout period. After the vernal bloom of phytoplankton, radionuclides were transported downwards by dead plankton algae. During the first half of May 1986, fresh fallout-nuclides were already detected in water at a depth of 100 m in the southern Baltic Proper, and by mid-June they were observed in the surface sediment layer at Station Teili1 (depth 166 m) in the northern Baltic Proper (Ilus et al., 1987).

After the initial fallout, several factors affected the concentrations of radionuclides in the water body, e.g. sea currents, mixing of water masses, river discharges from their catchment areas, the rate of sinking in the water column, deposition on the bottom and other processes in sediments. These factors were also reflected in the sedimentation rates of radionuclides.

Sedimentation rates (given as millimetres per year) and sediment accumulation rates (grams per square metre per year) vary greatly in the Baltic Sea, depending on local environmental factors. Rates can even differ considerably at sampling points situated very near to each other. A study carried out in 1995-1996 showed that sedimentation rates varied between 0.2 and 29 mm a^{-1} at different soft-bottom sampling sites in the Baltic Sea, depending on the sedimentation itself, and the method used in calculations (Ilus et al., 2001). The corresponding accumulation rates of dry matter ranged from 60 to 9,000 $\text{g m}^{-2} \text{a}^{-1}$. In sedimentation rate studies the importance of undisturbed and high-quality samples is especially crucial. The loss of soft surface sediments during sampling may significantly affect the results, at least where calculations are based on ^{137}Cs or $^{239,240}\text{Pu}$ peaks. Other factors which may disturb the integrity of sediment layers are bioturbation caused by benthic macrofauna, near-bottom currents, suspension and re-suspension processes, and the possible migration of radionuclides in sediments.

Sediment cores taken at a coastal station in the Gulf of Finland illustrate sedimentation rates in circumstances where bioturbation did not disturb sedimentation (Fig. 6.4.1.1). At this station, the sedimentation rate was estimated at 13-15 mm a^{-1} with the ^{137}Cs method, at 11-12 mm a^{-1} with the $^{239,240}\text{Pu}$ method, and at 12-13 mm a^{-1} with the ^{210}Pb CF:CS and CRS methods if porosities of the 0-1-cm sediment layer were used in the calculations (or at 4-6 mm if porosities of the 9-10-cm layer were used). The corresponding accumulation rates were 800-960, 620-670 and 720-820 g a^{-1} , respectively (Ilus et al., 2001).

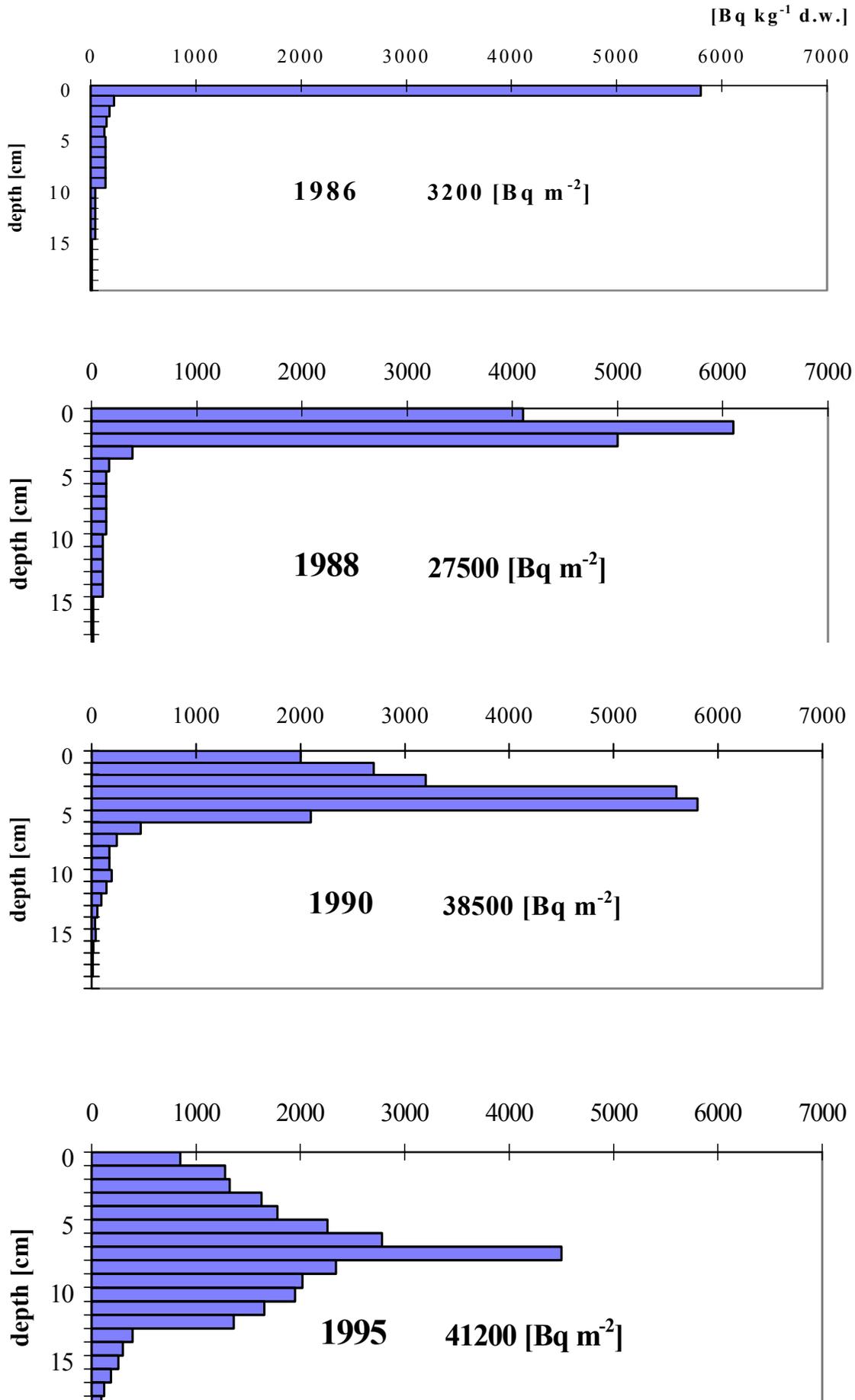


Figure 6.4.1.1 Vertical distributions and total amounts of ¹³⁷Cs at a coastal station on the Gulf of Finland, 1986-1995.

STUK and the Finnish Institute of Marine Research carried out a study on sedimentation rates in the Baltic Sea over the period 1995-1996. Sediment samples were taken from 56 sampling stations in different parts of the Baltic Sea. Sediment accumulation rates and sedimentation rates at each station were evaluated using four different methods: the ^{210}Pb based CF:CS and CRS mod-

els, the ^{137}Cs method and the $^{239,240}\text{Pu}$ method (Ilus et al., 2001). The results are summarised in Table 6.4.1.1.

	Bothnian Bay	Bothnian Sea	Gulf of Finland	Baltic Proper
Accumulation rate $\text{g m}^{-2}\text{a}^{-1}$	240-2,600	200-9,000	110-3,100	60-4,500
Sedimentation rate mm a^{-1} (calculations based on porosities in 0-1-cm sediment layer)	1.3-18	0.6-29	1.0-24	1.0-20
Sedimentation rate mm a^{-1} (calculations based on porosities in 9-10-cm sediment layer)	0.5-6.7	0.6-23	0.4-17	0.2-14

Table 6.4.1.1 Summarised results of a study on sedimentation rates in the Baltic Sea carried out during the period 1995-1996.

The results obtained in this study showed that in the Baltic Sea the use of more than one method in estimation of sedimentation rate is recommended. None of the methods is necessarily suitable for routine use, and it is always important to ensure that the basic preconditions for each method are met. In cases where the ^{137}Cs or $^{239,240}\text{Pu}$ peak is distinct, sharply defined and at sufficient depth, these respective methods may result in the best estimates. On the other hand, at stations where the peaks are spread out over a broad range of depths, the methods based on ^{210}Pb may give more accurate results if the preconditions for the ^{210}Pb model are fulfilled. In gen-

eral, all results should be presented as accumulation rates of dry matter (grams per square metre per year) to avoid the changes caused by compaction of the sediment (Ilus et al., 2001).

Sedimentation rates evaluated for various sub-regions and sampling stations of the Baltic Sea as collected from other publications are given in Table 6.4.1.2.

Table 6.4.1.2 Sedimentation rate values reported in other literature for various sub-regions and sampling stations around the Baltic Sea.

Sub-region	Station	Estimated sedimentation rate mm a ⁻¹	Reference
Bothnian Bay	general	0.6-1.3	Tulkki (1977)
	"	0.6-1.3	Niemistö (1986)
	"	0.6-1.9	Perttilä & Brüggmann (1992)
	C VI	1.3	Niemistö (1982)
	C VI	1.5-2	Tuomainen et al. (1986)
	BO 3	1.2	Niemistö 1982
	BO 3a	2.5	Tuomainen et al. (1986)
Bothnian Sea	general	0.27	Winterhalter (1972)
	"	0.1-0.8	Niemistö et al. (1978)
	"	0.9-2.4	Niemistö (1986)
	US 2b	0.92	Niemistö (1982)
	EB 1	2.4	Niemistö & Voipio (1981)
	EB 1	2-4	Tuomainen et al. (1986)
	Olk 2	10	Tuomainen et al. (1986)
Gulf of Finland	general	0.4	Perttilä et al. (1995)
	XV 1	7.4	Niemistö & Voipio (1981)
	Lov 1	5	Ilus, pers. comm.
Northern Baltic Proper	Teili 1	2.2	Niemistö & Voipio (1981)
Baltic Proper	BY 15	1.25	Niemistö & Voipio (1981)
	general	0.5-1.5	Niemistö (1986)
	"	0.5-4.2	Perttilä & Niemistö (1993)
Gulf of Riga	general	0.5-2.2	Kuptsov et al. (1984)
	"	2-10	Larsen (1995), Leivuori et al. (2000)
Bornholm Basin	general	0.5-1.5	Kögler & Larsen (1979)
Southern Baltic Proper	general	0.4-2.3	Pempkowiak (1991)
Gdansk Basin	general	0.5-4.0	Perttilä & Brüggmann (1992)
Western Baltic Sea	general	1-2	Brüggmann & Lange (1981)

6.4.2 Radionuclides in sediments and time trends

Since ¹³⁷Cs and ¹³⁴Cs were the most abundant radionuclides in the Chernobyl fallout, the main emphasis in monitoring has focused on them. Trends in the total amounts of ¹³⁷Cs over time in sediment profiles from 12 stations located in different regions of the Baltic Sea are given in Figures 6.4.2.1 and 6.4.2.2. These are stations

where monitoring was carried out every year, though they do not necessarily represent the maximum values in each region. Station C VI is situated in the northern Bothnian Bay and Station EB 1 in the southern Bothnian Sea. However, the highest total amounts per m² were detected in the southernmost part of the Bothnian Bay and in the northernmost part of the Bothnian Sea (Fig. 6.5.1), where the maximum value was 125,000 Bq m⁻². The highest activity concentration of ¹³⁷Cs found in a single sediment slice (1,740 Bq kg⁻¹ d. w.) was also from this station.

At Station C VI the total amount of ^{137}Cs has remained at the same, relatively low level, to which it rose in 1990-1991. In contrast, at Station EB 1 the amount of ^{137}Cs increased considerably in 1992 from the level of previous years (cf. Ilus et al., 1995, Fig. 6.5.1), but after that the values showed a declining trend (Fig. 6.4.2.1). At Station EB 1 the bottom is characterised by strong bioturbation caused by abundant amphipods, so the ^{137}Cs peak has remained in the uppermost surface layer of the sediment through the years, with peak concentrations decreasing evenly from $1,700 \text{ Bq kg}^{-1}$ in 1992 to $1,100 \text{ Bq kg}^{-1}$ d.w. in 1998.

In the Gulf of Finland, the maximum amount of ^{137}Cs ($58,000 \text{ Bq m}^{-2}$) was found at Station XV 1 in 1993 and the highest peak concentration ($4,020 \text{ Bq kg}^{-1}$ d.w.) was in the same sediment profile at a depth of 10-15 cm. The amounts of ^{137}Cs have varied considerably at Station XV 1 from year to year. This is probably due to the very soft and easily mobile surface sediments, which make sampling especially difficult at this location (Ilus et al., 2000). At Station LL3a the total amount of ^{137}Cs has been more stable after a notable increase in 1992. Since 1996 the ^{137}Cs peak has occurred at a depth of 15-20 cm. Both stations are situated in the eastern part of the Gulf of Finland.

At Stations Teili 1 (northern Baltic Proper) and P 1 (southern Baltic Proper) the amounts of ^{137}Cs increased slightly in the 1990s, but in the Gotland Deep (Station BY 15) the deposition of caesium has been very slow. At Station P 110 in the Gulf of Gdansk the total amounts of ^{137}Cs have been quite close to those at Station P 1, but clearly higher than those at stations in the Arkona Sea (ARKO 3) and the Belt Sea (FBELT 1, MEBU 2 and STOLGR). The maximum value reported from the southern part of the Baltic Sea was $7,600 \text{ Bq m}^{-2}$ at the Station P 110 in 1995. In the Belt Sea (Station KFOTN6) the maximum value was $13,000 \text{ Bq m}^{-2}$ in 1992.

This comparison of the ^{137}Cs concentrations in sediments from different sub-regions of the Baltic Sea shows that the strongest sedimentation of ^{137}Cs occurred in the northern Bothnian Sea and the eastern Gulf of Finland. This is in agreement with the distribution pattern of Chernobyl-derived caesium in the catchment area of the Baltic Sea. However, the highest concentrations in sediments are probably not caused by higher site-specific deposition values, but are due to particle transport patterns and the concentration of sinking, particle-bound radionuclides in the deepest points of accumulation basins. In addition to ^{137}Cs , the other important radionuclide in the Chernobyl fallout was ^{134}Cs . In the initial phase after the accident, the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio was about 0.5. Due to

its shorter half-life ^{134}Cs has disappeared from the environment more rapidly than ^{137}Cs . In 1998, the average $^{134}\text{Cs}/^{137}\text{Cs}$ ratio in the sediments of the Gulf of Finland was 0.015.

Concentrations of ^{103}Ru , ^{106}Ru , $^{110\text{m}}\text{Ag}$ and ^{125}Sb clearly increased in Baltic Sea sediments as a consequence of the Chernobyl accident, as well as those of caesium isotopes. The occurrence of these nuclides follows the distribution pattern of the caesium isotopes closely. They were generally most abundant in 1987, but due to their relatively short half-lives they started to decrease by 1988/1989 (Ilus et al., 1995). The nuclides with the shortest half-lives, ^{103}Ru and $^{110\text{m}}\text{Ag}$, were not reported in 1992-1998 and ^{106}Ru was detected only in 4 sediment cores taken in 1992-1994 from the Bothnian Sea, the Arkona Sea and the Belt Sea. ^{54}Mn was reported only from the sampling stations situated in the immediate vicinities of the Swedish NPPs.

In 1992, ^{125}Sb was detected at 10 sampling stations situated in the Bothnian Sea (EB 1), the Gulf of Finland (XV 1 and LL3a), the Northern Baltic Proper (Teili 1) and the Belt Sea (Fig. 6.4.2.3). Maximum concentrations in the sediment cores were about 40 Bq kg^{-1} d.w. at stations on the Gulf of Finland. In 1998, ^{125}Sb , with a half-life of 2.8 years, was still detected at 3 stations: LL3a, Teili 1 and LL23 (Landsort Deep), with a maximum concentration of 15 Bq kg^{-1} d.w..

Observations of ^{60}Co (half-life 5.3 years) in sediments also showed increases in different sub-regions of the Baltic Sea after 1986 (Ilus et al., 1995). In 1994, ^{60}Co was detected at 11 sampling stations (Fig. 6.4.2.4), with the highest concentrations occurring in surface sediments taken from the immediate vicinities of the Swedish Nuclear Power Plants. In the Belt Sea (6 stations), maximum concentrations varied between 0.7 and 2.3 Bq kg^{-1} d.w. In 1998, ^{60}Co was found at 6 stations, in the Northern Baltic Proper (e.g. Landsort Deep), Gotland West, the Belt Sea and the Sound.

^{226}Ra is of natural origin and is more or less evenly distributed in Baltic Sea sediments (Fig. 6.4.2.5). ^{238}Pu and $^{239,240}\text{Pu}$ (Fig. 6.4.2.6) mainly originate from the atmospheric nuclear weapons tests carried out in the late 1950s and early 1960s. Although Chernobyl fallout also included small amounts of plutonium, changes in the amounts of $^{239,240}\text{Pu}$ were small. Due to the different ratios of ^{238}Pu and $^{239,240}\text{Pu}$ in the two fallout types it is possible to detect even the tiny increase of plutonium caused by Chernobyl fallout in the Baltic Sea sediments.

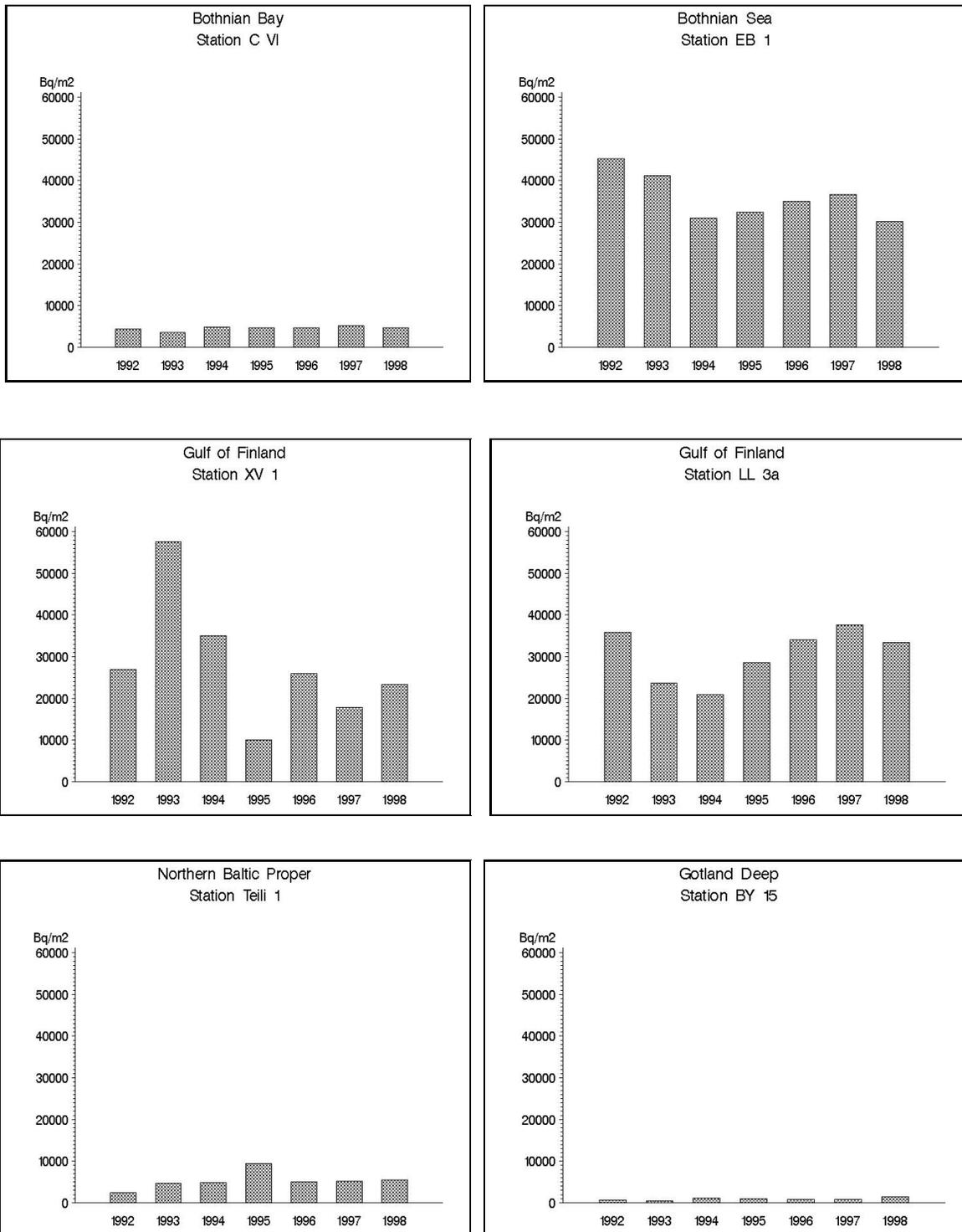


Figure 6.4.2.1 Total amounts of caesium-137 at sampling stations on the Gulf of Bothnia, the Gulf of Finland and the Baltic Proper, 1992-1998.

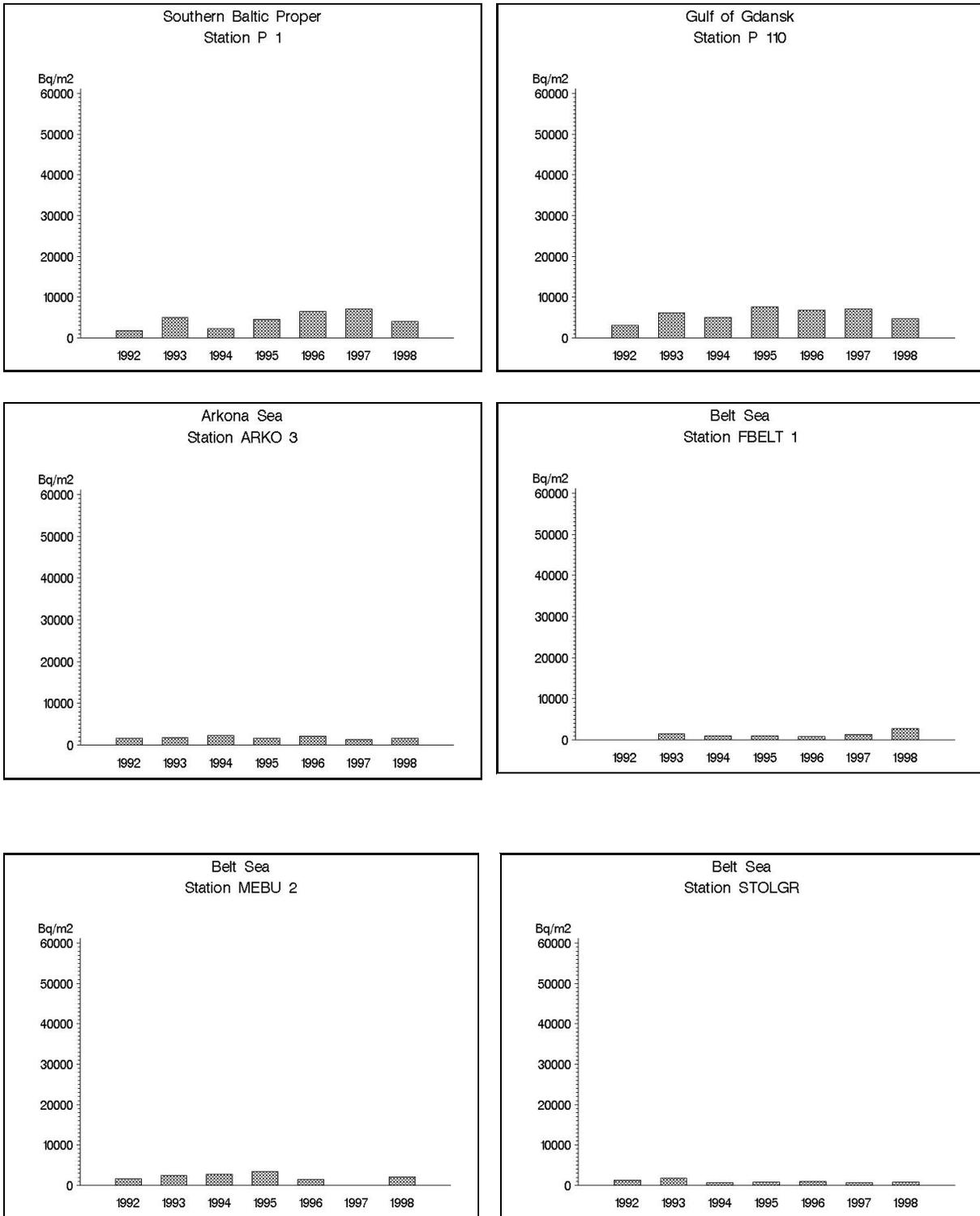


Figure 6.4.2.2 Total amounts of caesium-137 at sampling stations around the southern Baltic Sea, 1992-1998.

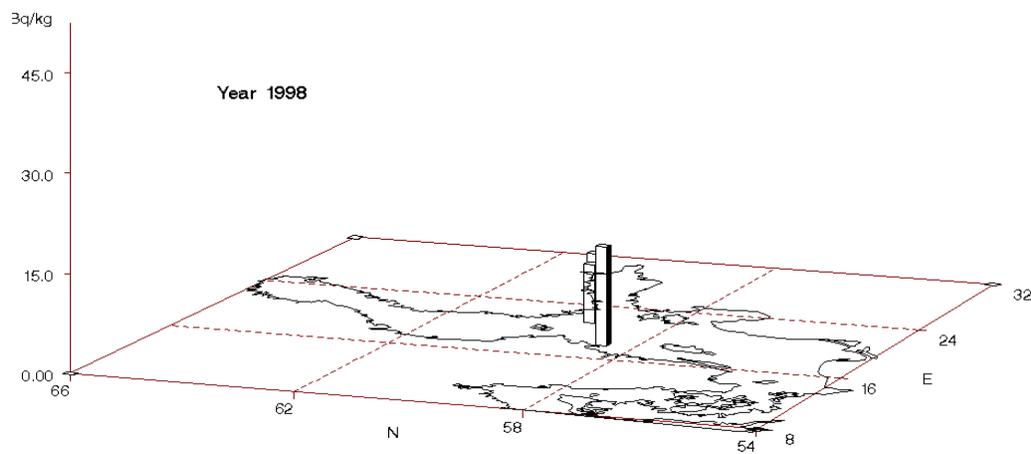
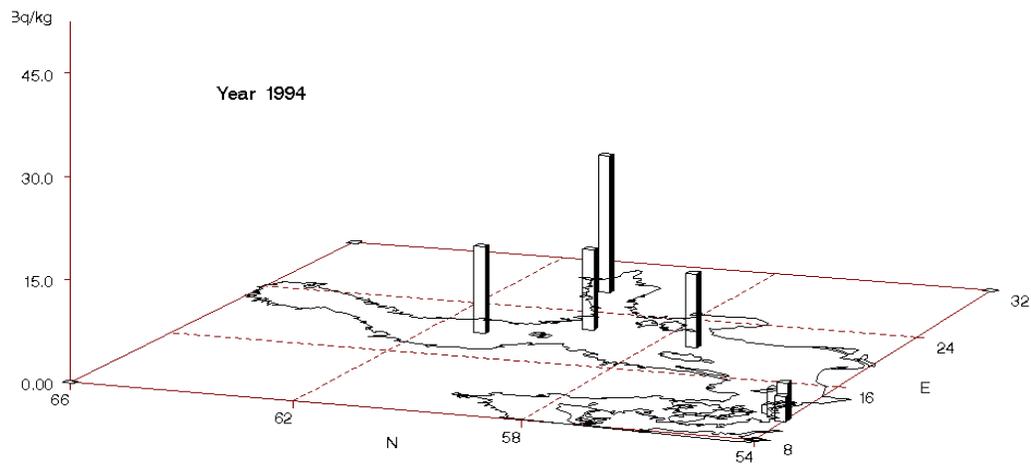
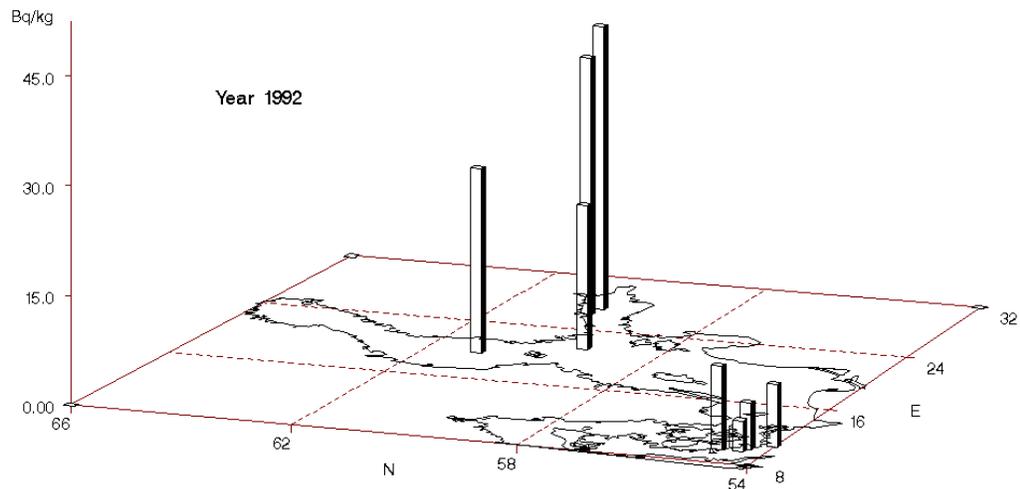


Figure 6.4.2.3 Concentrations of antimony-125 at different sampling stations in 1992, 1994 and 1998, with highest concentrations in sediment cores.

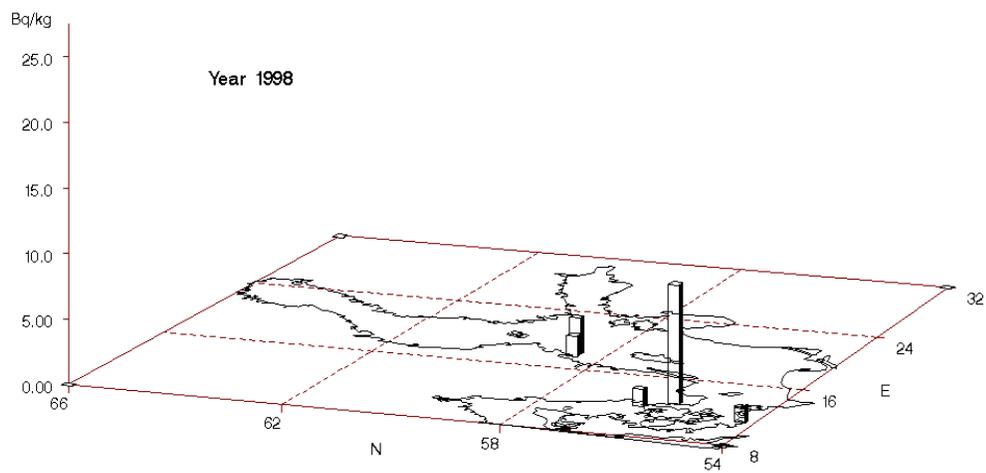
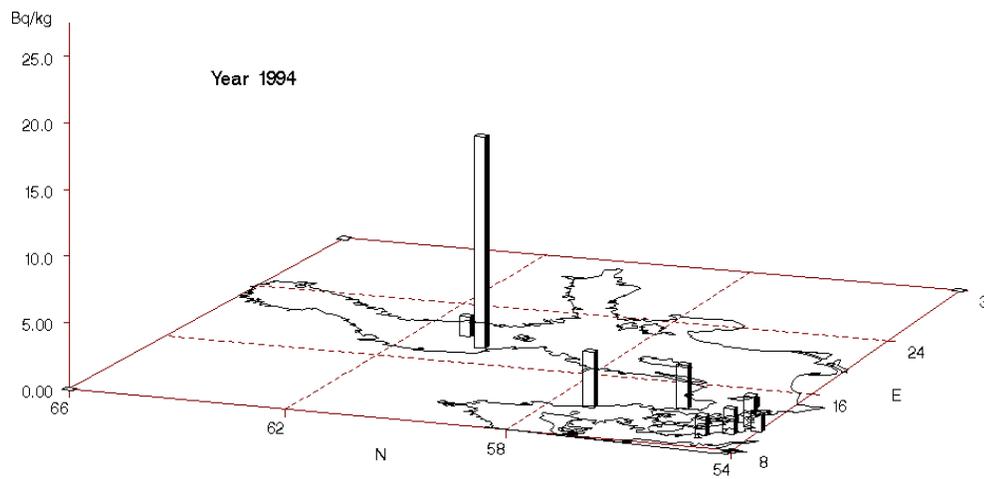
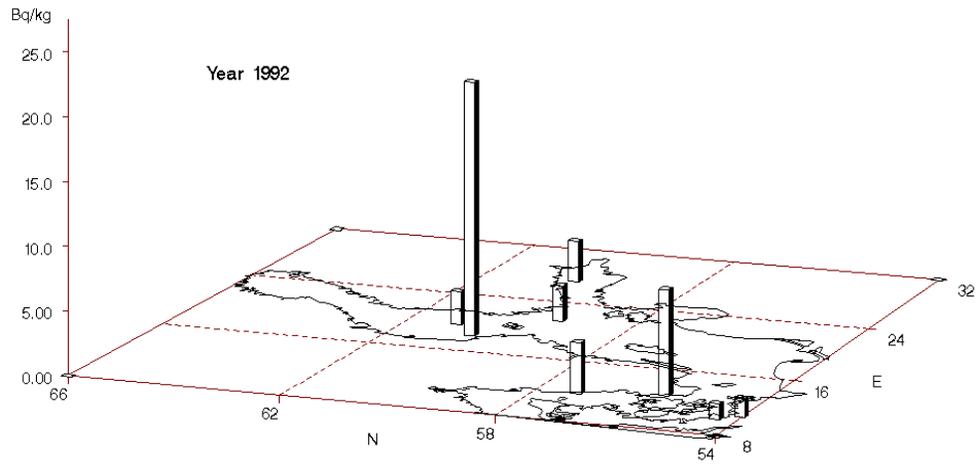


Figure 6.4.2.4 Concentrations of cobalt-60 at different sampling stations in 1992, 1994 and 1998, with highest concentrations in sediment cores.

Figure 6.4.2.5 Radium-226 in surface sediment layers at different sampling stations in 1992.

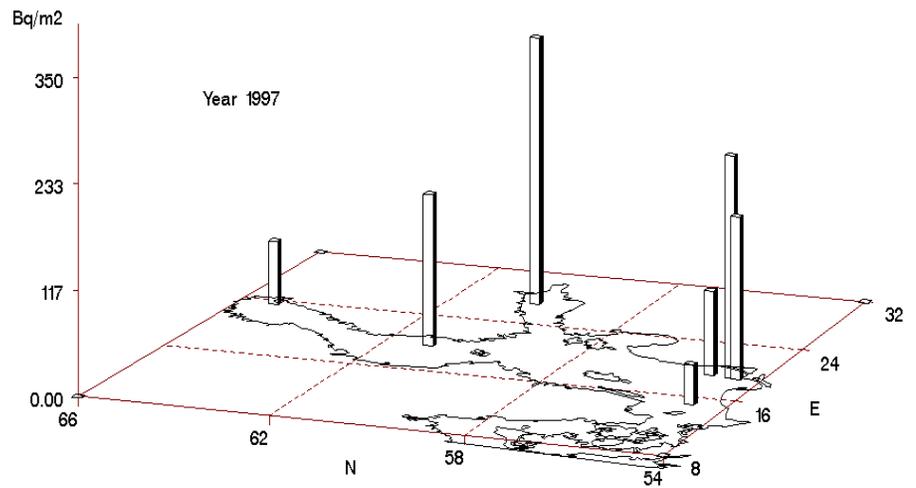
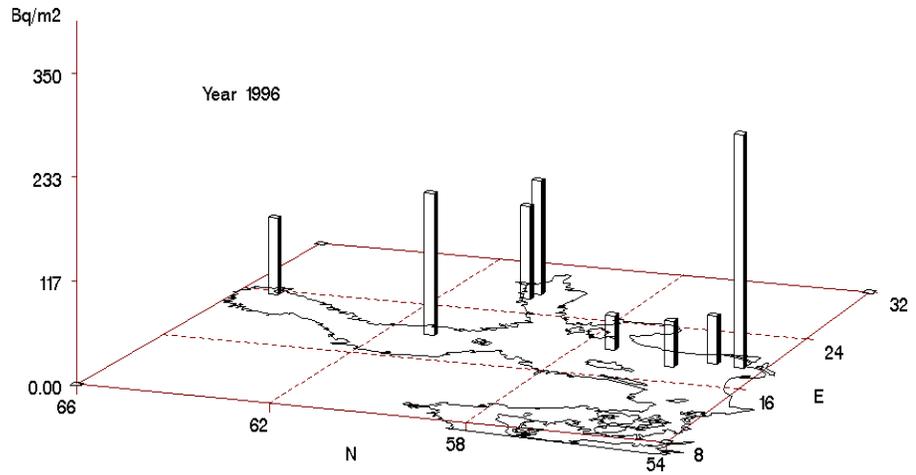
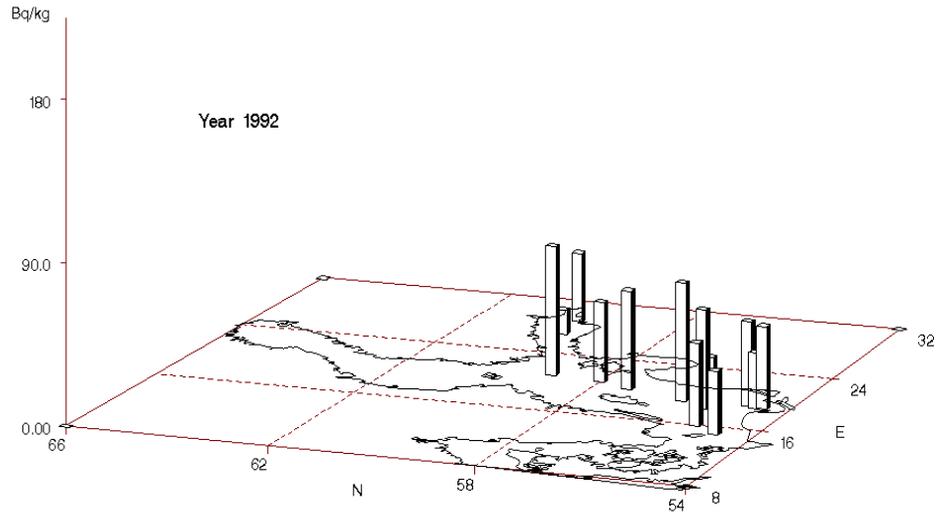


Figure 6.4.2.6 Total amounts of plutonium-239,240 at different sampling stations in 1996 and 1997.

6.5 Inventories in sediments

In recent years several investigations have been carried out to evaluate total inventories of certain long-lived radionuclides in Baltic Sea sediments. Salo et al. (1986) estimated the total amounts of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ bound to bottom sediments in the Baltic Sea at the beginning of the 1980s, using two different calculation methods. In the first case, they estimated the average contents of radionuclides (Bq m^{-2}) in soft and hard sediments in various sea areas and multiplied them by the areas of soft and hard bottoms in each. The second method was based on estimates of approximate mean concentrations of radionuclides in sinking matter and assumed sedimentation rates in different areas of the Baltic Sea. They concluded that at the beginning of the 1980s the total amounts of ^{137}Cs , ^{90}Sr and $^{239,240}\text{Pu}$ bound in Baltic Sea sediments were 277, 12 and 15 TBq, respectively.

The uneven distribution of Chernobyl fallout around the catchment area of the Baltic Sea has made calculations more difficult. Although the global fallout from the nuclear weapons tests was more or less evenly deposited across the northern hemisphere, the deposition of radionuclides in sediments was not evenly distributed at that time, either.

Prior to the evaluation presented here, the total inventory of ^{137}Cs in the Baltic Sea sediments after the Chernobyl accident was estimated through two separate calculations. The estimations were made by the first method used by Salo et al. (1986) based on average contents of radionuclides (Bq m^{-2}) in soft and hard sediments in various sea areas. In the first calculation (Ilus et al., 1995), it was estimated that the total amount of ^{137}Cs in Baltic Sea sediments was 1,400 TBq in 1990-1991 and that of $^{239,240}\text{Pu}$ was 18 TBq in 1987-1988. We were not able to estimate the total amount of ^{90}Sr , because very little data on strontium in sediments has been reported for the database since 1986. Salo et al. (1986) assumed that ^{90}Sr and ^{137}Cs are distributed between hard and soft bottoms in a ratio of 1/5, while for $^{239,240}\text{Pu}$ the ratio is about 1/10. Subsequent results have indicated that the ratio for ^{137}Cs may be much lower (1/20), which would lower the figure for the total inventory. By using this value, an estimate for ^{137}Cs in 1990-1991 of 1,200 TBq was calculated. All the above mentioned estimates were based on data from the HELCOM/MORS database.

Relatively few observations were used in the evaluation described above. In the second attempt (Ilus et al., 1999), the study material was more comprehensive, consisting of 129 sampling stations and 180 sediment cores taken by STUK and the Finnish Institute of Marine Research in

1993-1997 from different sub-regions of the Baltic Sea. In this evaluation activity concentrations were time-corrected to April 26th, 1996 (the 10th anniversary of the Chernobyl accident) and a ratio of 1/20 was used to calculate ^{137}Cs values for hard bottoms. According to this investigation, the total inventory of ^{137}Cs in Baltic Sea sediments was calculated as 2,140 TBq in 1996. The significant difference between this value and the value given above is presumed to result from the additional data available on ^{137}Cs in sediments, and the fact that Chernobyl-derived caesium had continued to be deposited in sea-bed sediments.

The present study is based on the ^{137}Cs data reported by the Contracting Parties to the HELCOM/MORS database, annexed with additional data from STUK and the Finnish Institute of Marine Research, which were not reported for the database. Before starting the calculations, data quality was controlled and obviously questionable values were omitted. These questionable values were identified by comparing the results given by different laboratories for samples from the same sampling stations, for instance. The latest observations reported by the laboratories for each station were then selected for manual checking of the results. After checking, the accepted values were used for calculating averages for each station. The sampling stations were grouped according to the respective sub-regions of the Baltic Sea, and the median value for each sub-region was chosen to represent the area in question. The median was used, because averages were dominated by a few, very high "hot spot" values. The spatial distribution of the total amounts of ^{137}Cs at different sampling stations in the Baltic Sea is shown in Figure 6.5.1.

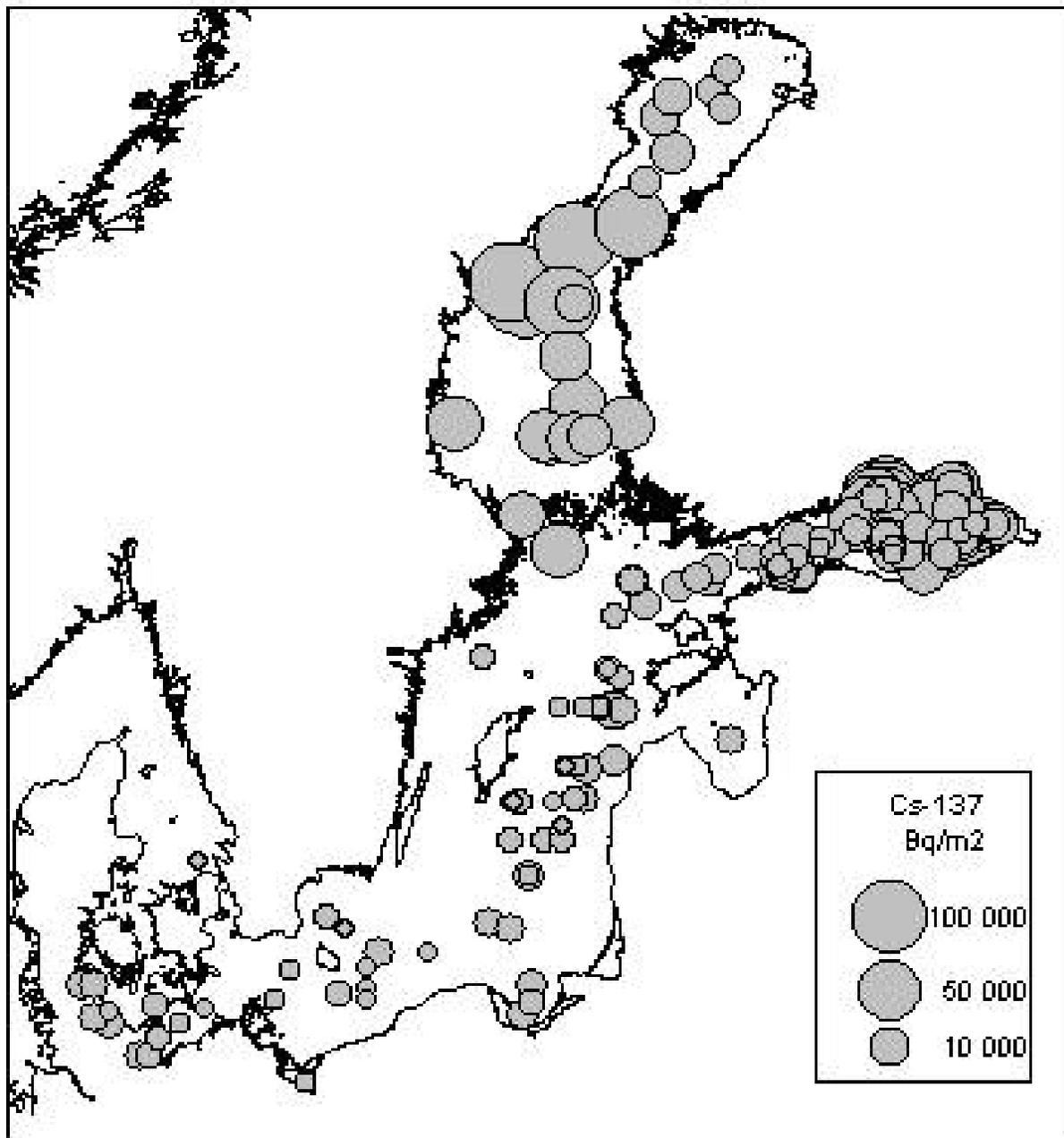


Figure 6.5.1 Total amounts of ^{137}Cs [Bq m^{-2}] at different sampling stations in 1998.

Sediment samples were usually taken from soft bottoms, i.e. from the sedimentation bottoms of deep basins. Soft bottoms very often act as "sinks" for radionuclides, whereas hard bottoms are regarded as transport bottoms with very low accumulation rates for sinking matter. However, erosion bottoms are very seldom uncontaminated, because bioturbation caused by benthic fauna may transfer contaminants and organic material into deeper sediment layers. Studies carried out on the Polish coast have shown that ^{137}Cs penetrates effectively into near-shore sandy sediments, and that rapidly accumulating sediments affected by river discharges have much higher contents of exchangeable radiocaesium than slowly accumulating marine sediments

(Knapinska-Skiba et al., 1994, 1995, 1997).

In this study, the two alternative ratios (1/5 and 1/20) were used to calculate ^{137}Cs values for hard bottoms. The values for hard bottoms were calculated from the above-mentioned median values for each sub-region. The content of ^{137}Cs (Bq m^{-2}) on soft and hard bottoms in different sub-basins was multiplied by the area of soft and hard bottoms in each, according to the values given by Salo et al. (1986). The values have been measured planimetrically from maps of Quaternary deposits in the Baltic Sea (Winterhalter et al., 1981).

Table 6.5.1 Areas of soft and hard bottoms [km²] in different regions of the Baltic Sea (Salo et al., 1986).

Sub-region	Total	Soft	Hard
Bothnian Bay	37,000	16,000	21,000
Bothnian Sea	79,000	40,000	39,000
Gulf of Finland	30,000	16,000	14,000
Baltic Proper	209,000	99,000	110,000
Gulf of Riga	19,000	7,000	12,000
Total	374,000	178,000	196,000

Depending on the method used, the total inventory of ¹³⁷Cs in the seabed of the Baltic Sea was estimated at 1,940 – 2,210 TBq in 1998. The inventories for different sub-regions are given in Table 6.5.2.

The Belt Sea, the Kattegat and the Sound were not included in the inventory, due to the lack of data on the proportion of soft and hard bottoms. In sediments within the Polish Economic Zone the total inventory of ¹³⁷Cs increased from 10 to 45 TBq as a consequence of the Chernobyl accident (Bojanowski et al., 1995a, b).

6.6 Conclusions

Bottom sediments play an important role in radioecological studies of the marine environment, because a large proportion of the radioactive substances entering the sea is adsorbed over time onto suspended particulate matter, and subsequently deposited in sediments. Sediments are generally a final sink for most of the organic material produced in the water phase, as well as for other particles transported by water currents from other parts of the sea and from adjacent terrestrial areas. During their slow settling, these particles tend to bind radionuclides from the water phase and drag them to the bottom. In favourable conditions the deposited particles form undisturbed laminae in a stratigraphic sequence on the seabed, and the bottom sediments create an archive recording the history of the area.

To date only the accident at the Chernobyl NPP in 1986 and the atmospheric nuclear weapons tests carried out in the 1950s and 1960s have significantly affected the amounts of artificial radionuclides in Baltic Sea sediments. During the reporting period 1992-1998, there were no such

Table 6.5.2 Inventories of ¹³⁷Cs in different sub-regions of the Baltic Sea in 1998 based on the areas of soft and hard bottoms in each.

Sub-region	Number of stations	Sum (1/20) TBq	Sum (1/5) TBq	Percent of total %
Bothnian Bay	8	140	165	7
Bothnian Sea	13	1,370	1,560	71
Gulf of Finland	78	230	255	12
Baltic Proper	35	175	205	9
Gulf of Riga	1	23	1	
Total	135	1,938	2,208	100

events with any notable impact on the total inventories of radionuclides. Thus the main emphasis was on following the occurrence and behaviour of fallout nuclides originating from these earlier events in sediments. In this sense, monitoring has focused on sedimentation processes and sedimentation rates for radionuclides, as well as on the accumulation of the fallout nuclides in sediments.

Since ¹³⁷Cs and ¹³⁴Cs were the most abundant radionuclides in the Chernobyl fallout, the main emphasis in monitoring has focused on them. Concentrations of ¹⁰³Ru, ¹⁰⁶Ru, ^{110m}Ag and ¹²⁵Sb also clearly increased in Baltic Sea sediments as a consequence of Chernobyl. The occurrences of these nuclides closely followed the distribution patterns of the caesium isotopes. They were generally most abundant in 1987, but due to their relatively short half-lives they already started to decrease by 1988/1989. Concentrations of ⁶⁰Co (half-life 5.3 years) observed in sediments also increased in various sub-regions of the Baltic Sea. In 1998, ⁶⁰Co was found at 6 stations, in the Northern Baltic Proper (e.g. Landsort Deep), Gotland West, the Belt Sea and the Sound.

The highest total amounts of ¹³⁷Cs per m² were detected in the southernmost part of the Bothnian Bay and in the northernmost part of the Bothnian Sea, where the maximum value was 125,000 Bq m⁻². The highest activity concentration of ¹³⁷Cs found in a single sediment slice (1,740 Bq kg⁻¹ d. w.) was also found here. The total inventory of ¹³⁷Cs in the seabed of the Baltic Sea was estimated at 1 940 - 2 210 TBq in 1998. This was about 8 times higher than the inventory made at the beginning of the 1980s (277 TBq) and about one and a half times higher than an earlier estimate made by this project group in 1990-1991.

6.7 Recommendations

This study has proved that our knowledge on distribution of radionuclides in Baltic Sea sediments is still insufficient. The Swedish waters of the Baltic Proper in particular have not yet been thoroughly investigated.

It must be remembered that long time-series of results are especially valuable in this monitoring context.

In recent years very little data on ^{90}Sr in Baltic Sea sediments has been reported for the HELCOM/MORS database. Updating this data is one of the most important challenges for the near future.

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7 RADIONUCLIDES IN BIOTA

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Levels of radionuclides in biota of marine origin are linked to the corresponding levels in sea water and sediment via accumulation through food chains. The complexity of food chains increases with the trophic level of the species considered. Fish, the most important biota type in the Baltic Sea for human consumption, accumulate most of the radionuclides from their food, rather than directly from the water.

Baltic Sea biota received the most important contribution to their radionuclide levels following the Chernobyl accident in 1986, predominantly in the form of ^{137}Cs and ^{134}Cs . The ratio $^{134}\text{Cs}/^{137}\text{Cs}$ in biota agrees very well with that of Chernobyl fallout. Time trends for ^{137}Cs in fish did not always closely follow the corresponding trends in sea water. The high trophic level species, including predators such as cod and pike, showed the highest ^{137}Cs levels, but with some delay compared to sea water in reaching maximum values after 1986. In the long term, ^{137}Cs time trends in biota evidently follow the trends in sea water.

The levels of ^{137}Cs in fish samples (mainly herring and pike) from the northern regions of the Baltic Sea, where the initial concentrations in sea water were the highest after Chernobyl, have decreased since the end of the 1980s. Levels of ^{137}Cs in herring and pike from the Bothnian Bay approached 20 and 40 Bq kg^{-1} , respectively. Slightly lower values were observed in the Gulf of Finland. Figure

7.1 contains data on ^{137}Cs in herring caught in the Baltic Sea during the period 1984-1998.

In the Baltic Proper, the area with the highest production of fish for human consumption, levels of ^{137}Cs in fish increased until the beginning of the 1990s and then decreased gradually. In more southerly waters, where the network of sampling stations is densest, ^{137}Cs levels in herring and cod varied between 10 and 20 Bq kg^{-1} in the second half of the 1990s. Benthic flat-fish and small sprats showed lower values of around 10 Bq kg^{-1} in the 1990s.

Fish from the Belt Sea showed lower values. Average ^{137}Cs levels in herring have been below 3 Bq kg^{-1} since 1990, at levels previously observed during the period 1965-1974. Similar trends were observed for plaice. ^{137}Cs levels in cod stayed at around 10 Bq kg^{-1} after 1987, at levels nearly twice as high as those observed during the period 1965-1974.

Fish from the Kattegat area showed the lowest ^{137}Cs levels: below 4 Bq kg^{-1} for herring and cod from about 1990 onwards, with no obvious impact from the Chernobyl accident.

Marine algae (*Fucus vesiculosus*) from nuclear power plant (NPP) monitoring stations in the Baltic Sea, used as a bio-indicator of radionuclides in the Baltic Sea, show low activities of radionuclides representing NPP discharges, such as ^{60}Co , ^{58}Co , ^{54}Mn , ^{65}Zn , and $^{110\text{m}}\text{Ag}$. Some of these radionuclides were also found in samples of benthic animals (e.g. *Mytilus edulis*, *Macoma baltica*, *Saduria entomon*). ^{137}Cs concentrations in *Fucus vesiculosus* from various parts of the Baltic Sea closely followed the ^{137}Cs time-trend for surface sea water.

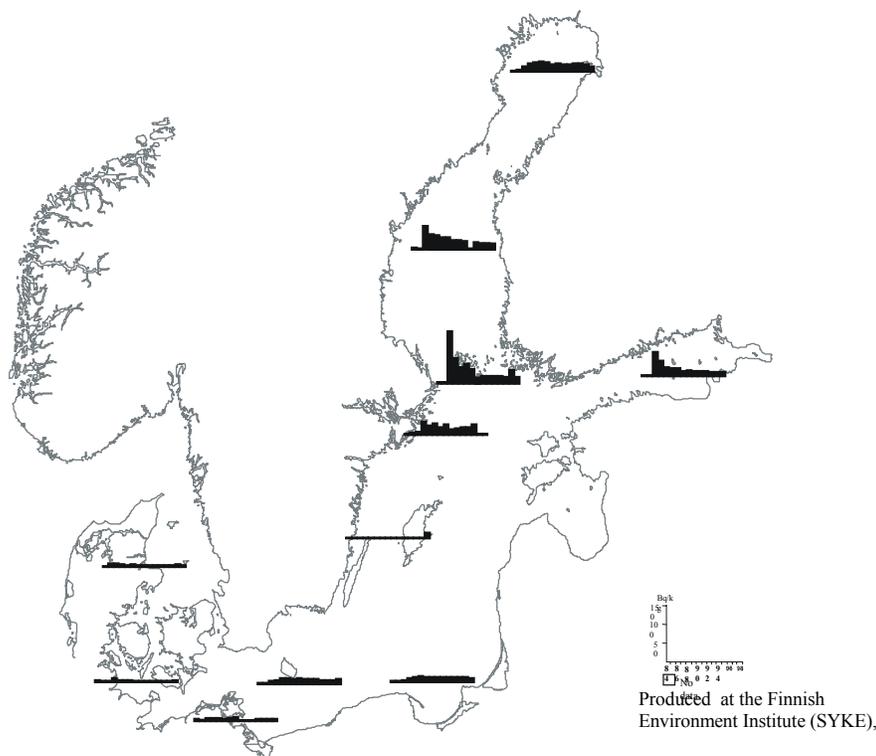


Figure 7.1 ^{137}Cs concentrations (Bq kg^{-1}) in herring, 1984-1998

8 MODELLING THE TRANSFER OF RADIONUCLIDES IN THE BALTIC SEA

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8.1 Introduction

The box model used here includes a description of the physical dispersion of radionuclides in the marine environment, a description of the transfer of radionuclides to biota, and calculations of doses to individuals and populations from a range of marine exposure pathways. The model covers the coastal waters of the North-East Atlantic, including the Baltic Sea. The whole model, rather than just the part covering the Baltic Sea, is used to take into account the transfer of radionuclides from the two European nuclear reprocessing plants, Sellafield and La Hague, into the Baltic Sea. This model is described in greater detail elsewhere (EC, 2000).

8.2 Model description

The model has been developed for the assessment of the radiological consequences of releases of radioactive material into the marine environment. It is applicable for European coastal waters, including the Baltic Sea. The model simulates the dispersion of radioactivity in water due to advective transport, including mixing caused by winds and tides. The association of radionuclides with suspended sediment material and their subsequent transfer into sediments through particle scavenging are also taken into consideration. Further mechanisms involved in the transfer of radionuclides between the water column and the sediments include diffusion, bioturbation and re-suspension. Given specified inputs of radioactivity into the marine environment, the model calculates time-dependent concentrations in sea water and sediments. This data is then used to calculate doses to man from a range of exposure pathways: ingestion of fish, crustaceans, molluscs or seaweed; external exposure on beaches; inhalation of sea spray and re-suspended beach sediments. Doses are calculated for individual members of critical groups and for wider populations. Compartment or box-model analysis is used to simulate the movement of radionuclides between different parts of the marine environment. Box-model analysis assumes instantaneous uniform mixing within each box, with rates of transfer across the boundaries of boxes being proportional to the inventories of material in the source boxes. The box-model analysis uses first order differential equations to describe the transfer of contaminant radionuclides between the boxes. Most coastal areas are represented with one-layer water boxes and underlying sediment

boxes, but some areas with stratification (e.g. the Baltic Sea Proper) include both surface and deep waters. Sediments are represented by two layers: a surface layer and a deeper layer. The model includes 92 water and surface-sediment boxes.

Figure 8.1 shows the regions used in the marine box model near the Baltic Sea. Each of the water compartments has associated suspended sediments and the water compartments in contact with the sea-bed have underlying sea-bed sediment compartments. The water compartments have odd numbers and the surface sediment compartments have even numbers.

The water compartment names, volumes and mean depths are given in Table 8.1.

Suspended sediment particles in coastal waters are partly maintained by a local depth-dependent re-suspension of surface-sediment particles to the water column, due to the mechanical transfer of energy via wind and tidal forces to surface sediments. The transfer of radioactivity from the top surface sediment to lower sediment layers is accounted for by assuming that the burial rate is equal to the flux of particles settling from overlying waters. Radioactive decay is accounted for in all boxes.

8.3 Testing the model's reliability

Data has been collected over several decades on environmental concentrations of the radionuclides ^{90}Sr and ^{137}Cs in the sea water, sediments and biota in the Baltic Sea (EC, 2000). This data is well suited for testing the reliability of model predictions.

8.3.1 Source terms

In order to compare predicted environmental levels of ^{137}Cs and ^{90}Sr with observations, the following major source terms were considered as input to the model: fallout from nuclear weapons tests, Chernobyl fallout, liquid discharges from the European reprocessing facilities at Sellafield and La Hague, and liquid discharges from nuclear installations bordering the Baltic Sea.

The fallout of ^{137}Cs and ^{90}Sr from atmospheric nuclear tests has been included based on measurements made in Denmark. This data, which covers the period from 1955 to 1996, is considered representative for the Baltic Sea area, and compares well with corresponding data collected near St. Petersburg. Each model surface compartment thus receives an annual input of ^{137}Cs and ^{90}Sr according to its surface area. The influence of the Chernobyl accident was eliminated from this source by extrapolating the fallout from nuclear weapons tests for the period 1986-1996 based on exponential regression of the data observed in the period 1970-1985.

The catchment area of the Baltic Sea ($1.7 \cdot 10^{12} \text{ m}^2$) is about four times as extensive as the sea itself ($4.0 \cdot 10^{11} \text{ m}^2$), and the transfer of terrestrial fallout in river water to the sea is an important contribution to

consider. Salo et al. (1985) investigated inventories of ^{137}Cs and ^{90}Sr in sea water and sediments in the Baltic Sea for the period 1961-1981 and produced data for the total runoff of ^{137}Cs and ^{90}Sr in the region's rivers. This data shows that the river runoff of ^{90}Sr for the period 1961-1981 constitutes an important contribution to the total input of ^{90}Sr into sea water; and since 1972 this river runoff has outweighed the direct fallout from the atmosphere into the sea. For ^{137}Cs , river runoff is less important - of the order of a few per cent of the atmospheric fallout into the water. Two simple sub-models have been constructed to incorporate this data, allowing extrapolations to be made for the runoff of ^{137}Cs and ^{90}Sr into the Baltic Sea. For the model calculations the total runoff for each radionuclide was split between the different Baltic Sea sub-regions according to the sizes of their corresponding catchment areas (EC, 2000).

The direct input of ^{137}Cs from the Chernobyl accident into the Baltic Sea has been estimated at 4.5 PBq (HELCOM, 1995). However, there are indications that the input to the Gulf of Finland was underestimated, so an adjustment has been made to account for this. The values used here for Chernobyl fallout in the various regions of the Baltic Sea are given in Table 8.2.

The values used for the amount and distribution of radiocaesium over the Baltic Sea are based on HELCOM information (HELCOM, 1995). Data from Denmark, Finland and Russia shows that the fallout of ^{90}Sr from the Chernobyl accident amounted to about 2% of the ^{137}Cs fallout.

The reported discharges of ^{137}Cs and ^{90}Sr from Sellafield (BNFL, 1996) into the Irish Sea, and La Hague (EC, 2000) into the English Channel, have been used as input in the model. However, only a small proportion of these discharges reaches the Baltic Sea. The transfer into the Baltic Sea has nevertheless been estimated through the model calculations, which indicate that about 4% of the discharges from Sellafield eventually reach the Kattegat, compared to about 8% of the discharges from La Hague. The relative transfer of ^{137}Cs is lower (about 10% relative difference) than that of ^{90}Sr because of chemical differences that result in higher transfer rates into sediments for ^{137}Cs than for ^{90}Sr . Due to the efficient mixing of the upper and lower waters in the Kattegat and the Belt Sea, most of the activity of these radionuclides from the two reprocessing plants returns to the Skagerrak, and only about 1% of the total discharges of ^{137}Cs and ^{90}Sr is estimated to be transferred into the Baltic Proper.

Table 8.3 presents an overview of the discharges into the Baltic Sea during the period 1950-1996 which have been considered for the present calculations.

8.3.2 Results for ^{137}Cs

Calculations were carried out according to the model for the time period 1950 to 2000 using the source terms specified in the previous section. The

calculated concentrations of ^{137}Cs in sea water were compared with observed levels. This comparison is illustrated in Figure 8.2, which shows a scatter graph for all ($n=254$) observed and predicted concentrations of ^{137}Cs in sea water. The line marks the ideal 1:1 relationship, and the points are scattered on both sides of the line. The geometric mean of the predicted-to-observed (P/O) ratios is 0.8, with a geometric standard deviation of 1.5. Thus the model on average tends to under-predict the concentrations of ^{137}Cs in sea water in the Baltic by about 20%.

The calculated concentrations of ^{137}Cs in fish were also compared with observed levels, as illustrated in Figure 8.3, which shows a scatter graph for all ($n=105$) observed and predicted annual mean concentrations of ^{137}Cs in fish. The line marks the ideal 1:1 relationship. The geometric mean of the predicted-to-observed (P/O) ratios is 0.9, with a geometric standard deviation of 2.2. Thus on average the model under-predicts the concentrations of ^{137}Cs in Baltic fish by about 10%.

A field study was carried out in 1996 on concentrations of ^{137}Cs in surface sediments from coastal areas around the Baltic Sea, in order to obtain experimental data for comparison with model calculations of ^{137}Cs concentrations in coastal sediments. This data was used in the model calculations for the assessment of doses from external exposure and inhalation, in connection with beach occupancy. In the Marina Study (CEC, 1990) reference was made to a similar investigation carried out in the UK, where calculated levels of radionuclides in coastal areas were found to over-predict the observed levels by an order of magnitude, and a corresponding correction was consequently applied in dose calculations. This comparison shows that the calculated concentrations are significantly higher than the observed concentrations – on average by a factor of 4. This correction factor has been applied in the model calculations of doses from beach occupancy.

8.3.3 Results for ^{90}Sr

The calculations carried out for ^{90}Sr were similar to those done for ^{137}Cs . Figure 8.4 shows a scatter graph of the observed and predicted levels of ^{90}Sr in sea water. The geometric mean of the P/O values ($n=261$) is 0.8, with a geometric standard deviation of 1.4. This corresponds to an average under-prediction of 20% by the model for concentrations of ^{90}Sr in sea water in the Baltic.

The calculated concentrations of ^{90}Sr in fish are compared with observed values in Figure 8.5, which shows a scatter graph for all ($n=80$) observed and predicted annual mean concentrations of ^{90}Sr in fish. The line gives the ideal 1:1 relationship. The geometric mean of the predicted-to-observed (P/O) ratios is 0.5, with a geometric standard deviation of 3.5. This shows that the model on the average under-predicts the concentrations of ^{90}Sr in Baltic fish by 50%.

8.4 Conclusions and recommendations

Transfers of radionuclides in the marine environment have been calculated with a model developed for the waters of the North Atlantic, including the Baltic Sea. The model accounts for the dispersion of radionuclides in the marine environment, the transfer of radionuclides to biota, and the calculation of doses to individuals and populations exposed to radionuclides in seafood.

The dispersion model is based on box-model analysis, and includes 12 water boxes and 12 sediment boxes for the Baltic Sea area. The physical processes covered by the model are net advection and the mixing of water between adjacent boxes, the sedimentation of particulate material from the water column into the top sediment, and the biological mixing of the top sediment. The model is intended for the prediction of annual average concentrations of radionuclides in the marine environment. Concentrations of radionuclides in biota are calculated from

concentrations of radionuclides in filtered seawater. The quality of the model predictions was investigated by comparing predicted levels of ^{137}Cs and ^{90}Sr in water and fish with observed levels. The predicted concentrations were generally in good agreement with the observations for both radionuclides. For the dominating pollutant radiocaesium, the model under-predicts the concentrations in fish by 10% on average, and shows individual agreement generally within a factor of 2. The reliability of the model predictions is therefore considered as satisfactory for radiological-assessment purposes.

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Box No.	Region	Volume (m ³)	Depth (m)
1	Other Oceans	1.0E+18	4.0E+03
3	Atlantic Ocean	3.0E+17	3.5E+03
5	North-East Atlantic	5.0E+16	3.5E+03
7	Arctic Ocean	1.7E+16	1.2E+03
9	Spitsbergen Waters	1.0E+14	1.2E+03
11	Barents Sea	3.0E+14	2.0E+02
13	Norwegian Coastal Waters	1.0E+15	1.2E+03
15	Scottish Waters West	1.0E+13	1.1E+02
17	Scottish Waters East	3.0E+12	1.1E+02
19	Irish Sea North-West	4.1E+11	9.3E+01
21	Irish Sea North	6.0E+10	3.4E+01
23	Irish Sea North-East	5.2E+10	2.4E+01
25	Irish Sea West	6.6E+11	6.3E+01
27	Irish Sea South-East	1.6E+11	3.1E+01
29	Cumbrian Waters	3.8E+10	2.8E+01
31	Irish Sea South	1.1E+12	5.7E+01
33	Liverpool + Morecambe Bay	3.2E+10	1.3E+01
35	Celtic Sea	2.0E+13	1.5E+02
37	Bristol Channel	1.0E+12	5.0E+01
39	Bay of Biscay	6.5E+14	4.0E+03
41	French Continental Shelf	3.5E+13	3.5E+02
43	Cantabrian Sea	3.0E+13	7.6E+02
45	Portuguese Continental Shelf	1.5E+13	4.9E+02
47	Gulf of Cadiz	2.3E+14	1.7E+03
49	Mediterranean Sea	4.0E+15	1.3E+03
51	English Channel West	3.2E+12	6.0E+01
53	English Channel South-East	6.5E+11	4.0E+01
55	English Channel North-East	6.5E+11	4.0E+01
57	North Sea South-West	4.5E+11	3.1E+01
59	North Sea South-East	9.5E+11	3.7E+01
61	North Sea Central	1.3E+13	5.0E+01
63	North Sea East	1.2E+12	2.2E+01
65	North Sea North	5.6E+13	2.4E+02
67	Skagerrak	6.8E+12	2.1E+02
69	Kattegat, deep	2.0E+11	1.0E+02
71	Kattegat, surface	3.2E+11	2.0E+01
73	Belt Sea, deep	1.4E+11	3.0E+01
75	Belt Sea, surface	1.5E+11	1.4E+01
77	Baltic Sea West, deep	7.7E+11	1.1E+02
79	Baltic Sea East, deep	1.5E+12	1.1E+02
81	Baltic Sea West, surface	3.8E+12	4.9E+01
83	Baltic Sea East, surface	7.0E+12	5.3E+01
85	Bothnian Sea	4.9E+12	6.2E+01
87	Bothnian Bay	1.5E+12	4.1E+01
89	Gulf of Finland	1.1E+12	3.7E+01
91	Gulf of Riga	4.1E+11	2.3E+01

Table 8.1 Water-compartment names, volumes and mean depths.

Table 8.2 Values for ^{137}Cs fallout from Chernobyl in different parts of the Baltic Sea, as used in this assessment.

Region	Chernobyl ^{137}Cs (TBq)
Belt Sea	60
West Baltic	1000
East Baltic	500
Bothnian Sea	2400
Bothnian Bay	200
Gulf of Finland	500
Gulf of Riga	40
Total	4700

Table 8.3 Discharges of ^{137}Cs and ^{90}Sr into the Baltic Sea, 1950-1996.

Source	^{137}Cs (TBq)	^{90}Sr (TBq)
Fallout from nuclear weapons tests; direct deposition into sea	1800	1130
Fallout from nuclear weapons tests; in river runoff	100	400
Chernobyl fallout	4700	100
European reprocessing plants	400	70
Total	7000	1700

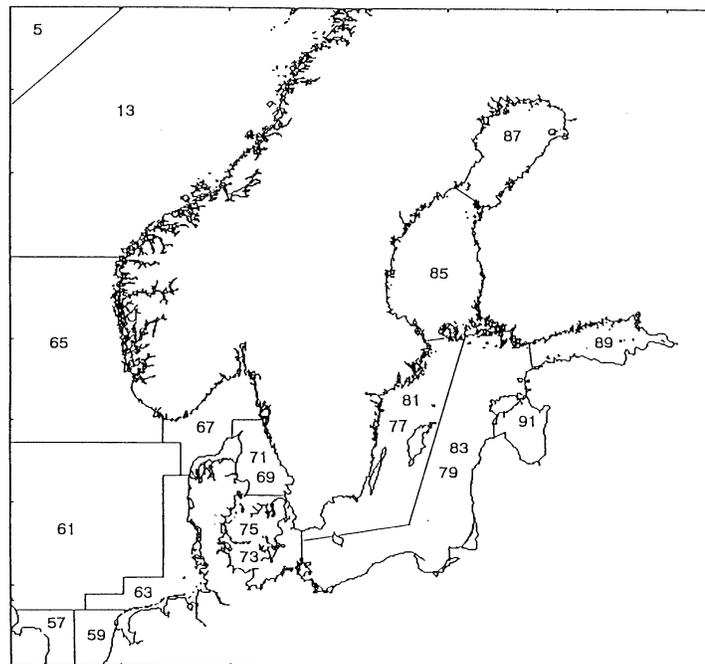


Figure 8.1 Regions covered by the marine box model. The numbers refer to the water boxes.

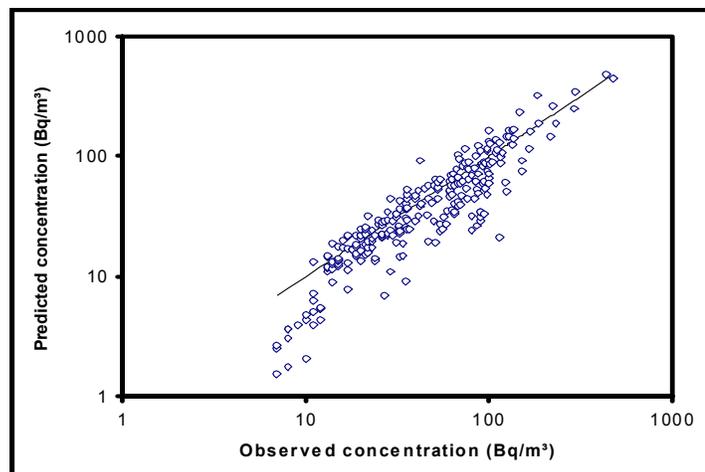


Figure 8.2 Scatter plot of observed and predicted annual mean concentrations of ^{137}Cs in Baltic sea water. The line indicates a 1:1 relationship.

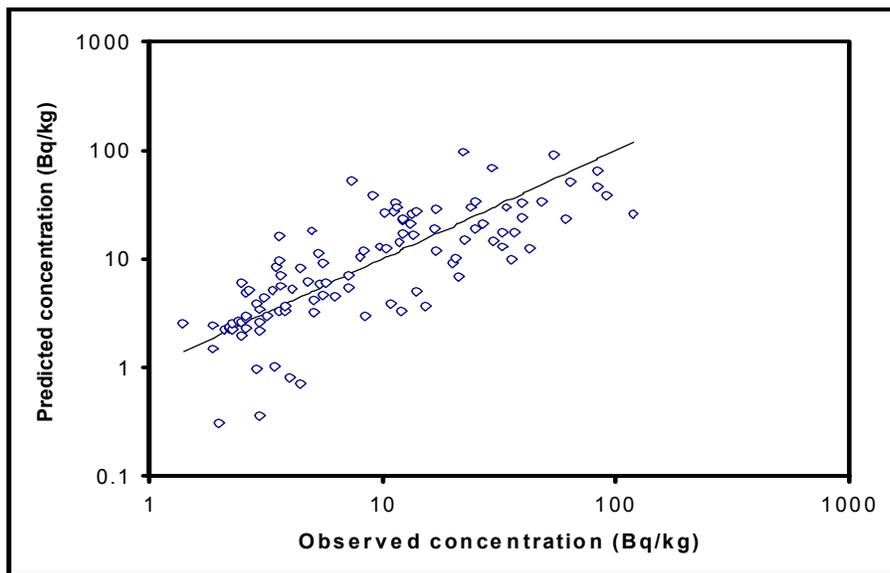


Figure 8.3 Scatter plot of observed and predicted annual mean concentrations of ^{137}Cs in Baltic fish. The line indicates a 1:1 relationship.

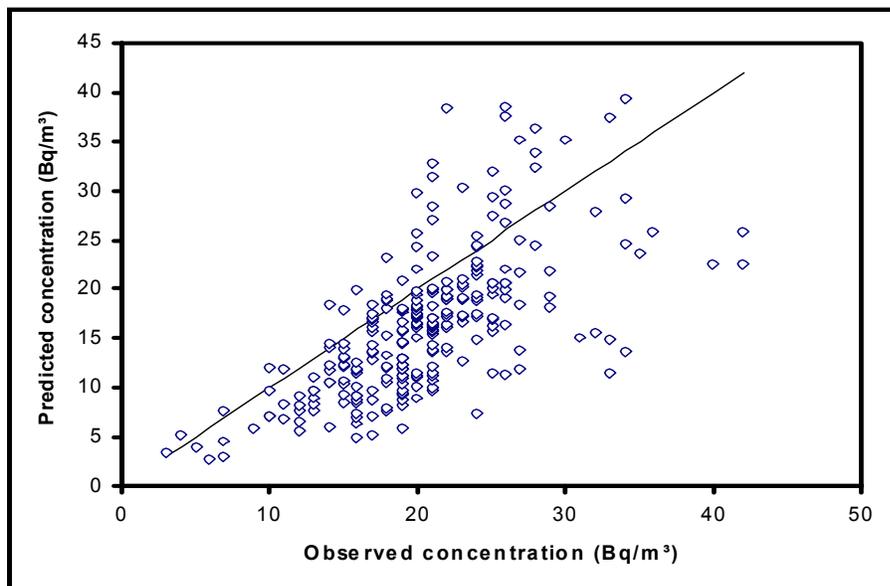


Figure 8.4 Scatter plot of observed and predicted annual mean concentrations of ^{90}Sr in Baltic sea water. The line indicates a 1:1 relationship.

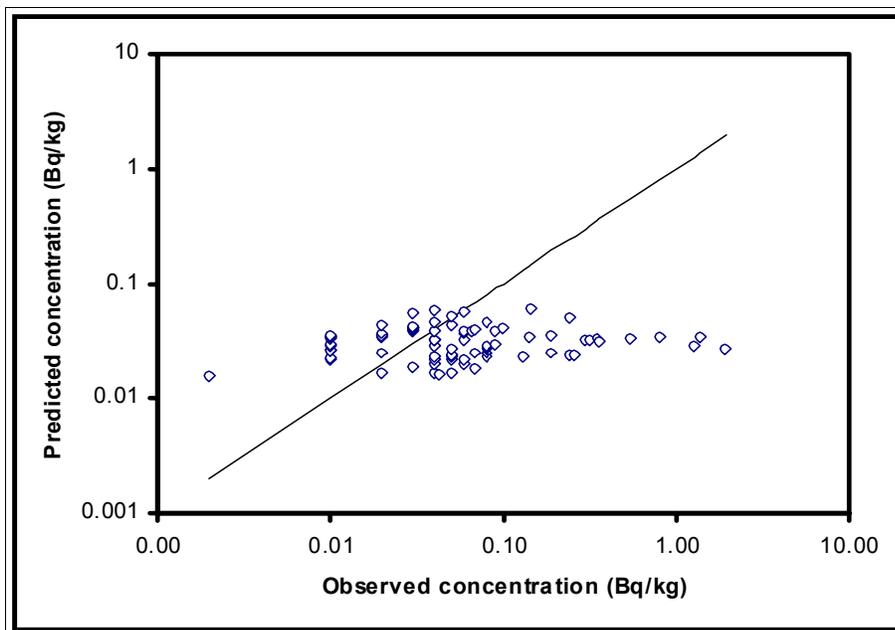


Figure 8.5 Scatter plot of observed and predicted annual mean concentrations of ^{90}Sr in Baltic fish. The line indicates a 1:1 relationship.

9 DOSE CALCULATIONS

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9.1 Pathways of human exposure to radioactivity in the Baltic Sea

Humans are exposed to radioactivity in the environment from a wide spectrum of exposure pathways. The importance of a particular pathway depends on various factors, e.g. the circumstances of the exposure, the short-term or long-term nature of exposure, radionuclide composition etc. The most important pathways are often the ingestion of contaminated food, and external exposure from the occupancy of contaminated land.

Some assessments of the consequences of radioactivity in the marine environment have included a wide range of exposure pathways (IAEA, 1986), but this present work has focused on marine pathways which are known to be most important (CEC, 1990) when radiocaesium is a major contributor. These pathways include the ingestion of contaminated fish, crustaceans and molluscs, the inhalation of resuspended contaminated coastal sediments and sea spray, and external exposure from the occupancy of contaminated coastal areas. More details are given elsewhere (EC, 2000).

9.1.1 Habits of critical groups

Doses to critical groups (i.e. individuals exposed significantly higher than the average) from routine discharges of radioactivity are typically highest close to the point of discharge. They are usually assessed on the basis of measured concentrations of radionuclides in the environment combined with knowledge of the habits of the most exposed group of people. Nuclear facilities and national authorities assess doses to local critical groups near nuclear installations. It has been beyond the scope of the present work to repeat this work. Instead doses to critical groups have been assessed on a regional scale from model calculations using the geographical resolution of the model and taking into consideration all major sources of radioactivity in the marine environment of the Baltic Sea.

Information was collected on the habitual behaviour of regional critical groups considering the marine exposure pathways in the Baltic Sea. It generally proved difficult to identify statistical information on the subjects, and data showed considerable variation. Furthermore, the available information was not considered to be detailed enough to justify using individual habit profiles for each country, so the following habits were taken from the upper end of the combined data and used for critical groups in all countries:

- Ingestion of fish – 90 kg y^{-1}
- Ingestion of crustaceans – 10 kg y^{-1}
- Ingestion of molluscs – 10 kg y^{-1}
- Beach occupancy times (inhalation of resuspended sediment and sea-spray, and external exposure) – 700 h y^{-1}

Information was collected on the habits of average individuals in each of the countries bordering the Baltic Sea, and for all countries the data shows values of about one order of magnitude below those given above for critical groups. Thus doses to average individuals will also range within one order of magnitude below doses to individuals from critical groups.

9.1.2 Habits of wider populations

The collective doses correspond to the sum of doses to individuals across populations, and consider the same exposure pathways as given above. But population habits are considered in a different way. Information on the amount of sea-food ingested by populations is based on the national fishery statistics available for most European countries. This information was collected for the Marina Balt Project (EC, 2000) and has resulted in a detailed survey of the quantities and utilisation of associated marine produce from the Baltic Sea across the European countries (Hagel, 2000). It has been assumed (conservatively) that the following fractions of the total landings of sea-food are used for human consumption: one half for fish, one third for crustaceans, and one sixth for molluscs.

The remaining exposure pathways considered (inhalation of sea-spray and resuspended sediment, and external exposure) all involve beach occupancy by populations. A collective occupancy factor of $50 \text{ h m}^{-1} \text{ y}^{-1}$ was used for North European marine waters in the Marina Project (CEC, 1990), and a factor of $75 \text{ h m}^{-1} \text{ y}^{-1}$ was used for the warmer conditions in the Mediterranean Sea in the Marina-Med Project (EC, 1994). The former value of $50 \text{ h m}^{-1} \text{ y}^{-1}$ was considered appropriate for the Baltic Sea. The collective occupancy expressed in person-hours per year for a given population is obtained by multiplying the occupancy factor with the length of the coastline where the population spends time.

9.2 Impact on man of radioactivity in the Baltic Sea

The model described in Chapter 8 was used for the assessment of doses to man from radioactivity in the Baltic Sea over the period 1950-1996. The assessment concerns dose rates (Sv y^{-1}) to individuals from critical groups and committed collective doses (manSv) to members of the public in the countries around the Baltic Sea. Doses to man from radiation exposure are indicators of the associated health risk. At low dose rates, the health risk is fatal cancer, and the risk factor is $5 \cdot 10^{-2} \text{ Sv}^{-1}$ (ICRP, 1991). The

dose calculations are based on dose-per-unit-intake factors from IAEA (1996).

The following sources of radioactivity in the Baltic Sea have been considered: fallout from atmospheric nuclear weapons testing, fallout from the Chernobyl accident, releases into the sea from European reprocessing facilities, and releases into the sea from nuclear facilities (nuclear power plants and nuclear research reactors) around the Baltic Sea. Furthermore, radiological consequences have been estimated for dumpings of radioactive waste in the Baltic Sea carried out in the 1960s by Sweden and the Soviet Union. The assessment is based on information on the sources of radioactivity in the Baltic Sea (Ilus, 2000), which covers data on 49 radionuclides released from 14 nuclear facilities during their entire operation.

Two nuclear facilities (the Ignalina nuclear power plant in Lithuania and the Salaspils research reactor in Latvia) are not located near the coast, but they discharge into lakes and rivers that ultimately flow into the Baltic Sea. The removal of radioactivity from lake and river water due to sedimentation processes before input to the Baltic Sea was taken into account. The methodology used here was adopted from a previous CEC study (1979), and is based on a classification of radionuclides into three categories based on knowledge of the freshwater concentration factors: radionuclides with strong interaction with sediments (Cr, Mn, Co, Zr, Ru, Cs, Eu, Np, Ce, Pu, Am, Cu); radionuclides with moderate interaction with sediments (C, Zn, Sr, Y); and radionuclides with weak interaction with sediments (H, Nb, Tc, Ag, Sb, Te, I). In both cases an average river flow of 2 m s^{-1} was assumed; while for Ignalina the distance to the Baltic Sea was taken as 600 km, and Salaspils was considered as being 18 km from the Gulf of Riga. For Salaspils the predicted removal was quite insignificant, but for Ignalina the predicted removal of the reactive elements amounted to a factor of 400.

Radioactive waste dumped into the Baltic Sea by Sweden and the Soviet Union is also included in the assessment. In 1960 the Soviet Union carried out dumpings of liquid radioactive waste near the Gogland Island in the Gulf of Finland (White Book No. 3, 1993). The amount of radioactivity was 0.2 Ci (7 GBq) with an unspecified nuclide composition. In 1958, 1961 and 1964, Sweden carried out dumpings of solid radioactive waste in Landsortsdjupet at the deepest location in the Baltic Sea (450 m) situated between Stockholm and Gotland (SSI, 1996). This waste was contained in cement in 200-L barrels, and the average concentrations were below 0.002 Ci g^{-1} and thus not considered as radioactive according to regulations in force at the time. A total of about 460 barrels, each weighing about 400 kg, were dumped, so the total activity is limited to no more than 0.4 Ci (14 GBq). There is no further information available on the amount of radioactivity or the nuclide composition of the waste. For the present assessment it has been assumed that the radioactivity of the waste dumped in both cases has

been equally divided between ^{90}Sr and ^{137}Cs . For Landsortsdjupet it has furthermore been assumed conservatively that equal amounts were dumped in each of the three years and that the waste was available for dispersion within the year of dumping.

The exposure pathways included in the assessment cover the ingestion of fish, crustaceans and molluscs, in addition to exposures due to the occupancy of coastal areas (inhalation of resuspended seawater and sediment, and external exposure). The statistical information collected on marine produce from the Baltic Sea (Hagel, 2000) concerning quantities and utilisation (including imports and exports of marine produce) has permitted estimates to be made of collective doses to populations in countries around the Baltic Sea and in other EU Member States. The annual catches of fish, crustaceans and molluscs used for the calculations of collective doses are shown in Table 9.1.

9.2.1 Doses to individuals

The results of the annual dose rates to individuals in the critical groups are shown for the time period 1950-2000 in Figures 9.1-9.8 for each region. Dose rates are given in terms of the source components: Chernobyl fallout, fallout from nuclear weapons tests, nuclear reprocessing facilities, nuclear power plants, and nuclear research facilities; and for comparison purposes the estimated upper bounds of the contribution from the dumpings in the 1960s are also included.

The contribution from nuclear weapons fallout affects all regions similarly, with an annual dose rate peaking at about 0.01 mSv y^{-1} around 1965 and then declining. The contribution from European reprocessing plants is highest close to the North Sea where the peak annual dose in the Kattegat region around 1980 is predicted at about 0.02 mSv y^{-1} . This contribution from reprocessing decreases further into the Baltic Sea with predicted peak annual doses around 1990 at $0.4 \mu\text{Sv y}^{-1}$ in the Bothnian Bay and $0.05 \mu\text{Sv y}^{-1}$ in the Gulf of Finland.

The fallout from the Chernobyl accident in 1986 is the dominant source component in annual doses to critical groups in every region in the Baltic Sea. The relative importance of the Chernobyl contribution to the maximum annual dose across the Baltic Sea regions during 1950 to 1996 ranges from 70% in the Kattegat to 99% in the Gulf of Finland. The maximum annual doses are listed in Table 9.2 for each region, with levels ranging from 0.2 mSv y^{-1} in the Bothnian Sea and the Gulf of Finland to 0.04 mSv y^{-1} in the Gulf of Riga and the Kattegat.

Figures 9.9-9.16 give for each region the individual contributions to dose rates to the critical groups from the two reprocessing facilities Sellafield and La Hague.

The remaining contributions from nuclear power plants, nuclear research facilities and dumpings of radioactive waste to the critical-group doses

rank lower than those mentioned above. In general, the annual doses to critical groups in the Baltic Sea from nuclear power plants are predicted to be higher than those from research facilities, which are themselves higher than the doses from the waste dumpings.

Figures 9.17-9.24 show for each region a further break-down of dose rates to critical groups from nuclear power plants by individual plant. The dose rates in the Kattegat region are about $0.1 \mu\text{Sv y}^{-1}$, and are dominated by releases from the Ringhals Nuclear Power Plant. The dose rates in the Belt Sea are about $0.1 \mu\text{Sv y}^{-1}$, and are dominated by releases from Barsebäck. The dose rates in the East and West Baltic region are between 0.01 and $0.1 \mu\text{Sv y}^{-1}$, and are dominated by releases from Oskarshamn until 1985, after which releases from Forsmark dominate. In the Bothnian Sea region dose rates are still lower: between 0.001 and $0.01 \mu\text{Sv y}^{-1}$, with dominating contributions from Oskarshamn until 1985 and subsequently from Olkiluoto. In the Bothnian Bay, dose rates are predicted to be below $0.001 \mu\text{Sv y}^{-1}$, and dominated by releases from Oskarshamn. In the Gulf of Finland, dose rates to critical groups are predicted to be below $0.01 \mu\text{Sv y}^{-1}$, and are dominated by releases from Loviisa. In the Gulf of Riga, dose rates to critical groups are predicted to be between 0.001 and $0.01 \mu\text{Sv y}^{-1}$, and are dominated by releases from Oskarshamn and Greifswald.

Finally, the individual contributions to the critical-group dose rates from nuclear research facilities are shown in Figures 9.25-9.32. For all regions the dose rates from discharges from Studsvik dominate, reaching levels between 0.1 and $0.01 \mu\text{Sv y}^{-1}$ at the most; while contributions from Risø and Salaspils range several orders of magnitude lower.

9.2.2 Doses to populations

The calculations of collective committed doses from radioactivity in the Baltic Sea are based on discharge data for the period 1950-1996. For the long-lived radionuclides (e.g. ^{14}C , ^{239}Pu , ^{241}Am , ^{99}Tc , ^{129}I) collective committed doses were truncated at year 2400.

The collective committed doses are illustrated in graphical form in Figures 9.33-9.36, which show the collective doses by source category, by country, by exposure pathway, and by radionuclide. The corresponding information is given in numerical form in Tables 9.3-9.5. The total collective dose is estimated at 2,560 manSv of which 1,700 manSv (67%) originates from Chernobyl fallout, 650 manSv (25%) from fallout from nuclear weapons tests, 200 manSv (8%) from European reprocessing facilities, 1 manSv (0.04%) from nuclear power plants, and 0.4 manSv (0.02%) from nuclear research facilities around the Baltic Sea. The collective doses from the dumping of radioactive waste in the Baltic Sea are estimated not to

exceed 0.004 manSv.

The distribution of the collective doses from radioactivity in the Baltic Sea across European countries shows that 2,400 manSv (94% of the total dose) is received by the countries around the Baltic Sea while the remaining 160 manSv (6%) is received by other EU Member States. Of the countries around the Baltic Sea, Sweden is predicted to receive the highest collective dose of about 560 manSv (22%) while Lithuania is predicted to receive the lowest dose of 50 manSv (2%). The reason for this difference lies in fishery statistics, which indicate a great difference between these two countries in terms of the amounts of Baltic fish available for human consumption.

The dominant exposure pathway is fish ingestion, which contributes about 2400 manSv (94%), while other pathways yield the rest, including 150 manSv (6%) from external radiation, 5 manSv (0.2%) from inhalation, 4 manSv (0.1%) from the ingestion of molluscs, and 2 manSv (0.07%) from the ingestion of crustaceans.

Radiocaesium is predicted to dominate in the collective doses: ^{137}Cs contributes with 2,200 manSv (88%) and ^{134}Cs with 300 manSv (12%). Only 20 manSv (0.8%) is delivered via other radionuclides, of which ^{90}Sr contributes with 15 manSv, ^{14}C with 2 manSv, and ^{106}Ru with 1 manSv. The total contribution from the remaining 57 radionuclides is about 1 manSv. The concept of collective dose is not meaningful at very low levels, and the data given on radionuclides is only shown to indicate their ranking.

The collective doses from European reprocessing facilities are shown in Table 9.6 by nuclide and facility. It should be noted that the release data from La Hague includes only ^{137}Cs and ^{90}Sr . Selafiel is predicted to contribute 190 manSv, and La Hague 10 manSv.

Table 9.7 shows the collective doses from nuclear research facilities by radionuclide and facility. It is notable that that Studsvik accounts for the entire collective dose, and that radiocaesium dominates. The collective doses from nuclear power plants are given in Table 9.8 by country and plant. The Swedish plants give the largest contributions, headed by Oskarshamn. Denmark and Sweden receive the highest collective doses from this source category. This data is illustrated in Figure 9.37.

A detailed break-down of the collective doses by radionuclide and exposure pathway is given for the nuclear power plants in Figures 9.38-9.46. The dominant radionuclides are corrosion products (^{60}Co and ^{65}Zn) for Barsebäck, Forsmark, Olkiluoto and Ringhals; and radiocaesium for Greifswald, Leningrad, Loviisa and Oskarshamn. For Ignalina, tritium is dominant, due to the removal of sediment-reactive nuclides during transportation to the coast.

The collective doses are also shown by nuclide and pathway in Figures 9.47-9.49 for the nuclear

research facilities at Salaspils and Studsvik; and for Sellafield. In all these cases radiocaesium is the dominant radionuclide.

9.2.3 Doses from natural radioactivity

Naturally occurring radionuclides in sea water are incorporated into marine organisms, and give rise to radiation exposure to humans from the ingestion of seafood. Polonium-210 is the major contributor in this context, due to its high bioaccumulation rate in marine organisms, and the relatively high dose factor. Typical ^{210}Po concentrations in fish from the Baltic Sea are 0.8 Bq kg^{-1} (Bojanowsky, 1998; Dahlgard, 1996; Holm, 1994), in crustacea 20 Bq kg^{-1} (Swift et al., 1994) and in molluscs 30 Bq kg^{-1} (Dahlgard, 1996). Using the same assumptions on seafood intake as in the preceding sections, with a dose factor of $1.2 \cdot 10^{-6} \text{ Sv Bq}^{-1}$ (IAEA, 1996), we obtain a dose rate for ^{210}Po of 0.7 mSv y^{-1} to individuals within critical groups. This value is a factor of 3–4 higher than the peak dose rates for individuals from any critical group exposed to anthropogenic radioactivity in the Baltic Sea during the period 1950–1996. Similarly, the collective dose rate from ^{210}Po may be calculated using fisheries data, resulting in a value of about 400 manSv y^{-1} . For the time period considered in this assessment, the collective dose from ^{210}Po thus amounts to about $20,000 \text{ manSv}$. This is about an order of magnitude higher than the collective dose from marine pathways and all man-made radionuclides in the Baltic Sea.

9.3 Conclusions

An assessment of the radiological consequences of radioactivity in the Baltic Sea has been carried out. The assessment is based on detailed information on inputs, and on levels of radioactivity observed in the Baltic Sea during the period 1950–1996. Doses to man have been calculated with a computer model designed to cover the waters of the North Atlantic, including the Baltic Sea.

Doses have been calculated to members of the public from the ingestion of radionuclides in seafood from the Baltic Sea, and from exposure to radioactivity in coastal areas. These calculations cover the period 1950–1996, and include source contributions from nuclear weapons testing, the Chernobyl accident, the two European reprocessing plants Sellafield and La Hague, and nuclear installations around the Baltic Sea.

Dose rates from anthropogenic radioactivity to individual members of the public (critical groups) have been calculated on the basis of specified rates of annual intake (90 kg of fish, 10 kg of crustacea and 10 kg of molluscs) and beach occupancy (700 h y^{-1}). The dose rates to individuals from the regions of the Bothnian Sea and the Gulf of Finland are predicted to be higher than for any other area in the Baltic Sea, due to the pattern of fallout from the Chernobyl accident. These dose rates are predicted to have peaked in 1986 at a value of 0.2 mSv y^{-1} .

Collective committed doses to members of the public have been calculated based on predicted concentrations of radionuclides in biota and coastal sediments. The total collective dose from anthropogenic radioactivity in the Baltic Sea is estimated at $2,600 \text{ manSv}$, of which about two-thirds originates from Chernobyl fallout, about one quarter from fallout from nuclear weapons testing, about 8% from European reprocessing facilities, and about 0.04% from nuclear installations around the Baltic Sea.

A radiological assessment of the dumpings of low-level radioactive waste in the Baltic Sea in the 1960s by Sweden and the Soviet Union indicates that doses to man are negligible.

Doses from naturally occurring radioactivity in seafood (polonium-210) have been calculated on a similar basis and compared with the doses from anthropogenic radioactivity received through marine pathways. The results of this comparison show that dose rates and doses from natural radioactivity dominate, except for the year 1986, when individual dose rates from Chernobyl fallout approached those received from natural radioactivity in some regions around the Baltic Sea.

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Table 9.1 Annual catches of fish, crustaceans and molluscs (metric tonnes per year) in regions of the Baltic Sea as used for calculations of collective doses

Region	Fish (t y ⁻¹)	Crustaceans (t y ⁻¹)	Molluscs (t y ⁻¹)
Kattegat	5.9E+04	4.7E+03	5.7E+03
Belt Sea	4.8E+04	3.2E+02	6.7E+03
Baltic Sea West	1.6E+05		
Baltic Sea East	2.6E+05		
Bothnian Sea	3.8E+04		
Bothnian Bay	8.9E+03		
Gulf of Finland	3.2E+04		
Gulf of Riga	2.3E+04		

Table 9.2 Maximum annual doses (mSv y⁻¹) to individuals in critical groups in the Baltic Sea during the period 1950-1996.

Region	Max. annual dose rate
Kattegat	0.04
Belt Sea	0.09
West Baltic	0.1
East Baltic	0.05
Bothnian Sea	0.2
Bothnian Bay	0.06
Gulf of Finland	0.2
Gulf of Riga	0.04

Table 9.3 Collective dose (manSv) by country and source category.

Country	Cher-nobyl	Nu-clear Tests	Reproc-essing	Nuclear Power	Re-search Reactors	Total
Sweden	366	139	50	0.26	0.11	556
Finland	285	81	9	0.05	0.03	375
Poland	243	101	21	0.09	0.07	366
Den-mark	199	83	51	0.27	0.06	333
Russia	178	65	11	0.05	0.03	254
Latvia	130	59	11	0.04	0.03	200
Ger-many	98	41	17	0.11	0.03	156
Estonia	80	29	4.6	0.021	0.013	113
France	33	14	6.0	0.025	0.009	52
Lithua-nia	34	14	3.1	0.012	0.009	51
Nether-lands	21	8.5	3.9	0.017	0.006	33
Italy	12	5.3	2.6	0.011	0.003	20
United Kingdom	11	4.5	2.1	0.009	0.003	17
Spain	8.9	3.7	1.8	0.008	0.002	14
Belgium	5.1	2.1	1.0	0.004	0.001	8.2
Portugal	4.3	1.8	0.8	0.004	0.001	6.9
Greece	0.9	0.4	0.2	8.E-04	2.E-04	1.4
Ireland	0.9	0.4	0.2	7.E-04	3.E-04	1.4
Total	1708	652	197	1.0	0.4	2558

Table 9.4 Collective dose (manSv) by country and exposure pathway.

Country	Fish	Exter-nal	Inhala-tion	Mol-luscs	Crusta-ceans	Tot-al
Sweden	536	18	0.34	0.32	0.58	556
Finland	303	67	3.9	0.006	0.018	375
Poland	342	24	0.06	0.005	0.019	366
Denmark	318	11	0.30	3.1	0.13	333
Russia	251	2.6	0.04	0.003	0.025	254
Latvia	197	2.4	0.06	0.001	0.0003	200
Germany	146	8.9	0.22	0.11	0.33	156
Estonia	105	7.6	0.15	0.004	0.006	113
France	52			0.047	0.17	52
Lithuania	49	2.0	0.01	0.001	0.008	51
Nether-lands	33			0.052	0.11	33
Italy	20			0.021	0.086	20
United Kingdom	17			0.018	0.060	17
Spain	14			0.016	0.060	14
Belgium	8.2			0.010	0.027	8
Portugal	6.9			0.013	0.028	7
Greece	1.4			0.001	0.006	1
Ireland	1.4			0.004	0.004	1
Total	2403	145	5.1	3.8	1.7	2558

Table 9.5 Collective dose (manSv) by nuclide and source category.

	Chernobyl	Nuclear Tests	Reprocessing	NPP	Research Reactors	Dumping	Total
¹³⁷ Cs	1419	639	185	0.31	0.31	0.004	2243
¹³⁴ Cs	288		6.5	0.10	0.05		295
⁹⁰ Sr	1.0	13	1.2	1.E-04	0.008	1.E-04	15
¹⁴ C			2.0				2.0
¹⁰⁶ Ru			1.5	2.E-05	7.E-05		1.5
²³⁹ Pu			0.40	2.E-06			0.40
⁶⁰ Co			0.0004	0.37	0.025		0.39
²⁴¹ Pu			0.22				0.22
⁶⁵ Zn				0.17	0.001		0.17
⁹⁹ Tc			0.17				0.17
²³⁸ Pu			0.07	5.E-06			0.071

Table 9.5 Collective dose (manSv) by nuclide and source category. (continued)

	Chernobyl	Nuclear Tests	Reprocessing	NPP	Research Re-actors	Dumping	Total
¹²⁹ I			0.02				0.018
⁵⁸ Co				0.014	3.E-05		0.014
³ H			0.003	0.002	0.004		0.008
^{110m} Ag				0.006	3.E-06		0.006
¹²⁴ Sb				0.004	0.001		0.005
⁵⁴ Mn				0.003	0.001		0.004
⁵⁹ Fe				0.004	1.E-04		0.004
¹²⁵ Sb				0.002	2.E-04		0.002
¹¹³ Sn				0.002			0.002
²⁴¹ Am			0.001	2.31E-07			0.001
¹⁵² Eu					8.E-04		0.001
¹⁵⁴ Eu					3.E-04		3.E-04
¹³¹ I				1.E-04	7.E-06		1.E-04
¹⁴⁴ Ce			2.E-05	1.E-05	7.E-05		1.E-04
⁵¹ Cr				8.E-05	2.E-06		8.E-05
⁹⁵ Zr			1.E-06	5.E-05	3.E-06		5.E-05
¹⁴¹ Ce				4.E-05	7.E-09		4.E-05
⁹⁵ Nb			2.E-07	3.E-05	2.E-06		3.E-05
¹³⁶ Cs				9.E-06	3.E-06		1.E-05
⁵⁷ Co				1.E-05	8.E-08		1.E-05
⁷⁵ Se				1.E-05			1.E-05
¹⁴⁰ Ba				3.E-06			3.E-06
¹⁵⁵ Eu					3.E-06		3.E-06
^{117m} Sn				2.E-06			2.E-06
¹⁰³ Ru				1.E-06	6.E-08		1.E-06
²⁴⁴ Cm				4.E-07			4.E-07
^{123m} Te				3.E-07			3.E-07
²⁴² Cm				8.E-08			8.E-08
¹³³ Ba				5.E-10	6.E-08		6.E-08
⁸⁹ Sr				4.E-08			4.E-08
²⁴³ Cm				3.E-08			3.E-08
¹³¹ Ba				3.E-08			3.E-08
²⁴³ Am				2.E-08			2.E-08
¹⁵³ Gd					5.E-09		5.E-09
²² Na				3.E-09			3.E-09
⁴⁶ Sc				1.E-09			1.E-09
¹⁸¹ Hf				3.E-10			3.E-10
⁸⁵ Sr				2.E-11			2.E-11
Total	1708	652	197	1.0	0.41	0.004	2558

Table 9.6 Collective doses originating from European reprocessing facilities received via the Baltic Sea shown by nuclide and facility.

Nuclide	Sellafield	La Hague	Total
¹³⁷ Cs	176	8.8	185
¹³⁴ Cs	6.5		6.5
¹⁴ C	2.0		2.0
¹⁰⁶ Ru	1.5		1.5
⁹⁰ Sr	0.88	0.37	1.2
²³⁹ Pu	0.40		0.40
²⁴¹ Pu	0.22		0.22
⁹⁹ Tc	0.17		0.17
²³⁸ Pu	0.071		0.071
¹²⁹ I	0.018		0.018
³ H	0.003		0.003
²⁴¹ Am	0.001		0.001
⁶⁰ Co	4.E-04		4.E-04
¹⁴⁴ Ce	2.E-05		2.E-05
⁹⁵ Zr	1.E-06		1.E-06
⁹⁵ Nb	2.E-07		2.E-07
Total	188	9	197

Table 9.7 Collective doses originating from re-search reactors by nuclide and facility.

Nuclide	Studsvik	Risø	Salaspils	Total
¹³⁷ Cs	0.31		3.E-05	0.31
¹³⁴ Cs	0.051		5.E-06	0.051
⁶⁰ Co	0.025		1.E-07	0.025
⁹⁰ Sr	0.008		7.E-07	0.008
³ H	0.004	5.E-05		0.004
⁶⁵ Zn	0.001			0.001
⁵⁴ Mn	8.E-04			8.E-04
¹⁵² Eu	8.E-04		3.E-07	8.E-04
¹²⁴ Sb	5.E-04			5.E-04
¹⁵⁴ Eu	3.E-04			3.E-04
¹²⁵ Sb	2.E-04		5.E-08	2.E-04
⁵⁹ Fe	1.E-04			1.E-04
¹⁴⁴ Ce	7.E-05		2.E-09	7.E-05
¹⁰⁶ Ru	7.E-05		7.E-09	7.E-05
⁵⁸ Co	3.E-05			3.E-05
¹³¹ I	7.E-06			7.E-06
¹³⁶ Cs	3.E-06			3.E-06
¹⁵⁵ Eu	3.E-06			3.E-06
^{110m} Ag	3.E-06		2.E-07	3.E-06
⁹⁵ Zr	3.E-06			3.E-06
⁹⁵ Nb	2.E-06			2.E-06
⁵¹ Cr	2.E-06			2.E-06
⁵⁷ Co	8.E-08			8.E-08
¹⁰³ Ru	6.E-08		5.E-11	6.E-08

Table 9.8 Collective doses (manSv) from nuclear power plants around the Baltic Sea by country and plant.

Country	Barsebäck	Forsmark	Greifswald	Ignalina	Lenin-grad	Loviisa	Olkiluoto	Oskars-hamn	Ringhals	Total
Denmark	0.098	0.029	8.E-04	6.E-06	1.E-07	0.001	6.E-04	0.061	0.078	0.27
Sweden	0.026	0.057	0.001	1.E-05	2.E-07	0.002	0.002	0.11	0.068	0.26
Germany	0.053	0.013	5.E-04	3.E-06	8.E-08	8.E-04	3.E-04	0.027	0.021	0.11
Poland	0.003	0.026	0.002	1.E-05	2.E-07	0.002	6.E-04	0.058	0.003	0.094
Finland	9.E-04	0.009	6.E-04	6.E-06	1.E-06	0.008	0.005	0.024	0.001	0.048
Russia	0.001	0.007	7.E-04	5.E-06	7.E-07	0.007	4.E-04	0.027	0.002	0.045
Latvia	0.001	0.007	7.E-04	5.E-06	1.E-07	0.001	3.E-04	0.026	0.001	0.037
France	0.003	0.003	2.E-04	1.E-06	2.E-08	2.E-04	9.E-05	0.009	0.010	0.025
Estonia	6.E-04	0.004	3.E-04	2.E-06	5.E-07	0.004	1.E-04	0.011	8.E-04	0.021
Netherlands	0.002	0.002	9.E-05	6.E-07	2.E-08	2.E-04	6.E-05	0.006	0.007	0.017
Lithuania	4.E-04	0.003	2.E-04	1.E-06	3.E-08	3.E-04	9.E-05	0.007	6.E-04	0.012
Italy	0.001	0.001	6.E-05	4.E-07	9.E-09	9.E-05	3.E-05	0.003	0.005	0.011
United Kingdom	0.001	9.E-04	5.E-05	3.E-07	1.E-08	1.E-04	3.E-05	0.003	0.004	0.009
Spain	0.001	8.E-04	4.E-05	3.E-07	9.E-09	9.E-05	2.E-05	0.002	0.003	0.008
Belgium	5.E-04	4.E-04	2.E-05	2.E-07	4.E-09	4.E-05	1.E-05	0.001	0.002	0.004
Portugal	5.E-04	4.E-04	2.E-05	1.E-07	5.E-09	5.E-05	1.E-05	0.001	0.002	0.004
Greece	1.E-04	8.E-05	4.E-06	3.E-08	6.E-10	6.E-06	2.E-06	2.E-04	3.E-04	8.E-04
Ireland	8.E-05	1.E-04	3.E-06	2.E-08	6.E-10	5.E-06	3.E-06	3.E-04	3.E-04	7.E-04
Total	0.19	0.17	0.007	5.E-05	3.E-06	0.028	0.010	0.37	0.21	0.98

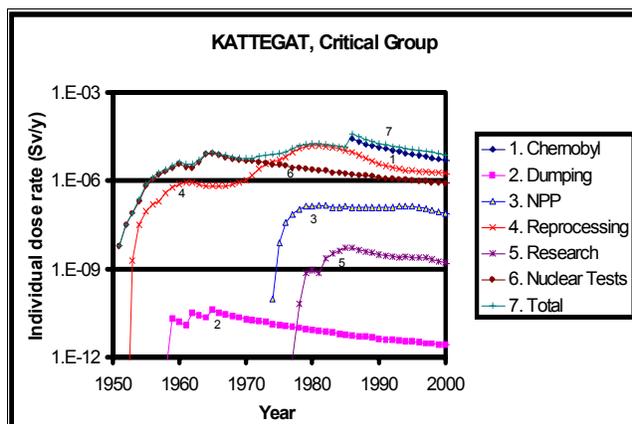


Figure 9.1 Predicted annual doses (Sv y^{-1}) to individuals in critical groups in the Kattegat area by source category.

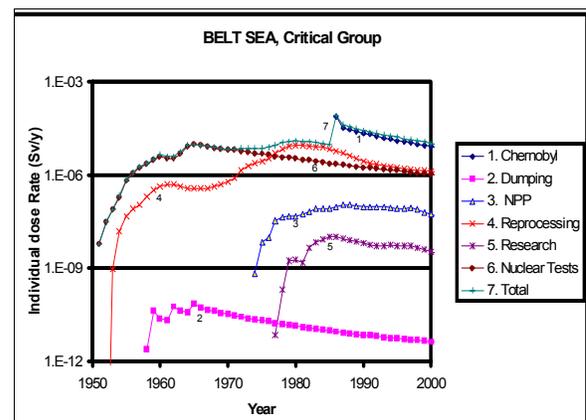


Figure 9.2 Predicted annual doses (Sv y^{-1}) to individuals in critical groups in the Belt Sea area by source category.

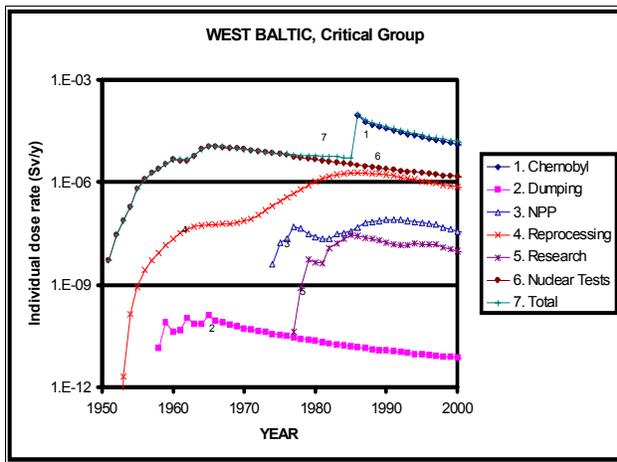


Figure 9.3 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the West Baltic area by source category.

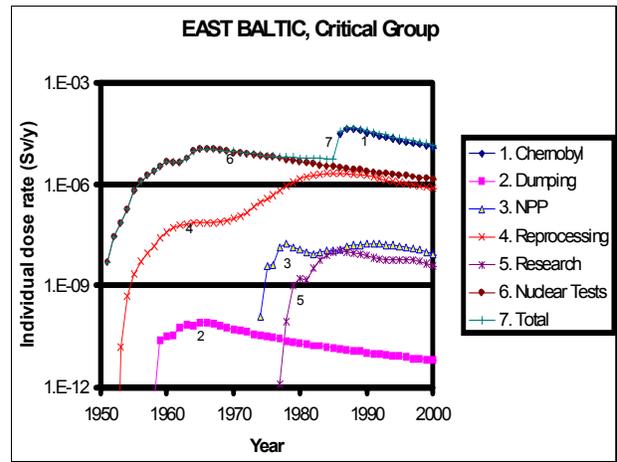


Figure 9.4 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the East Baltic area by source category.

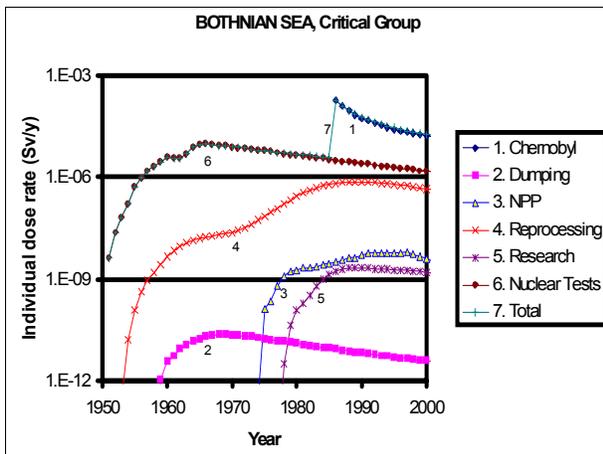


Figure 9.5 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the Bothnian Sea area by source category.

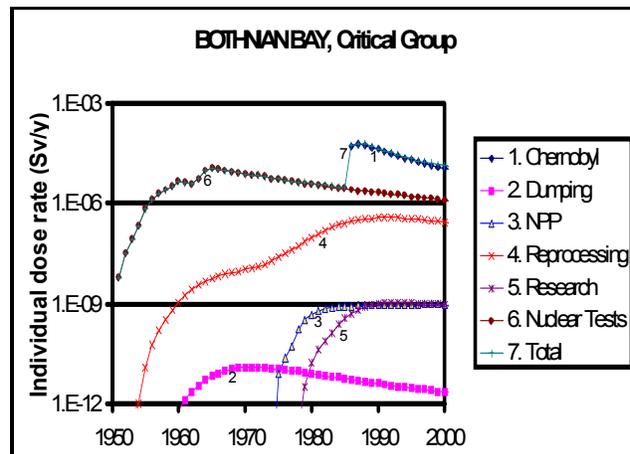


Figure 9.6 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the Bothnian Bay area by source category.

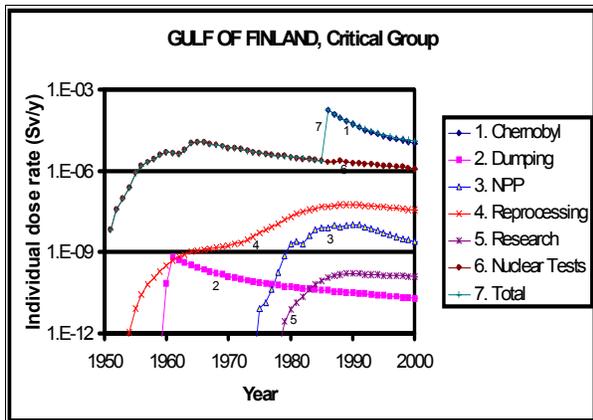


Figure 9.7 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the Gulf of Finland area by source category.

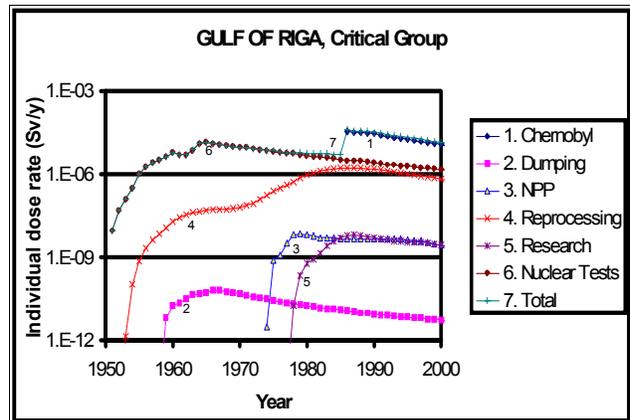


Figure 9.8 Predicted annual doses ($Sv\ y^{-1}$) to individuals in critical groups in the Gulf of Riga area by source category.

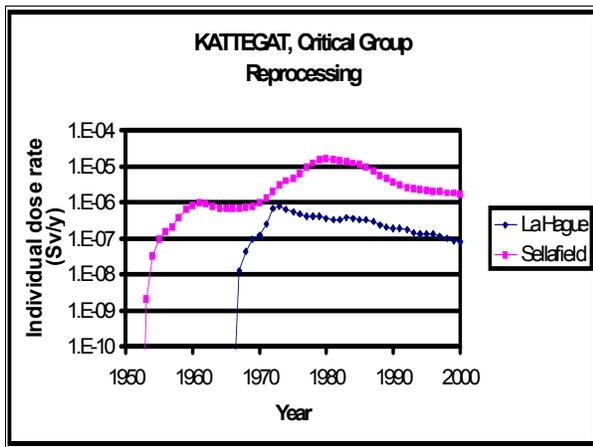


Figure 9.9 Predicted annual doses (Sv y^{-1}) from European reprocessing plants to individuals of critical groups in the Kattegat area by reprocessing plant.

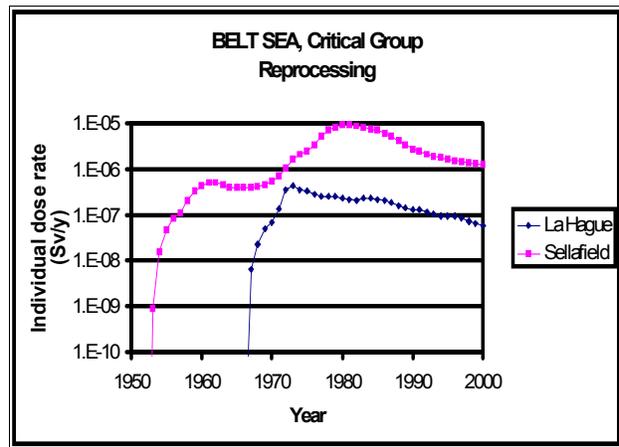


Figure 9.10 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the Belt Sea area by reprocessing plant.

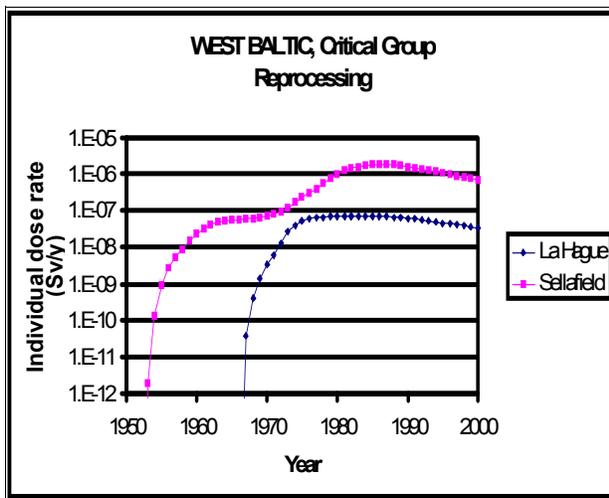


Figure 9.11 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the West Baltic area by reprocessing plant.

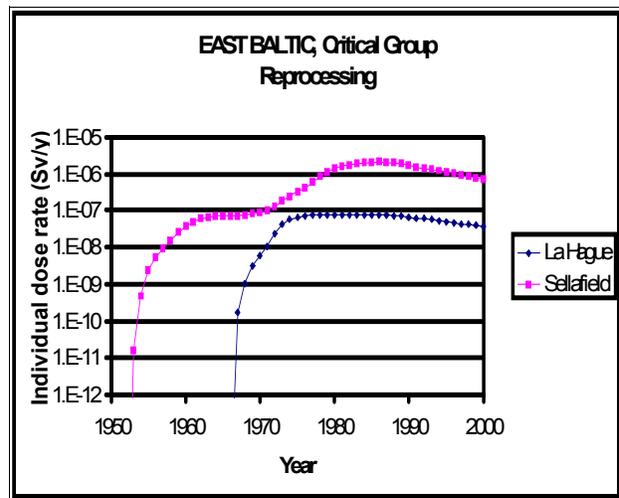


Figure 9.12 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the East Baltic area by reprocessing plant.

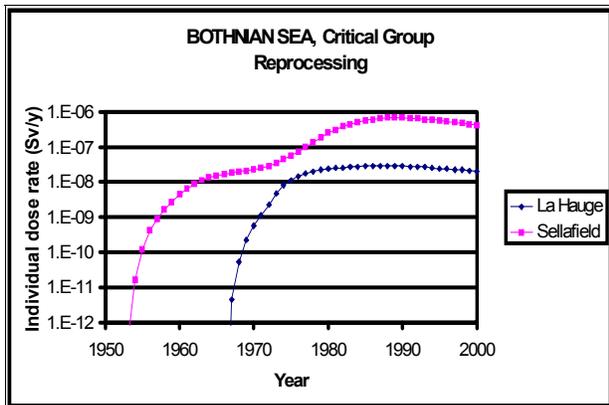


Figure 9.13 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the Bothnian Sea area by reprocessing plant.

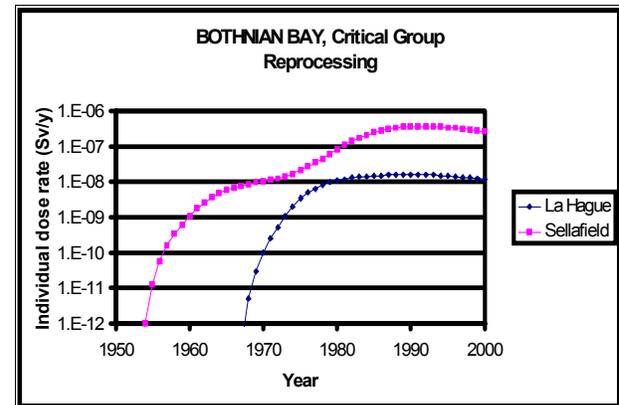


Figure 9.14 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the Bothnian Bay area by reprocessing plant.

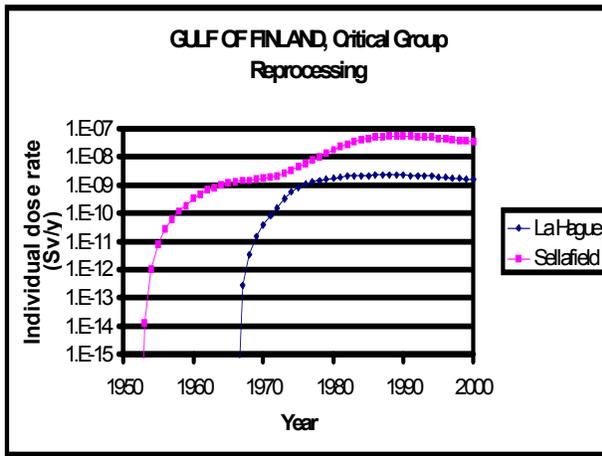


Figure 9.15 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the Gulf of Finland area by reprocessing plant.

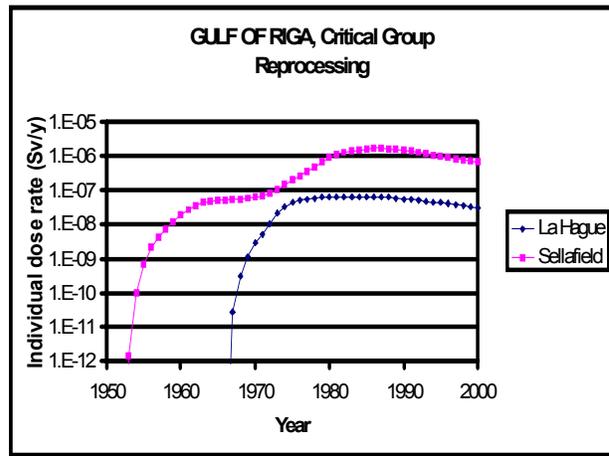


Figure 9.16 Predicted annual doses (Sv y^{-1}) from European reprocessing to individuals in critical groups in the Gulf of Riga area by reprocessing plant.

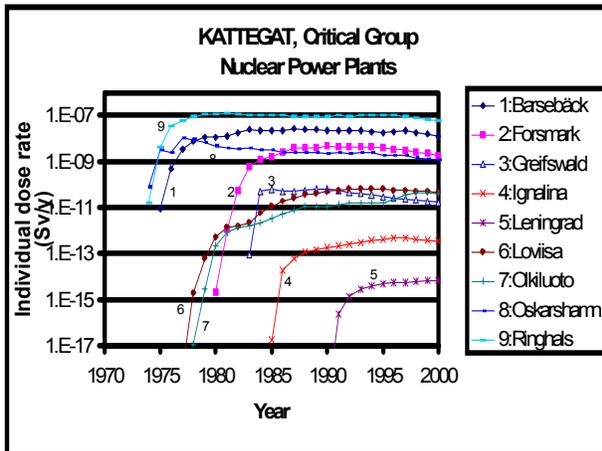


Figure 9.17 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Kattegat area by power plant.

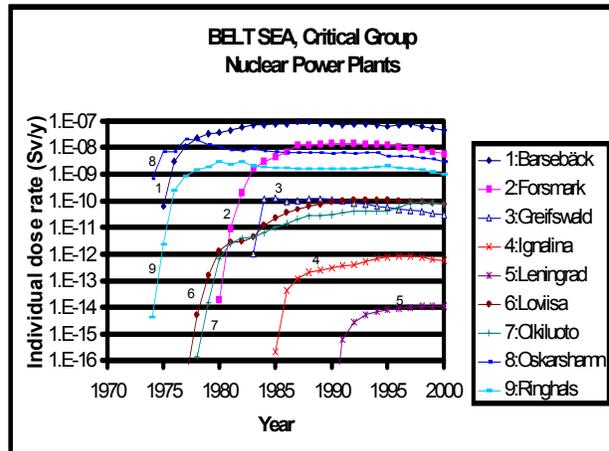


Figure 9.18 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Belt Sea area by power plant.

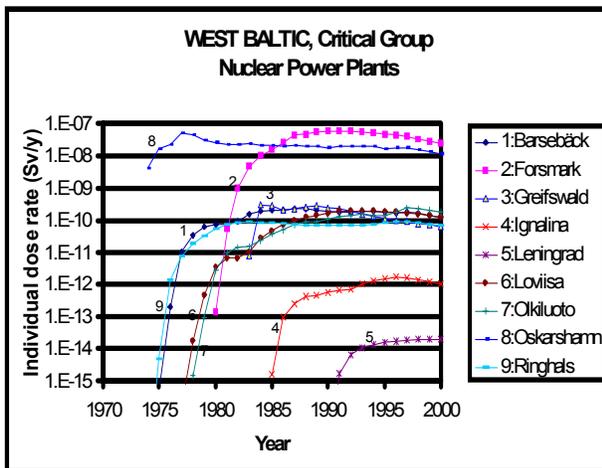


Figure 9.19 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the West Baltic area by power plant.

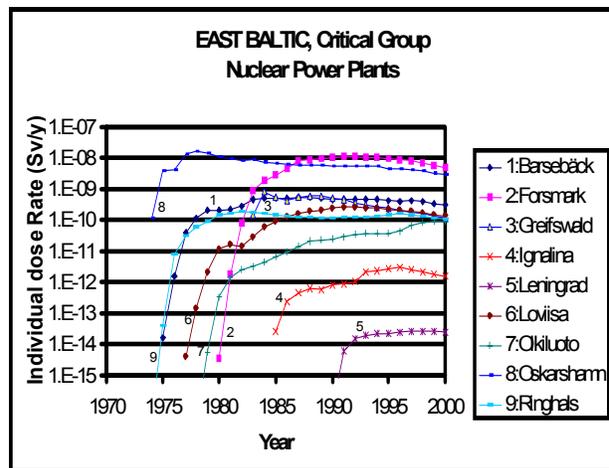


Figure 9.20 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the East Baltic area by power plant.

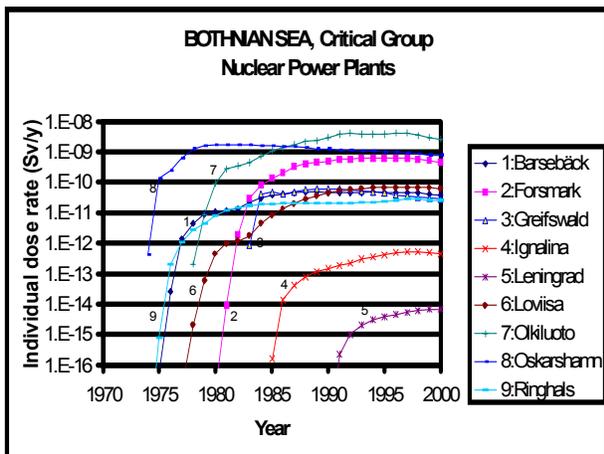


Figure 9.21 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Bothnian Sea area by power plant.

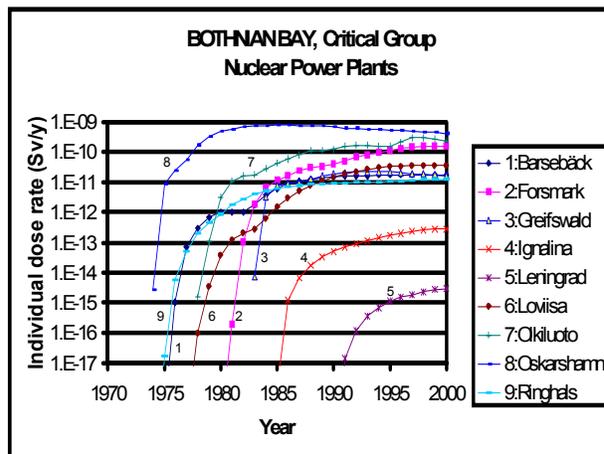


Figure 9.22 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Bothnian Bay area by power plant.

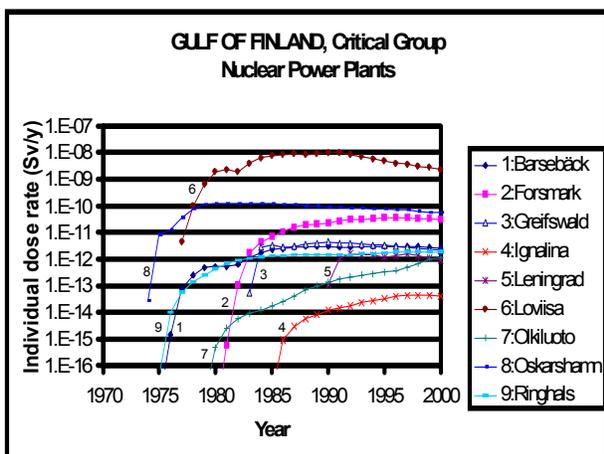


Figure 9.23 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Gulf of Finland area by power plant.

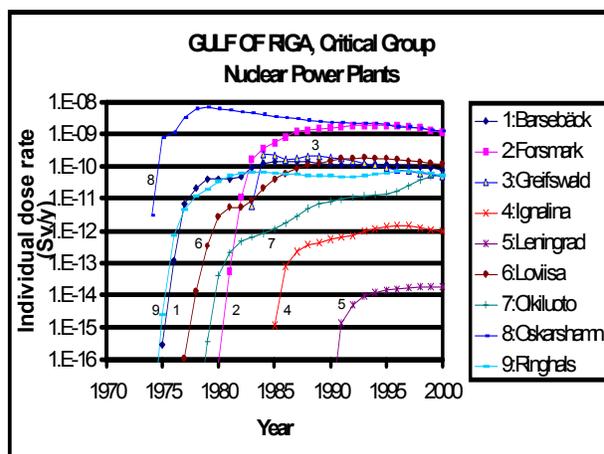


Figure 9.24 Predicted annual doses (Sv y^{-1}) from Baltic nuclear power plants to individuals in critical groups in the Gulf of Riga area by power plant.

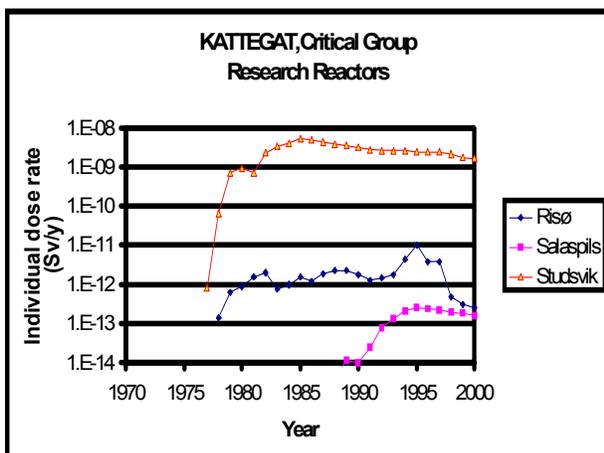


Figure 9.25 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Kattegat area by research facility.

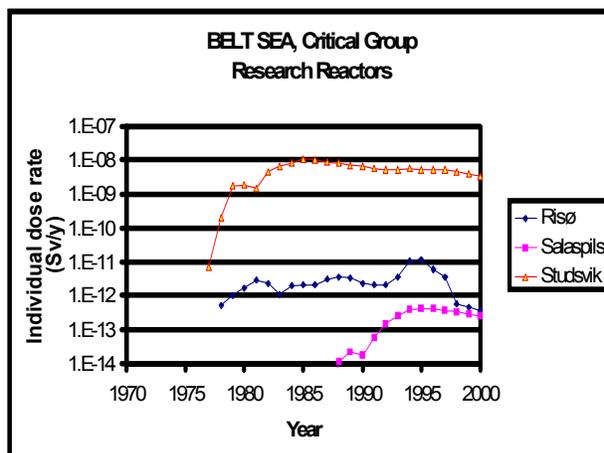


Figure 9.26 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Belt Sea area by research facility.

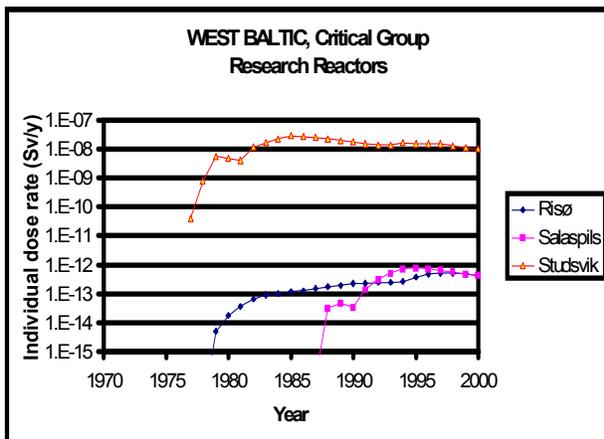


Figure 9.27 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the West Baltic area by research facility.

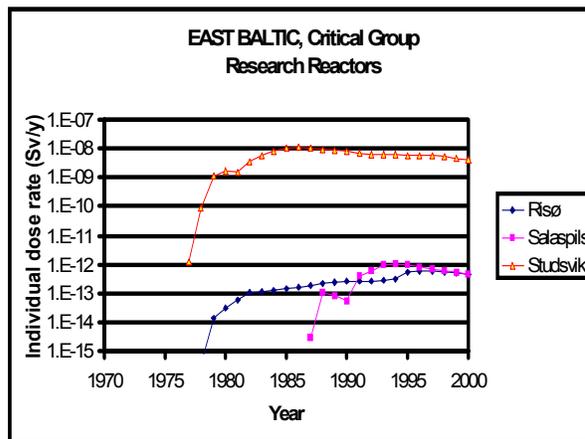


Figure 9.28 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the East Baltic area by research facility.

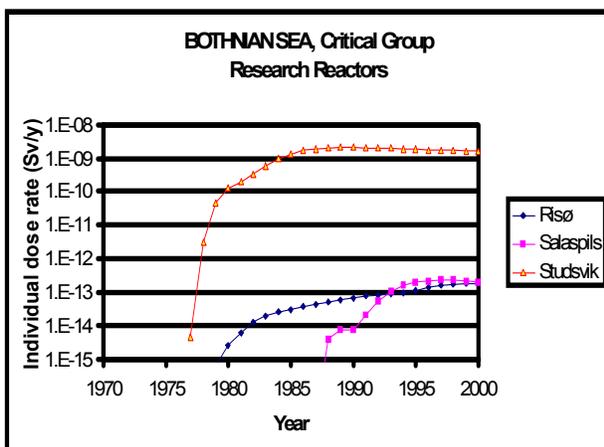


Figure 9.29 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Bothnian Sea area by research facility.

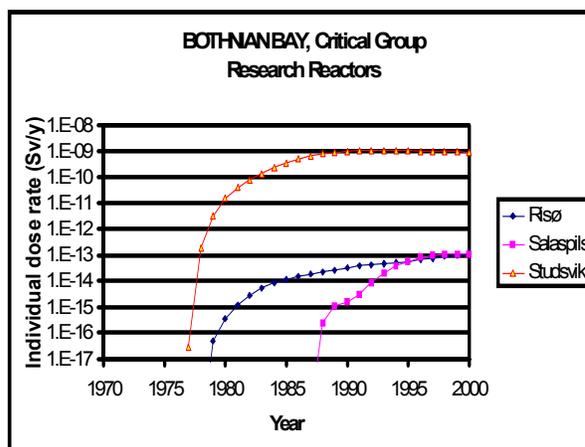


Figure 9.30 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Bothnian Bay area by research facility.

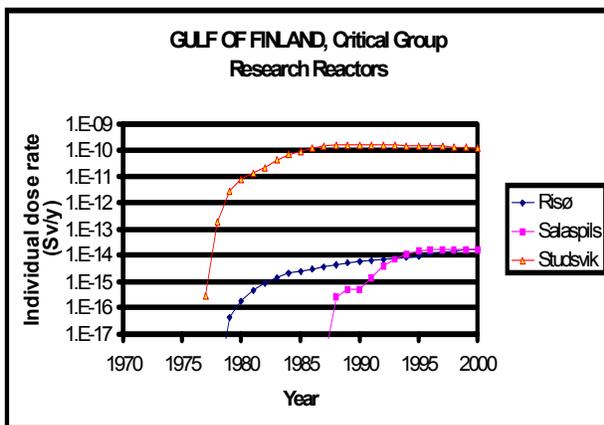


Figure 9.31 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Gulf of Finland area by research facility.

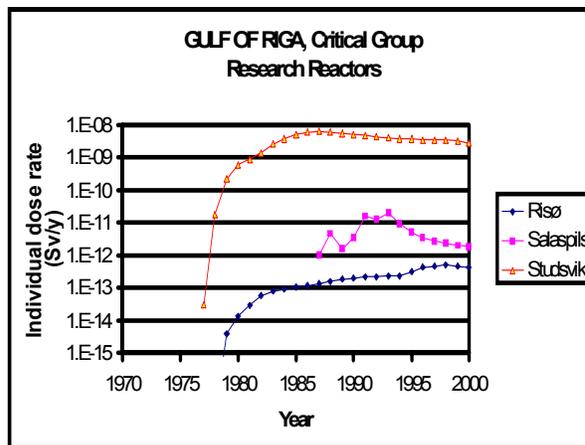


Figure 9.32 Predicted annual doses (Sv y^{-1}) from Baltic nuclear research facilities to individuals in critical groups in the Gulf of Riga area by research facility.

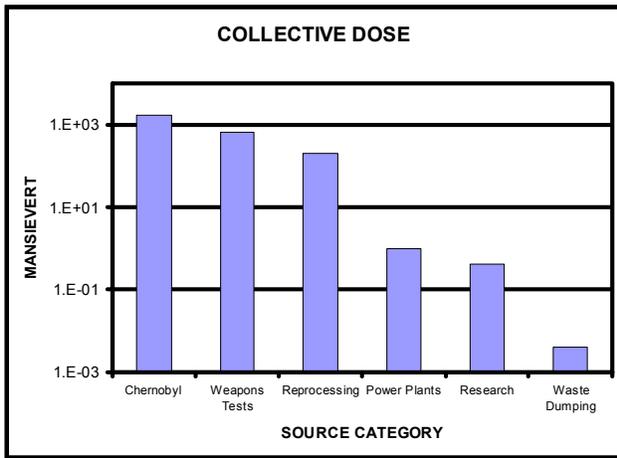


Figure 9.33 Collective doses from anthropogenic radioactivity in the Baltic Sea by source category.

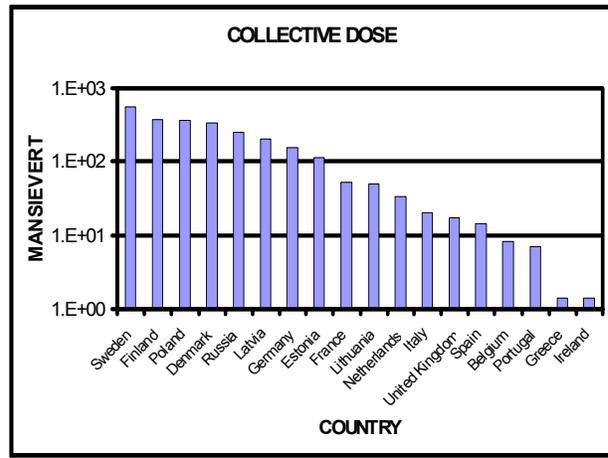


Figure 9.34 Collective doses from anthropogenic radioactivity in the Baltic Sea by receiving country.

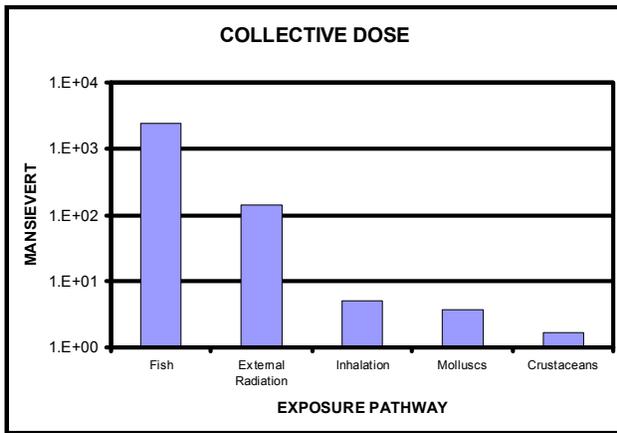


Figure 9.35 Collective doses from anthropogenic radioactivity in the Baltic Sea by exposure pathway.

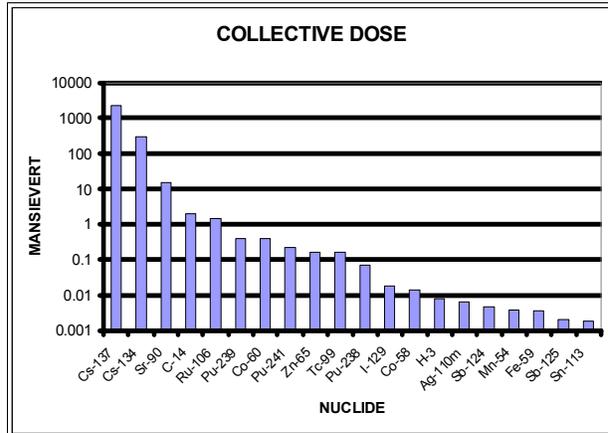


Figure 9.36 Collective doses from anthropogenic radioactivity in the Baltic Sea by radionuclide.

Figure 9.37 Collective doses from discharges into the Baltic Sea from nuclear power plants by power plant and receiving country.

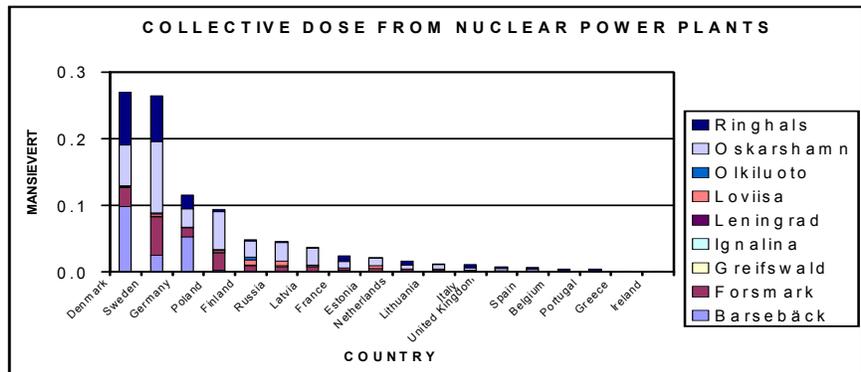
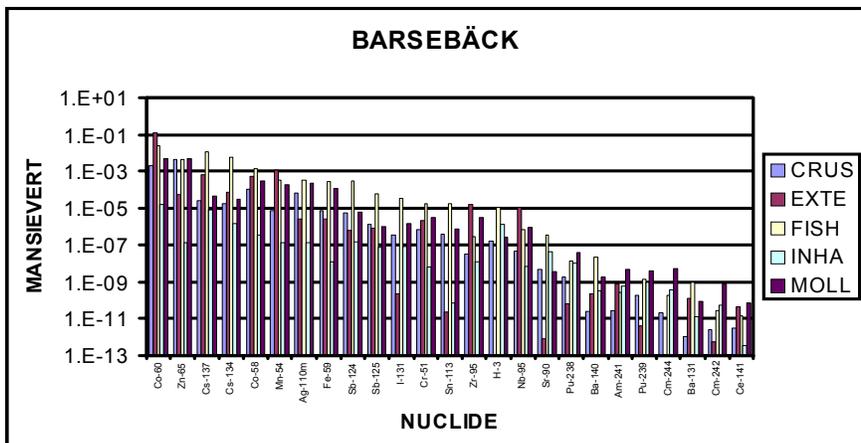


Figure 9.38 Collective doses from discharges into the sea from Barsebäck nuclear power plant by exposure pathway and radionuclide.



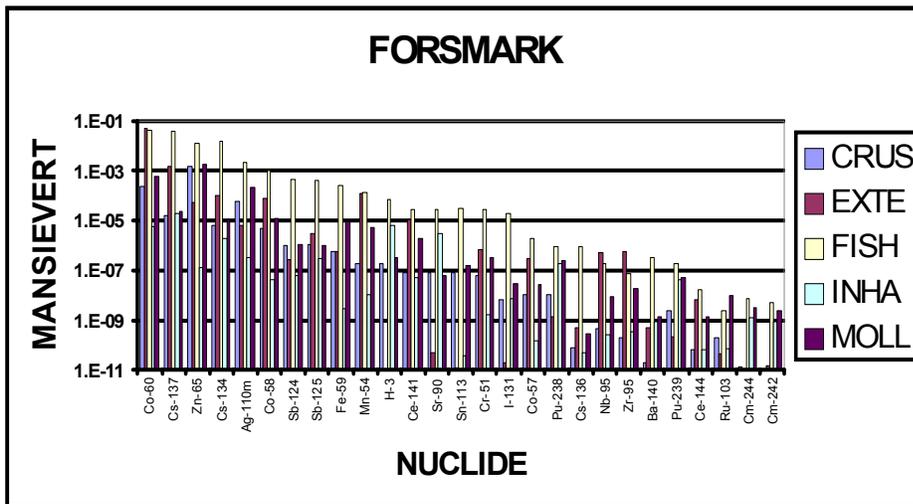


Figure 9.39 Collective doses from discharges into the sea from Forsmark nuclear power plant by exposure pathway and radionuclide.

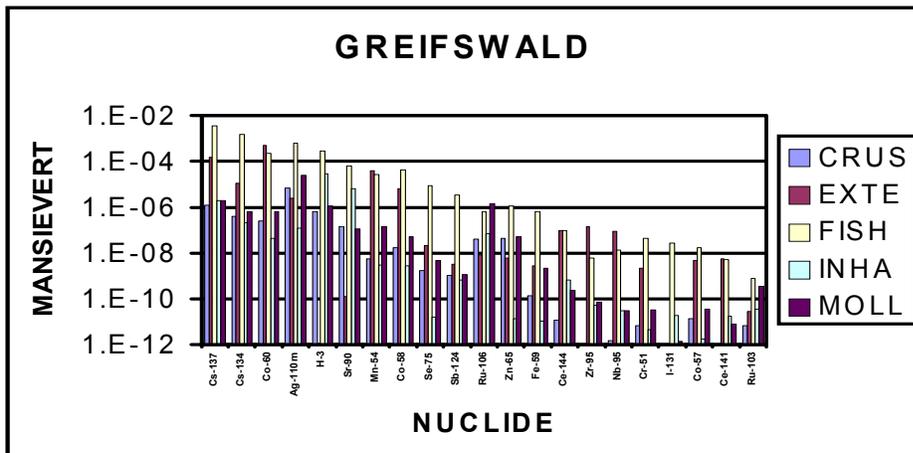


Figure 9.40 Collective doses from discharges into the sea from Greifswald nuclear power plant by exposure pathway and radionuclide.

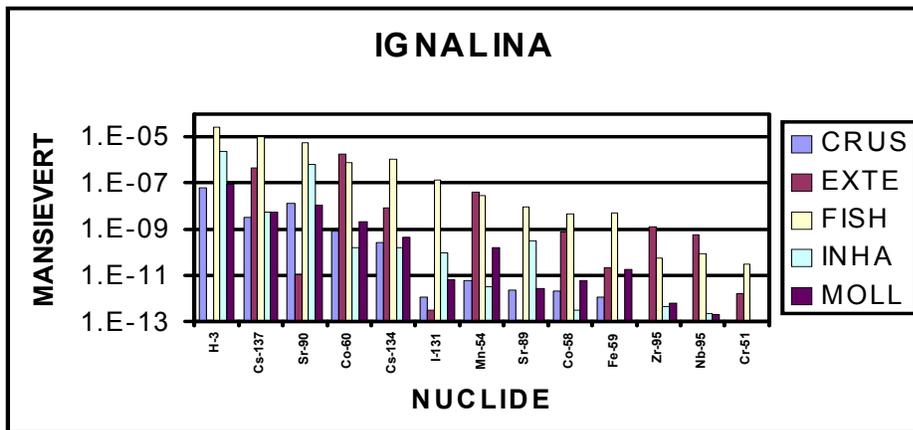


Figure 9.41 Collective doses from discharges into the sea from Ignalina nuclear power plant by exposure pathway and radionuclide.

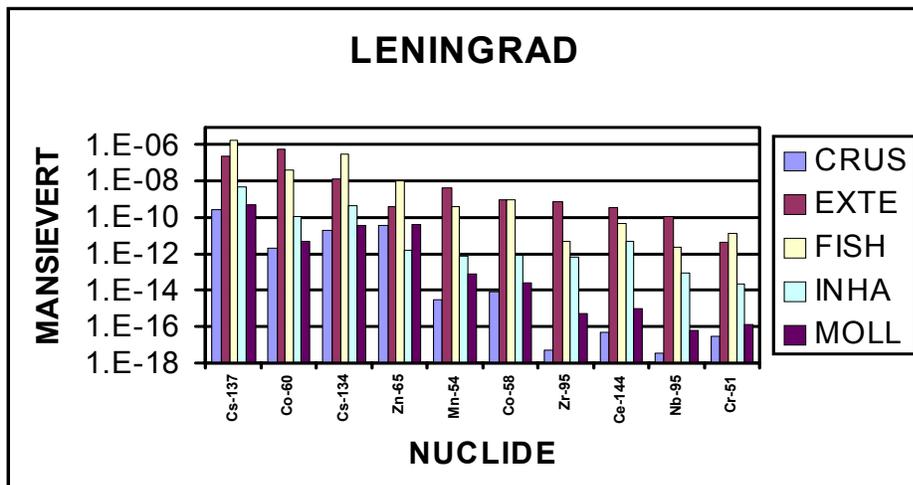


Figure 9.42 Collective doses from discharges into the sea from Leningrad nuclear power plant by exposure pathway and radionuclide.

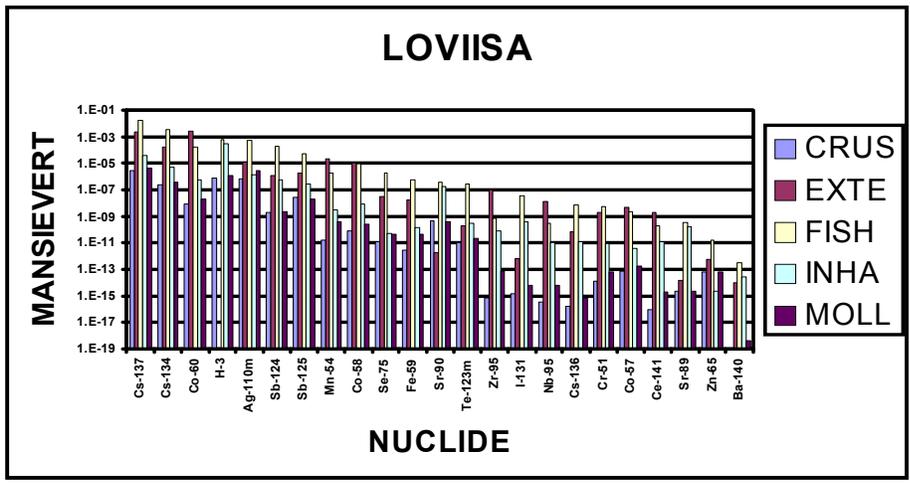


Figure 9.43 Collective doses from discharges into the sea from Loviisa nuclear power plant by exposure pathway and radionuclide.

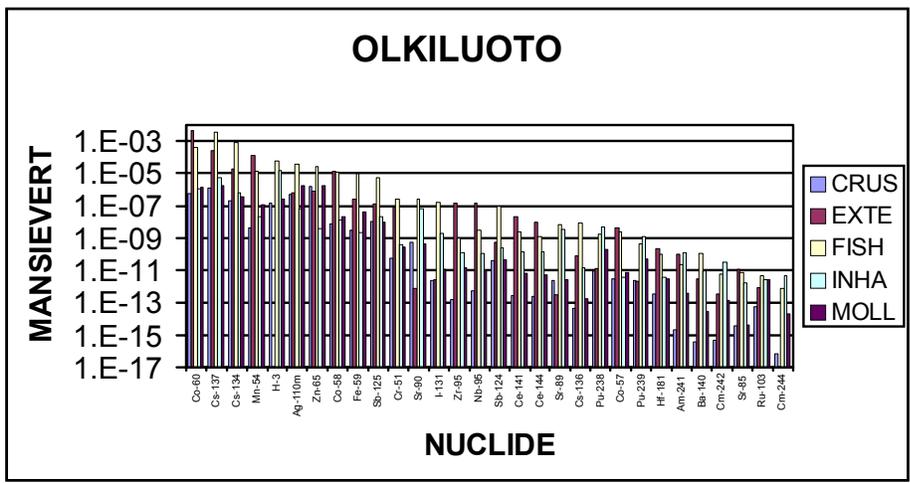


Figure 9.44 Collective doses from discharges into the sea from Olkiluoto nuclear power plant by exposure pathway and radionuclide.

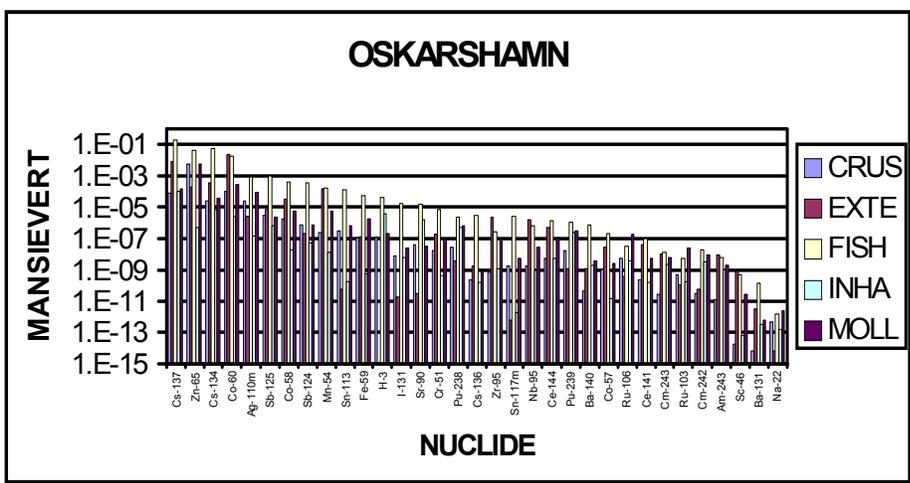


Figure 9.45 Collective doses from discharges into the sea from Oskarshamn nuclear power plant by exposure pathway and radionuclide.

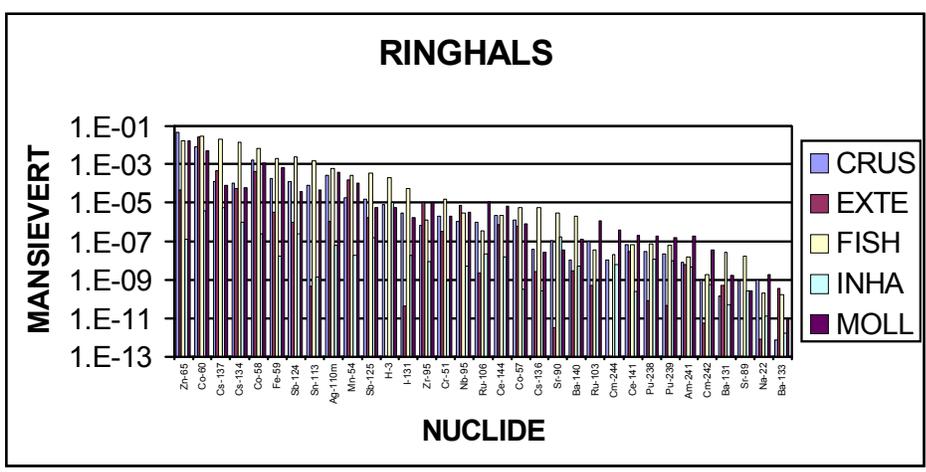


Figure 9.46 Collective doses from discharges into the sea from Ringhals nuclear power plant by exposure pathway and radionuclide.

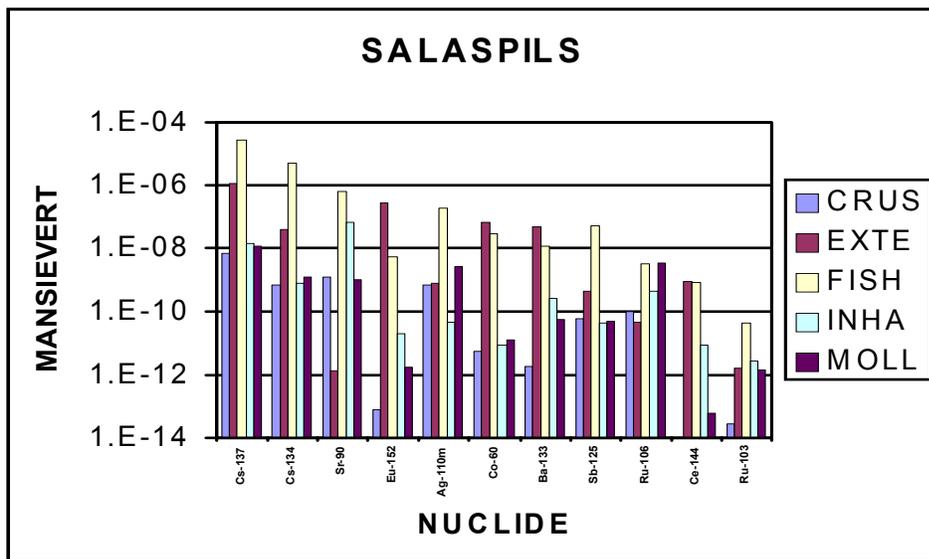


Figure 9.47 Collective doses from discharges into the sea from Salaspils nuclear research facility by exposure pathway and radionuclide.

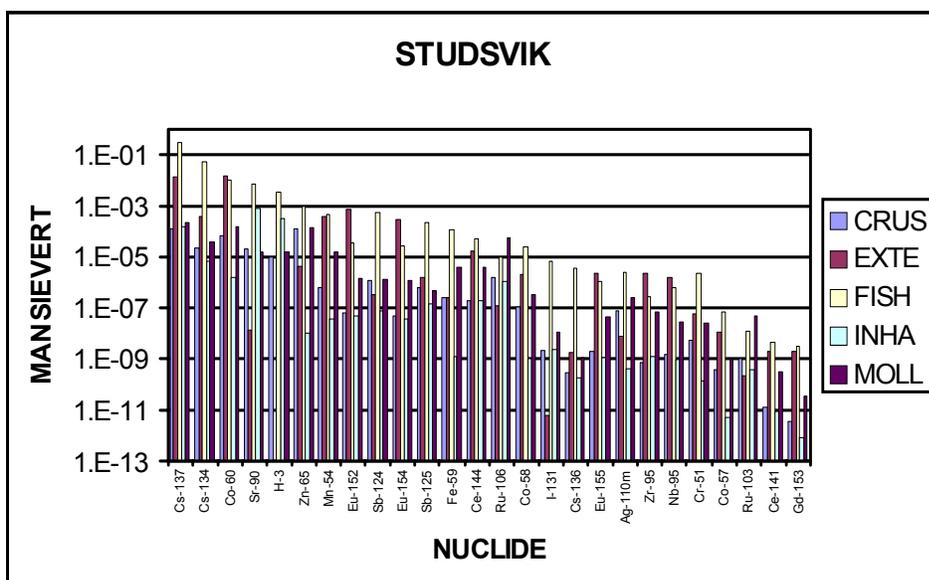


Figure 9.48 Collective doses from discharges into the sea from Studsvik nuclear research facility by exposure pathway and radionuclide.

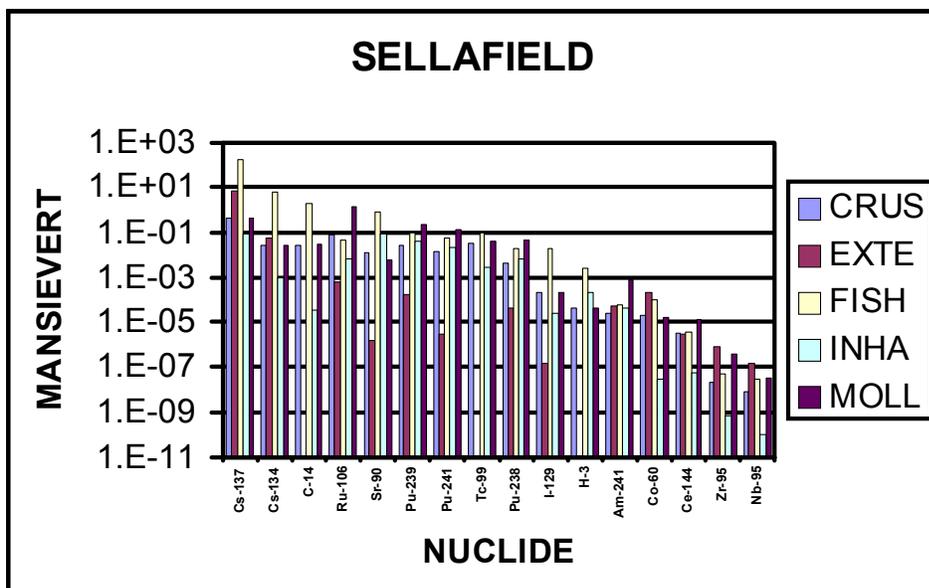


Figure 9.49 Collective doses via Baltic marine exposure pathways from discharges into the sea from Sellafield reprocessing facility by exposure pathway and radionuclide.

10 CONCLUSIONS

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The most significant source of artificial radionuclides in the Baltic Sea is the fallout from the accident at the Chernobyl nuclear power plant in 1986. The most important radionuclides present in this deposition were ^{137}Cs and ^{134}Cs . The total input of ^{137}Cs from Chernobyl into the Baltic Sea has been estimated at 4,700 TBq. Post-Chernobyl river discharges of ^{137}Cs have been calculated in the Marina Balt Study to be 300 TBq, comprising 6-7% of the total input.

The second most important source is global fallout from the atmospheric nuclear weapons tests carried out during the late 1950s and early 1960s. The predominant radionuclides in this global fallout were ^{137}Cs and ^{90}Sr in an activity ratio of about 1.6. During the late 1990s the decay-corrected amounts of weapons-test ^{137}Cs and ^{90}Sr in the Baltic Sea were evaluated at 800 and 500 TBq, respectively.

The corresponding decay-corrected total inputs of ^{137}Cs and ^{90}Sr originating from nuclear reprocessing plants in Western Europe (Sellafield and La Hague) have been estimated at 250 and 40 TBq, respectively. This source is now only of minor importance, due to significant reductions in discharges from Sellafield in recent years.

The predominant radionuclide in the discharges from the nuclear power plants and research reactors in the Baltic Sea region is ^3H . Total discharges of ^3H from these local sources have amounted to 2,400 TBq, while discharges of other beta-gamma nuclides amount to about 20 TBq. Total discharges of alpha nuclides have amounted to some 0.0001 TBq.

For ^{137}Cs , the main sources were fallout from Chernobyl (82%) and from nuclear weapons tests (14%). For ^{90}Sr , the main source of contamination was the fallout from nuclear weapons tests (81%), while the proportion from Chernobyl fallout was lower (13%).

The HELCOM Contracting Parties have carried out monitoring of radioactive substances in the Baltic Sea according to HELCOM Recommendation 18/1, which covers radionuclides in sea water, sediments, fish, aquatic plants and benthic animals. The data from these monitoring programmes and data on discharges of radionuclides have been submitted for the HELCOM databases. Environmental data is managed by the Finnish Environment Institute, while the discharge data is managed by STUK – the Radiation and Nuclear Safety Authority, Finland.

The Contracting Parties have made significant efforts to verify the analytical quality of the environmental data submitted for the database. During the period 1992-1998 the IAEA organised 7 intercomparison exercises covering radionuclides

in sea water, sediment and fish. The results have demonstrated that the quality of environmental data from the MORS-PRO group has been very good.

Transfers of radionuclides in the marine environment have been calculated using a box model designed for assessing the waters of the North Atlantic, including the Baltic Sea. This model includes the dispersion of radionuclides in the marine environment, the transfer of radionuclides to biota, and the calculation of doses to individuals and populations exposed to radionuclides in seafood.

The quality of the model's predictions was investigated by comparing predicted levels of ^{137}Cs and ^{90}Sr in water and fish with observed levels from the HELCOM database. The predicted concentrations were generally in good agreement with the observations for both radionuclides. For the dominating pollutant radiocaesium, the model under-predicts the concentrations in fish by 10% on average, and shows individual agreement generally within a factor of 2. The reliability of the model predictions is considered as satisfactory for radiological-assessment purposes.

An assessment of the radiological consequences of radioactivity in the Baltic Sea was carried out based on information on inputs and observed levels of radioactivity in the Baltic Sea for the period 1950-1996. Doses to members of the public were calculated on the basis of rates of ingestion of radionuclides in seafood from the Baltic Sea and rates of exposure to radioactivity in coastal areas. The calculations cover the period from 1950 to 1996, and include source contributions from nuclear weapons testing, the Chernobyl accident, the two European reprocessing plants at Sellafield and La Hague, and nuclear installations around the Baltic Sea.

Dose rates from anthropogenic radioactivity to individual members of the public (and individuals in critical groups) have been calculated on the basis of rates of annual intake and beach occupancy. The dose rates to individuals from the Bothnian Sea and Gulf of Finland regions are predicted to be larger than for any other area around the Baltic Sea, due to the pattern of fallout from the Chernobyl accident. These dose rates are predicted to have peaked in 1986 at a value of 0.2 mSv y^{-1} .

Collective committed doses to members of the public were calculated according to predicted concentrations of radionuclides in biota and coastal sediments. The total collective dose from anthropogenic radioactivity in the Baltic Sea is estimated at 2,600 manSv, of which about two-thirds originates from Chernobyl fallout, about one quarter from fallout from nuclear weapons tests, about 8% from European reprocessing facilities, and about 0.04% from nuclear installations around the Baltic Sea.

An assessment of the dumping of low-level radioactive waste in the Baltic Sea in the 1960s by Sweden and the Soviet Union has shown that the related doses to man are negligible.

Doses from naturally occurring radioactivity in seafood were calculated on a similar basis, and com-

pared with doses of anthropogenic radioactivity obtained via marine pathways. The results of this comparison show that dose rates and doses from natural radioactivity dominate, except for the year 1986 when individual dose rates from Chernobyl fallout approached those received from natural radioactivity in some regions around the Baltic Sea.

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