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RADIOACTIVITY IN THE BALTIC SEA 1984 - 1991

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Errata to BALTIC SEA ENVIRONMENT PROCEEDINGS No. 61

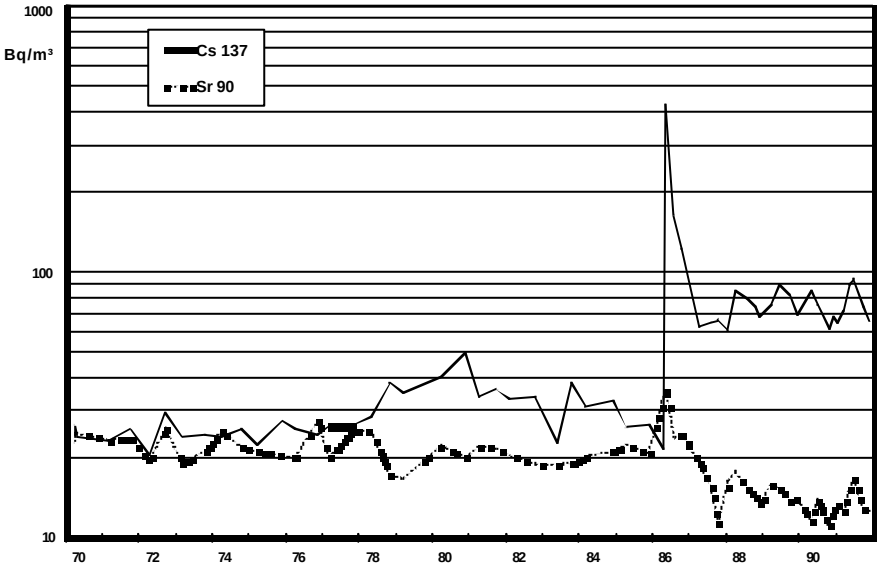
Due to technical errors, Table 2.1.2 (page 8) and figure 5.3.7 (page 66) have been misprinted in above mentioned publication. Please replace accordingly.

Table 2.1.2. Cumulative deposition of Sr-90 and Cs-137 based on national measurements (kBq m^{-2}).

	Denmark		Jutland		Bornholm		Finland		Average ± 1 F	
Period	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137
1950 -1985	1.59	2.60	1.77	2.89	1.41	2.31	1.1	1.8	1.5 ±0.3	2.4±0.5
- 1991	1.41	3.36	1.57	3,74	1.26	2.99	1.0 -1.6	1.8 - 32#	1.4 ±0.15	*

The data are based on the measurements of 17 sampling stations.
* Due to the very uneven distribution averages cannot be calculated.

Figure 5.3.7 Cs-137 and Sr-90 concentrations (Bq/m^3) in surface waters at a site in the Baltic Belt Sea, Schleimündung, in 1970-1991.



RADIOACTIVITY IN THE BALTIC SEA 1984 - 1991

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PREFACE

In 1980 the International Atomic Energy Agency (IAEA) initiated a Coordinated Research Programme entitled "Study of Radioactive Materials in the Baltic Sea". This study contained evaluation of the long-term behaviour of radionuclides entering the Baltic Sea, including their transport back to man and it was carried out by scientists from all countries bordering the Baltic Sea and from the International Laboratory for Marine Radioactivity in Monaco during the years 1981-1984. The results of this research programme provided an excellent knowledge about the levels and the behaviour of artificial radioactivity in the Baltic Sea prior to the reactor accident at Chernobyl.

In 1985 the Helsinki Commission decided to continue the work of IAEA under the auspices of HELCOM and a Group of Experts on Monitoring of Radioactive Substances in the Baltic Sea (EC MORS) was established.

Within the framework of the Helsinki Commission - Baltic Marine Environment Protection Commission - monitoring data on radioactivity have been collected since 1984 under the supervision of EC MORS. The first report of the Group "Three Years Observations of the Levels of Some Radionuclides in the Baltic Sea after the Chernobyl Accident" was published in 1989 in the Baltic Sea Environment Proceedings (No. 31).

The report "Radioactivity in the Baltic Sea in 1984-1991" has been prepared by experts as indicated under each Chapter. The Chairman of EC MORS, Mr. Hartmut Nies, acted as coordinator for the work. A special workshop for the preparation of the report took place in Hamburg, Germany, 21-25 March 1994.

In preparing the report, the experts were assisted by the consultants of the Commission, the Environment Data Centre (EDC) of the National Board on Waters and the Environment of Finland and the Finnish Centre for Radiation and Nuclear Safety (STUK) as well as by the following experts: Ms. Vija Bute (Latvia), Ms. Anita Skujina (Latvia), Ms. Danuta Grzybowska (Poland) and Ms. Maria Suplinska (Poland).

The draft versions of this report have been considered and amended by relevant experts within the framework of EC MORS prior to release. In the Helsinki Commission Secretariat the editorial work has been coordinated by the Environment Secretary, Ms. Eeva-Liisa Poutanen and her assistant Ms. Teija-Liisa Lehtinen. A working group at the Federal Maritime and Hydrographic Agency, Germany, has digitized and postprocessed all graphs under the supervision of Mr. Jürgen Herrmann.

Furthermore, due to the wide time frame covered by the present report and the involvement of several scientists in the MORS work at an earlier stage, especially the contributions of the former Chairmen of EC MORS, Mr. Aarno Voipio, Finland, and Ms. Anneli Salo, Finland, are cordially appreciated.

EXECUTIVE SUMMARY

Within the framework of the Helsinki Commission (HELCOM) monitoring data on radioactivity in the Baltic Sea have been collected since 1984. These data comprise both radioactivity in different compartments of the Baltic marine environment (water, sediment and biota) and data on discharges from nuclear installations (power stations, research facilities etc.) on the drainage area of the Baltic Sea.

All the data collected have been evaluated by experts in order to provide the scientific community and the public with an assessment about the situation of the radioactive burden of the Baltic Sea. The Baltic Sea received a dramatic change of its radioactive inventory in April/May 1986 due to the Chernobyl accident. The Baltic Sea is still today the marine area together with the Irish Sea and the Black Sea where the levels are highest compared to other sea areas in the world. Besides this source, previous sources of artificial radioactivity in the Baltic Sea were global fallout and transport of contaminated seawater from western European reprocessing plants, primarily Sellatield, by the prevailing current systems through the Danish Straits. Compared to the sources

global fallout,
water transport from Sellafield and La Hague or
Chernobyl fallout,

the input from nuclear power stations or research centres on the drainage area is extremely low.

The dominating radionuclide is Cs-137 with a half-life of 30 years. The concentration of the two years half-life nuclide Cs-134 which was also deposited into the Baltic after the Chernobyl accident meanwhile decreased significantly by radioactive decay. The highest deposition occurred in the southern part of the Bothnian Sea and in the Gulf of Finland.

The monitoring of radioactivity has always been supported by an intense quality assurance programme in order to assure the comparability of the data. Several intercalibrations of samples of seawater, sediment and biota were carried out by the International Atomic Energy Agency (IAEA) with its Marine Environmental Laboratory in Monaco. These attempts showed that the data produced by the laboratories responsible for monitoring are of very good quality.

The Baltic Sea is a semi-enclosed and shallow brackish water body. Consequently, it can be expected that the Cs-137 deposited into the Baltic Sea very inhomogeneously in 1986, will remain for longer periods in the water column. The inventory of Cs-137 in seawater was estimated up to 320 TBq in 1984 and about 4600 TBq in October 1986. In 1991 the highest levels in seawater were still observed in the Bothnian Sea, however, during 1991 a more homogeneous distribution in seawater was ascertained due to mixing and horizontal transport of water masses. The concentration of Cs-137 decreased in the water phase also by adsorption to particles and subsequent deposition to the sediment. Levels of artificial radionuclides other than Cs-134 , Cs-137 and Sr-90 are extremely low. The main source for Sr-90 and Pu-isotopes is still the global fallout from atmospheric nuclear weapon tests during the fifties and sixties.

The levels of Cs-137 and Cs-134 in sediment reflect in general the deposition pattern of the

activity deposited to the Baltic Sea initially and on its drainage areas by the Chernobyl fallout. However, the levels are also very much depending on the sediment type and sedimentation rate at various locations. About 20 to 25 % of the total Cs-137 inventory in the Baltic Sea is meanwhile located in the sediments. In the vicinity of nuclear installations low levels of some further radionuclides from the fuel cycle could be detected.

The concentrations measured in biota show the same pattern in time and space as the distribution of radionuclides in seawater. The dominating artificial radionuclides in biota are Cs-137 and Cs-134 from the Chernobyl accident. The concentration factors water/fishflesh for radiocaesium varies between 30 and 500 for typical marine fish species.

A box model was used to calculate Cs-137 and Sr-90 distribution in the Baltic Sea taking into account the above mentioned four main sources. The model also predicts the evolution of the radionuclide concentrations in seawater, sediment and biota to the year 2000. The comparison between measured and calculated values are sufficient. The results show that the accuracy of the model predictions is better than a factor of two.

The box model was used to estimate the radiation exposure from Cs-137 and Sr-90 of the population in the riparian states from the four main sources by consumption of marine food. The calculations show that the dose rates are clearly dominated by the nuclide Cs-137 which is about one to two orders of magnitude higher than the dose caused by Sr-90. This is due to a much higher level of contamination in Baltic seawater of Cs-137 (about 100 Bq m”) than of Sr-90 (about 10 Bq m”) and a higher biological uptake of radiocaesium in fish from seawater than of radiostrontium. The maximum individual dose rate was calculated for 1986 at a value of 0.2 mSv per year. The corresponding annual dose rate caused by natural radionuclides in seafood (Polonium-210) is about 1.2 mSv, or six times higher. Comparing the dose rate received by marine contamination with that from terrestrial pathways which range from two to five mSv per year in the riparian countries, one can conclude that the additional exposure from marine pathways to a potential critical group was less than 10 % in 1986. At present radiation doses to man from marine pathways are only in the order of 1 % of those from terrestrial pathways of the population in the countries bordering the Baltic Sea.

INTRODUCTION

Natural and artificial radioactive substances are present in all compartments of the environment. However, due to their potential property as extremely harmful substances to humans and biota the use and release of radionuclides are restricted to a significant extent. These restrictions are regulated within national and international conventions. HELCOM established a group of experts for the Monitoring of Radioactive Substances (EC MORS) in the Baltic Sea in 1985. The first meeting of this group of experts took place in Helsinki in April 1986 just three weeks before the accident at Chernobyl.

The group MORS has carried out a comprehensive investigation and monitoring of the Baltic Sea marine environment. Numerous measurements have been performed on seawater, sediment, and various species of biota over the period of time of the existence of that group. Furthermore, the radiological situation was well documented before the Chernobyl accident by a Coordinated Research Project funded by the International Atomic Energy Agency between 198 1 and 1985. The monitoring data gained since 198 1 to 1991 from the laboratories in the Baltic Sea states provided an excellent knowledge about the spatial and temporal distribution of various radionuclides in the Baltic Sea.

The measurements have always been certified by an intensive programme for quality assurance for the data. This part was mainly carried out by the Marine Environment Laboratory (MEL) of the IAEA at Monaco. The results produced by the participating laboratories within these intercomparison exercises showed a very good quality of determination of radionuclides compared to other world wide participants. Therefore, the data presented in this report "Radioactivity in the Baltic Sea 1984 - 1991" are reliable.

This report comprises all aspects on artificial radionuclides in the Baltic Sea including pre-burden from atmospheric nuclear weapon tests in the sixties, inflow of water contaminated by Sellafield and La Hague through the Danish Straits; the report compares the levels derived from discharges from nuclear installations in regular operation around the Baltic Sea, it shows the significant change of the inventory primarily of the radiologically most significant radionuclide Cs-137 after the deposition from the Chernobyl accident and its subsequent change; the assessment about the dose to man completes the report.

The document presents the actual knowledge about the behaviour of radionuclides in the largest brackish water area of the world, the Baltic Sea.

The following division of the sub-basins of the Baltic Sea is used in this report, except for model and dose calculations in Chapters 8 and 9:

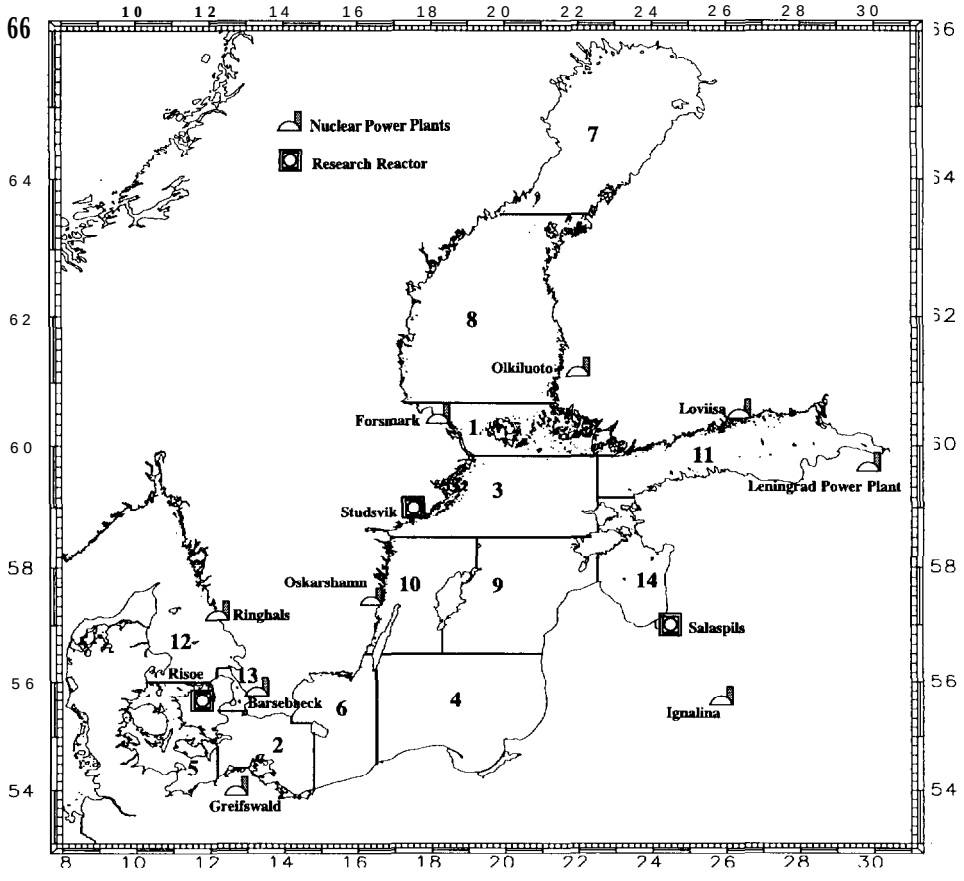
1. Archipelago Sea and Åland Sea
2. Arkona Sea
3. Northern Baltic Proper
4. Southern Baltic Proper
5. Belt Sea
6. Bornholm Sea
7. Bothnian Bay

8. Bothnian Sea
9. Central Baltic Proper (Gotland East)
10. Central Baltic Proper (Gotland West)
11. Gulf of Finland
12. Kattegat
13. Sound
14. Gulf of Riga

The geographical extent of the sub-basins is shown in Figure 1.1.

For the assessment of radioactivity in the Baltic Sea, the key question is the radiation dose to man by the consumption of marine food or other pathways. Man-made radioactivity is to be compared to naturally occuring radioactivity. The final results of this report show that, on account of the environmental levels detected up to now, the contribution of the first is relatively low compared to the latter. However, the use of nuclear power in one country may also mean a potential impact to areas in neighbouring countries. The work of the Group of Experts MORS has been and will be of significant value for better understanding the dispersion of radionuclides and other pollutants in the Baltic Sea. The Group cooperated in excellent manner during the previous years and represents an important working body in nuclear expertise for the Contracting Parties of the Baltic Sea.

Figure 1.1. Division of the Baltic Sea into sub-basins and the location of the nuclear power plants and research reactors.



2. SOURCES OF RADIOACTIVITY IN THE BALTIC SEA

Hartmut Nies [1], Ryszard Bojanowski [2], Olof Karlberg [3], Sven P. Nielsen [4]

1) Federal Maritime and Hydrographic Agency, Germany
2) Institute of Oceanology, Polish Academy of Sciences, Poland
3) Swedish Radiation Protection Institute, Sweden
4) Risø National Laboratory, Denmark

Artificial radioactivity in the Baltic Sea area originates mainly from four sources:

I **Global fallout**
from the nuclear weapon tests primarily during the sixties, whereas, part of the radionuclides deposited directly to the sea surface and part of it being discharged into the sea indirectly via river run-off from land territory.

II **Fallout from the accident at the Chernobyl nuclear power plant in April 1986.**

III **Discharges from western European nuclear reprocessing plants**
at La Hague (F) and Sellafield (UK) by means of the transport of contaminated water through the Belt Sea.

IV **Direct discharges into the Baltic Sea**
from nuclear power plants and other nuclear facilities such as research centres, located within the drainage area of the Baltic Sea.

Other sources, e.g. application of short lived nuclides in hospitals, are considered to be negligible in relation to the above mentioned ones and are therefore not included in this study.

Efforts to quantify these sources have been carried out by several authors. In the following chapter, a discussion of these efforts is made and the relative contribution of these sources is shown.

2.1. Fallout

The term "fallout" is commonly applied to substances which are released to the atmosphere and settle onto the ground by means of both dry or wet ("rainout") deposition. In the domain of radioactivity studies it is generally used to describe debris ejected into the atmosphere as a result of nuclear weapon testing. Atmospheric nuclear tests giving rise to radioactive pollution of air and, subsequently to the earth's surface, have been conducted since 1945. With a few exceptions which were mainly on local impact, e.g. the Windscale reactor fire in 1957 (UK) (CEC, 1991), there were no other sources of radioactivity that would contribute noticeably to atmospheric fallout prior to the accident at Chernobyl. Due to the different nature of these sources it is reasonable to treat them separately.

2.1.1. Global Fallout

The total fission yield of atmospheric nuclear tests performed until the end of 1980 has been estimated at 217.2 Mt. Nearly 90 % of the resulting releases occurred before 1963 and only about 1 % originated from tests between 1976 and 1980 (UNSCEAR, 1982). Depending on the type of the test up to 50 % of the radioactive debris were deposited in the vicinity of testing sites giving rise to local fallout. The remainder was injected into the various atmospheric strata and were entrained in the global circulation of air masses. The portion injected into the stratosphere (i.e. at the altitude above 9 - 17 km) represents a major reservoir of world-wide (global) fallout. It accounts for most of the world-wide contamination of long-lived fission products (UNSCEAR, 1983).

The mean residence time of aerosols in the lower stratosphere ranges from a few months to about two years and the most rapid passage through the tropopause takes place in the Northern Hemisphere in spring, giving rise to a characteristic time-dependent fallout pattern over the year.

The distribution of global fallout on the earth surface is uneven. The majority of large yield explosions were carried out in the Northern Hemisphere within the 30-70" latitude belt. The subsequent atmospheric transport of fallout debris and its deposition generated the most intense deposition in the belt between 40" to 60" northern latitude.

2.1.1.1. Sr-90 and Cs-137 Deposition in the Baltic Sea Area

Based on the values given in the UNSCEAR Report (1982) one can calculate the accumulative deposition of Sr-90 and Cs-137 onto the Baltic Sea area. The Baltic Sea extends from 54" to 66" N which means that it lies within the zone of the most intensive global fallout. In order to estimate the total amount of representative radionuclides from nuclear explosions that fell on the surface of the Baltic Sea, the area was divided into two zones, to account for the latitude dependent difference in fallout density. The parallel of 60" N was arbitrarily chosen as a division line. Accordingly, the weighted integrated average fallout density was then estimated for each zone. The resulting values were then recorded for decay to the date of Chernobyl accident (26 April 1986).

Table 2.1.1. Cumulative deposition of Sr-90 and Cs-137 fallout radionuclides on the Baltic Sea surface to the end of 1980 based in UNSCEAR data (decay corrected to 26 April 1986).

Sea area	Surface area 10^3 km^2	Deposition density 10^3 Bq·m^{-2}		Deposition PBq	
		Sr-90	Cs-137	Sr-90	Cs-137
< 60 ° N	285.5	1.54	2.46	0.44	0.70
> 60 ° N	130.8	1.10	1.76	0.14	0.23
Total average	415.3	1.41	2.25	0.58	0.93

The product of deposition rates and the respective sea surfaces gave a total deposition of the global fallout in these zones. Strontium-90 was chosen as a representative radionuclide, because it is one of the most important fission nuclides, has a sufficient long half life, can be determined with high accuracy and has the longest measurement record dating back to the early 1950's. The deposition values for Cs-137 were obtained by multiplying the Sr-90 data by a factor of 1.6 representing the almost constant activity ratio of these two nuclides in global fallout.

The deposition values calculated from UNSCEAR data can be compared with the results of direct measurements at the stations situated around the Baltic basin to check if the data are mutually consistent.

Among the Baltic countries Denmark maintains the longest record of Sr-90 deposition values which date back to the early 1950's (Aarkrog at al., 1992). Apart from the annual deposition values, available are also decay-corrected cumulative deposits for each consecutive year. The results were obtained from three stations. In Finland the accumulated deposition of Sr-90 and Cs-137 was estimated since 1955, regular measurements began in 1960 (Salo et al., 1984; Saxén, pers. corn.; STUK, annual reports). The pre-Chernobyl values for Sr-90 and Cs-137 and accumulated deposition values after the Chernobyl accident until 1991 are shown in Table 2.1.2.

Fable 2.1.2. Cumulative deposition of Sr-90 and Cs-137 based on national measurements ($kBq \cdot m^{-2}$).

	Denmark		Jutland		Bornholm		Finland		Average ± 1 a	
Period	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	Cs-137	Sr-90	cs-137
1950 - 1985	1.59	2.60	1.77	2.89	1.41	2.31	1.1	1.8	1.5 ±0.3	2.4±0.5
- 1991	1.41	3.36	1.57	3.74	1.26	2.99	1.0 - 1.6	1.8 - 32 #	1.4 ±0.15	*

The data are based on the measurements of 17 sampling stations.
* Due to the very uneven distribution averages cannot be calculated.

In comparison with the results given in Table 2.1.1. the average values do not differ significantly.

The maximum cumulative deposit of Sr-90 was recorded in the Northern Hemisphere in 1966. Since then a continuous decrease of activity supply was observed as a result of the predominance of physical decay over fresh fallout input. According to the Danish data, the maximum for both Sr-90 and Cs-137 appeared in the Baltic basin only in 1971, i.e. five years later (Aarkrog et al., 1988b and 1992).

The period of steady decrease of Cs-137 fallout activity was interrupted by the Chernobyl fallout in 1986. Sr-90 was present in the Chernobyl fallout on the Baltic Sea area about a

factor of hundred lower than Cs-137. As a result, the cumulative Sr-90 deposition was at the end of 1991 about the same level as in 1985.

From 1981 to the accident at Chernobyl in 1986, there were no remarkable releases of fission products to the atmosphere. Since the stratospheric reservoir is nearly exhausted, the recently observed changes in the intensity of fallout are practically entirely due to physical decay. As a consequence, Sr-90 and Cs-137 activities in the global fallout continue to decrease at a rate of 2.4 and 2.3 % per year, respectively.

2.1.1.2. Contents of Sr-90 and Cs-137 in Soil

Soils are capable of retaining Cs and Pu very effectively by adsorption to clay minerals. Strontium, however, is adsorbed less strongly and penetrates to greater depths in soil profiles but, in principle, can also be quantified provided sufficiently long cores have been taken. It is also more accessible to river runoff water. Therefore, a continuous contribution of weapon's fallout Sr-90 was detectable in river water. There exist a few soil profiles at a few stations around the Baltic Sea where the integrated activities of Cs-137, Sr-90, and Pu-239,240 were measured (Table 2.1.3).

Table 2.1.3. Cumulative soil contents of Sr-90 and Cs-137 at some locations around the Baltic Sea.

Sampling region	**Year of sampling**	**Sr-90** ($kBq \cdot m^{-2}$)	cs-137 ($kBq \cdot m^{-2}$)	Reference
Germany				BMU,
Kiel	1985	2.13	3.98	1985
Bad Bramstedt	1985	1.75	3.73	
Finland				STUK,
Loviisa	1983	0.82	2.0	1987
Olkiluoto	1985	0.60	1.7	

Considering inherently lower accuracy of deposition values derived from soil profiles, their agreement with the results in Table 2.1.1 is satisfactory.

2.1.2. **Chernobyl** Fallout

The amounts of radioactive substances (excluding noble gases) released to the atmosphere as a result of the Chernobyl accident were estimated to l-2-10" Bq (IAEA, 1986). Seo et al. (1987) calculated a value of $4.8 \cdot 10^{18}$ Bq by integrating the available data of deposited activity within a 3000 km radius around the point of release which is more than twice as high than the total amount of the estimated figure from the USSR State Committee given in the IAEA

report. The relative activity contribution of the most important radionuclides at various locations in Europe is given by Aarkrog (1988a). Most of the nuclides released are short-lived and their impact on the environment was negligible.

Among the longer-lived radionuclides Cs-137 is the most important one because of its long half life and its relevance for dose contribution. Its total amount released to the environment has been estimated from 38 PBq ± 50 % (IAEA, 1986) to 167 (Seo, 1987). The most frequently used value is 70 PBq corresponding to ca. 25 % of the Cs-137 calculated as the inventory in the reactor core (Bennet, 1990). The Cs-137/Sr-90 ratio measured on air dust collected at Warsaw (Poland) in the period of 29 April to 8 May 1986 was found to have varied from 80 to 270 with an average value of 170 ± 70 (Bojanowski, priv. corn.). In Nurmijärvi (Finland) the mean Cs-137/Sr-90 ratio measured in rainwater samples from the months April and May 1986 was 79 (STUK, 1992). At Roskilde (Denmark) the mean Cs-137/Sr-90 ratio measured in air samples from May 1986 was 53, and for the peak sample from 27-28 April 1986 the ratio was 42 (Aarkrog et al., 1988b).

Measurements surveys in order to map the fallout have been carried out by many countries around the Baltic Sea. These surveys were used to draw a map showing the fallout density for Cs-137 on the drainage area of the Baltic Sea (Fig. 2.1.1).

2.1.2.1. Source Estimation for Cs-137 Based on Cs-134 Measurements

Due to the pre-contamination of the Baltic Sea by weapon fallout and the additional input from west European reprocessing plants it is not simple to determine the contribution of Cs-137 and Sr-90 from the Chernobyl fallout in the Baltic Sea. However, the fairly constant activity ratio of Cs-134/Cs-137 in the Chernobyl fallout permits to give an estimate of the Chernobyl contribution of Cs-137 based on the Cs-134 measurements. This nuclide was not detectable before the Chernobyl accident in the Baltic Sea.

Table 2.1.4 shows an estimation of the Cs-137 input after the Chernobyl accident, based on measured concentrations of Cs-134 (Nies and Wedekind, 1988; Nies, 1989) in the water column on more than 90 positions in October 1986. From these water data the apparent area1 deposition was calculated. The values were obtained from averages of measured depth profiles in respective areas.

Figure 2.1.1. Terrestrial deposition of caesium-137 in the Baltic Sea drainage area (kBq/m²); compiled by STUK, Helsinki.

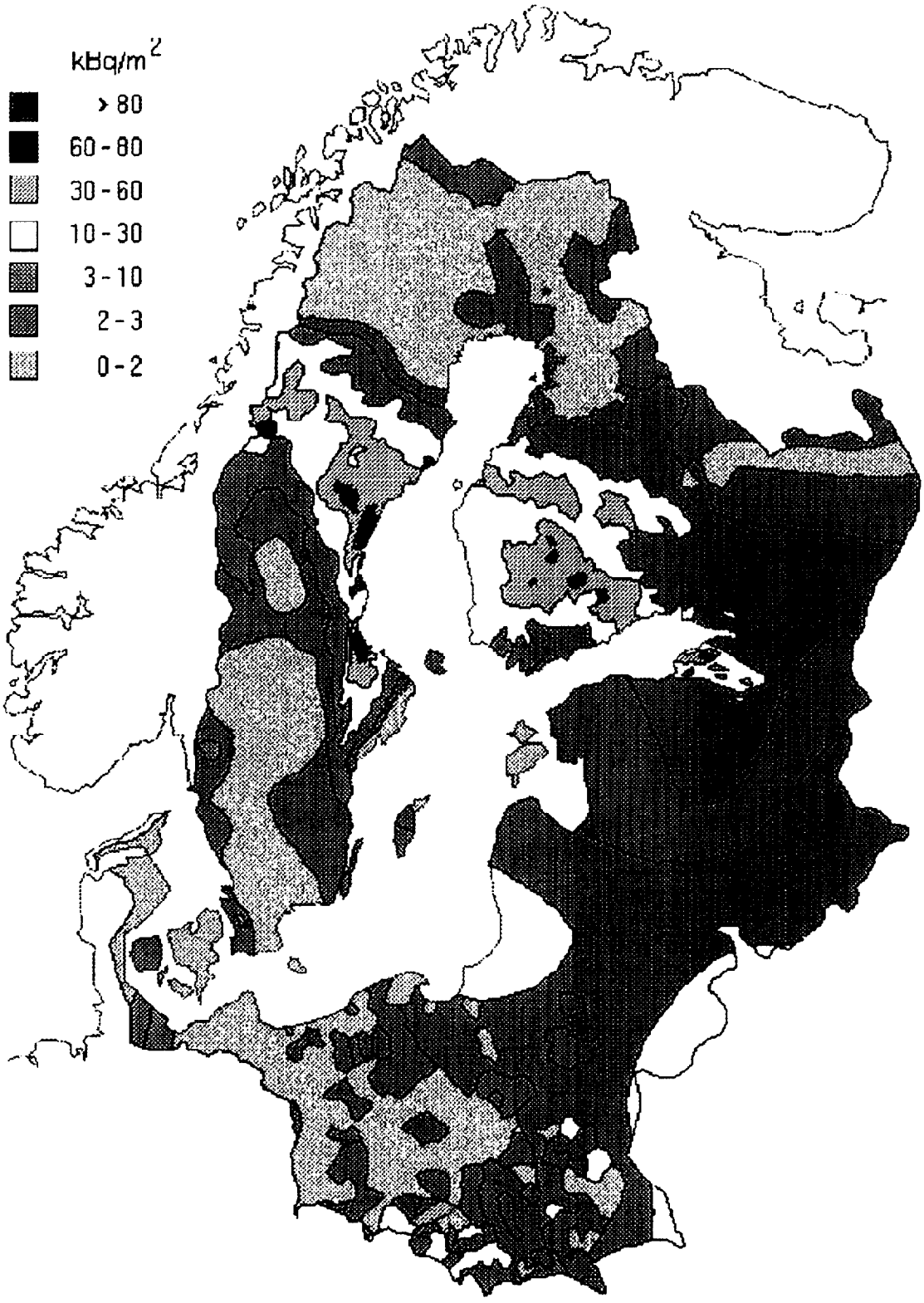


Table 2.1.4. Apparent deposition and inventory of Chernobyl derived Cs-137 in seawater of the Baltic Sea based on Cs-134 concentrations in October 1986; the data are decay corrected to 1 May 1986. The median value of the area1 deposition was calculated in different subareas.

Basin	Area km²	Deposition density kBq·m^{-2}	Inventory PBq (= 10^{15} Bq)
Bothnian Bay	37200	6.9	0.29
Bothnian Sea	65600	35.0	2.33
Archipelago Sea	8300	17.3	0.15
Åland Sea	5200	72.5	0.38
Gulf of Finland	29498	8 *)	0.31 *)
Gulf of Riga	17913	4 8	0.05
Baltic Proper	209930	2.7 - 7.5 #	0.58 - 1.57
Belt Sea	18900	1.8	0.03
Kattegat	22287	1.7	0.04
Sund	2300	0.4	0.00
Total	417128	12	4.16 - 5.16

§ Estimated value

Due to the very uneven distribution with a large range of data, it is very difficult to calculate a reliable mean value. In the area of the Baltic Proper the geometric mean represents the lower value. The upper value is based on the arithmetic mean of the measured data.

*) probably underestimated due to insufficient number of data

As can be seen by this estimation of the Chernobyl derived Cs-137 inventory about half of the Chernobyl fallout was deposited in the Bothnian Sea and about 10 % reached the Gulf of Finland. A significant amount was also deposited to the largest area, the Baltic Proper (between 15 and 30 %). However, the very uneven distribution of a large range of data does not permit to give a single central estimate.

2.1.2.2. Source **Estimation for Cs-137 Based on Profiles in Seawater in 1986 and from Cs-137 Increments in Sediments**

These values are yet to be estimated from the data in Chapter 5 and 6, provided that the number of suitable results are sufficient for this purpose. Estimation of Chernobyl fallout from Cs-137 concentrations in seawater should yield essentially the same result as the one derived from Cs-134 concentrations, if allowance is made for the pre-Chernobyl Cs-137 in Baltic waters. The latter can be taken from the paper by Salo and Tuomainen (1986). After correction for decay to May 1986 the total Cs-137 inventory in seawater was, at that date, $0.32 \cdot 10^{15}$ Bq (0.32 PBq). The corresponding result for Sr-90 would be $0.43 \cdot 10^{15}$ Bq. The first estimate following the Chernobyl accident gave an increase up to between 4.26 and 4.62 PBq (Chapter 5, Table 5.1). The net value that can be ascribed to the Chernobyl input would thus lie in the range of 3.94 to 4.30 PBq, i.e. very similar to the values obtained from the two preceeding estimations.

In considering the total input of Cs-137 due to Chernobyl fallout it must be borne in mind that a fraction of radioactivity carried by airborne particles was not dissolved in seawater, but reached the bottom, giving rise to enhanced accumulation in sediments (Chapter 6). The total input of Cs-137 and other radionuclides should be calculated by summing up the inventories in both seawater and sediments. We may expect that a significant amount of the deposited radiocaesium settled quickly to the bottom and was thus excluded from the inventory calculations based on Cs- 134 and Cs-137 concentrations in water.

2.1.3. River Run-off

Besides the direct fallout onto the Baltic Sea surface, radioactive material was also transported from the whole catchment area by river water with so-called river run-off. The river run-off is considered to be more important for strontium than caesium due to its higher solubility and the higher adsorption affinity of caesium to clay minerals. However, the input of river run-off was mainly significant during 1986, directly after the deposition.

The river run-off cannot be directly estimated in a straightforward way. Model approaches, based indirectly on measurements, could however be used to estimate the relative contribution of this process, and such an analysis is carried out in model analysis Chapter 8.2.

2.2. Discharges from **Western European Nuclear Reprocessing Plants**

Discharges from the reprocessing plants at Sellafield and, of minor importance from La Hague, influenced the concentration in the Baltic Sea by the inflow of saline water through the Danish Straits. The transport time is about four to five years after the discharge into the Irish Sea or about two years after the release into the Channel, respectively. Cs-137 was the dominating nuclide from Sellafield and combined with the fact that caesium is transferred more rapidly from the water column to the sediments in lower salinity water, an almost linear Cs-137/salinity relationship was ascertained in the Baltic Sea before 1986. However, this source became meanwhile of minor importance due to significant reductions of the discharges at Sellafield during recent years.

To estimate the contribution from this source in a quantitative way, again model approaches must be used as described in Chapter 8.

2.3. **Discharges from Nuclear Installations in the Baltic Sea Area**

Liquid radioactive discharges into the Baltic Sea originate from a number of nuclear installations on the catchment area as shown in Fig. 1.1. The installations comprise nuclear power plants as well as nuclear research reactor sites with different types of laboratories. Table 2.3.1 shows the present installations.

Table 2.3.1. Nuclear installations on the drainage area of the Baltic Sea (locations given in Fig.1.1).

Installation	Country	Type of installation; number of units	Remarks
Greifswald	Germany	Power plant; 2 PWR	Shut down in 1990
Risø	Denmark	Research reactor	Tritium discharges only
Ringhals	Sweden	Power plant; 3 PWR, 1 BWR	
Barsebäck	Sweden	Power plant; 2 BWR	
Oskarshamn	Sweden	Power plant; 3 BWR	
Forsmark	Sweden	Power plant; 3 BWR	
Studsvik	Sweden	Research reactor	
Olkiluoto	Finland	Power plant; 2 BWR	
Loviisa	Finland	Power plant; 2 PWR	
Leningrad	Russia	Power plant; 4 RBMK	Discharges not included
Salaspils	Latvia	Research rector	"
Ignalina	Lithuania	Power plant; 2 RBMK	"
Paldiski	Estonia	Training centre	Shut down 1995?

The discharges from some of the installations are, as indicated in the table, not included in the quantitative analysis due to missing or partially missing information. However, the discharges from these installations are expected to be small and will not influence the results significantly.

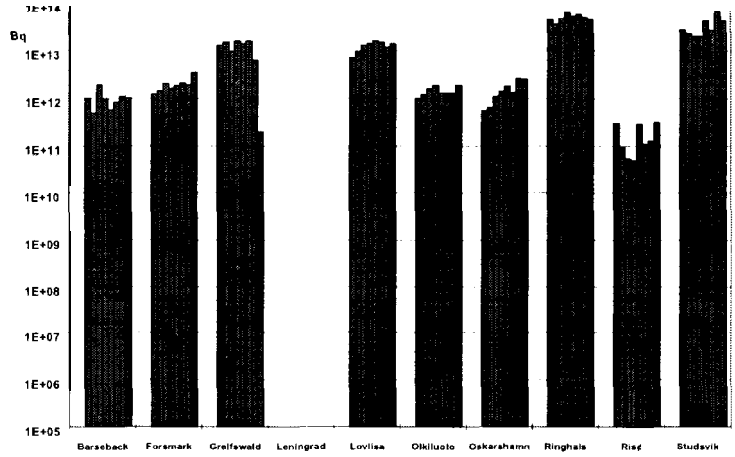
There also exist other installations in the area such as fuel factories, university laboratories, old closed power reactors etc., but the recipient of their discharges are mainly inland lakes and the contribution to the Baltic Sea from these installations are negligible. Discharges from hospitals and other institutions working with radioactive materials have also been excluded due to the extreme short half-lives of the applied radionuclides.

The release procedures used by the installations in Table 2.3.1 were investigated within the MORS group by means of a questionnaire to the authorities in the contracting countries. The questionnaire concerned release quantities, frequencies, total volumes, analysis methodologies, equipment etc. The main conclusion from the questionnaire is that the discharge measurement procedures seem to be reliable in general and that the accuracy of the values reported are acceptable with respect to the radiological consequences originating from the discharges.

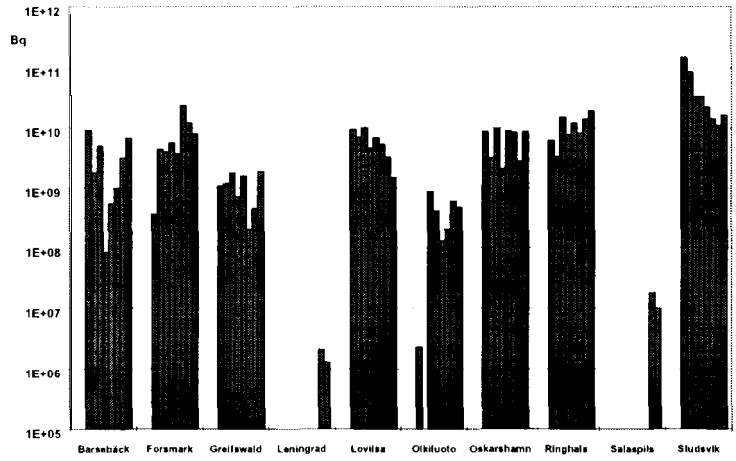
The information on the reported discharges has been stored in a database by the Finnish Centre for Radiation and Nuclear Safety (STUK) in form of annual values for a selection of the most important nuclides. The data for some longer lived radionuclides are summarized in Table 2.3.2 which shows the total input to the Baltic Sea per year as well as the decay corrected input to the year 1991 from these installations. As can be seen, there is no general trend of either increasing or decreasing values. Figure 2.3.1 shows the annual discharges of three nuclides as reported to the HELCOM database from each installation in the period 1984-1991.

Figure 2.3.1. Annual discharges 1984-1991 from nuclear installations into the Baltic Sea.

a. Tritium



b. Cs-137



c. Co-60

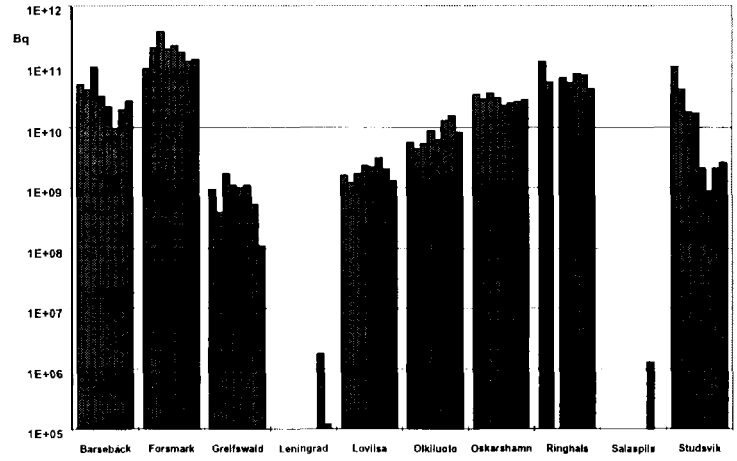


Table 2.3.2. Total discharges of some longer lived radionuclides to the Baltic Sea from all nuclear installations as reported to HELCOM

Discharges during the year/
Discharges **decay corrected to** 1991

(Bq/year)

Nuclide / half-live (years)	**1984**	**1985**	**1986**	**1987**	**1988**	**1989**	**1990**	**1991**	**Total**
Co-58 / (0.195)	1.86E+11 **2.92E+00**	1.37E+11 **7.55E+01**	1.49E+11 **2.86E+03**	1.09E+11 **7.34E+04**	1.32E+11 **3.09E+06**	1.49E+11 **1.22E+08**	2.13E+11 **6.10E+09**	1.15E+11 **1.15E+11**	**1.21E+11**
Co-60 / (5.3)	4.05E+11 **1.62E+11**	3.74E+11 **1.71E+11**	5.29E+11 **2.75E+11**	3.47E+11 **2.06E+11**	3.31E+11 **2.23E+11**	2.99E+11 **2.30E+11**	2.59E+11 **2.27E+11**	2.40E+11 **2.40E+11**	**1.73E+12**
Cs-134 / (2.06)	6.47E+10 **6.16E-09**	3.35E+10 **4.46E+09**	2.49E+10 **4.63E+09**	1.68E+10 **4.38E+09**	2.43E+10 **8.88E+09**	4.05E+10 **2.07E+10**	2.35E+10 **1.68E+10**	1.39E+10 **1.39E+10**	**7.99E+10**
cs-137 / (30.17)	1.85E+11 **1.57E+11**	1.07E+11 **9.32E+10**	7.96E+10 **7.09E+10**	5.42E+10 **4.95E+10**	5.59E+10 **5.22E+10**	6.03E+10 **5.76E+10**	4.74E+10 **4.63E+10**	6.23E+10 **6.23E+10**	**5.89E+11**
H-3 / (12.35)	9.81E+13 **6.63E+13**	8.82E+13 **6.30E+13**	9.64E+13 **7.28E+13**	1.21E+14 **9.66E+13**	1.33E+14 **1.12E+14**	1.22E+14 **1.09E+14**	1.43E+14 **1.36E+14**	1.12E+14 **1.12E+14**	**7.68E+14**
Mn-54 / (0.855)	2.57E+10 **8.84E+07**	3.17E+10 **2.45E+08**	6.05E+10 **1.05E+09**	2.52E+10 **9.86E+08**	1.93E+10 **1.69E+09**	1.80E+10 **3.55E+09**	2.21E+10 **9.84E+09**	2.07E+10 **2.07E+10**	**3.82E+10**
Sb-125 / (2.77)	5.80E+09 **1.01E+09**	4.13E+09 **9.21E+08**	5.44E+09 **1.56E+09**	6.64E+09 **2.44E+09**	8.43E+09 **3.98E+09**	8.05E+09 **4.88E+09**	1.03E+10 **8.05E+09**	9.92E+09 **9.92E+09**	**3.28E+10**
Sr-90 / (29.1)	1.82E+11 **2.18E+10**	1.73E+10 **2.75E+10**	1.33E+10 **5.37E+10**	5.18E+09 **2.29E+10**	2.57E+09 **1.80E+10**	8.23E+10 **1.72E+10**	1.20E+10 **2.16E+10**	7.69E+10 **7.69E+10**	**2.60E+11**

2.4. Conclusions

The most relevant source with respect to total inventory of artificial radionuclides in the Baltic Sea was the fallout after the Chernobyl accident in April and May 1986. The direct deposition of the most important nuclides Cs-137 and Cs-134 to the sea surface as well as river run-off and currents in the sea were the dominant factors affecting the distribution pattern of concentrations. The subsequent input from river run-off after 1987 was of minor importance.

The second most important source of radionuclides was the fallout from atmospheric weapon tests during the sixties. This source gave more homogeneous distribution to the Baltic Sea. The dominating nuclides were Cs-137 and Sr-90 in an activity ratio of about 1.6.

Discharges from the reprocessing plants at Sellafield and, of minor importance from La Hague, influenced the concentration in the Baltic Sea by the inflow of saline water through the Danish Straits about five years after their discharge into the Irish Sea or about 2 years after the release into the Channel, respectively. Cs-137 was the dominating nuclide from Sellafield and therefore, combined with geochemical conditions an almost linear Cs-137/salinity relationship in seawater was ascertained before 1986. However, due to the significant reductions of discharges at Sellafield in recent years this source became of minor importance.

The least significant source of artificial radionuclides are the discharges from regular operation of nuclear installations on the drainage area of the Baltic Sea. Most of the nuclides from these sources are only detectable in the vicinity of the discharge area.

These four relevant sources of radioactivity for the nuclides Cs-137 and Sr-90 are given as quantitative total injections into the Baltic Sea in Table 2.4.1. A decay correction to the year 1991 was applied.

Table 2.4.1. Quantitative total injections of Cs-137 and Sr-90 into the Baltic Sea (decay corrected to 1991).

	cs-137 PBq	Sr-90 PBq
Global Fallout	0.9	0.6
Chernobyl Fallout	4.1 - 5.1	0.08
Western European Reprocessing Plants	0.25	0.04
Nuclear Installations	0.0006	0.0002

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3. MONITORING NETWORK

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1) Helsinki Commission - Baltic Marine Environment Protection Commission, Finland
2) Finnish Centre for Radiation and Nuclear Safety, Finland
3) Finnish Environment Agency, Finland

3.1. Stations

To fulfil the duties given by the Commission to the Group of Experts on Monitoring of Radioactive Substances in the Baltic Sea (EC MORS) the Contracting Parties have set up a network of monitoring stations, some of them being the same stations as are used within the Baltic Monitoring Programme. The monitoring programme of radioactive substances is based on HELCOM Recommendation 10/3 adopted by the Commission in 1989 (to supersede HELCOM Recommendation 8/1 adopted in 1987). The environmental monitoring covers water, sediment, fish, aquatic plants, and benthic animals in all sub-areas of the Baltic Sea. Also sinking matted has been included in the latest version of the programme.

According to HELCOM Recommendation 10/3 "each Baltic Sea State should have environmental stations on the sea or on the coast, and the Commission will be informed about the positions of the stations". The routine station network for the regular monitoring programme is shown in Figures 3.1.1 - 3.1 .5. Additional stations are recommended to be sampled and the data to be reported accordingly.

Figure 3.1.1. **The** sampling stations of Finland, the former German Democratic Republic, Sweden and the former USSR for seawater.

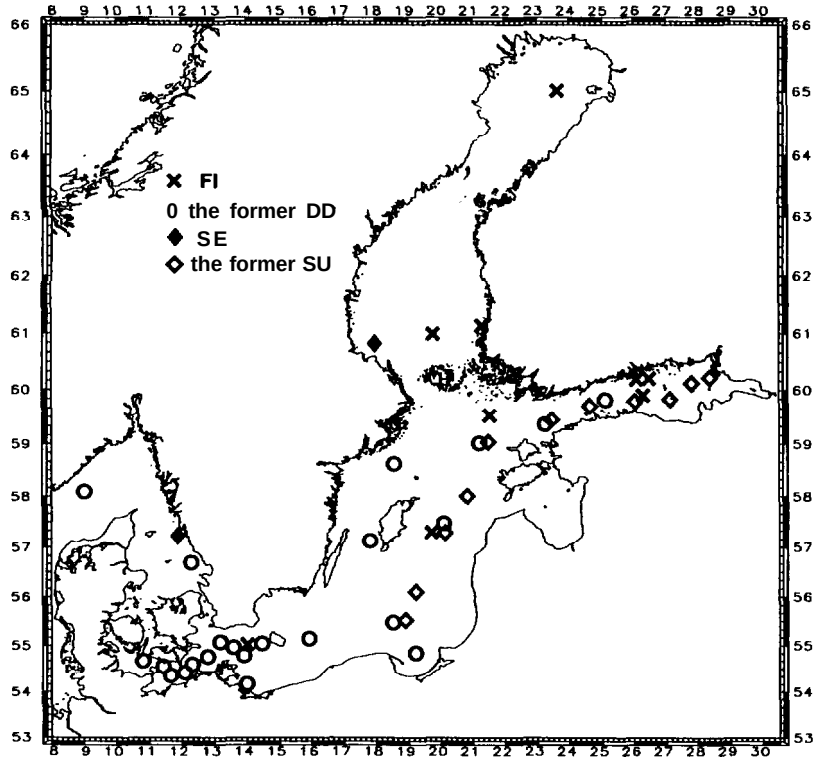


Figure 3.1.2. The sampling stations of Denmark, Federal Republic of Germany and Poland for seawater.

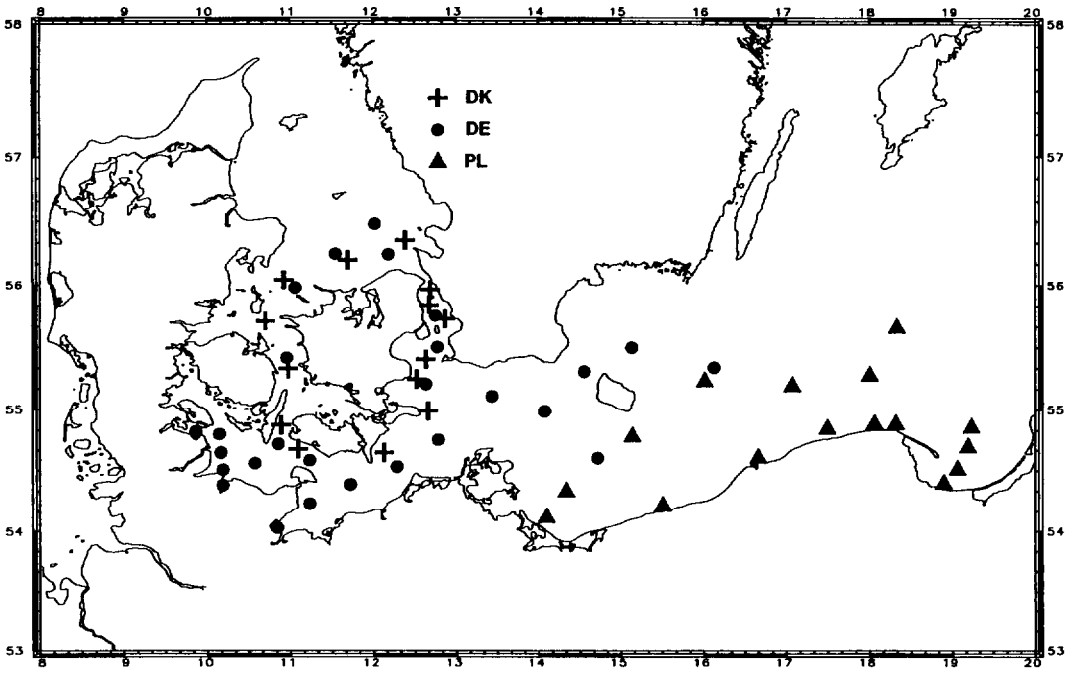


Figure 3.1.3. The sampling stations for sediment / sinking matter.

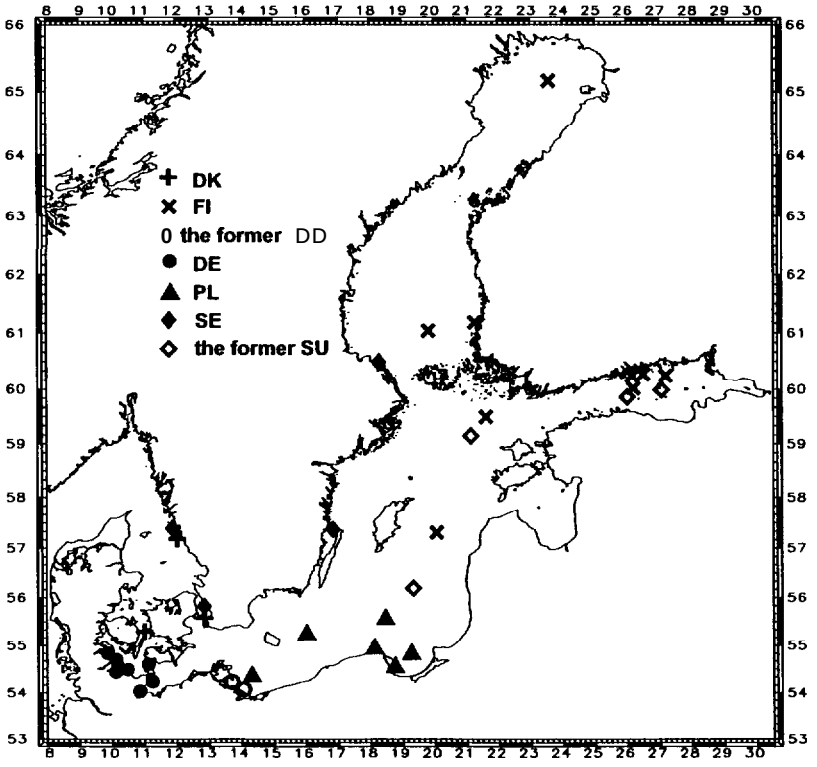


Figure 3.1.4. The sampling stations for aquatic plants and benthic animals.

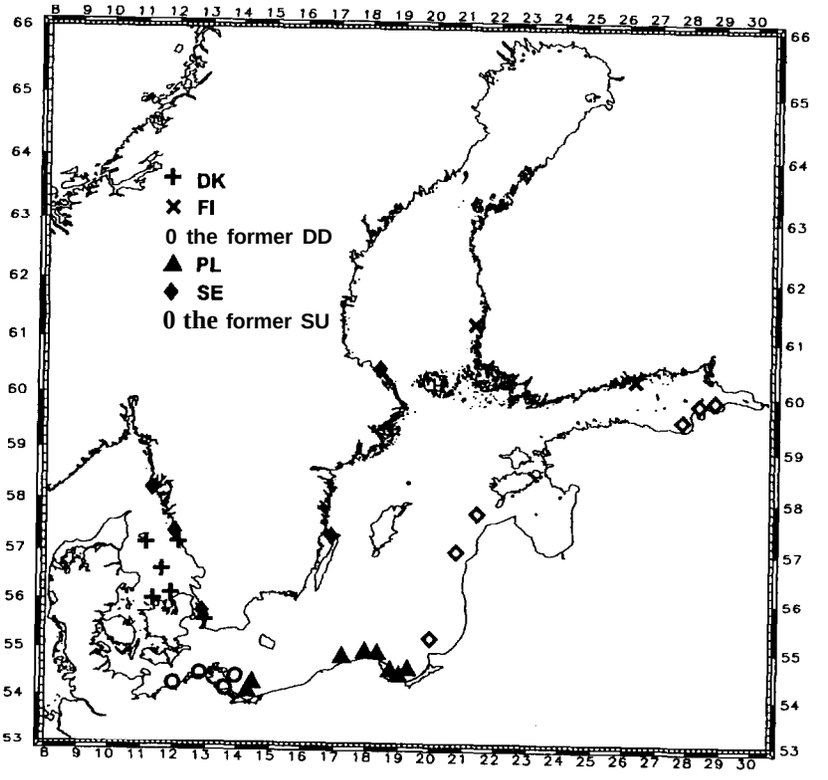
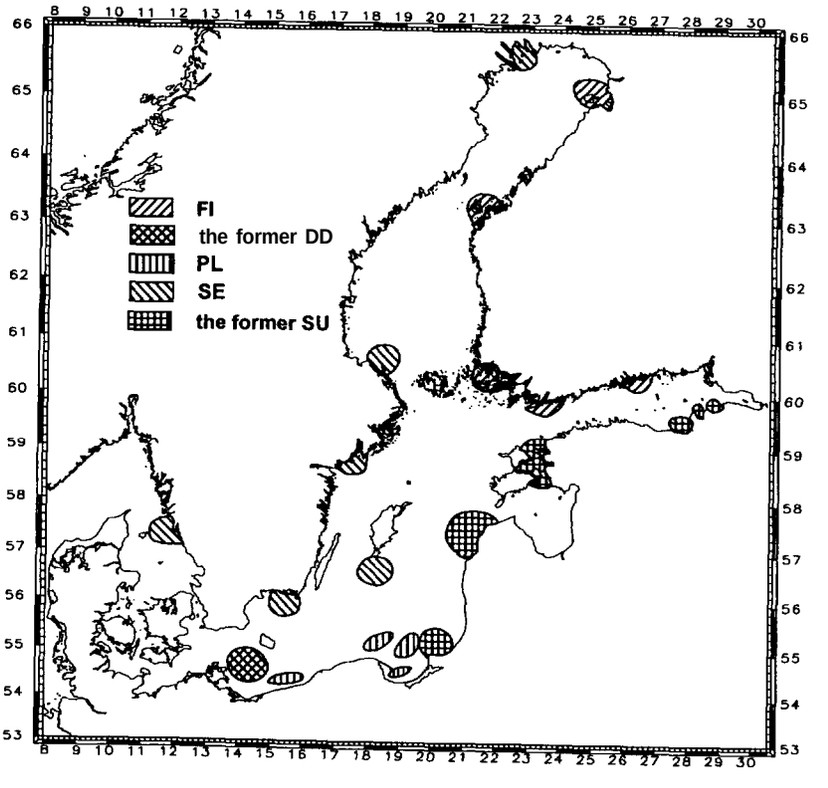


Figure 3.1.5. The sampling areas for fish.



The monitoring of the radionuclides in the Baltic Sea is carried out in each country according to its technical capability and equipment. Finland, Germany, Poland and Russia carry out monitoring also at locations remote from land, whereas Denmark and Sweden put more emphasis on coastal stations.

The laboratories involved in the work of EC MORS in 1984-1991 were the following:

Denmark: - Risø National Laboratory, Roskilde

Finland: - Finnish Centre for Radiation and Nuclear Safety, Helsinki

Germany: - Federal Maritime and Hydrographic Agency, Hamburg
- Federal Centre for Fisheries Research, Hamburg
- National Board for Atomic Safety and Radiation Protection (before the re-unification of Germany in October 1990)

Poland: - Central Laboratory for Radiological Protection, Warsaw
- Institute of Meteorology and Water Management, Gdynia

Sweden: - National Swedish Environmental Protection Board, Coastal Research Laboratory
- National Institute of Radiation Protection, Stockholm
- Environmental Protection Agency, Stockholm

Russia: - V.G. Khlopin Radium Institute, St. Petersburg (responsible for the data from the former USSR)

In addition to the environmental data on radionuclides, data are collected on releases from nuclear power plants. The location of the nuclear power plants in the drainage area of the Baltic Sea is shown in Figure 1.1. The programme covers discharges into the aquatic environment on obligatory basis as well as discharges into air on voluntary basis. Only nuclides with a half-life longer than one week should be reported and other necessary monitoring is encouraged to be carried out, e.g. related to airborne pollution and river discharges.

In addition to the location of the nuclear power plants also the location of the research reactors within the drainage area of the Baltic Sea are shown in Figure 1.1, although they are not included in HELCOM Recommendation 10/3.

In this report the data of the German Democratic Republic and the Federal Republic of Germany are given separately for the time period before the unification on 3 October 1990, and the data for this evaluation period from the present areas of Estonia, Latvia, Lithuania and Russia have been submitted by the former USSR.

3.2. Measured Parameters

The monitoring programme is based on measurements of obligatory and voluntary radionuclides as shown in Table 3.2.1. Further details on the investigated species of biota are

given in Chapter 7. In addition to the data on radionuclides supporting information e.g. on water temperature, salinity, water content of sediment and size of fish is submitted to the database as indicated in Table 3.2.1.

Table 3.2.1. Radionuclides to be monitored in the Baltic Sea.

SAMPLE	OBLIGATORY	VOLUNTARY	DESIRABLE REMARK
WATER	Radiocaesium' Sr-90"	H-3; Tc-99, Pu-239, 240; Am-241; y-emitters	- Bq/m^3, - sal., - temperat., - sample depth, - total depth
SEDIMENT	y-emitters"	Sr-90; Pu-239, 240; Am-24 1; natural radionuclides (e.g. Po-2 10)	- type of sediment (mud/sand), - grain size, - water content, - oxic/anoxic, - density, - mass depth(kg/m^2), - Bq/kg d.w,. - sample treatm. and storage, - sed. rates
FISH	y-emitters"	Sr-90; natural radionuclides (e.g. Po-210)	- species, - total fish or organ (fract.) - size, age, sex, Bqlkg w.w.
SESTON/ AQUATIC PLANTS	γ-emitters***	Sr-90; Tc-99; Pu-239, 240; Am-241 ; natural radionuclides	- Bqlkg d.w., - species
BENTHIC	γ-emitters***	Sr-90; Tc-99; natural radionuclides (e.g. Po-210); Pu-239, 240; Am-241	- Bq/kg d.w., - fraction, - species
SINKING MATTER	cf. sediment	cf. sediment	dry weight (%), ignition loss (%)

*) Cs-137 and Cs-134, if possible
**) regularly, on a carefully selected number of samples
***) K-40, Cs-137 and other gamma-emitters identified in the gammaspectrum

As shown in Table 3.2.2 there was a substantial increase in number of analyses in 1986 in all compartments. The main emphasis has been put on the analyses of y-emitters in seawater, sediment and fish. No data on Tc-99 in seawater, sinking matter and benthic animals and Am-241 in aquatic plants, benthic animals and sinking matter have been submitted. Only one Contracting Party has submitted data on sinking matter.

Table 3.2.2. **The** amount of data of different constituents in the HELCOM data bank for 1984-1991.

Constituent	Number of analyses							
	1984	1985	1986	1987	1988	1989	1990	1991
A. WATER								
Obligatory								
cs-137	213	168	402	444	217	371	248	279
Cs-134	2	2	361	438	211	362	244	273
Sr-90	192	116	276	215	13.5	186	148	174
Voluntary								
H-3	81	74	85	70	57	77	55	47
Tc-99	-	-	-	-	-	-	-	-
Pu-239,240	9	13	54	49	17	33	7	27
Am-241	-	-	50	48	17	32	-	24
other γ-emitters (excluding Cs)	13	9	173	108	37	42	47	74
B. SEDIMENT								
Obligatory								
y-emitters	77	174	261	1665	840	560	512	596
Voluntary								
Sr-90	10	22	6	9				
Pu-239,240	11	26	5	56	25	6	29	77
Am-241							3	31
natural radionuclides	56	110	65	676	359	249	377	562
C. FISH								
Obligatory								
y-emitters	87	54	332	1034	292	367	295	382
Voluntary								
Sr-90	32	23	61	91	31	49	28	43
natural radionuclides	41	30	80	221	79	75	64	97

Table 3.2.2. **The** amount of data of different constituents in the HELCOM data bank for 1984-1991 (continued).

D. AQUATIC PLANTS								
Obligatory								
γ-emitters	55	41	65	52	49	44	57	74
Voluntary								
Sr-90	4	3	2	3	4	3	3	4
Tc-99	-	-	-	-	-	-	10	10
Pu-239,240	-	-	-	1	2	1	1	-
Am-241	-	-	-	-	-	-	-	-
natural radionuclides	11	6	6	9	10	15	6	9
E. BENTHIC ANIMALS								
Obligatory								
y-emitters	23	21	42	54	61	148	56	61
Voluntary								
Sr-90	3	5	2	4	4	16	6	8
Tc-99	-	-	-	-	-	-	-	-
Pu-239,240	1	1	-	-	1	8	2	3
Am-241	-	-	-	-	-	-	-	-
natural radionuclides	19	14	4	12	16	81	15	12
F. SINKING MATTER								
Obligatory								
y-emitters	27	30	204	88	68	56	59	88
Voluntary								
Sr-90								
Pu-239,240								
Am-241								
natural radionuclides	6	8	17	11	11	10	13	21

The environmental data on radionuclides in seawater and sediments submitted to the HELCOM database are quite evenly distributed throughout the Baltic Sea although the number of sampling locations for radionuclides in sediments is much smaller than that for water, as shown in Figures 3.2.1 and 3.2.2. Most of the sampling locations of fish, aquatic plants and benthic animals are concentrated to the Southern regions (Figures 3.2.3 - 3.2.5).

Figure 3.2.1. Sampling locations of seawater for measurements of radioactive substances, 1984-1991.

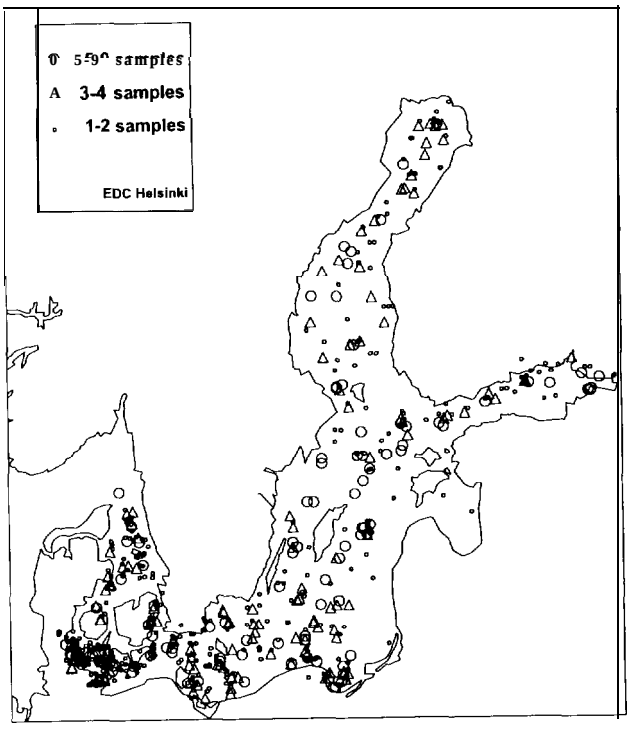


Figure 3.2.2. Sampling locations of sediment for measurements of radioactive substances, 1984-1991.

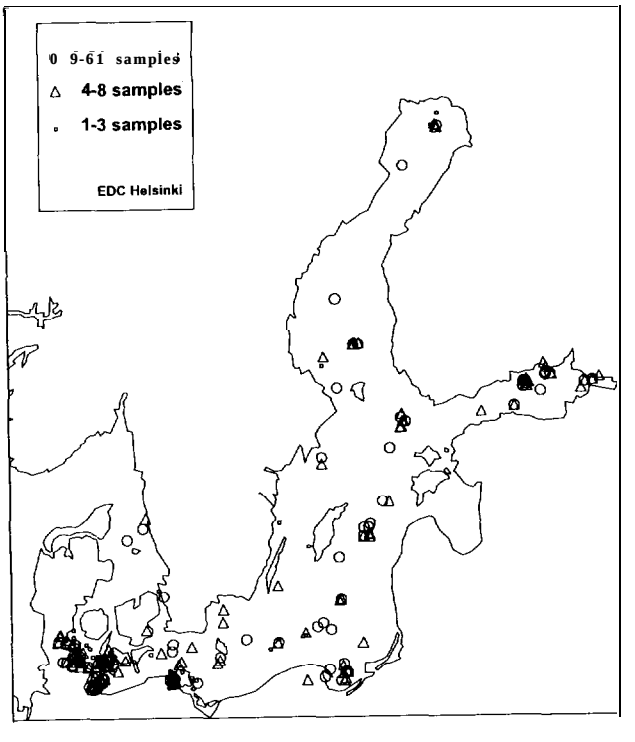


Figure 3.2.3. Sampling locations of fish for measurements of radioactive substances, 1984-1991.

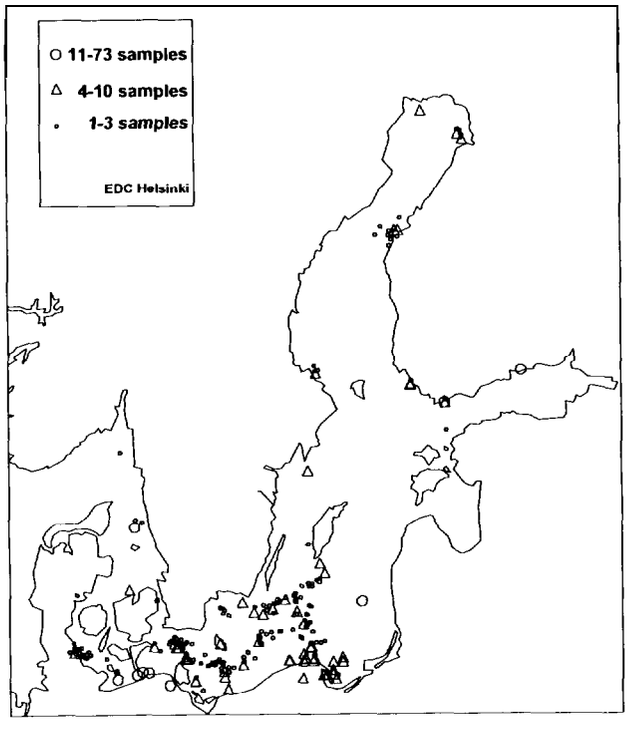


Figure 3.2.4. Sampling locations of *Fucus vesiculosus* for measurements of radioactive substances, 1984-1991.

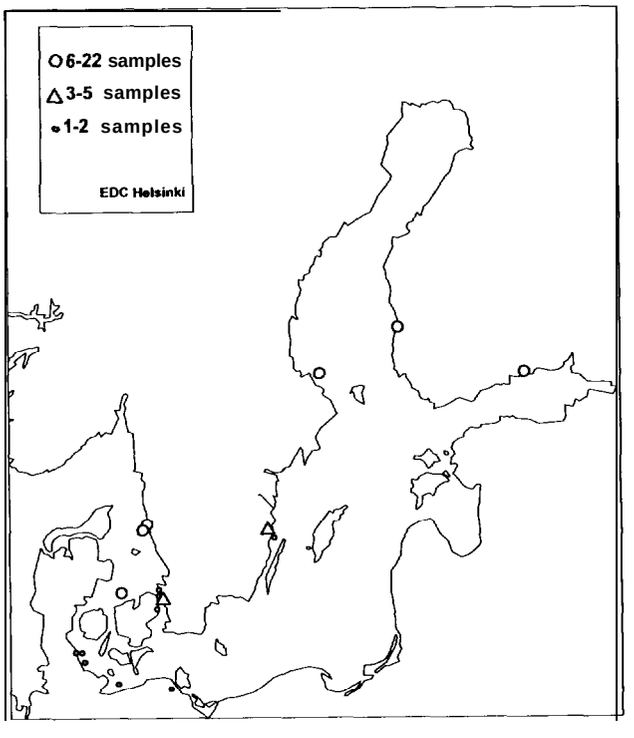
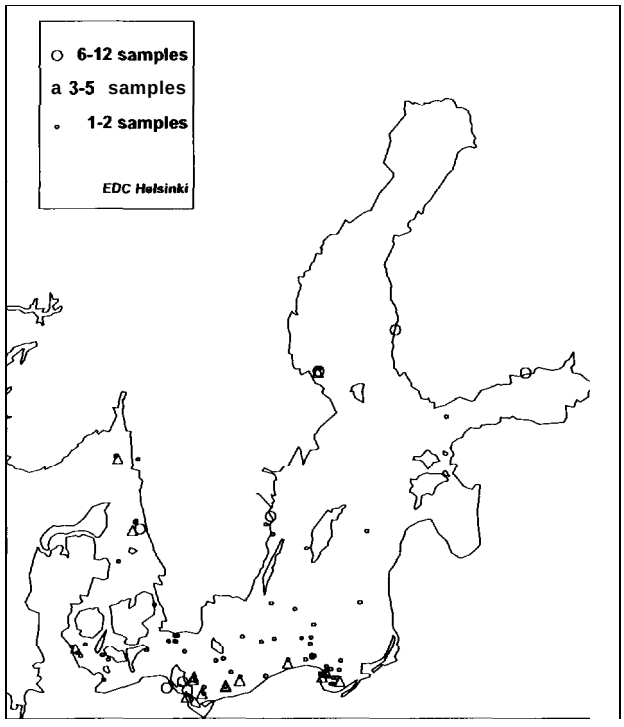


Figure 3.2.5. Sampling locations of benthic animals for measurements of radioactive substances, 1984- 1991.



3.3. Description of the HELCOM Databases

The results of the monitoring programme are submitted to the HELCOM databases run on consultant basis. The environmental data on radionuclides are managed by the Environment Data Centre of the National Board of Waters and the Environment in Finland (EDC) and the discharge data by the Finnish Centre for Radiation and Nuclear Safety (STUK).

HELCOM MORS environmental database

Technical environment

The database on environmental radioactivity has been developed using the tools available in the Ingres product family. The main computer at present is DEC-system 5500 with the Ultrix operating system. The applications can be run and interactive use of the database can also be done in the VMS operating system computers. The incoming data are normally in the ASCII-format or in dBase-tables and are converted into a format suitable for Ingres tables using a series of Fortran-programs. The outputting routines have been set up using Ingres ABF and Report Writer. The maps are printed using ArcInfo-software.

Logical structure

The design of the logical structure of the MORS database follows quite strictly the structure used in the input programme PMENU and the reporting guidelines. Only some minor changes have been made, mainly in order to make classification and analyses of data easier. This means that a small number of new fields has been added to the original version.

In each of the four reporting mediums the data are organized in two tables, one describing the sample and the other bearing the results of the analyses. In addition to this, there are also code tables with the codes and their explanations.

Coding principles

In the present guidelines there are codes for the following data items: country, laboratory, filtration, sampling device, sediment type, oxic conditions, plant or animal species, plant or animal tissue, radionuclide, analyse method and weight basis. The codes for sampling device and analyse method are laboratory specific.

Database and its contents

There are eight main data tables: two for each reporting medium: seawater, sediment, biota and sinking matter. The number of data rows covering the years 1984- 1991 is the following:

	seawater	sediment	biota	sinking matter
sample records	**2707**	**1966**	**1138**	**88**
analyse records	**7080**	**7467**	**5152**	**717**

HELCOM MORS discharge database

As a consultant of HELCOM the Finnish Centre for Radiation and Nuclear Safety keeps a register for radioactive discharges from nuclear power plants and other nuclear facilities in the Baltic Sea area. Aquatic discharges from 11 facilities since 1984 have been reported to the database. Reporting of airborne discharges is on voluntary basis.

The discharge database has been built using the tools available in the SAS product family under VMS in VAX system. SAS is also available in other systems such as Windows and Unix. The incoming data are in ASCII-format, which is simple to convert into SAS-format. All output routines are made using the SAS.

There is only one table with six variables in the database: name of the power plant, type of discharge (aquatic/airborne), year, name of radionuclide, annual discharge in Bq and notes. Only nuclides with a longer half-life than one week are stored in the database. The number of records from 1984-1991 is 1351.

4. QUALITY OF DATA

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2) Federal Maritime and Hydrographic Agency, Germany
3) Finnish Centre for Radiation and Nuclear Safety, Finland

4.1. Analytical Procedures Used by Laboratories Involved in MORS

4.1.1. Pretreatment of Marine Samples

Pretreatment methods of biota, fish and sediment samples vary somewhat in the Baltic Sea countries. Samples are usually stored frozen. Drying is done with drying at air, ovens (105 1 10°C) or freeze-drying. Ashing of biota and fish samples is also done in some cases before gamma measurement. Samples are homogenized before measurements. Fish samples are cleaned before drying. All eatable parts or only flesh are measured. A description of the methods of sediment and biota sampling is given in Chapters 6 and 7, respectively.

Pretreatment methods of seawater samples vary significantly in the Baltic Sea countries. Caesium is adsorbed on AMP (Ammonium phosphomolybdate) on board or the concentration of the samples is done in the laboratory by evaporation. Two laboratories coprecipitate strontium as oxalate on board after addition of carriers and yield monitors, whereas some laboratories only acidify the water samples for later analysis in the land-based laboratories.

Detailed pretreatment methods

Denmark (DK)
Biota: - Drying, ashing in oven or wet ashing
Seawater: - For Cs: Adsorption of caesium on AMP on board
- For Sr: Acidifying, adding of carriers and yield monitors at laboratory

Finland (FI)
Biota: - Cleaning of fishes to eatable parts, drying of other biota samples at air
- Drying at 105°C, homogenizing
- Ashing of fish samples at 450°C
Seawater: - Acidifying, adding of carriers, evaporation to 400-500 ml at laboratory

Germany (DE) / Federal Fis eries Research Institute (BFAFi)
Biota: - Cleaning of fishes
- Drying at 1 10°C
- Ashing at 460°C

Germany (DE) / Federal Mari ime and Hydrographic Agency (BSH)
Seawater - Acidification to pH 2 on board

IAEA/MEL

Biota: - Drying, grinding and homogenizing, ashing at 450°C

Seawater: - For Cs, adsorption on AMP on board; for preliminary results: Evaporation of 5-10 1 water
- For Sr: Acidification, addition of carriers and yield monitors on board

Poland (PL)

Biota: - Drying, ashing or wet ashing

Seawater: - For Cs, adsorption on AMP on board
- For Sr, precipitation with oxalic acid on board

Russia (RU)

Biota: - Cleaning of fishes to eatable parts, ashing at 450°C

Seawater: - Acidifying and adding of carriers on board

Sweden (SE)

Fish: - Cleaning of fishes, drying and ashing

Other biota: - Drying at air, homogenizing

Seawater: - Adsorption of Cs on AMP

4.1.2. Analytical Methods

4.1.2.1. Gamma Spectrometry

Measuring geometry

A number of cylindrical containers with different filling volumes (several standard geometries or all volumes between known geometries), and OS-l.0 1 Marinelli geometries are used.

Measuring

Most of the detectors are high purity Ge, some are Ge(Li) detectors. The samples are measured in background shields to reduce the background and are usually placed directly on the detectors to increase the detection efficiency. Measuring times vary according to samples and nuclides.

Analyses of spectrum

The gamma spectra are analysed with the assistance of computers. The software is specially designed for this purpose. The programmes can contain modules as:

peak-search
calculation of total peak area either according to manual calculation or applying a peak fitting procedure
adaptation of energy and efficiency calibrations to the spectrum
identification of nuclides in the spectrum
background correction
half-life correction of the nuclide
taking into account of self-absorption in the sample
correction for coincidence summing
reporting of nuclide concentrations and the uncertainty estimations

Calibration

Different mixed standard solutions or/and solutions of single nuclides are used. Water is the most common basic matrix, but other matrices with a higher or lower density are also used. Frequencies of calibrations vary from several times a year to once in 2-3 years in some laboratories.

Detailed methods of gamma measurements in the Baltic Sea countries

* Equipment

DK
Ge detectors, efficiency 15-40%, resolution 2-3 keV
Multichannel analysers (4000 channels)
Energy range O-3 MeV.

FI
Ge detectors, efficiency 15-41%, resolution 1.75-2.1 keV
Multichannel analysers (4000 or 8000 channels)
Energy range lo-2700 keV

DE/BFAFi
Ge and Ge(Li) detectors, efficiency 20-48 % , resolution 1 .8-2.3 keV
Multichannel analysers (4000 channels)
Energy range 30-2000 keV

DE/BSH
Ge detectors, efficiency 18.5-44%, resolution 1.78-l .88 keV
Multichannel analysers (8000 channels)
Energy range 45-2000 keV

IAEA/MEL
Ge detectors, efficiency 19-30%, resolution 1.75-2.1 keV
Multichannel analysers (4000 channels)
Energy range 20-2000 keV

PL
Ge(Li) detectors, efficiency 5.5 % , resolution 3.4-4.02 keV or Ge detector, efficiency 33 % , resolution 1.8 keV
Multichannel analysers (4000 channels)
Energy range 8 l-2 185 keV

RU
Ge(Li) detectors, resolution 1.8-2.8 keV
Multichannel analyser
Energy range 50-2500 keV

SE
Ge and Ge(Li) detectors, efficiency 18%, resolution 2 keV
Multichannel analysers (4000 channel)
Energy range O-2200 keV

* Geometry

DK 200 ml cylinder filled from 10-200 ml
1 1 Marinelli for 0.8-l 1, well detector O-2 ml

FI Two cylindrical containers, all volumes up to maximum of 35 ml and 1 1
0.5 1 Marinelli

DE/BFAFi Two cylindrical containers, 50 ml and 200 ml, with variable degrees of filling

DE/BSH One 200 ml cylinder with variable degrees of filling
1 1 Marinelli

IAEA/MEL Two cylindrical containers, fixed volumes up to 50 ml and 200 ml,
Well detector O-5 ml

PL Cylindrical containers, several standard geometries

RU Different cylindrical containers, all volumes up to 1 1

SE Beaker with 5, 35, 60, 90, 180, 500 ml filling 1 1 Marinelli

* Analyses of spectra

DK Main frame computer using software developed at Risø including:
corrections for sample densities and coincidence losses
calculation of analytical error from counting statistics
nuclide library
graphics

FI GAMMA-83 computer code developed at STUK including:
corrections of height and density of sample and coincidence losses
nuclide library of 92 nuclides, 486 gamma lines
calculation of analytical error from counting statistics and calibration uncertainty
high resolution graphic print of the spectra for visual inspection

DE/BFAFi FORTRAN programme based on the principles of SAMPO80 developed at BFAFi including:
corrections of coincidence losses for some nuclides
nuclide library of 55 nuclides and 522 gamma lines
calculation of analytical error from counting statistics
fitting procedure contains a graphical package, showing graphically each multiplet together with the fitted total function

DE/BSH Omnigam programme by EG&G, ORTEC, including:
nuclide library of 25 nuclides and 79 gamma lines
calculation of analytical error from counting statistics
correction of self-adsorption by special procedure, if required
high resolution graphical display of fitting function

IAEA/MEL Intergamma computer code developed by Intertechnique (France) including:
nuclide library of 91 nuclides, 239 gamma lines
calculation of analytical error from counting statistics
high-resolution graphic print of the spectra for visual inspection

PL Software by Canberra including corrections of height

RU Computer programme SeDeAc developed at RI including:
nuclide library of 100 nuclides and 2000 gamma lines
graphics, showing each multiplet together with the fitted total function
corrections of height, density, self-absorption and coincidence

SE Evaluation software of ORTEC and Nuclear Data, including:
graphics
corrections are calculated manually

* Calibration

DK Calibration with mixed liquid solutions carried out for all geometries
Frequency: Low
Error of calibration about 5%
Frequency of energy calibration control: Weekly

FI Peak and total efficiency calibration with separate single-line nuclides in water solutions
Frequency: Once in every 2-3 years
Control of energy resolution of each detector is done weekly
Error of single calibration about 4%, total error of calibration less than that
Frequency of energy calibration control with every measurement

DE/BFAFi Two different calibration procedures:
* calibration using ashes and sediment of different densities and several nuclides (old detectors), error of single calibration 7%, total error 5 %
* calibration with liquid solutions (new detectors)
Frequency: less than once a year
Frequency of energy calibration controlled weekly

DE/BSH Calibration of new detectors with mixed radionuclides in aqueous solution
Error of calibration about 5%
Frequency of energy and efficiency calibration controlled weekly

IAEA/MEL Calibrations with mixed radionuclides (liquid standards) and with reference materials of different composition in levels, density and isotopes
Frequency: 2/year
Control of energy resolution of each detector weekly
Error of single calibration 5%
for Cs measurements only (AMP), calibration with standard source of known activity and same counting geometry
Frequency of energy calibration controlled weekly

PL Calibration with mixed nuclide standards in water solution
Frequency: 1 /year
Error of calibration: 5%

RU Calibration with mixed standard nuclides, point sources
Frequency of calibration: 2/year
Total error of calibration about 2%
Frequency of energy calibration control with every measurement

SE Calibration with mixed nuclide standards in water solution
Calibration with solutions of single nuclides
Frequency of calibration for at least one of the geometries: 6/year
Error of calibration: About 5%

4.1.2.2. Radiochemical Determination of Caesium

Three different methods are used for the radiochemical determination of caesium in seawater. In 3 laboratories (2 in Poland, and occasionally IAEA/MEL), Cs-137 is measured after a radiochemical separation from seawater samples. Caesium is adsorbed on AMP, which is dissolved with NaOH and after purification, precipitated as caesium chloroplatinate (Cs_2PtCl_6). Chemical yield is determined gravimetrically. The activity is counted with a low background gas-flow beta counter. Mean analytical error of this method is about 10% (PL). Beta counters are calibrated with known Cs-137 standards, monthly in IAEA/MEL and 2-3/year in Poland.

In Russia, caesium is coprecipitated from seawater with potassiumferrocyanate. After purification, caesium is precipitated as caesiumtetraphenylboron and measured by gamma spectrometry. The chemical yield is determined gravimetrically.

At the German BSH laboratory, caesium is coprecipitated with potassium hexanitrocobaltate, purified and finally precipitated as caesium iodobismutate. The chemical yield is determined gravimetrically. Both gamma spectrometry and low-level gas-flow beta anticoincidence counters are applied.

4.1.2.3. Radiochemical Determination of Strontium-90

Methods

Two different methods have been used for the determination of Sr-90 combined with several kinds of pretreatment.

* Classic nitric acid method

This method is used in Denmark, IAEA/MEL, Finland, Germany (BFAFi), Poland and Russia.

After first precipitations with oxalate, ferrocyanate or phosphoric acid, strontium is separated from calcium and partly from other interfering ions by precipitation as nitrate several times with 70-77% nitric acid. Barium, radium and lead are separated by precipitation as chromates. Thorium, daughters of radium and rare earth metals, and in particular yttrium, are removed by ferric hydroxide scavenging. After two weeks, ingrowth period of the short-lived daughter of Sr-90, Y-90, yttrium is separated from strontium and precipitated as oxalate to obtain the sample for beta counting. Chemical yield is determined by gravimetry or titration of the yttrium carrier added. The chemical yield of strontium is determined by Atomic Absorption Spectroscopy, X-ray fluorescence or with gamma counting of Sr-85.

* Extraction method

This method is used in Germany for fish flesh samples (BFAFi) and seawater (BSH).

Yttrium-90 is extracted with HDEHP (hydroxy-diethyl-hexyl-phosphoric acid) in n-heptane from dissolved ashed sample or seawater, respectively. After purification, yttrium is re-extracted with hydrochloric acid. After washing, yttrium is precipitated as hydroxide and finally as oxalate and counted like in the nitric acid method. The method is applied on seawater before the determination of caesium on the same sample.

Detailed methods for measuring Sr-90 in Baltic Sea countries

DK
Classic nitric acid method
Y-90 counted with anti-coincidence low-level beta counters
Yield of Sr-90 by gamma counting of Sr-85
Yield of Y-90 by gravimetry
Calibration for Y-90 occasionally with Sr-90 standard solution
Stability of detectors is checked weekly with Cl-36 source

FI
Classic nitric acid method
Y-90 counted with low-level gas-flow beta counters
2 measurements of each sample for following the possible interferences
Yields: Sr-90 by AAS, Y-90 by titration
Measurement of stable Sr from chlorinity of seawater sample
Calibration: For Y-90, 4/year, two preparates from Sr-90 standard solution, measured several times to check the purity of Y-90

DE/BFAFi Two methods: Classic nitric acid method and extraction method
Y-90 counted with gas-flow anticoincidence beta counters
Four measurements of each sample for following possible interferences
Yields: Sr-90 by gamma counting of Sr-85, Y-90 by titration
Calibration: For Y-90, once a year; few samples from Sr-90 solution

DE/BSH Extraction method
Y-90 counted with gas-flow anticoincidence beta counters
Yield determined by gravimetry
Purity control by determination of half-life of Y-90
Calibration once a year

IAEA/MEL Classic nitric acid method
Y-90 counted with low-level gas-flow beta counter
Yields: Sr-90 by gamma counting of Sr-85, Y-90 by gravimetry
Calibration: For Y-90 several times a year, two samples from Sr-90 standard solution, measured twice to check the purity of Y-90

PL Classic nitric acid method
Y-90 counted with low-level anticoincidence beta counter or with gas-flow beta counter
Yields: Y-90 by gravimetry
Calibration: For Y-90, 2-3 times a year, from Sr-90 standard solution

RU Y-90 counted with low-level gas-flow beta counters
Yields: Sr-90 with X-ray fluorescence, Y-90 by gravimetry
Calibration: For Y-90, 4/year from Sr-90 standard solution,
measured several times to check the purity of Y-90

Eauioment and calibration

Low-background gas-flow anticoincidence beta counters are used for measuring the beta activity of Y-90. The chemical purity is controlled by determination of Y-90 decay. Beta counters are calibrated l-4 times a year with Sr-90 standard solutions.

4.1.2.4. Radiochemical Determination of Transuranium Elements

Methods for plutonium

Two types of analytical methods are used for the determination of plutonium in marine samples: Ion-exchange method and extraction method combined to anion-exchange. Plutonium is finally electrodeposited for alpha spectrometry.

* Anion-exchange method

Pu-242 is used as an internal tracer for counting alpha activity of plutonium isotopes. After

ashing, wet ashing or concentrating with coprecipitations (seawater) plutonium is oxidized to valence +4 with sodium nitrite or hydrogen peroxide or with both. Plutonium is separated using anion exchanger either in nitric or hydrochloric acid media. After washing, plutonium is reduced to +3 valence and eluted from exchanger. Solution is evaporated, and plutonium is electrodeposited on stainless steel disc from diluted nitric acid or hydrochloric acid media.

* Extraction method coupled with anion-exchange method

In this method, plutonium is extracted at +4 valence state with TOPO (trioctylphosphin oxide) in cyclohexane. Plutonium is re-extracted and coprecipitated with lanthanide fluoride. After dissolution, it is adsorbed on an anion-exchange material and separation is continued like for the ion-exchange method.

Methods for americium and curium

Methods for americium and curium determinations were reported by 3 laboratories. The methods consist in several ion exchanges (anion and cation-exchange resins), coprecipitation with calcium oxalate and extraction with DDCP or HDEHP before final anion exchange in nitric acid-methanol medium. Am-243 is used as an internal tracer. Electrodeposition and alpha spectrometry are performed like for plutonium.

Detailed methods for transuranic analysis

DK
Ion-exchange method for plutonium
Semiconductor detectors and multichannel analyser
Internal tracers of Pu-242 and Am-243
Detection limit about 0.2 mBq/sample for 6500 min counting time
Calculation by computer programme
Calibration occasionally with sources produced from standard solutions by electrospraying

FI
Ion-exchange method for plutonium
Coprecipitation, several ion exchanges and HDEHP extractions for americium and curium
Internal tracers of Pu-242 and Am-243
PIPS no-alpha semiconductor detectors and multichannel analyser
Detection limit about 0.1 mBq/sample for 5000 min counting time
Results calculated by self-written FORTRAN programme
Calibration of detectors by mixed nuclide standard source (Pu-239, Am-241, Cm-244) and by Am-241 standard source: 6/year

DE/BFAFi
Extraction method with ion exchange for plutonium
Ruggedized type surface barrier and ion implanted detectors and multichannel analysers
Internal tracer of Pu-242
Detection limit in fish: 20-50 $\mu Bq\,kg^{-1}$ w.w.

Results calculated by self-written computer programme
Calibration of detectors by mixed nuclide standard source (Pu-239, Am-241, Cm-244) twice a year

DE/BSH Combined ion-exchange/extraction method after coprecipitation for plutonium and americium
PIPS no-alpha semiconductor detectors and multichannel analyser
Detection limit: Seawater 1 mBq m"; sediment 1 μBq kg^{-1}, 6600 min counting time
Results calculated by computer programme provided by BFAFi
Internal tracers of Pu-242 and Am-243
Calibration of detectors by mixed nuclide standard source (Pu-239, Am-241, Cm-244) or (Am-243,Pu-242) twice a year. Efficiency calibration is performed with single Am-241 source, certified by PTB,Braunschweig.

IAEA/MEL Ion-exchange method for plutonium
Coprecipitation, several ion exchanges and DDCP extractions for americium
Surface barrier silicon detectors and multichannel analysers
Internal tracers of Pu-242 and Am-243
Detection limit 0.2-0.5 mBq/sample for 10 000 min counting time
Results calculated with self-written computer programme
Calibration of detectors by mixed nuclide standard source (Pu-239, Am-241, Cm-244) or by single ,Pu-239,240 source: 6/year

PL Ion-exchange method for plutonium
Surface barrier detectors and multichannel analyser
Internal tracer of Pu-242
Detection limit about 0.2 mBq/sample
Calibration by Ra-226 standard solution twice a year and with mixed standard sources (Th-230, Pu-239, Am-241)

RU Ion-exchange method for plutonium
Silicon surface barrier detectors and multichannel analyser
Internal tracer of Pu-236 or Pu-242
Detection limit about 0.1 mBq/sample
Results calculated by self-written computer programme
Calibration with standard Pu isotopes S/year

Equipment and calibration

Several types of surface barrier detectors and multichannel pulse height analysers are used for measuring the alpha activities. Results are calculated manually or by computer programme. Calibration of detectors are done 2-6 times per year. Standards used are either mixed transuranic standards and single sources or Ra-226 standard solution.

4.2. Quality Assurance, Internal and External Checking

To confirm the precision and the long-term repeatability of the analyses, several routines of internal and external checking procedures are performed by the MORS laboratories. Parallel samples are analysed occasionally. Reference materials are analysed regularly or occasionally. Long-term background measurements are performed at least once a year. The radiological purity of reagents and glassware is checked by analysing reagent blanks. Blind samples are also analysed. The participation in national and international intercomparison exercises is one of the most important parts of quality assurance. In general, all MORS laboratories are encouraged to participate in these exercises.

Detailed checking procedures in Baltic Sea countries

DK
Analysis of reference materials in connection with intercomparison exercises
Analysis of blind and blank samples occasionally
Analysis of parallel samples in connection with intercomparison exercises
Long-term background measurements:
gamma: 2-3 times/year;
beta: between all measurements
alpha: monthly
Participation in international intercomparison exercises (IAEA, WHO/IRC), tends to take part in all possible intercomparison exercises

FI
Analysis and measurement of different kinds of reference materials by gamma, beta and alpha at least once a year
Analysis and measurement of parallel samples occasionally
Long-term background measurements:
gamma: 2-3/year,
beta: once a month
alpha: 6 times/year
Analysis of reagent blanks (beta, alpha) 2-4 times/year and analysis of blind samples (beta, alpha) occasionally
Participation in international intercomparison exercises (IAEA, Nordisk Kontaktorgan för Strålsäkerhetsfrågor, Environment Protection Agency in USA)

DE/BFAFi
Analysis of reference samples about twice a year
Long-term background measurements:
gamma: 2-3 times/year
beta: twice a year
alpha: l-2 times/year
Analysis of blind samples (alpha): 10-20% of samples per year.
Participation in national and international intercomparison exercises.

DE/BSH
Analysis of reference samples about twice a year
Long-term background measurements:
gamma: twice a year

beta: 2-4 times/year
alpha: l-2 times/year

Analysis of reagent blanks (beta) 25 times/year and blind samples (alpha):once/year.

Participation in national and international intercomparison exercises (IAEA).

IAEA/MEL

Analysis and measurement of different kinds of reference materials by gamma, beta and alpha with each set of samples

Long time background measurements:

gamma: monthly
beta: monthly
alpha: 6 times/year

Analysis of reagent blanks with each set of samples

Participation in international intercomparison exercises

PL

Analysis and measurement of different kinds of reference materials by gamma, beta and alpha occasionally

Analysis and measurement of parallel samples, (alpha) occasionally, at least twice a year

Long-term background measurements:

gamma: monthly
beta: between all measurements
alpha: monthly

Analysis of reagent blanks occasionally

Participation in international intercomparison exercises (IAEA, Environment Protection Agency in USA)

RU

Analysis of different kind of reference materials by gamma, beta and alpha at least once a year

Analysis of blind samples (alpha, beta) occasionally

Analysis of reagent blanks (alpha, beta) regularly with each set of samples

Analysing of parallel samples once with each set of samples

Long-term background measurements:

gamma: 3-4 times/year
beta: monthly
alpha: monthly

Participation in national and international intercomparison exercises (IAEA, Environment Measurement Laboratory in USA)

SE

Analysis of reference materials occasionally

Long-term background measurements by gamma twice a year

Participation in national and international intercomparison exercises

4.3. Intercalibrations Carried out

4.3.1. Historical Background

The Baltic Sea countries have long traditions in oceanographic investigations in the Baltic Sea area. Radiological investigations in the Baltic Sea have been carried out by all Baltic countries for already a few decades. Since 1985 the study of radioactive material in the Baltic has been an integral part of the work of the Helsinki Commission (HELCOM). Most of the results were given in national annual reports only and were in many cases not available on an international level.

In 1980 the International Atomic Energy Agency has organized a research programme with the responsibility of the coordination of the following tasks:

> Intercalibration of sampling and analytical procedures needed for the monitoring activities.
> Coordination of the radiological monitoring programme of the Baltic Sea.
> Improvement of the exchange of information of the radiological studies in the Baltic Sea including data on releases into the Baltic Sea.

The outcome of this coordinated research programme "Study of radioactive materials in the Baltic Sea" was published in IAEA-TECDOC-362, 1986. The importance of analytical quality control in co-operative research involving environmental radioactivity measurements was recognized and the Monaco Laboratory was requested to provide such services to the laboratories of the Baltic Sea countries participating in the Coordinated Research Programme. Four intercalibration exercises were organized during the period 1981-1984, using two seawater samples of different salinity and of different radionuclides' contents (SW-N-2, seawater sample collected in North Sea and SW-B-l collected in Baltic Sea), one sediment sample (SD-N-l sample collected in North Sea), and one seaweeds sample (AG-B-1, collected in southwest part of Baltic Sea).

Eleven laboratories from the 7 Baltic Sea countries and Monaco Laboratory participated in intercalibration exercises. The results of these intercomparisons were very satisfactory. Since the CRP was achieved, the final meeting proposed the following: "Since radioactive materials are included in Annex II of the Helsinki Convention the meeting recommended that the IAEA should request the Helsinki Commission to study the possibility of coordinating the study of radioactive materials in the Baltic Sea as performed today in the Coordinated Research Programme of the Agency. The IAEA should be associated with the programme, i.e. as an observer. In order to obtain comparable results, further intercomparison work on various samples is needed. The intercomparison should be performed at an interval of 3-5 years and should be carried out under the auspices of the IAEA. Special attention should be paid to the intercomparison of sampling techniques".

The IAEA-MEL, therefore, decided to continue with intercalibration exercises for radionuclides as it was done during the CRP. Depending on their availability, the samples were only sent to Baltic Sea countries. In other cases, a more global intercalibration was envisaged giving the opportunity to the Baltic Sea countries to take part.

The objectives of these exercises were to offer the laboratories a possibility of cross-checking their analytical results, to discover and eliminate possible systematic errors and to give the data a common basis in order to ensure the overall comparability. It has been agreed from the beginning that analytical quality control would be absolutely necessary to the studies in order to take full advantage of the data accumulated through the concerted efforts of all participants over the period 1984-1991.

4.3.2. **List of Exercises**

During the period 1984-1991, 10 intercomparison exercises were organized. The characteristics of the different exercises are presented in Table 4.3.1. Three samples, MA-B-3 (fish flesh *Bellone bellone),* IAEA-306 (sediment) and IAEA-299 (seawater) were collected in the Baltic Sea while the others were collected in North Sea, Atlantic Ocean, Mediterranean Sea and Pacific Ocean. None of these exercises, except IAEA-299 exercise, was designed specifically for the purposes of the Baltic Sea countries programmes but these laboratories have always been encouraged to participate. The IAEA-299 Baltic seawater exercise was specially designed for the Baltic countries. The decision was taken during the fourth MORS meeting and implemented during the fifth MORS meeting in Stockholm. It was agreed that the participants will measure Sr-90 and Cs-134,137 isotopes during a period of 5 years, each laboratory being requested to do analysis of the same sample every year. The participants were also provided each year a set of standard solutions of Sr-90 and Cs-137 in order to check their equipment and chemical procedures.

Table 4.3.1. Marine samples for Radionuclides' Intercomparison exercises.

Sample code	Sample type	Source of material	Year of collect.	Year of exercise	Participants World	Baltic
SD-N-2	Sediment	North Sea	1979	1986	21	5
MA-B-3	Fish fl.	Baltic Sea	1984	1987	43	4
SD-A- 1	Sediment	Atlantic	1985	1988	34	8
IAEA-306	Sediment	Baltic Sea	1986	1989	84	7
IAEA-307	Sea plant	Medit. Sea	1986	1989	66	7
IAEA-308	Seaweeds	Medit. Sea	1986	1989	67	6
IAEA-352	Tuna fl.	Medit. Sea	1988	1990	63	7
IAEA-367	Sediment	Marshall Isl	1982	1991	81	8
IAEA-368	Sediment	Mururoa Atoll	1989	1991	89	8
IAEA-299	Seawater	Baltic Sea	1991	1991-1996	-	8

During that period the Baltic countries participating in the HELCOM/MORS Programme were: Denmark, Finland, Germany, Poland, Russia (formerly USSR) and Sweden. Denmark, Finland and Russia were always represented by one laboratory while Germany, Poland and Sweden were represented by 2-3-4, 2-3 and 2-3-4 laboratories, respectively.

A complete list of laboratories which participated in one or more intercomparison exercises organized by IAEA-MEL during this period is given in Table 4.3.2.

The rate of participation, i.e. the number of results reported by the Baltic countries laboratories in relation to the total number of results received in the worldwide exercise, of the different laboratories at the 10 exercises conducted during that period is quite good for Cs-137 and Pu-239,240, 38% and 50% respectively, but less better for Sr-90, 21% only. Nevertheless, it can be noted that the Central Laboratory for Radiological Protection (Warsaw, Poland) and the V.G. Khlopin Radium Institute (St. Petersburg, Russia) have participated in only one exercise. Four laboratories have participated in more than 80% of the exercises (RISØ, BFAFi, BSH and SOPOT).

The results of each exercise were published under an IAEA-AQCS (Analytical Quality Control Services) Report where the quality of the data reported by each participant was fully documented (IAEA, 1986-1991b).

Table 4.3.2. List of Laboratories which have participated in the IAEA-MEL Intercomparison Programme during the period 1984-1991.

Main Investigator	Laboratory #	Participation
Aarkrog/Nielsen (DK)	RISØ	All except MA-B-3
Salo/Ilus (FI)	STUK	SD-N-2, SD-A- 1, IAEA-352 and IAEA-299
Kanisch (DE)	BFAFi	All except IAEA-299
Nies/Herrmann (DE)	BSH	All except SD-N-2 and MA-B-3
Moldenhawer/Weiss(DE) Siebert/Thiele(DE)	SAAS	All except SD-N-2, IAEA-352 and IAEA-299
Bojanowski (PL)	Sopot	All except SD-N-2 and MA-B-3
Grzybowska (PL)	CLOR	Only SD-A- 1
Tomczak (PL)	Gdynia	MA-B-3, IAEA-306, IAEA-307 and IAEA-299
Panteleev (RU)	Khlopin	Only IAEA-299
Melin (SE) NIRP/Stockholm Neumann (SE)		SD-N-2, IAEA-352, IAEA-367 and IAEA-368
NSEPB/Uppsala		MA-B-3, SD-A-l, IAEA-367, IAEA-368 and IAEA-299
Sandell (SE)	Studsvik	Only IAEA-299

#) For laboratory abbreviations, see p. 58.

4.3.3. Data Treatment

All data submitted by the Baltic countries over the whole study period of 1984-1991 were incorporated in the data compilation of the different exercises.

Following past practice, the data were those selected from individual reports with minor alteration comprising mainly the removal of excessive digits, standardizing activity units and matching the activities to reference date.

All laboratories from Baltic countries which have participated in worldwide intercomparison exercises agreed not to keep anonymity for the treatment of the data for the present evaluation.

4.3.4. **Discussion**

Identification of outlying data

One of the main objectives of the intercalibration was to check if data produced within the Baltic group are mutually consistent and can be regarded as belonging to the same population. Several criteria have been proposed for the rejection of suspect values, none are fully satisfactory because of various assumptions which must be made "a priori". To partially overcome this problem we have been using for some years a non parametric procedure developed by Veglia (1981) and modified by Pszonicki et al. (1983). We have recently adopted the Box and Whisker plot test (Tukey, 1977). These tests are based on the assumption of non parametric distribution of data to which distribution-free statistics are applicable. The data treatment consists of identifying and eliminating the outlying values, calculating the median and setting the confidence intervals (Gilbert, 1987).

Many laboratories among the Baltic countries participants in the IAEA-MEL AQCS Programme have tried to measure as many radionuclides as possible, especially when using gamma spectrometry. We have considered that for the purpose of the present evaluation it would be sufficient to examine Sr-90, Cs-137 and Pu-239,240. These isotopes represent alpha, beta and gamma emitting radionuclides and they are considered as the most important ones in terms of doses to man.

Evaluation of the data

The results of Sr-90, Cs-137 and Pu-239,240 reported by the Baltic countries laboratories during the period 1984-1991 are presented in Tables 4.3.3 to 4.3.8.

The scatter of the values reported to the median values chosen as the reference values is rather small and this results in a number of outlying values which is also very small. The percentage of outliers is 4.3% (1/23), 9% (5/54) and 7% (3/41) for Sr-90, Cs-137 and Pu-239,240, respectively. Although successful gamma measurements depend largely on the concentration of radionuclides in samples, most of the participants were able to detect and measure activities as low as a few Bq kg^{-1} with satisfactory precision using intact samples. Nevertheless, it is worth mentioning that among the 5 outlying values observed for Cs-137, 3 were obtained with samples with levels close to zero or blank (SD-A-l and IAEA-352), one with a sample having a concentration of 4.9 Bq kg^{-1} (IAEA-307) while the fifth one was obtained after applying a radiochemical procedure followed by beta counting (IAEA-306) sample having a concentration of 201 Bq kg-'.

As far as the Pu-239,240 measurements are concerned, while the activity levels in the sample varied between 0.0 (or blank) and 38 Bq kg^{-1}, the results are of excellent quality after the removal of 3 outliers.

While this report was intended to cover the period 1984-1991, it was found of interest to study the evolution of the performance of the Baltic countries laboratories since the IAEA-MEL initiated its Analytical Quality Control Programme in the early 1970's. All participants of the Baltic countries laboratories agreed not to keep the anonymity for this special purpose. The matching ratio has been calculated for each participation of a laboratory at a given exercise: reported value divided by the reference value obtained after statistical treatment for Sr-90, Cs-137 and Pu-239,240. The results presented in the Tables 4.3.9, 4.3.10 and 4.3.11 are self explanatory and they give the possibility to follow the performance, whenever possible, of any laboratory of the Baltic countries. The magnitude of deviation between the values of Cs-137 and Pu-239,240 reported by the Baltic laboratories and the reference values was also represented in Figures 4.3.1 - 4.3.4.

Comnarison with other intercalibration data

The accuracy of analytical results obtained by the Baltic countries laboratories on the different samples listed in Table 4.3.1 can be checked directly by comparing them with the reference values established for these samples through the worldwide intercalibration. When sufficient results were available, average values were calculated and compared with reference values. While for Cs-137 and Pu-239,240 the comparison is very satisfactory in all cases except in one case for Cs-137 in IAEA-368, it has been more difficult to draw any conclusion for Sr-90 because of the small number of results reported.

Table 4.3.3. Sr-90 measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples				
		SD-N-2	MA-B-3	SD-A- 1	IAEA-306	IAEA-307
RISO	DK				5.6±0.1	0.15 f0.03
STUK	FI		-			
BFAFi	DE	0.28±0.08	-	-0.047 ±0.040	3.5±0.2	0.28±0.06
BSH	DE					
SAAS	DE		-		6±2	<7.4
Sopot	PL				4.4±0.1	0.33f0.06
CLOR	PL					
Gdynia	PL		0.7±0.2	-	6±1	1.6±1.0
NIRP	SE			-		
NSEPB	SE		-	-		
Khlopin	RU		-			
IAEA-MEL	MC		0.13±0.05			
AQCS values		-	0.7	0.0	4.8	0.72
Range of accepted values		-			2.1-6.1	0.28-l .6
Conf. interval		-	0.13-1.01		4.0-5.1	0.28-l .6

[1]) For laboratory ablbreviations, see p. 58

Table 4.3.4. Sr-90 measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples			
		IAEA-308	IAEA-352	IAEA-367	IAEA-368
RISØ	DK	0.4±0.1	<0.014	129±13	0.45 ±0.14
STUK	FI				
BFAFi	DE	0.3±0.1	-		
BSH	DE				
SAAS	DE	0.8f0.7			
Sopot	PL	0.22 f0.05	0.03 f0.02		0.18±0.03
CLOR	PL	-	-		
Gdynia	PL	-	-		
NIRP	SE	-	-		
NSEPB	SE	-	-		
Kblopin	RU	-	-		
IAEA-MEL	MC	-	-		
AQCS values		0.4	0.19	102	1.8
Range of accepted values		0.22-0.8	0.03-0.5	50-132	0.18-8.6
Conf. interval		0.22-0.8	0.03-0.5	62-129	O-45-6.9

#) For laboratory abbreviations, see p. 58.

Table 4.3.5. Cs-137 measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples				
		SD-N-2	MA-B-3	SD-A- 1	IAEA-306	IAEA-307
RISØ	DK	1.00f0.14			199f 1	4.40f0.26
STUK	FI			-0.04±0.04		
BFAFi	DE	0.90f0.08	14.6±0.1	0.08 f0.05	179±3	4.22 f0.15
BSH	DE			<0.34	183±3	4.73 f0.35
SAAS	DE		14.1±0.1	1.7±0.6*	230±2	4.66 ±0.47
Sopot	PL				105 ±4*	3.3f0.2
CLOR	PL			9.3 ±0.2*		
Gdynia	PL		14.8f0.4		174±11	9.3±1.1*
NIRP	SE					
NSEPB	SE		14.6f0.9	0.4±0.1	-	
Kblopin	RU					
IAEA-MEL	MC	0.54 ±0.09	13.8±0.1	0.2 ±0.1	204±10	5.8f1.2
AQCS values		0.8	14.2	0.2	201	4.9
Range of accepted values		0.5-l .o	10.5-17.3	-0.56/+0.70	170-232	2.3-7.4
Conf. interval		0.5-l .o	13.7-15.3	0-0.6	194-206	4.5-5.2

* Outlying values

#) For laboratory abbreviations, see p. 58.

Table 4.3.6. Cs-137 measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples			
		IAEA-308	IAEA-352	IAEA-367	IAEA-368
RISØ	DK	5.4f0.3	2.5 f0.2	187±9	<0.5
STUK	FI		3.0±0.6		
BFAFi	DE	5.1±0.2	2.6 f0.2	175±3	
BSH	DE	4.81 f0.52	2.3±0.2	186±1	
SAAS	DE	4.5 f0.4		214±33	<0.8
Sopot	PL	4.7±0.2	2.1±0.2	201±10	1.6±0.1*
CLOR	PL				
Gdynia	PL				
NIRP	SE		2.9kO.6	170±20	
NSEPB	SE			220f 10	
Khlopin	RU				
IAEA-MEL	MC	4.4±0.8	2.7 f0.3	192±6	<0.9
AQCS values		5.6	2.7	195	0.34
Range of accepted values		3.9-7.7	1.9-3.5	162-230	0.12-0.5
Conf. interval		5.3-6.0	2.5-2.8	190-201	0.2-0.44

Outlying values

#) For laboratory abbreviations, see p. 58.

Table 4.3.7. Pu-239,240 measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples				
		SD-N-2 ●	MA-B-3 .	SD-A-l .	IAEA-306	IAEA-307
RISØ	DK	6.5f1.6		0.8±2	5.7±0.3	0.76 f0.07
STUK	FI	6.2±0.6				
BFAFi	DE	6.0±0.8	-0.10±0.40	-10± 14"	5.7f0.2	0.79f0.01
BSH	DE				5.1f0.3	
SAAS	DE				6.0f0.3	2.84±0.23*
Sopot	PL	-			5.4±0.2	0.66 f0.06
CLOR	PL					
Gdynia	PL		-	-		
NIRP	SE	14±4				
NSEPB	SE				-	
Khlopin	RU				-	
IAEA-MEL	MC	8.5 i-2.2	-0.28±0.18	1.4±0.9	5.7±0.3	0.82±0.10
AQCS values		8.8	0.000	1.4	5.7	0.72
Range of accepted values		6.0-19.8		-0.6/+2.9	4.7-6.6	0.41-0.94
Conf. interval		6.5-14.0	(<0.001)	-0.6/+2.9	5.5-6.3	0.66-0.79

* Outlying values ● mBq kg" dry weight

#) For laboratory abbreviations, see p. 58.

Table 4.3.8. **Pu-239,240** measurements reported by the Baltic countries in intercomparison samples (Unit: Bq kg^{-1} dry weight).

Laboratory #		AQCS Samples			
		IAEA-308	IAEA-352	IAEA-367	IAEA-368
RISØ	DK	0.53 f0.03	< 0.005	37.8f2.3	30.5 ±2.1
STUK	FI				
BFAFi	DE	0.50f0.01		34.3f1.6	28.3±0.7
BSH	DE			48.2f2.6	36.4f4.6
SAAS	DE	0.5 f0.2			
Sopot	PL	0.46 ±0.02	$(0.0\pm0.5)10^{-3}$	38 ±2	29 ±2
CLOR	PL				
Gdynia	PL				
NIRP	SE			36 ±8	54 ± 10"
NSEPB	SE				
Khlopin	RU				
IAEA-MEL	MC	0.47 f0.02	<0.001	42 ±3	32 ±2
AQCS values		0.50	-	38	31
Range of accepted values		0.38-0.57	-	24-5 1	18.5-39.2
Conf. interval		0.46-0.52	-	34.4-39.8	29-34

: Outlying values

#) For laboratory abbreviations, see p. 58.

Table 4.3.9. Sr-90 activity ratios (x/median value) reported by experts of MORS Group participating in IAEA-MEL AQCS Programme. (For laboratory abbreviations, see p. 58)

Sample	Type	Year	RISØ	BSH	BFAFi	IAEA	Sopot	STUK	Khlopin	NSEPB	NIRP	Gdynia	Median (Bq kg^{-1})
IAEA-367	Sediment	91	1.26										102
IAEA-368	Sediment	91	0.25				0.10						1.8
IAEA-352	Biota	90					0.16						0.19
IAEA-306	Sediment	89	1.17		0.73		0.92					1.25	4.8
IAEA-307	Biota	89	0.21		0.39		0.46					2.22	0.72
IAEA-308	Biota	89	1.0		0.75		0.55						0.4
MA-B-3	Biota	87				0.19						1.0	0.7
AG-B- 1	Biota	85	1.32			0.97							10.6
SW-N-2	Seawater	85	1.10	0.99				1.10	1.02			0.83	0.026
SD-N- 1	Sediment	84				0.57		0.85				2.06	0.92
SW-B- 1	Seawater	84	1.40”	1.04				1.47*	0.97			0.94	0.018
SD-B-3	Sediment	78	0.97					1.25					292
MA-B-l	Biota	76	1.58		2.70”		1.31				1.12		273.8
SW-A-l	Seawater	76	1.10		12*	1.00	0.90				1.3		0.0037
MA-B-2	Biota	76	0.24		1.35		0.23						11.5
SW-I-3	Seawater	76	0.77		1.16	1.07					1.16		1.1
SD-B- 1	Sediment	73	0.91			1.11	1.20	1.15					555
AG-I- 1	Biota	73	0.93			1.02	1.03	0.95					370
SW-I-1	Seawater	71	0.96	0.96		1.03	0.45	1.10					185
SW-I-2	Seawater	71	1.23	1.03		0.97	0.40	1.07					2042
Laboratory Mean			0.96	1.01	2.73	0.88	0.64	1.12	1 .oo	-	1.19	1.38	
Standard deviation (All values)			0.40	0.04	4.16	0.30	0.41	0.19	0.04	-	3.09	0.60	
Laboratory Mean			0.94	1.01	0.88	0.88	0.64	1.07	1.00	-	1.19	1.38	
Standard deviation (Outliers not included)			0.40	0.04	0.38	0.30	0.41	0.13	0.04	-	3.09	0.60	

* Outlying values

Table 4.3.10. Cs-137 activity ratios (x/median value) reported by experts of MORS Group participating in IAEA-MEL AQCS Programme. (For laboratory abbreviations, see p. 58)

Sample	Type	Year	RISØ	BSH	BFAFi	IAEA	Sopot	STUK	Khlopin	NSEPB	NIRP	Gdynia	Median (Bq kg-')
IAEA-367	Sediment	91	0.96	0.95	0.90	0.98	1.04			1.13	0.87		195
IAEA-368	Sediment	91	1.00			1.00	4.70*						0.3
IAEA-352	Biota	90	0.93	0.85	0.96	1.00	0.78	1.11			1.07		2.7
IAEA-306	Sediment	89	0.99	0.91	0.89	1.01	0.52*					0.87	201
IAEA-307	Biota	89	0.90	0.97	0.86	1.18	0.67					1.90*	4.9
IAEA-308	Biota	89	0.96	0.86	0.91	0.79	0.84						5.6
MA-B-3	Biota	87			1.00	0.97				1.03		1.04	14.2
SD-N-2	Sediment	86	1.25		1.13	0.68							0.8
AG-B-1	Biota	85	1.00	1.26	0.88	0.93		1.02	1.20	0.84	0.66		16.7
SW-N-2	Seawater	85	1.08	1.04		1.01		0.94	0.98	1.07		0.98	0.159
SD-N-l	Sediment	84	0.95	0.96		0.93	0.93	0.84	0.86		0.95	0.90	14
SW-B-l	Seawater	84	1.02	0.95		0.97		0.95	0.89	1.02		0.87	0.0157
SD-B-2	Sediment	78	1.14			1.01	0.87	1.04					3367
SD-B-3	Sediment	78	0.99			1.05	1.02	1.33					6700
MA-B-l	Biota	76	1.11		1.01		0.93				1.22		599
SW-A-l	Seawater	76	0.76	0.64	2.36*	0.64	0.58						0.0093
MA-B-2	Biota	76	1.17		1.29		1.06						152
SW-I-3	Seawater	76	0.99	1.04	1.09	0.97					1.02		2.90
SD-B-l	Sediment	73	0.99	0.90		0.96	0.95	0.46*					13949
AG-I-1	Biota	73	0.91			0.97	1.03	0.80					2775
SW-I-1	Seawater	71	0.44*	0.94		0.81		1.05					0.592
SW-I-2	Seawater	71	1.09	1.03		0.93		1.11					7.36
Laboratory Mean			0.98	0.95	1.11	0.94	1.14	0.97	0.98	1.02	0.97	1.09	
Standard deviation (All values)			0.16	0.14	0.41	0.12	1.04	0.22	0.15	0.11	0.19	0.40	
Laboratory Mean			1.01	0.95	0.99	0.94	0.86	1.02	0.98	1.02	0.97	0.93	
Standard deviation (Outliers not included)			0.11	0.14	0.13	0.12	0.18	0.15	0.15	0.11	0.19	0.08	

* Outlying values

Table 4.3.11. Pu-239,240 activity ratios (x/median value) reported by experts of MORS Group participating in IAEA-MEL AQCS Programme. (For laboratory abbreviations, see p. 58)

Sample	Type	Year	RISØ	BSH	BFAFi	IAEA	Sopot	STUK	Khlopin	NSEPB	NIRP	Gdynia	Median (Bq kg^{-1}) (♦mBq kg^{-})
IAEA-367	Sediment	91	0.99	1.27	0.90	1.10	1.00				0.95		38
IAEA-368	Sediment	91	0.98	1.17	0.91	1.03	0.94				1.74*		31
IAEA-306	Sediment	89	1.00	0.89	1.00	1.00	0.95						5.7
IAEA-307	Biota	89	1.06		1.10	1.14	0.92						0.72
IAEA-308	Biota	89	1.06		1.00	0.94	0.92						0.50
SD-N-2	Sediment	86	0.74		0.68	0.97		0.70			1.64		8.8♦
AG-B-1	Biota	85	1.06	1.00	0.50	0.92		1.06			1.52		0.066
SW-N-2	Sea water	85	1.09	0.90		0.97		0.97			1.23	3.31*	0.124♦
SD-N-1	Sediment	84	0.95	1.04		1.05		1.00			1.11		0.56
SW-B-1	Sea water	84	0.70	1.07		0.60		1.16					4.3♦
SW-N-1	Sea water	82	0.97	1.04		0.92							0.070♦
SD-B-3	Sediment	78	0.85			1.14							21.1
SW-A-1	Sea water	76				0.82							0.041♦
MA-B-1	Biota	76	1.36			0.82							1.6
MA-B-2	Biota	76	18.20*			0.91							0.041
SW-I-3	Sea water	76				1.14							3.81♦
SD-B-1	Sediment	73	1.05			0.98							35.5
AG-I-1	Biota	73	0.77			0.89	1.01						999
SW-I-1	Sea water	71	0.92			1.15							3.1♦
SW-I-2	Sea water	71	0.59			0.91							8.1♦
Laboratory Mean			1.96	1.05	0.87	0.97	0.96	0.98	-	-	1.37	3.31*	-
Standard deviation (All values)			4.06	0.13	0.21	0.14	0.04	0.17	-	-	0.31	0.00	-
Laboratory Mean			0.95	1.05	0.87	0.97	0.96	0.98	-	-	1.29	-	-
Standard deviation (Outliers not included)			0.18	0.13	0.21	0.14	0.04	0.17	-	-	0.29	-	-

* Outlying values

Figure 4.3.1. Deviation of individual Cs-137 measurements from the reference value (For laboratory abbreviations, see p. 58).

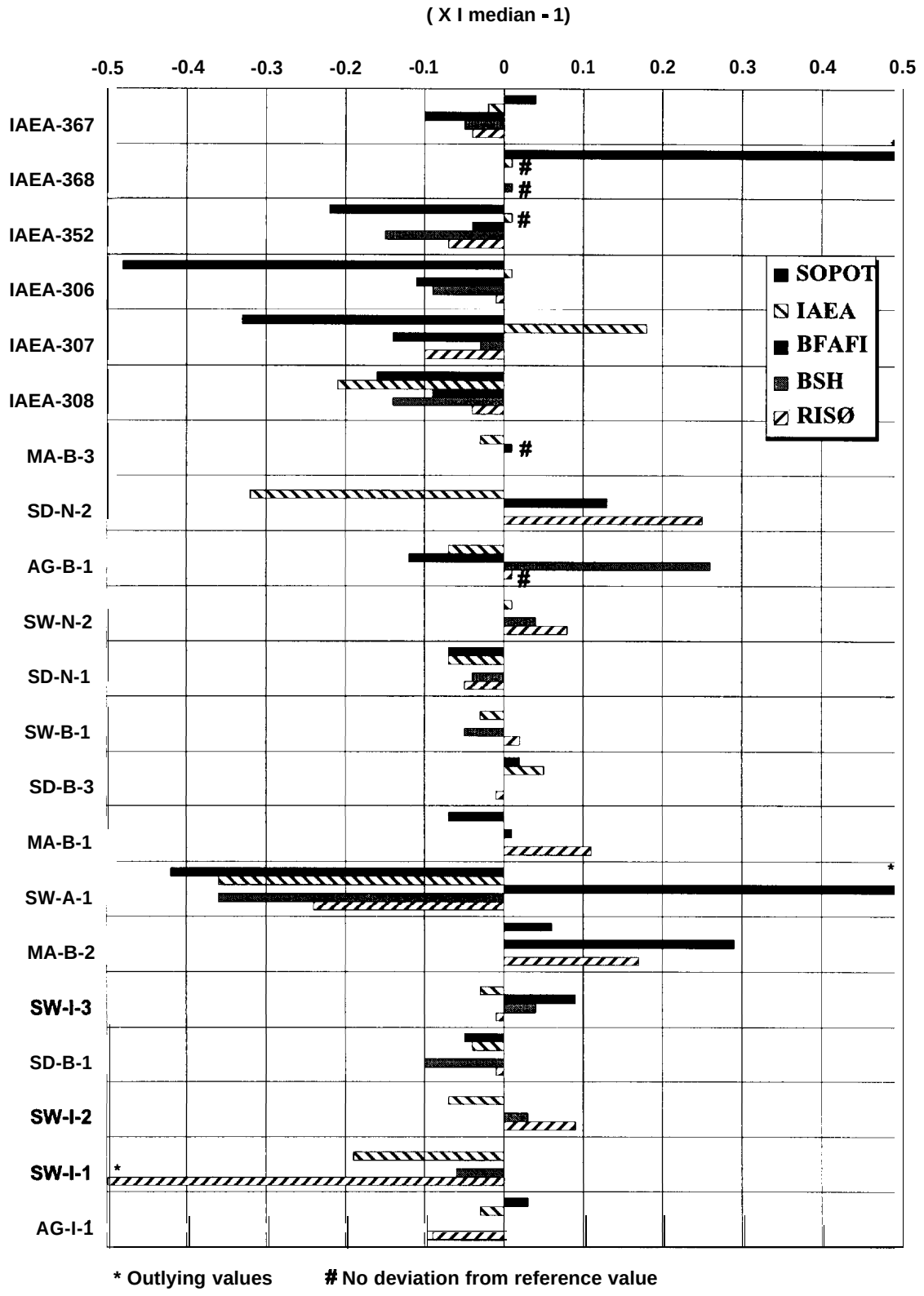


Figure 4.3.2. Deviation of individual Cs-137 measurements from the reference value (For laboratory abbreviations, see p. 58).

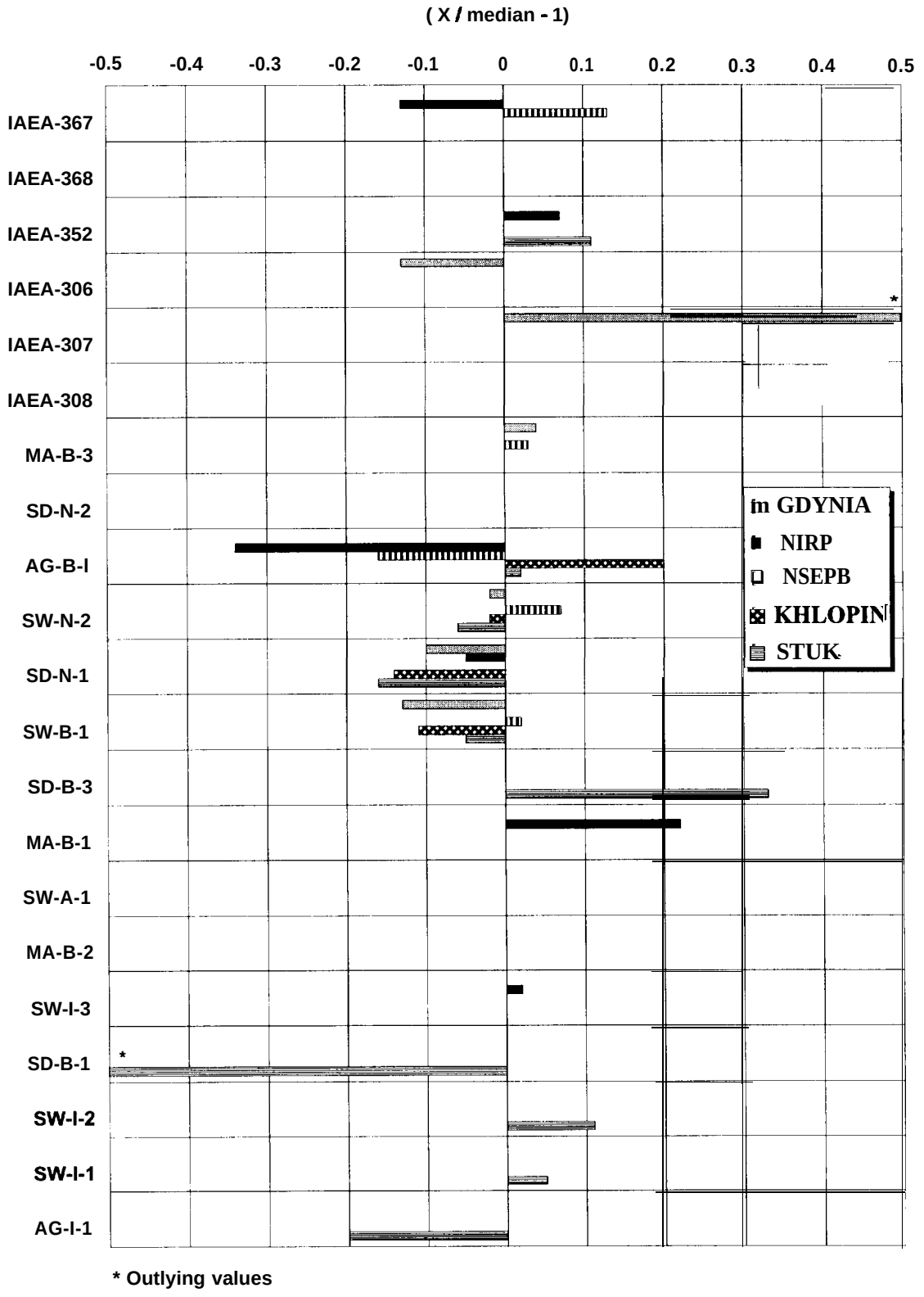


Figure 4.3.3. Deviation of individual **Pu-239,240** measurements from the reference value (For laboratory abbreviations, see **p.** 58).

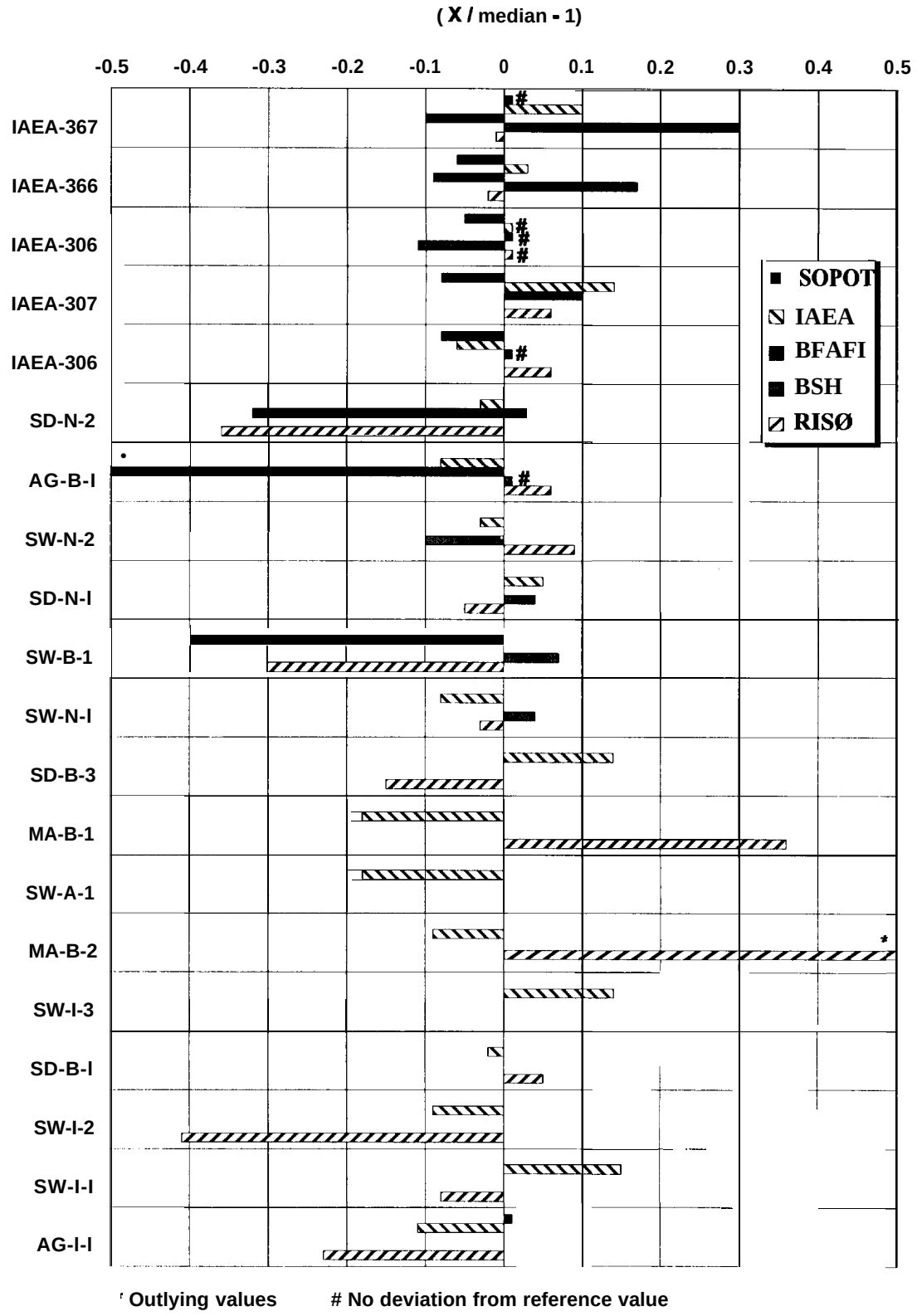
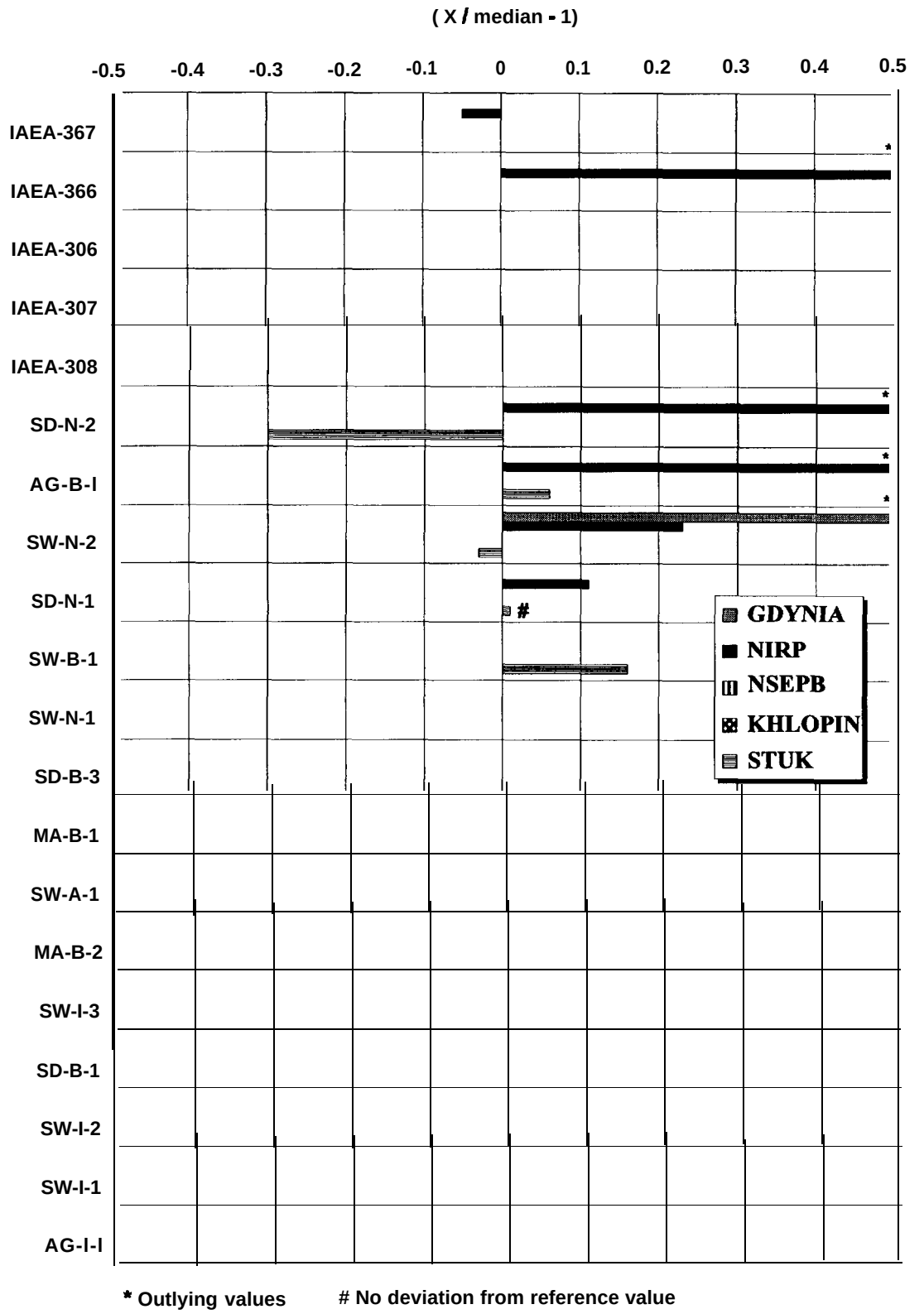


Figure 4.3.4. Deviation of individual Pu-239,240 measurements from the reference value (For laboratory abbreviations, see p. 58).



4.4. Conclusion

During the period 1984-1991, 10 intercomparison exercises were organized by IAEA-MEL. The IAEA-299 Baltic seawater exercise was specially designed for the laboratories of the HELCOM-MORS group while others were organized on a worldwide scale.

Laboratories from Baltic countries have always been encouraged to take part and the rate of participation has been quite good for Cs-137 and Pu-239,240, 38% and 50%, respectively, but less better for Sr-90 with 21% only.

The radiochemical procedures and gamma spectrometry techniques used by the different laboratories for measurements of the most important radionuclides Sr-90, Cs-134,137 and Pu-239,240 are well documented and correspond to those used by the majority of the laboratories, especially by laboratories having good reputation.

The number of outlying data reported by the laboratories was very small: 4.3%, 7% and 9% for Sr-90, Pu-239,240 and Cs-137, respectively; it is worth mentioning that most of the outlying data reported for Cs-137 were obtained with samples with levels close to zero or blank. The comparison of the data reported by the Baltic countries laboratories as a group with the reference values established through the worldwide intercomparison exercises is very satisfactory, especially for Cs-137 and Pu-239,240. This shows that the data produced by laboratories of the MORS group are of very good quality. The small number of Sr-90 data reported did not allow to draw any conclusion. It is recommended that all laboratories from the Baltic countries try to analyse Sr-90 in the future.

Laboratories from countries who joined lately the HELCOM-MORS group, i.e. Estonia, Latvia and Lithuania are kindly invited to participate in the intercomparison exercises organized by IAEA-MEL.

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Laboratory abbreviations:

RISØ	Risø National Laboratory, Denmark
STUK	Finnish Centre for Radiation and Nuclear Safety, Finland
BFAFi	Federal Fisheries Research Institute, Germany
BSH	Federal Maritime and Hydrographic Agency, Germany
SAAS	National Board for Atomic Safety and Radiation Protection, Germany (in the former GDR)
Sopot	Institute of Oceanology, Poland
CLOR	Central Laboratory for Radiological Protection, Poland
Gdynia	Institute for Meteorology and Water Management, Poland
Khlopin	V.G. Khlopin Radium Institute, Russia
NIRP	National Institute of Radiation Protection, Sweden
NSEPB	National Swedish Environmental Protection Board, Sweden
Studsvik	Studsvik Nuclear AB, Sweden
IAEA-MEL	International Atomic Energy Agency - Marine Environment Laboratory, Monaco

5. RADIONUCLIDES IN SEAWATER

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5.1. Introduction

A detailed description of the Baltic Sea area was given in the first joint report "Study of Radioactive Materials in the Baltic Sea" (IAEA, 1986a).

Water body is one of the most important components in the marine environment of the Baltic Sea. Marine dynamic processes control the distribution of radionuclides and water is the source for supply of radionuclides to bottom sediments and biota.

The Baltic Sea basin has been intensively investigated in a monitoring network in the sea area comprising over 200 sampling stations. The bulk of the data incorporated in the database refer to the seawater. All the analytical data of MORS participants are based on monitoring of unfiltered seawater.

Seawater contains both natural and artificial radionuclides. This chapter deals only with artificial radionuclides.

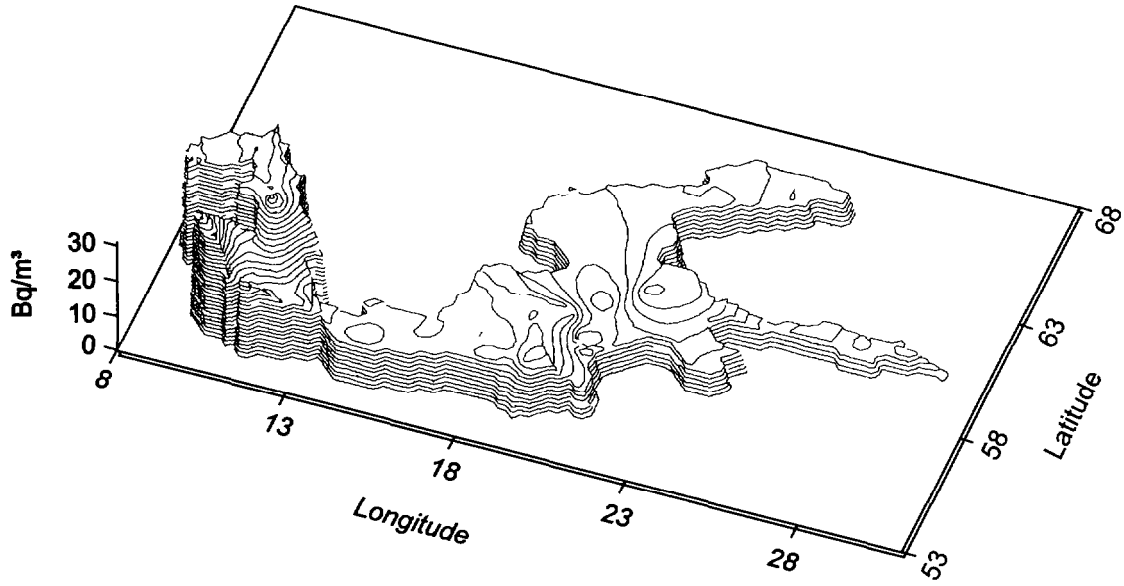
5.2. Origin and Distribution of Radionuclide Concentrations in the Baltic Sea

The Baltic Sea is a semi-enclosed and shallow brackish water body that has received artificial radioactivity from the sources mentioned in Chapter 2. Before 1986, the main sources of contamination by tritium, Sr-90, Cs-137 and plutonium isotopes were global fallout. Since the 1970s the inflow of contaminated water from the North Sea made contamination from the Sellafield nuclear reprocessing plant more important. Combined with the fact that the rate of transfer of caesium from the water column to sediments is higher in brackish waters than in high-salinity water, this led to an almost linear correlation between the Cs-137 activity concentrations and salinity (IAEA, 1986b). The distribution of the Cs-137 activity concentrations in 1985 is shown in Figure 5.2.1. The Cs-137 concentrations generally increased with depth, being higher in bottom waters in correspondence with the vertical stratification of saline water in the Baltic Sea. This again reveals that the source was saline water from the North Sea.

When the Cs-134/Cs-137 activity ratios are taken into account in the releases of reprocessing plants, the actual concentrations at the entrance to the Baltic should not exceed 1 Bq/m³. It requires about five years for contaminated seawater to be transported from the Irish Sea to the entrance of the Kattegat.

Discharges of Sr-90 from the La Hague and Sellafield plants were much lower and, consequently, the salinity relationship was much less pronounced in the Belt Sea.

Figure 52.1. Distributions of Cs-137 activity concentrations in 1985 in the Baltic Sea.



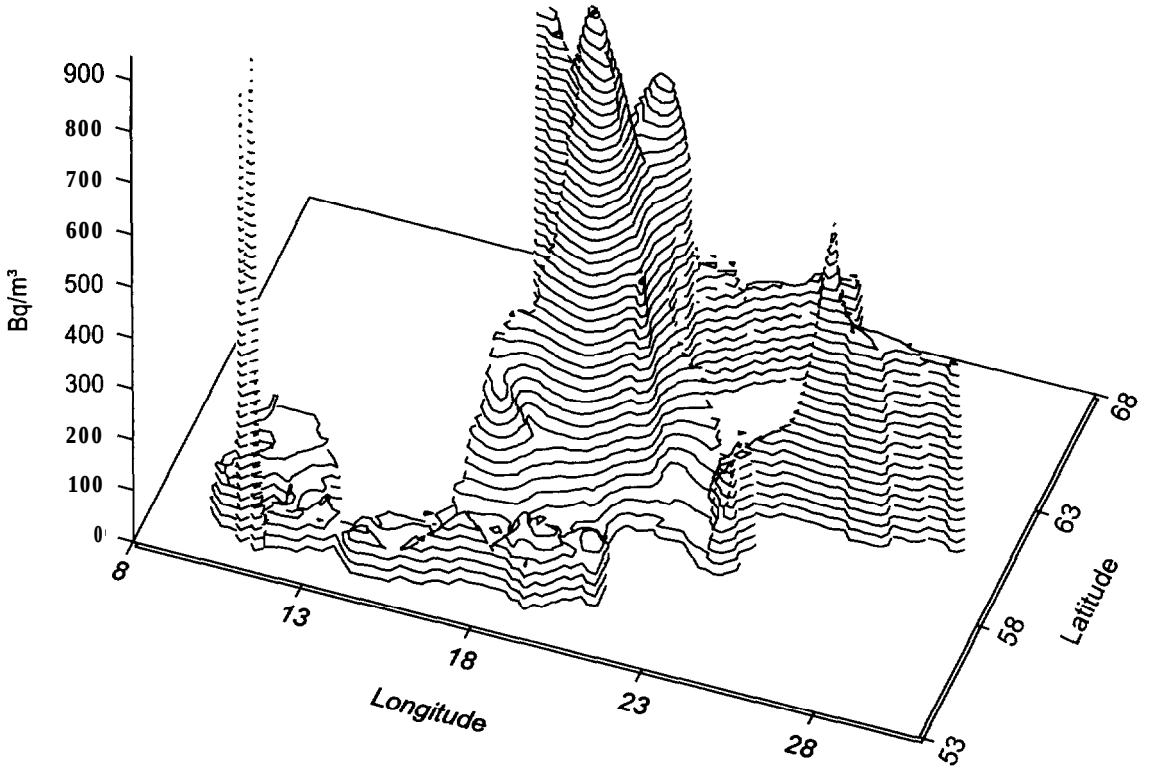
Tritium was mainly deposited in the Baltic Sea by direct atmospheric fallout or by river runoff from the drainage area after nuclear weapons tests. Near the nuclear power stations and other nuclear facilities tritium was detected at higher levels than in seawater generally. Very low concentrations of certain nuclides e.g. Mn-54 and Co-60 were also detected in the vicinity of nuclear power stations and other nuclear facilities.

During the Chernobyl accident in April 1986 plumes of contaminated air masses moved from the site of the accident to the Baltic region. Radioactive contamination of Baltic seawater since the accident were dominated by the caesium isotopes 134 and 137. In addition, more than 20 different radionuclides were detected in the fallout soon after the accident. Many radionuclides, such as Sr-89, Zr-95/Nb-95, I-131, Te-132/I-132, Ba-140/La-140, were also detected in seawater but due to their short half-lives, decayed within several days or months (Ilus et al. 1987, Saxén et al. 1987). Small increases in Sr-90 and alpha-emitting plutonium isotope activity concentrations were observed within a year of the accident in those parts of the Baltic Sea where deposition was high. By 1991 the Sr-90 and Pu-239,240 concentrations in the Baltic Sea were low, on average about 20 Bq/m³ for Sr-90 and less than 0.01 Bq/m³ for Pu-239,240 (Ikaheimonen et al. 1995; V.G. Khlopin Radium Institute, 1992).

The Chernobyl fallout was scattered very unevenly over the Baltic Sea area (HELCOM, 1989). The range of the Cs-137 concentrations measured in May and June 1986 was between 40 Bq/m³ (Arkona Sea) and 5200 Bq/m³ (northern coastal area of the Gulf of Finland). In

general, the radionuclide concentrations were lowest in the southern Baltic Sea and the Bothnian Bay. The most contaminated seawater areas were in the Gulf of Finland, the Bothnian Sea and the Mecklenburg Bight (Figure 5.2.2). There was a marked difference between coastal and open sea areas, the concentrations being higher near the coast. The Cs-134/Cs-137 activity ratio measured after the accident was in the range of 0.5 corresponding to the theoretical value for the fuel of the Chernobyl nuclear power plant (IAEA, 1986a).

Figure 5.2.2. Distributions of Cs-137 activity concentrations in 1986 in the Baltic Sea.



5.3. Temporal Evolution

The initial contamination of the surface layer rapidly penetrated into deeper waters in areas lacking a stable halocline. In 1991, the Cs-137 concentrations were still lower in the deep water layer of the Baltic Proper than in surface water. For example, at the Gotland Deep, the Cs-137 activity concentrations in surface and deep waters were 120 and 25 Bq/m³, respectively.

The temporal evolution of Cs-137 concentrations in surface water is depicted in Figure 5.3.1 and that in near-bottom water in Figure 5.3.2.

Figure 5.3.1 Temporal evolution of Cs-137 concentrations in surface water 1984-1991 in Baltic Sea (Annual mean O-10 m, Bq/m³).

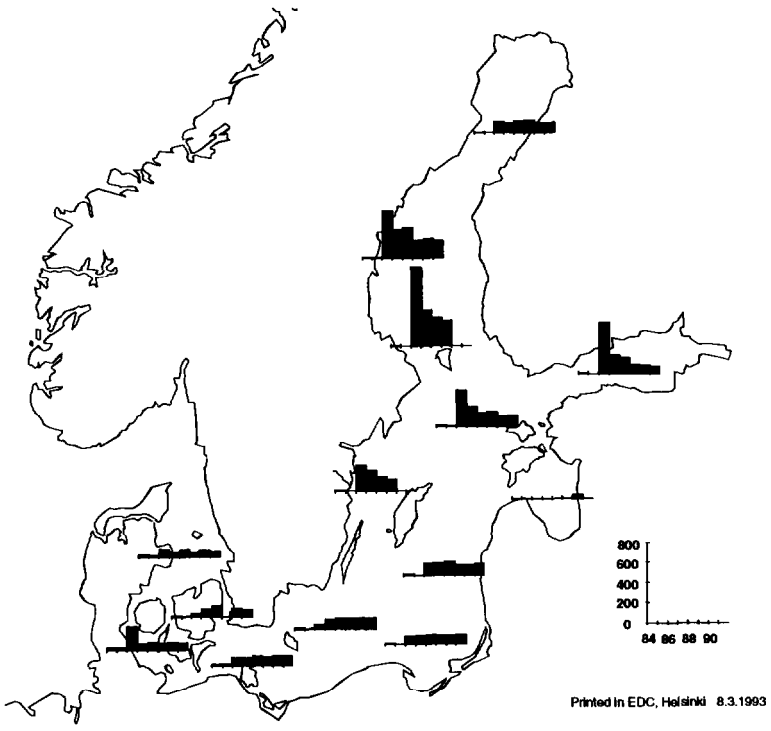
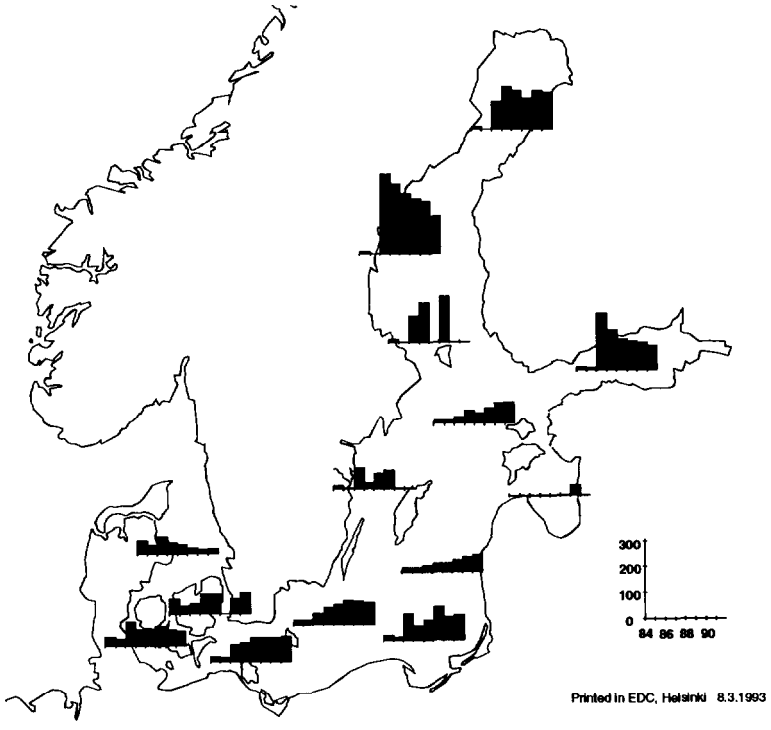
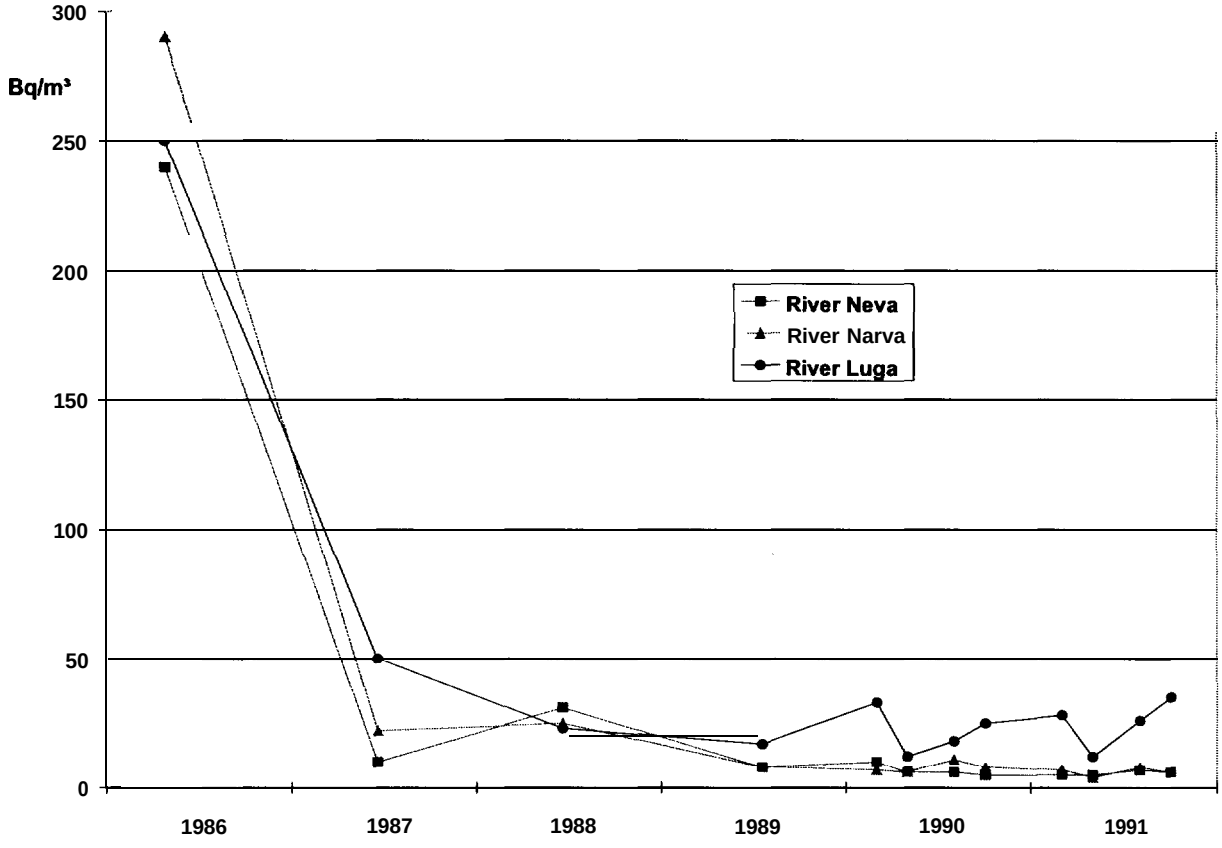


Figure 5.3.2 Temporal evolution of Cs-137 concentrations in near-bottom water 1984-1991 in Baltic Sea (Annual mean 10 m above bottom, Bq/m³).



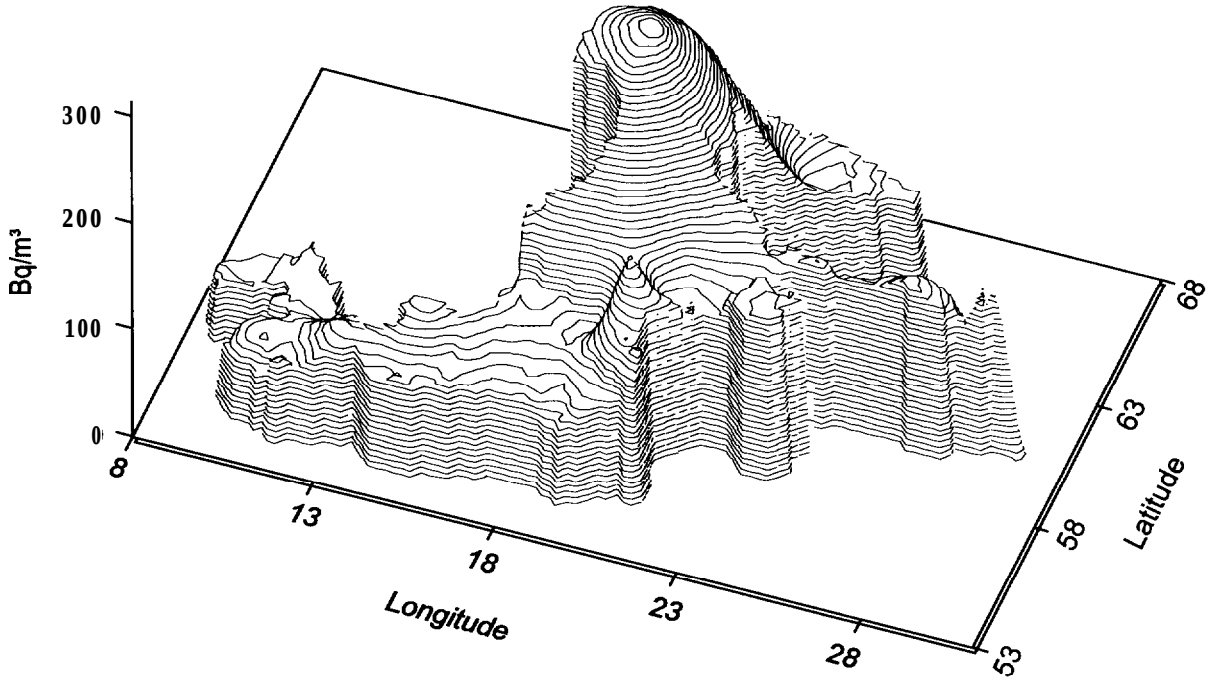
In the years following 1986, the contaminated waters in the Gulf of Finland were renewed more rapidly than those of the Bothnian Sea (Figure 5.3.3) (Ikäheimonen et al. 1995). This is due both to runoff from the Neva, Luga, and Narva rivers from less contaminated land areas (Figure 5.3.3) and a higher water exchange rate between the Gulf of Finland and the Baltic Proper than between the Gulf and the Bothnian Sea (Anisimov et al. 1991, Gavrilov et al. 1990, Ilus et al. 1993, Lazarev et al. 1988, V. G. Khlopin Radium Institute, 1991).

Figure 5.3.3 **Cs-137** concentrations (Bq/m^3) in waters of Narva and Luga rivers in 1986-1991.

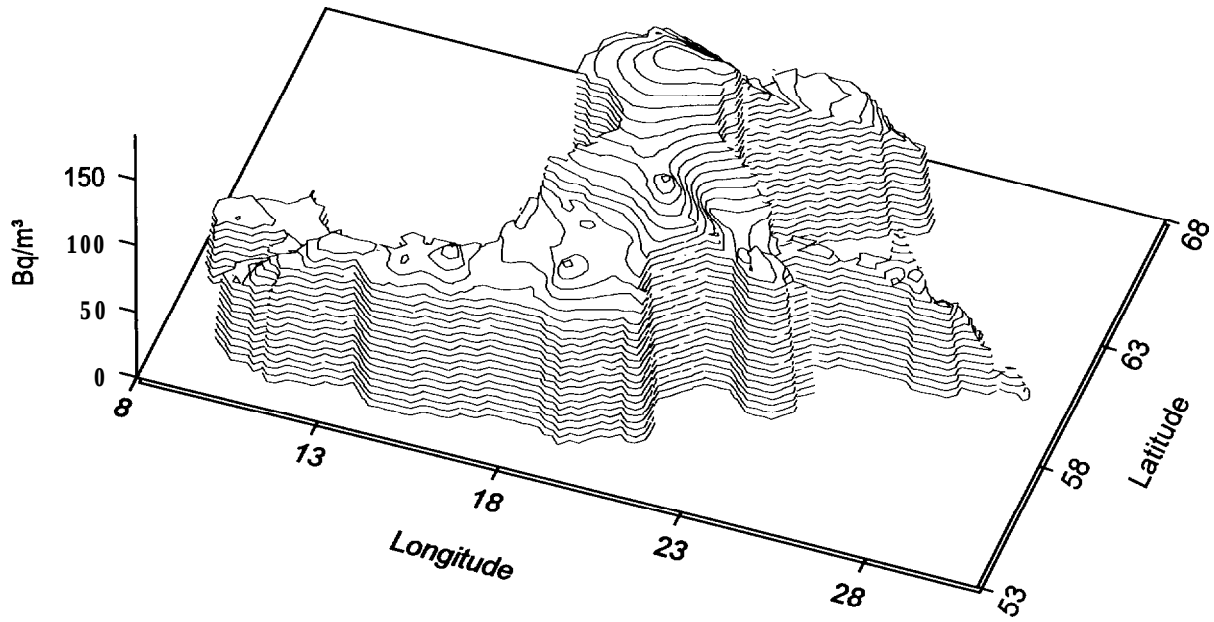


The contaminated water migrated southwards, which led to increasing Cs-134 and Cs-137 activity concentrations in these areas. The general distribution of Cs-137 concentrations in various regions of the Baltic Sea for 1986, 1988 and 1991 are shown in Figures 5.2.2, 5.3.4 and 5.3.5.

Figure 5.3.4 Distributions of Cs-137 activity concentrations in 1988 in the Baltic Sea.



Fiire **5.3.5** Distributions of Cs-137 activity concentrations in 1991 in the Baltic Sea.



By 1988, the caesium concentrations had largely smoothed out due to hydrological processes in the Baltic Sea and subsequent river discharge. Nevertheless, there is a marked difference between Cs-137 concentrations in different sea areas. This is seen in Figure 5.3.6 which illustrates the Cs-137 distribution in 1989. In 1991, Cs-137 concentrations in the surface waters of the eastern Baltic Proper ranged from 100 to 130 Bq/m^3, being (120 $\pm$ 10) Bq/m^3 on average.

Figure 5.3.6 Distribution of Cs-137 concentrations in different Baltic Sea areas in June 1989.

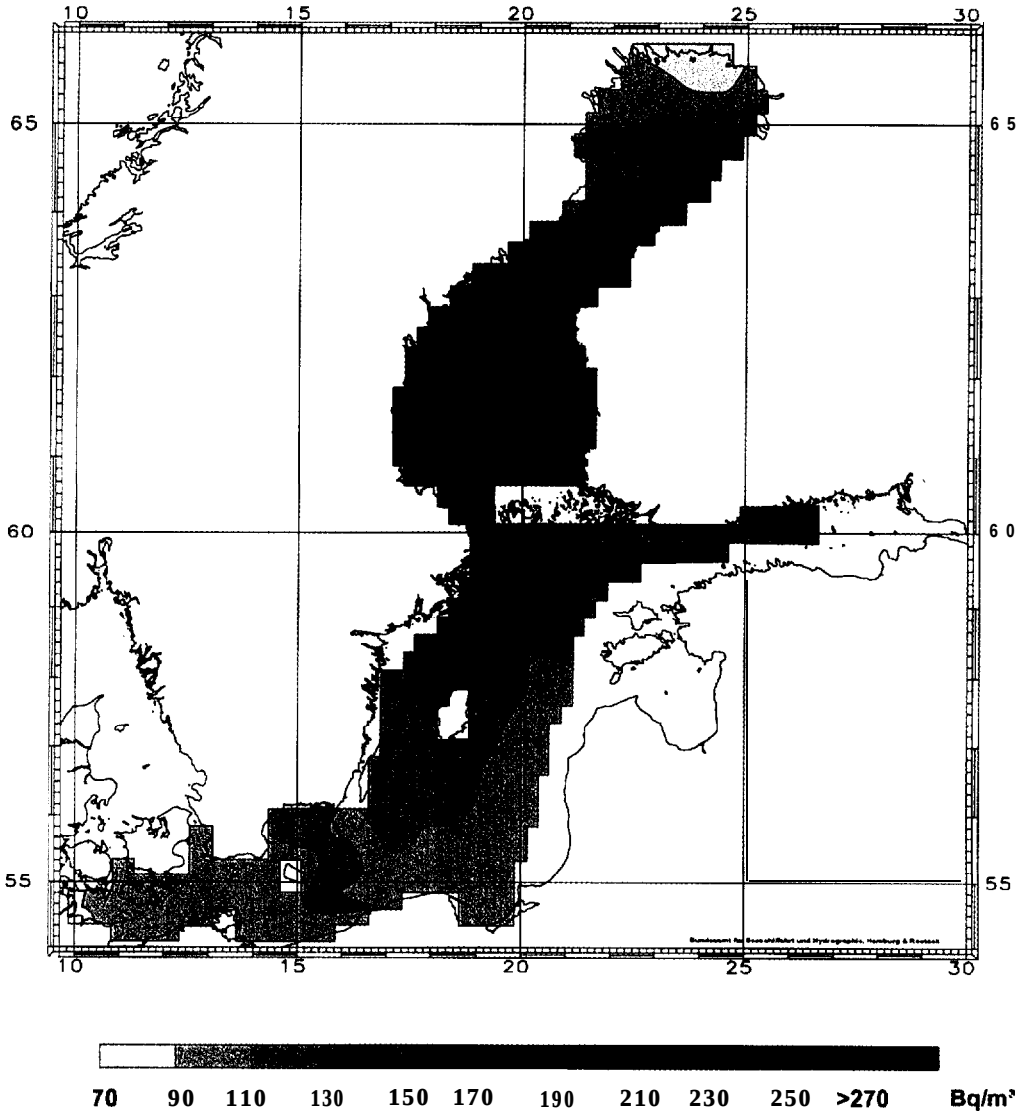


Figure 5.3.7 depicts the trend over time of Cs-137 and Sr-90 activity concentrations in surface water at a site in the Belt Sea (Schleimündung) from 1970 to 1992. The first Cs-137 maximum in 1980 was due to the maximum annual discharge at Sellafield in 1975, revealing the time it required contaminated water to be transported to this site. The concentrations of Cs-137

changed dramatically after the Chernobyl accident. Those of Sr-90, however, increased only slightly in 1986, and soon returned to the pre-Chernobyl level (of about 20 Bq/m³).

Figure 5.3.7 Cs-137 and Sr-90 concentrations (Bq/m³) in surface waters at a site in Baltic Belt Sea, Schleimündung, in 1970-1991.

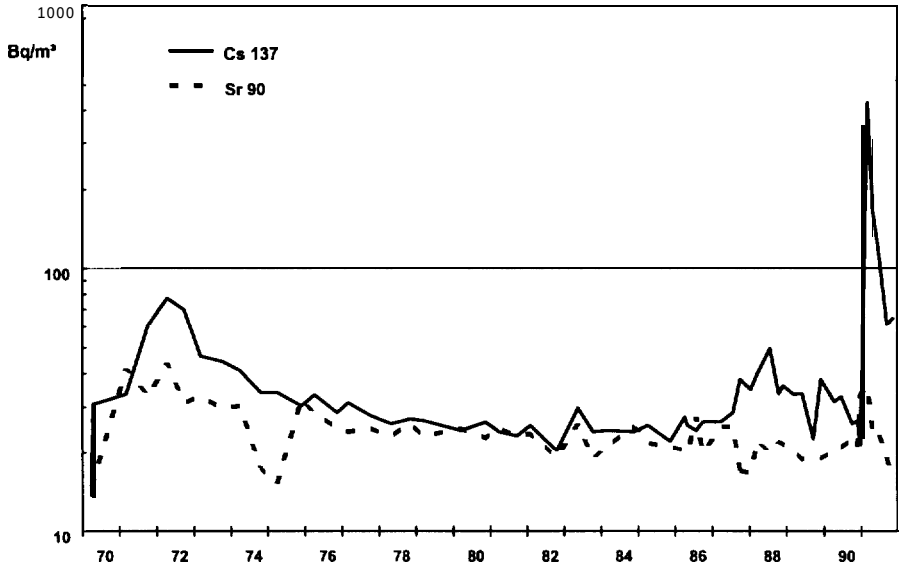
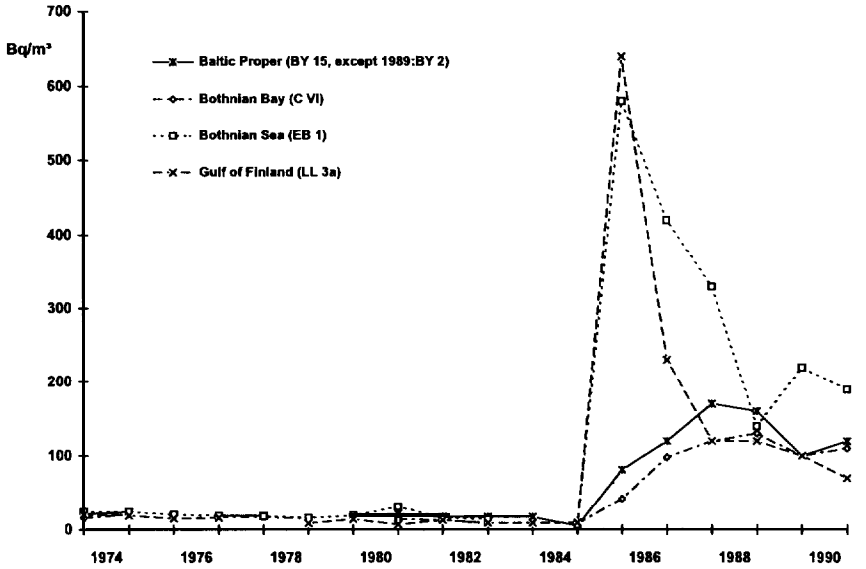


Figure 5.3.8 gives the trend in Cs-137 concentrations from 1974 to 1991 at four sites in the Baltic Sea. The trend shows a decreasing trend but still the mean concentrations of Cs-137 are about 10-20 times higher in 1991 than before the Chernobyl accident.

Figure 5.3.8 Cs-137 concentrations (Bq/m³) of seawater at four Baltic Sea sampling stations in 1974- 1991.



The concentrations in the Bothnian Sea were at a somewhat higher level than in other parts of the Baltic Sea. As a whole, the Cs-137 concentrations are currently at a significantly higher level in the Baltic Sea than in seas elsewhere in the world.

5.4. Inventories in Seawater

Attempts were made to compile inventories of the main radionuclides in the water of the Baltic Sea on the basis of observation data. Several results were received at Risø in 1989 (Dahlgaard, 1989). Those estimates showed that the inventory of Cs-137 in the Baltic water mass was 324 TBq in 1983, 4620 TBq by September 1986 and 3400 TBq in August 1987. The Sr-90 inventory was 416 TBq in 1983, 460 TBq in October 1986, and 391 TBq in August 1987. Later on, some estimates were made by experts of the MORS Group. These results are given in Table 5.4.1. The procedure for calculating the inventory is based on the rather simplified assumption that the estimated mean concentration of Cs-137 is the same for the entire Baltic Sea. The unusual decrease in 1988 was probably caused by the lack of observation data on the most contaminated areas during that period. It is worth to note that the estimates were made using a much simplified approach and need to be further refined to yield more accurate results. Problems related to the calculation of inventories, e.g. contributions from different sources, will be discussed in detail in Chapter 8.

Table **5.4.1.** Cs-137 Inventories in the Baltic seawater.

Year	Inventories TBq	By H. Dahlgaard (1989) TBq
1985	325	324 (in 1983)
1986	4260	4620
1987	2700	3400
1988	1790	
1989	2320	
1990	2060	
1991	2330	

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6. RADIONUCLIDES IN SEDIMENT AND SUSPENDED PARTICULATE MATTER

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3) Institute of Oceanology, Polish Academy of Sciences, Poland

6.1. Introduction

During the period 1984-1991 the most important event which had an impact on radioactivity in Baltic Sea sediments was the accident at the Chernobyl Nuclear Power Plant (NPP) in April 1986. Global fallout during the 1960s and discharges from reprocessing plants in western Europe and nuclear facilities in the Baltic Sea region have had markedly smaller impacts on radionuclide concentrations, especially in respect to the northern Baltic Sea.

In this chapter, most of the attention has been paid to Cs-137, because it was the main long-lived radionuclide in Chernobyl fallout. However, certain amounts of Cs-137 already occurred in Baltic Sea sediments before the Chernobyl accident, mainly as a consequence of the fallout caused by atmospheric nuclear weapons testing during the 1960s. The role of Cs-137 is especially important in sedimentological studies, because the affinity of caesium for clay particles is well known.

Baltic Sea sediments offer an exceptionally good opportunity for conducting sedimentological studies, since the average rate of sedimentation is much higher there than in the oceans and in most other coastal seas. The appearance of anoxic conditions in the near-bottom water of the Baltic Proper and hence a lack of benthic animals on large bottom areas results in the formation of an undisturbed sedimentary package, enabling the sampling of sediment laminae in an undisturbed historical order. Modern dating methods provide reliable timescales in which time-dependent changes in radionuclide contents can be recorded.

Bottom sediments play an important role in radioecological studies of the marine environment, because a large proportion of radioactive substances entering a body of water is adsorbed over time onto suspended particulate matter and deposited in sediments. However, it is often problematic to take reliable samples from bottom sediments, due to the many sources of error associated with sampling techniques. Sediment sampling is much more sensitive to errors than any other sampling technique in the marine environment, and these errors can cause remarkable differences in results. This fact should always be taken into account when evaluating sediment results.

6.2. Sampling Techniques

Sediment sampling techniques and sampling devices differ widely in many aspects. Even within the HELCOM/MORS Group the variability of sampling devices is large. Almost every laboratory has developed its own type of device, with individual sampling area, hydrodynamic

features, slicing systems, etc. Most probably, this variability also causes differences in results. Very often, large differences appear between results expressed in Bq m^{-2}, although the values expressed as Bq kg^{-1} dry weight are similar.

During recent years several intercomparisons were conducted between most sampling devices used in the MORS Group (Nies et al. 1990, Klemola et al. 1991, Anisimov et al. 1991). In 1992 an extensive intercomparison exercise was carried out at four Baltic Sea stations with different bottom types. The aim was that all sampling devices used in the MORS Group should be tested during the exercise. The results of this intercomparison will be published in the near future.

Description of sampling devices used in the MORS Group

Denmark

The HAPS sampler used by Risö National Laboratory is a frame-supported cylindrical corer with a 13.6 cm core diameter. After sampling, the stainless steel coring tube is removed from the sampler and the sediment is extruded through the tube into a slicing device fixed to the top of the tube. The slicing device is normally used to cut 3 cm sections. One core represents a sampling area of 145 cm^2 (Nies et al. 1990).

Federal Republic of Germany

The regular method of the BSH in the collecting of sediment samples is the use of a 50 x 50 cm box corer which takes the sediment core with its overlying water (Nies et al. 1990). The water is removed from the surface by careful pumping and sub-samples are taken by means of perspex tubes at locations which seem to be appropriate for the penetration of the tubes. The diameter of the cylindrical tubes was mostly 94 mm. The tubes are fitted to a gum collar and pressed downwards onto the top of a piston and the sediment can be sliced into 2 cm or 3 cm vertical sections, as desired. The slices from two cylinders are combined into one sample representing a total area of 139 cm^2. The samples are stored in plastic boxes and deep-frozen, then freeze-dried in the laboratory and homogenized.

Finland

The ordinary sediment samples of the STUK were taken aboard the Finnish Research Vessel ARANDA using an AQUARIUS box corer described in Klemola et al. 1991. From 1990 onwards, the corer was equipped with a steel frame to avoid listing on the bottom and to limit corer penetration into the sediment. The sampling area of the corer is 182 x 183 mm. An iron framework surrounds an inner plexiglass "aquarium" 30 cm high. The entire device is 82 cm high and weighs 25 kg. The principle of the slicing apparatus is similar to that of the NIEMISTÖ corer described below. Normally, one core/station is taken with the AQUARIUS corer (area 333 cm^2) and sectioned into 5-cm-thick subsamples.

A NIEMISTÖ corer (Niemistö 1974) was used mostly in special samplings when very fine slices are needed from the uppermost layers of the surface sediment. The very precise sectioning apparatus allows the taking of subsample slices only 2-mm-thick.

The sampler consists of a plexiglass tube (inner diameter 50 mm) in a stainless steel body. The corer is 140 cm long and weighs 25 kg when loaded with three lead weights. In special studies at the Teili-1 Station 10 parallel samples were taken with the NIEMISTÖ corer; the total sampled area was 196 cm^2. The cores were sectioned into l-cm-thick parallel subsamples which were combined for analyses. After slicing, the samples were rinsed in plastic bags or boxes and stored frozen, then freeze-dried and homogenized before analysis.

German Democratic Republic

For most of the soft sediment samples a NIEMISTÖ-type corer with a 54 mm inner diameter was used. The corer was used six times in parallel to obtain one sample representing an area 6 x 22.9 cm^2 = 137 cm^2. The perspex tube was fitted to a sectioning apparatus in which the sediment core was extruded by a piston and sliced with a sliding sheet. Then the samples were deep-frozen and freeze-dried.

Some samples were collected by means of a NIEMISTG-type corer with an 82 mm inner diameter. The corer was used twice to obtain one sample 105.6 cm^2 in area. This corer showed less vertical disturbance than the smaller-diameter corer (Nies et al. 1990).

Poland

The sediment samples of the CLOR were taken aboard the Polish research vessel HYDROMET using two types of gravity corers. During 1984-1986 a NIEMISTÖ-type corer was used and since 1987 the sediment samples were taken with a gravity corer constructed by the Radium Institute, St. Petersburg.

The NIEMISTÖ corer consisted of a plexiglass tube (inner diameter 46 mm) in a stainless steel body. In general, three parallel samples were taken which totalled 50 cm^2 in area. The sediment cores were sectioned into 5-cm-thick subsamples in 1984 and into 3-cm-thick subsamples in 1985 and 1986.

The corer constructed by the Radium Institute consists of a plexiglass tube (inner diameter 90 mm) in a stainless steel body.

In general, two parallel samples were taken with this corer which total 127 cm^2 in area. During 1987-1990 the cores were sectioned into 3-cm-thick subsamples. Since 1991 the cores were sectioned into l-cm-thick subsamples at depths up to 5 cm and into 2-cm-thick subsamples at depths of 5-2 1 cm.

The parallel subsamples were combined for analyses. After slicing, the samples were placed in plastic boxes and stored frozen, then air-dried and homogenized before analysis.

Soviet Union

Sediment samples were taken with the corer constructed by the V.G. Khlopin Radium Institute, which consists of several modifications. The corer has a Plexiglas tube with a 64 cm^2 sampling area surrounded by a stainless steel framework and penetrates into the bottom

sediment by gravity. It is possible to control the depth of penetration by changing the weight of the corer.

The height of the sample core normally varies from 25 to 36 cm. After lifting the corer aboard, the plexiglass tube is removed; usually l-2 parallel samples are taken at each sampling station. The cores are sectioned into 2-cm-thick subsamples at depths up to 10-15 cm, either manually or with a special device.

Sweden

The sampling methods used within the HELCOM/MORS sediment programme (as well as in the environmental monitoring programmes of Swedish NPPs) are described in detail by Notter (1987).

There are three different types of corer which were alternatively used:

1. The "topplod" corer (core diameter 2 or 3 cm). Fifteen parallel cores were taken at each site; the upper 2 cms from each were collected and pooled to constitute one sample for measurements.

2. The WILLNER- and NIEMISTÖ-corers (diameters 6.5-7.5 cm and 5 cm, respectively). Four to five cores were taken at each site and the upper 2 cms of each were used for a pooled sample. At greater depths the NIEMISTÖ corer was preferred.

6.3. Description of Sediment Types

The uneven distribution of different sediments in the Baltic Sea reveals the dynamic nature of the sedimentation processes. The present sedimentation shows a wide range of processes, from erosion and transportation to accumulation. The different regions of the Baltic Sea are characterized by rapid land upheaval in the North and by differences in seabed geology. These differences range from the rugged forms of crystalline bedrock in the Gulfs of Bothnia and Finland to the smoother sedimentary formations in the southern Baltic Sea.

The southwestern Baltic Sea is characterized by vast areas with sandy bottoms. The southern and central regions expose large accumulation basins with fine-grained, soft silty and muddy sediments. The northern Baltic Proper is characterized by an irregular topography partly dominated by a mosaic of small-scale sedimentation basins and partly by a few large basins, the Gotland Basin being the largest in the Baltic Sea.

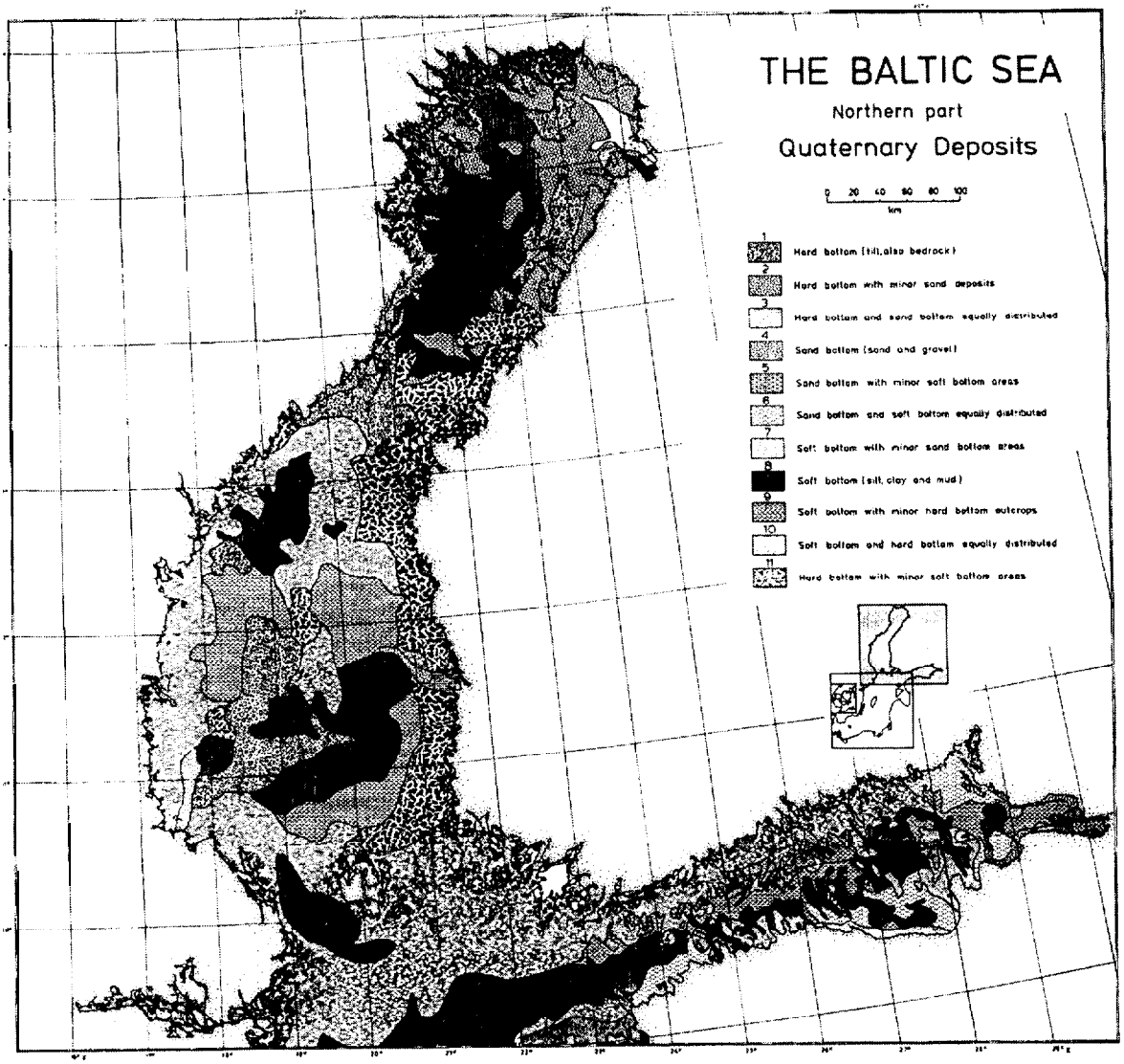
Rugged forms of bedrock create a series of very small, irregular separate basins in the Gulf of Finland. The largest sedimentation basins are situated in the southern Gulf, while the eastern Gulf is dominated by the estuary of the Neva River. Sedimentation in the eastern region may be very high as a result of the river transport of particulate matter.

The sediments in the Gulf of Bothnia are characterized by the “basin-filling nature” of sedimentation in the deepest areas. The rapid land upheaval continuously reveals sediments for

erosion caused 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 surfaces.

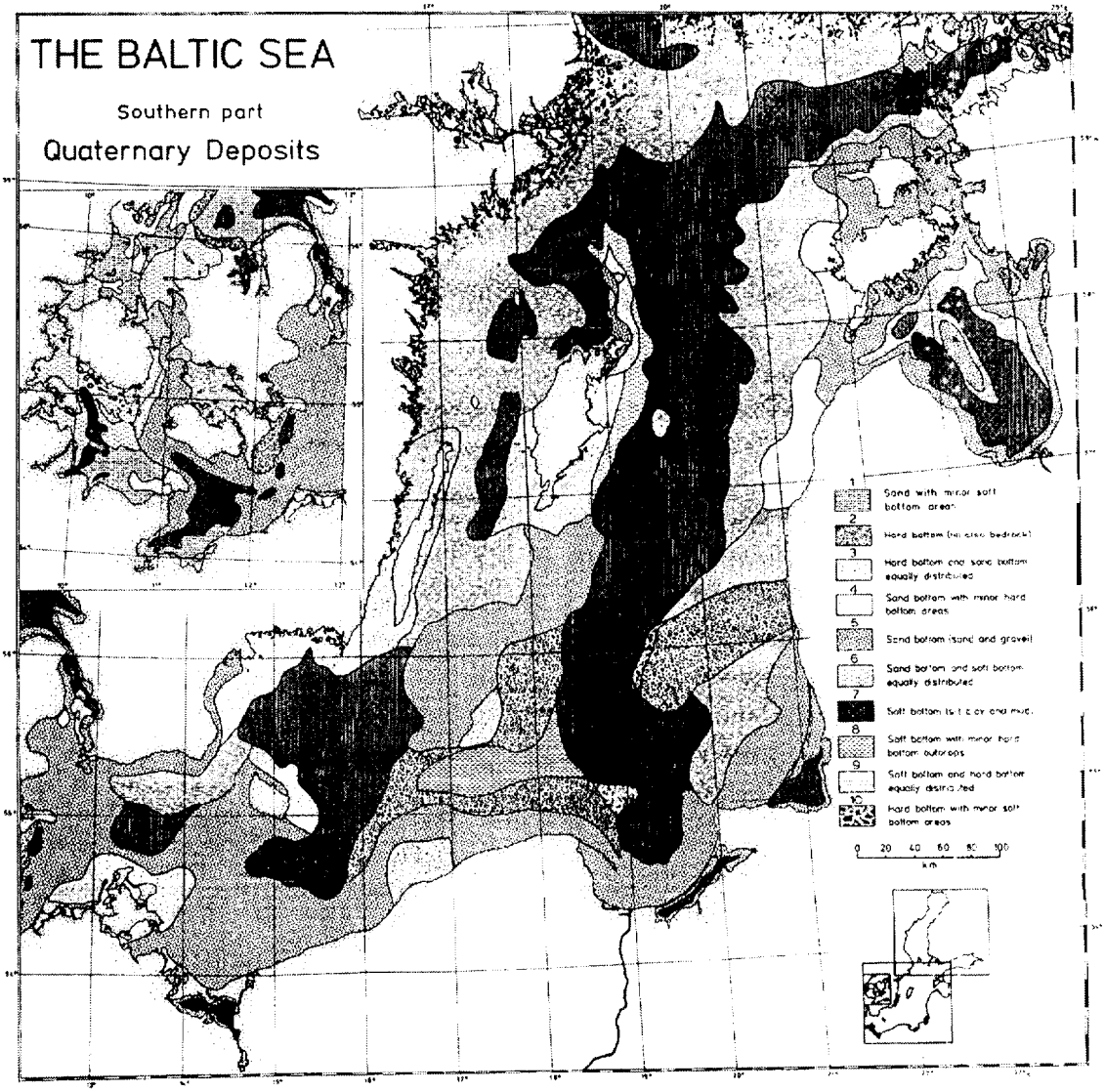
The making of total inventories of radionuclides in sediments of the Baltic Sea requires a knowledge of the distribution of different sediment types in the area. A useful description of the Quarternary deposits in the Baltic Sea has been published by Winterhalter et al. (198 1); (Figures 6.3.1 and 6.3.2). Based on these maps, Salo et al. (1986) made a planimetric calculation of the proportion of soft and hard bottoms in the Baltic Sea (Table 6.3.1). This calculation shows that over 50% of the total area can be classified as hard bottom.

Figure 6.3.1. Distribution of Quaternary deposits in northern Baltic Sea (Winterhalter et al. 1981).



In most cases sediment samples were taken from soft bottoms, i.e. from real sedimentation bottoms. Soft bottoms very often act as “sinks” for radionuclides, whereas hard bottoms are regarded as transport bottoms with very low accumulations of sinking matter (and radionuclides). However, even erosion bottoms very seldom have uncontaminated surfaces, because bioturbation caused by benthic fauna mixes contaminants and organic material into the surface sediment layers.

Figure 6.3.2. Distribution of Quaternary deposits in southern Baltic Sea (Winterhalter et al. 1981).



Salo et al. (1986) assumed that Sr-90 and Cs-137 are distributed between hard and soft bottoms in a ratio of 1/5, while for Pu-239,240 the ratio is about 1/10. Recent results from the coastal area adjacent to the Olkiluoto NPP show that the ratio for Cs-137 may be even smaller: 1/20; which would lower the total inventory of Cs-137.

Table 6.3.1. Areas of soft and hard bottom in different regions of the Baltic Sea (Salo et al. 1986).

Sea area	Soft bottom km^2	Hard bottom km^2
Gulf of Bothnia		
Bothnian Bay	16 000	21 000
Bothnian Sea	40 000	39 000
Gulf of Finland	16 000	14 000
Baltic Proper	9 000	110 000
Gulf of Riga	7 000	12 000
Total	178 000 +	196000 = 374000

6.4. Sedimentation Rates

In 1986, soon after the deposition of Chernobyl fallout on the surface of the Baltic Sea, sinking of fresh fallout through the water layers was observed to be quite rapid. During the first half of May, fresh fallout nuclides were already detected in water at a depth of 100 m in the southern Baltic Proper (Ilus et al. 1987). It was found that airborne particles from Chernobyl, after deposition on the sea surface, released only a part of their caesium content into the water, while the other part settled on the bottom rather rapidly (Bojanowski, unpubl.).

Most probably, the sinking of the fallout nuclides was also accelerated by phytoplankton, which was at the end phase of its spring maximum during the main fallout period. After the vernal bloom of phytoplankton, radionuclides were transported downwards by dead plankton algae (Ilus et al. 1987). In mid-June fresh fallout nuclides were already detected in the surface sediment layer at Station Teili-1 (depth 166 m) in the northern Baltic Proper.

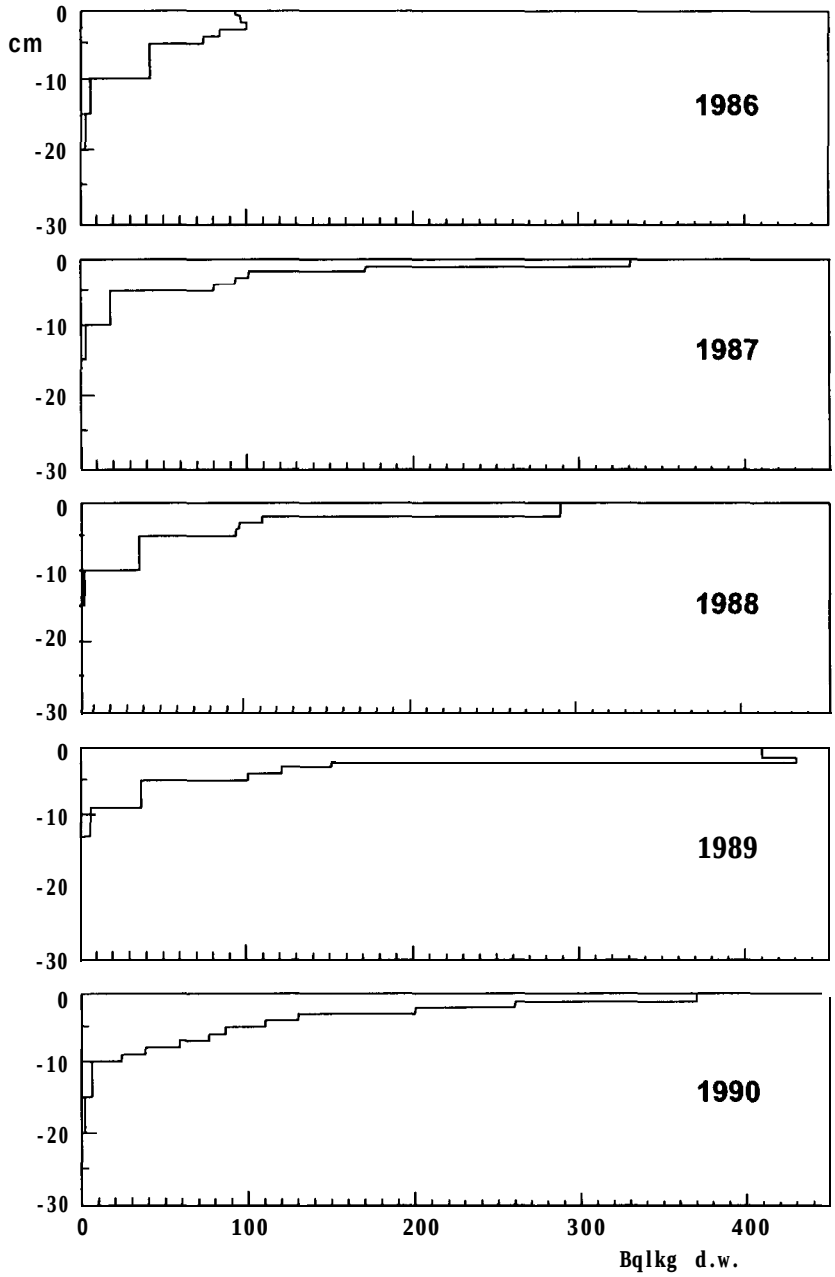
After the initial fallout situation, several factors affected the concentrations of radionuclides in the water body, e.g. sea currents, mixing of water masses, river discharges from the catchment areas, sinking in the water column and deposition on the bottom. These factors were also reflected in the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

The Chernobyl fallout, as well as the global fallout during the 1960s, offer an excellent opportunity to sedimentologists for evaluating sedimentation rates in the Baltic Sea. The concentration peaks of long-lived radionuclides remain as signs of maximum fallout periods in sediment layers. Many factors, however, can weaken the value of these concentration peaks in this respect. One of the main factors is associated with errors in sampling techniques, i.e. how undisturbed are the samples we can get with our sampling device. Other factors

disturbing the integrity of sediment layers are bioturbation caused mainly by benthic macrofauna, near-bottom currents, suspension-resuspension processes, eventual movements of radionuclides in sediments, etc.

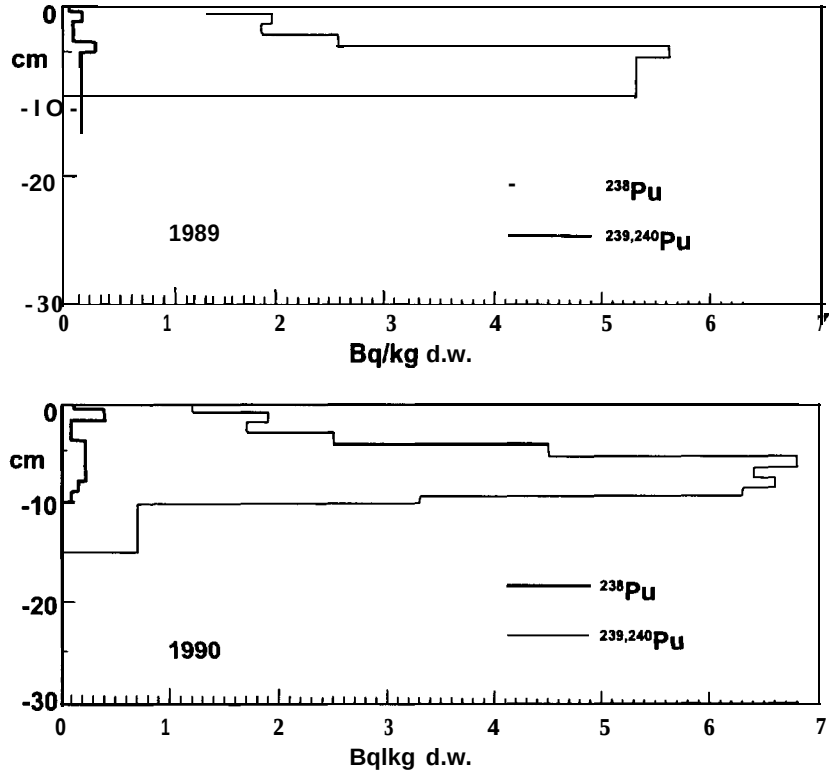
Sediment cores taken at Station Teili-1 in the northern Baltic Proper (Figures 6.4.1 and 6.4.2) illustrate sedimentation rate in a basin where bioturbation does not disturb the sedimentation process due to the absence of benthic animals from anoxic conditions.

Figure 6.4.1. Vertical distribution of caesium-137 (Bq kg^{-1} dry wt.) in sediment samples taken at Teili-1 Station in 1986-1990 (Ilus et al. 1993b).



In 1990 the lower edge of the elevated Cs-137 concentration was at a depth of 2-3 cm and that of Pu-239,240 (originally from the maximum period of global fallout during 1962-1965) at 9-10 cm. By means of these peaks, the sedimentation rate at the station could be evaluated at about 3.7-5 mm a^{-1}. These values are slightly higher than that evaluated earlier for this station with the Pb-210/Po-210 method: 2.3 mm a^{-1} (Niemistö and Voipio, 1981). However, at least the higher value, which is based on the Cs-137 peak may be overestimated, because in 1990 the sediment layers concerned had not yet become compact.

Figure 6.4.2. Vertical distribution of plutonium-238 and plutonium-239,240 (Bq kg^{-1} d.w.) in sediment samples taken at Teili-1 Station in 1989 and 1990 (Ilus et al. 1993b).



Sedimentation rates evaluated with different methods in various subregions of the Baltic Sea are presented in Table 6.4.1.

Table 6.4.1. List of reported sedimentation rates in various sub-regions of the Baltic Sea.

Subregion	Station	Estimated sedimentation rate mm a^{-1}	Reference
Bothnian Bay	general	0.6-1.3	Tulkki (1977)
	-"-	0.6-1.3	Niemistii (1986)
	-"-	0.6-1.9	Perttili & Brügmann (1992)
	C VI	1.3	Niemistii (1982)
	c VI	1 s-2	Tuomainen et al .(1986)
	BO 3	1.2	Niemistii (1982)
Bothnian Sea	general	0.27	Winterhalter (1972)
	-"-	0.1-0.8	Niemistii et al.(1978)
	-"-	0.9-2.4	Niemistö (1986)
	US 2b	0.92	Niemistii (1982)
	EB 1	2.4	Niemistii & Voipio (1981)
	EB 1	2-4	Tuomainen et al. (1986)
	BO 3a	2.5	Tuomainen et al. (1986)
	Olk 2	10	Tuomainen et al. (1986)
Gulf of Finland	general	0.4	Perttili et al.(1995)
	XV1	7.4	Niemistb et al.(1981)
	Lov 2	5	Ilus, pers. comm.
Northern Baltic Proper	Teili 1	2.2	Niemistii & Voipio (1981)
Baltic Proper	BY 15	1.25	Niemistii & Voipio (1981)
	general	0.5-1.5	Niemistii (1986)
	-"-	0.5-4.2	Perttilä & Niemistii (1993)
Gulf of Riga	general	0.5-2.2	Kuptsov et al.(1984)
Bornholm Basin	general	0.5-1.5	Kögler & Larsen (1979)
Southern Baltic Proper	general	0.4-2.3	Pempkowiak (199 1)
Gdansk Basin	general	0.5-4.0	Perttilä & Brügmann (1992)
Western Baltic Sea	general	l-2	Brügmann & Lange (198 1)

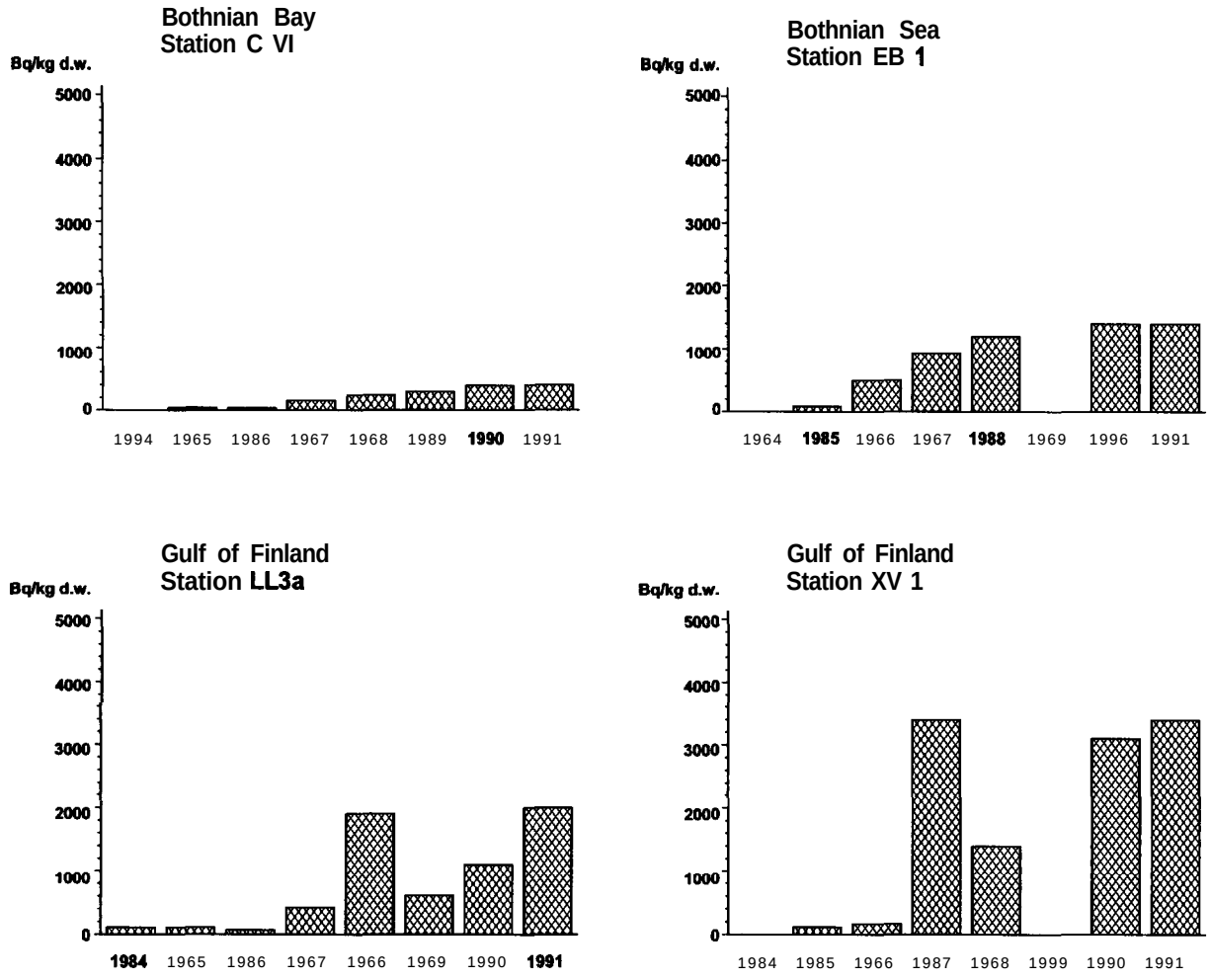
6.5. Activity Concentrations in Sediments and Time Trends

This Evaluation Report is based on data reported by the Contracting Parties to the HELCOM/MORS database. Sediment data from the period 1984-1991 were reported by seven countries as follows: DE 157, DD 49, DK 7, FI 43, PL 43, SE 12 and SU 33, a total of 344 sediment profiles. When all slices are included the material consists of 998 samples. Prior to spring 1986 eleven radionuclides were reported in sediment samples and 23 thereafter.

The activity concentrations of Cs-137 in the surface sediment layer (Bq kg^{-1} dry weight) and

total amounts of this nuclide in all layers (Bq m^{-2}) at various sampling stations are presented in Figures 6.5.1-6.5.4. As early as 1986, a clear increase of Cs-137 was observed in the sediments at some sampling stations. During 1987 elevated levels of caesium were observed, especially in the eastern Gulf of Finland (e.g. station XV 1). Thereafter, the Cs-137 concentration in the surface sediment remained at the same level at station XV 1, but the total amount (Bq m^{-2}) attained its maximum only during 1990.

Figure 6.5.1. Caesium-137 in surface sediment layer (O-5 cm) at some sampling stations of Gulf of Bothnia and Gulf of Finland in 1984-1991.

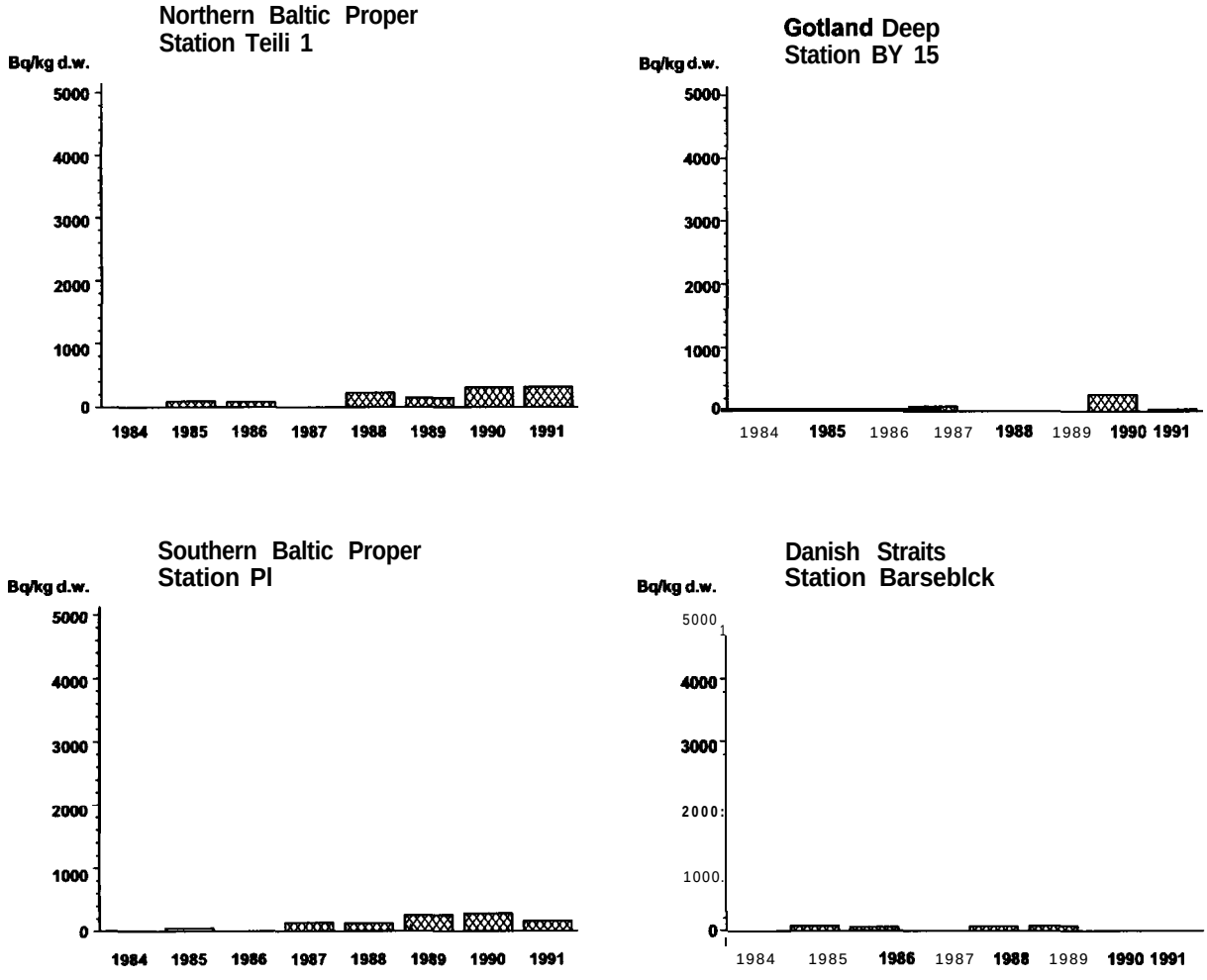


At many other stations, Cs-137 values have since continued to increase, indicating that settling of caesium from the water phase into the seabed continues.

A comparison of different subregions of the Baltic Sea shows that the largest sedimentation of Cs-137 occurred in the Gulf of Finland and the Bothnian Sea. This is in good agreement with the distribution pattern of Cs-137 deposition in the drainage area of the Baltic Sea. The highest concentration in the surface sediment layer, 3 400 Bq kg^{-1} dry weight, was observed during 1987 at station XV 1 in the eastern Gulf of Finland and the highest total amount,

43 000 Bq m^{-2}, at the same station during 1990. At station EB 1 in the mid-Bothnian Sea, the maximum values were 1400 Bq kg^{-1} and 21 000 Bq m^{-2} during 1990. The highest amounts in sediments are most probably not caused by higher site-specific deposition values, but are due to particle transport and the focusing of sinking radionuclides in the deepest areas of the basins. In the Baltic Proper and Bothnian Bay, the amounts of caesium in sediments were smaller, although a clear increase in concentration values was also observed in these areas since 1986.

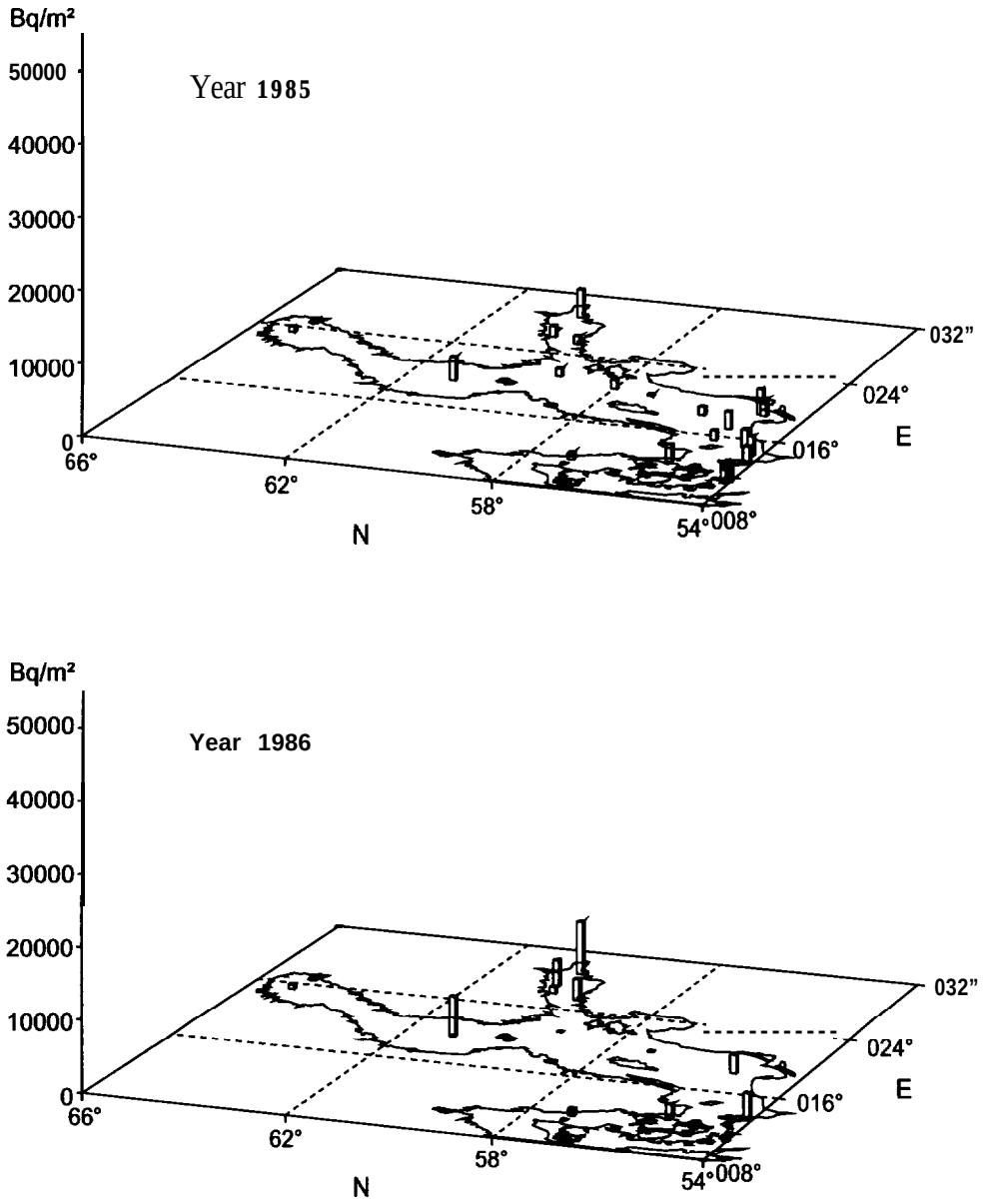
Figure 6.5.2. Caesium-137 in surface sediment layer (O-5 cm) at some sampling stations of Baltic Proper and Danish Straits in 1984-1991.



In addition to Cs-137, the other important radionuclide in the Chernobyl fallout was Cs-134. In the HELCOM/MORS database there are no data on Cs-134 in sediments prior to 1986, although it had been detected earlier in large water samples taken from the southern Baltic (Saxen et al. 1989). In the initial phase after the accident, the Cs-134/Cs-137 ratio was about 0.5. Due to its shorter half-life Cs-134 has disappeared from environmental samples more rapidly than Cs-137.

In addition to the caesium isotopes, the amounts of Ru-103, Ru-106, Ag-110m and Sb-125 clearly increased in Baltic Sea sediments as a consequence of the Chernobyl fallout. The occurrence of these nuclides followed rather well the distribution pattern of the caesium isotopes (Figures 6.5.3. and 6.5.4.). In general, these nuclides were most abundant during 1987, but due to their relatively short half-lives they already started to decrease by 1988/1989.

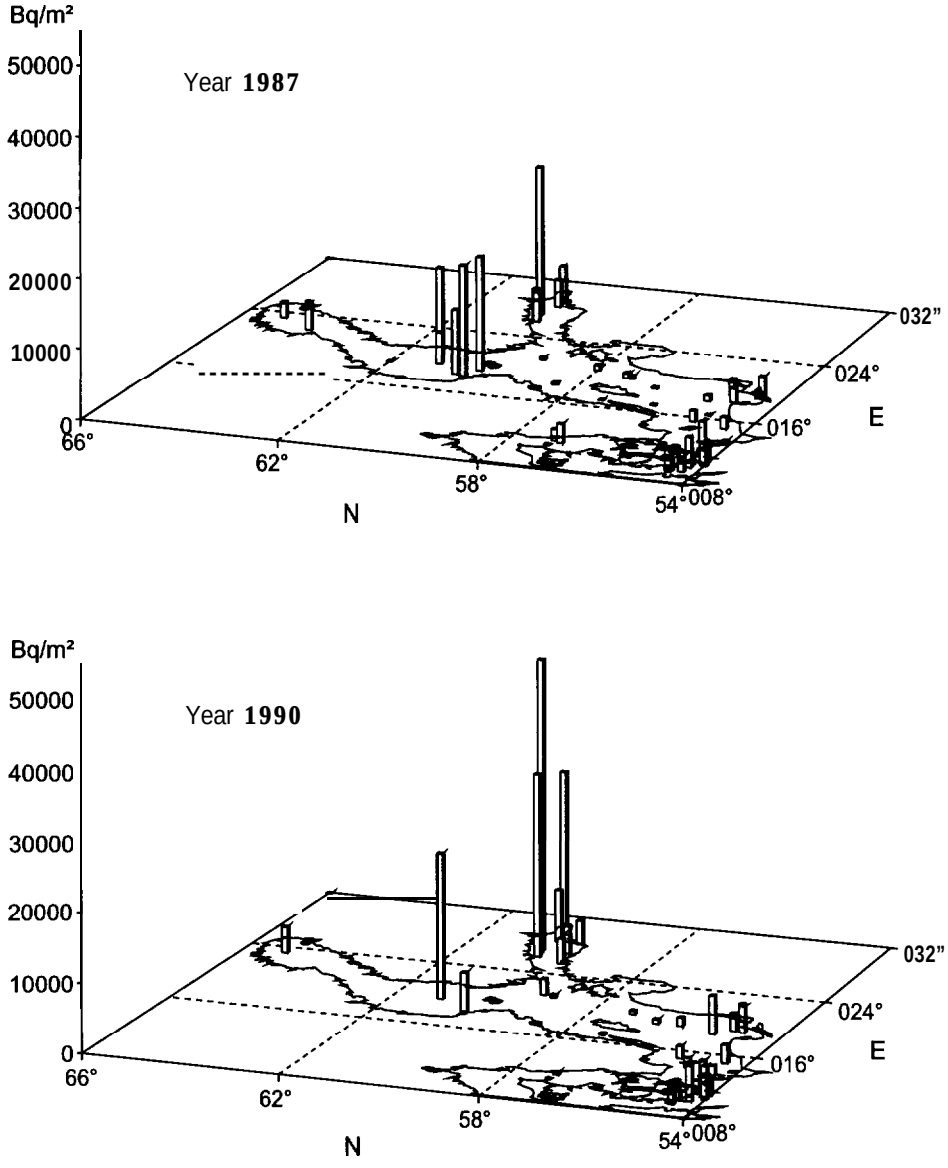
Figure 6.5.3. Total amount of caesium-137 at different sampling stations in 1985 and 1986.



During 1985 Co-60 was detected in sediment samples taken from the vicinity of the NPPs situated on the west coast of Sweden. During 1986 it was also detected in some shallow stations close to the Danish Straits. However, during 1987 observations of Co-60 were made at several stations covering the entire Baltic Sea area (Figure 6.5.9). This might be an

additional indication that small amounts of Co-60 were present in the Chernobyl fallout. Similar indications were found earlier, e.g. in algae samples taken from the Bothnian Sea coasts of Sweden and Finland (Grim&s et al. 1986, Ilus et al. 1988) and in some sediment samples taken from Finnish lakes (Ilus et al. 1993a).

Figure 6.5.4. Total amount of caesium-137 at different sampling stations in 1987 and 1990.



The concentrations of Ra-226 are of natural origin; thus they are very evenly distributed in Baltic Sea sediments (Figure 65.8). Pu-238 and Pu-239,240 mainly originate from the fallout caused by nuclear weapons testing during the 1960s. Although the Chernobyl fallout also included small amounts of plutonium, the changes caused by the fallout in the Fu-239,240 concentrations of surface sediments were small (Figure 6.5.10). Due to the different ratios of

Pu-238 and Pu-239,240 in the two fallouts it is possible to detect even the tiny increase of plutonium caused by the Chernobyl fallout in Baltic Sea sediments.

Figure 6.5.5. Silver-110m in surface sediment layer at different sampling stations in 1987.

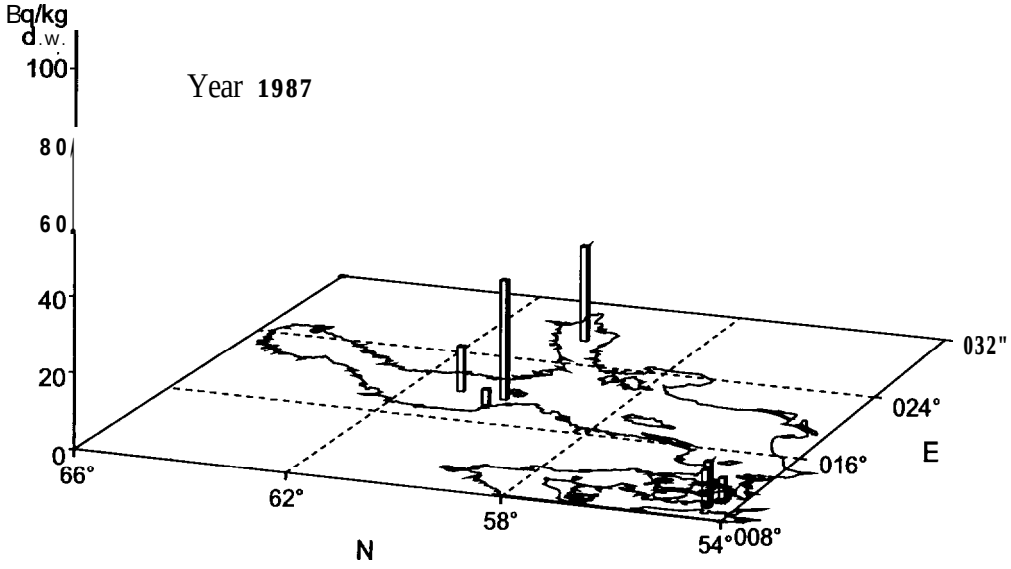
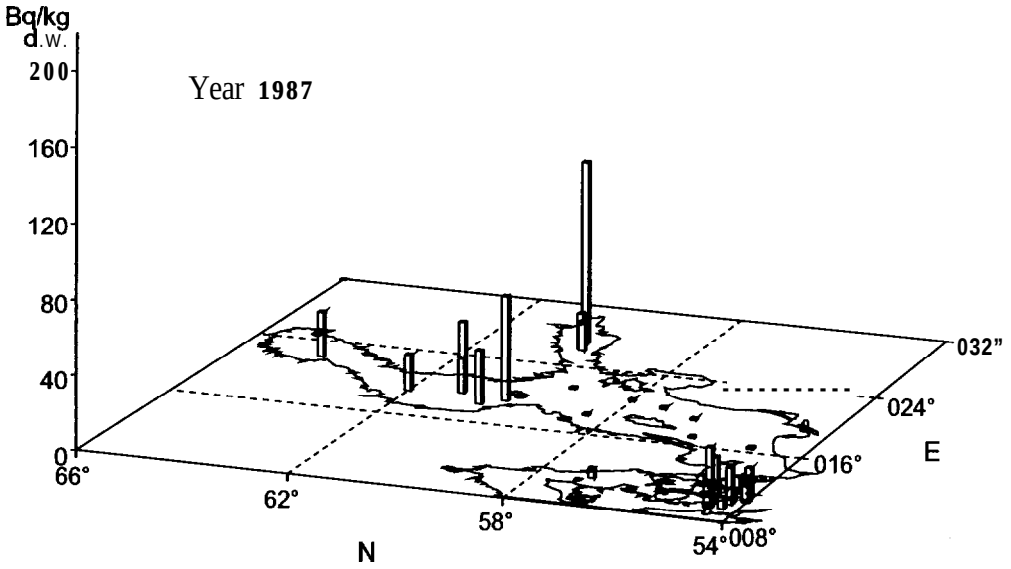


Figure 6.5.6. Antimony-125 in surface sediment layer at different sampling stations in 1987.



Fii 6.5.7. Ruthenium-106 in surface sediment layer at different sampling stations in 1987.

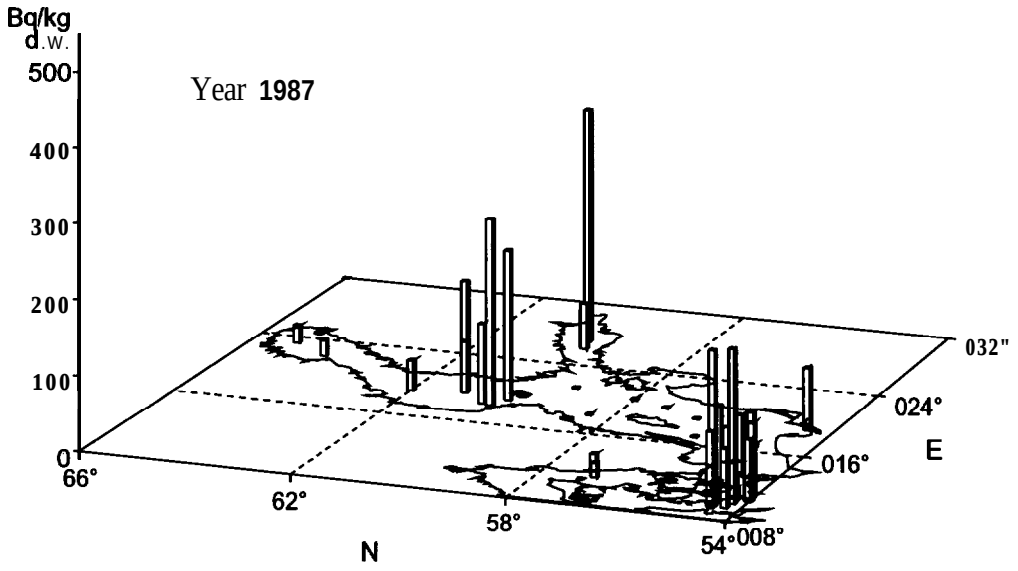


Figure 6.5.8. Radium-226 in surface sediment layer at different sampling stations in 1987.

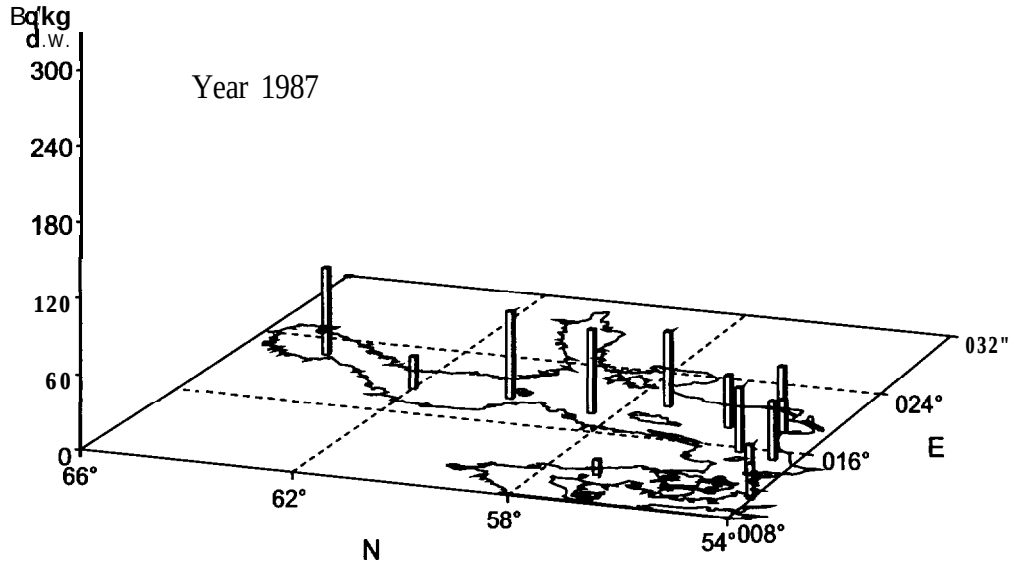


Figure 6.5.9. Cobalt-60 in surface sediment layer at different sampling stations in 1986 and 1987.

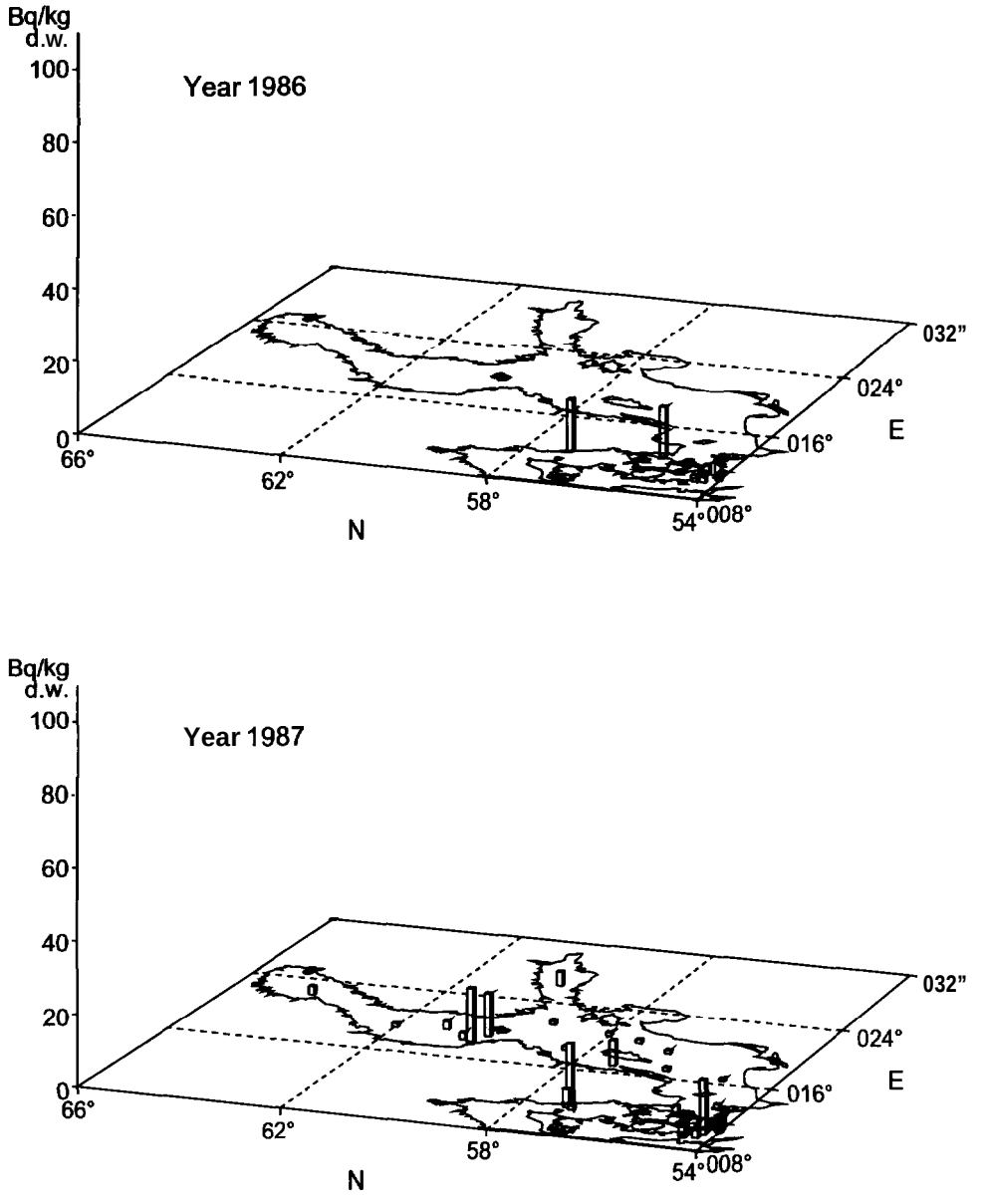
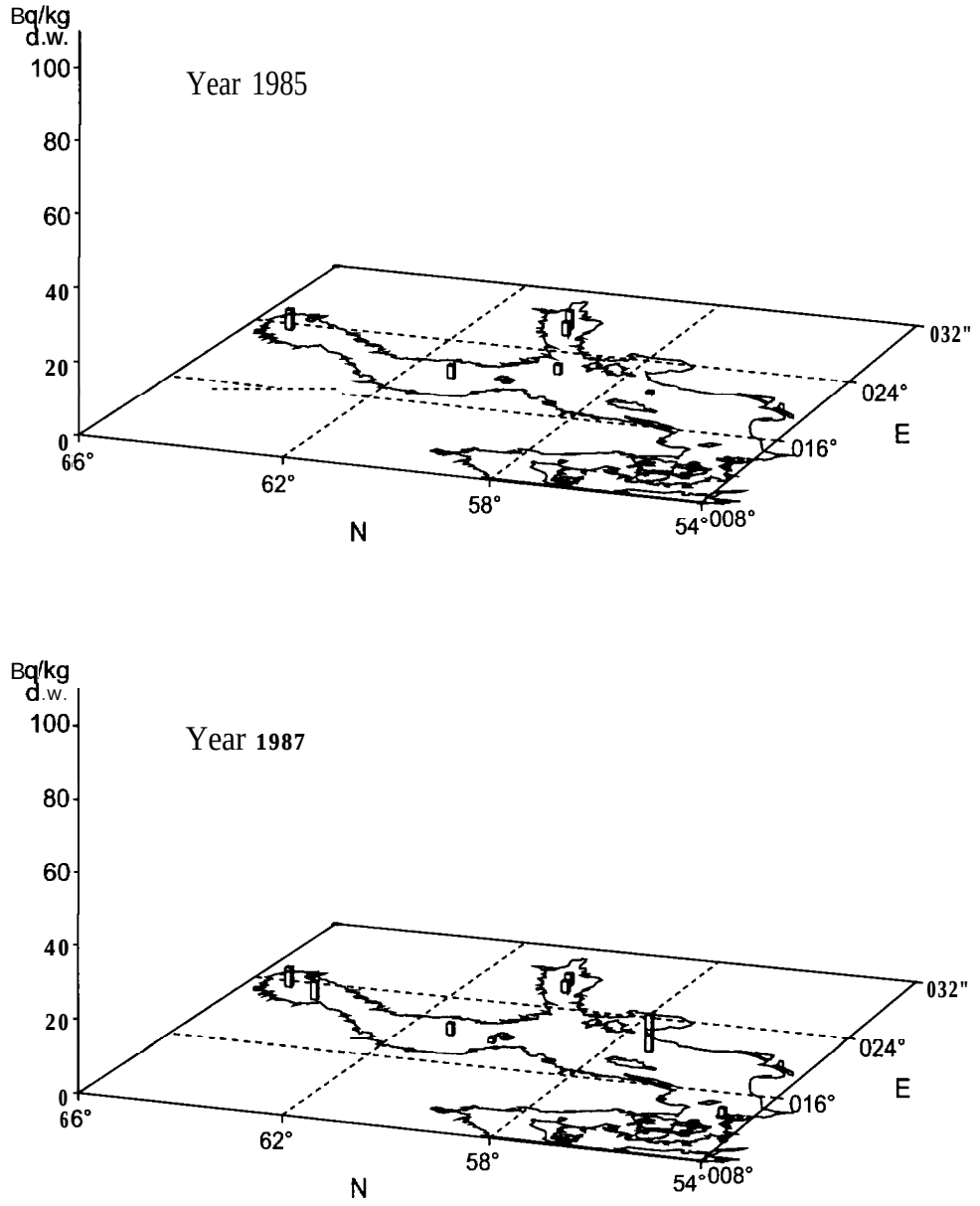


Figure 6.5.10. Plutonium-239,240 in surface sediment layer at different sampling stations in 1985 and 1987.



6.6. Activity Concentrations in Suspended Particulate Matter

Only Finland has reported data on suspended particulate matter to the HELCOM/MORS database. The material was collected from two coastal areas close to the Finnish NPPs Loviisa and Olkiluoto; thus, the results include both Chernobyl nuclides and nuclides originating from the local NPPs. However, the proportion of the Chernobyl-derived caesium isotopes, Cs-137 and Cs-134, is highly dominant in the samples, due to the affinity of caesium for particulate matter.

Sinking matter is an excellent sampling object in environmental monitoring of NPPs. Since there are many difficulties in sampling of recently (weeks or months) settled particles from the surface of the sediment, the proper sediment samples can be replaced by collecting sinking matter. In the environs of the Finnish NPPs sinking matter is collected continuously at four stations and proper sediment samples are taken only once every four years.

Theoretically, the activity concentrations in suspended particulate matter should be about the same as in the surface layer of sediment. Table 6.6.1 indicates, however, that sinking matter in general gives higher concentration values and may be more sensitive in detecting some radionuclides at low concentrations.

Table 6.6.1. Gamma-emitting radionuclides in sinking matter and surface sediment layer (Bq/kg dry weight) at four sampling stations in the vicinity of Loviisa NPP in 1989-90.

	Sampling depth m	Dry weight g	K-40	Mn-54	Co-58	Co-60	Ru-106	Ag-110m	Sb-125	cs-134	Cs-137	Ce-144
Loviisa 1	7											
15.11.88-5.5.89		23.0	890	1.4	0	40	120	19	30	630	2700	40
5.5.89-13.6.89		11.2	690	0	0	16	0	0	0	510	2200	0
13.6.89-1.9.89		41.9	830	1.5	0	21	74	10	35	470	2300	0
9.1.90-3.5.90		18.7	690	2.3	0	28	17	17	18	300	1700	0
3.5.90-13.6.90		2.7	630	0	0	9.3	0	0	0	230	1400	0
13.6.90-31.8.90		27.4	770	0	0	17	0	16	0	290	1900	0
31.8.90-6.11.90		38.1	820	0	0	18	0	23	0	280	1900	0
SEDIMENT (O-5 cm) 20.6.90			760	0	0	13	42	5	17	320	2000	0
Loviisa 2	11											
15.11.88-5.5.89		15.5	830	4.7	0	100	140	58	35	700	2900	0
5.5.89-15.6.89		5.9	630	0	0	21	50	10	21	450	2100	0
15.6.89-1.9.89		10.8	750	7.0	6	47	100	22	31	480	2300	0
1.9.89-19.12.89		37.6	770	0	0	40	66	36	25	510	2600	0
19.12.89-3.5.90		17.8	740	22	0	220	41	84	22	360	2100	0
3.5.90-13.6.90		5.0	580	0	0	22	0	7.1	17	200	1300	0
13.6.90-31.8.90		7.9	730	0	12	58	0	68	35	250	1600	0
31.8.90-6.11.90		21.7	730	5.9	0	39	42	36	20	300	2000	15
SEDIMENT (O-5 cm) 19.6.90			840	0	0	44	73	9	33	420	2600	0

0 = below the detection limit

Table 6.6.1. continued.

	Sampling depth m	Dry weight g	K-40	Mn-54	co-60	Ru-106	Ag-110m	Sb-125	cs-134	cs-137
Loviisa 3	16									
15.11.88-5.5.89		20.4	860	7.9	78	120	24	47	710	3000
5.5.89-14.6.89		8.1	480	0	12	0	0	0	450	2000
14.6.89-1.9.89		14.7	720	0	26	95	15	34	470	2300
1.9.89-19.12.89		26.3	830	0	34	70	32	40	500	2500
19.12.89-3.5.90		15.1	740	16	190	43	80	17	360	2000
3.5.90-13.6.90		13.7	520	20	48	0	12	15	210	1300
13.6.90-31.8.90		10.5	640	0	68	0	25	20	270	1700
31.8.90-6.11.90		25.1	870	0	33	39	40	27	280	1900
SEDIMENT (O-5 cm) 28.6.90			710	0	42	69	12	42	580	3500
Loviisa R1	12									
15.11.88-4.5.89		25.5	990	0	0	60	0	20	560	2400
4.5.89-13.6.89		21.0	930	0	0	0	0	0	430	2000
13.6.89-1.9.89		111.4	950	0	0	55	0	26	420	2000
1.9.89-9.1.90		54.1	920	0	1	29	2	17	340	1900
9.1.90-3.5.90		23.8	860	0	1.8	0	0	0	310	1800
3.5.90-13.6.90		51.5	1100	0	0	0	0	0	230	1500
13.6.90-31.8.90		73.7	920	0	0	0	0	0	240	1600
31.8.90-6.11.90		75.7	950	0	0	0	0	12	220	1600
SEDIMENT (O-5) 15.6.90			830	0	0	27	0	13	290	1800

0 = below the detection limit

6.7. Inventories in Sediments

Salo et al. (1986) estimated the total amounts of Sr-90, Cs-137 and Pu-239,240 bound to bottom sediments of the Baltic Sea at the beginning of the 1980s. They used two different methods in the calculations. Firstly, the estimated contents of radionuclides (Bq m^{-2}) in soft and hard sediments from various sea areas were multiplied by the areas of soft and hard bottoms (Table 6.3.1). The second method was based on the estimation of approximate mean concentrations of radionuclides in sinking matter and assumed sedimentation rates in different areas of the Baltic Sea.

Since very few data are available on radionuclide concentrations in sinking matter and the use of concentrations observed in the surface sediment layer may be doubtful for this purpose, we used only the first method to estimate the total amounts of Cs-137 and Pu-239,240 in the bottom sediments after the Chernobyl accident.

By using exactly the same method as Salo et al. (1986), we estimated that the total amount of Cs-137 in Baltic Sea sediments was 1 400 TBq in 1990-1991 and that of Pu-239,240 was 18 TBq in 1987-1988. The corresponding values estimated by Salo et al. at the beginning of the 1980s were 277 and 15 TBq, respectively, and show that the Chernobyl fallout increased the total amount of Cs-137 by a factor of 5, whereas the increase of Pu-239,240 was negligible. By using the ratio 1/20 for Cs-137 values on hard/soft bottoms, instead of 1/5 (p. 73), the total amount of Cs-137 was estimated at 1 200 TBq.

We were not able to estimate the total amount of Sr-90, because very few data on strontium in sediments have been reported to the database since 1986. However, it is well known that the proportion of Sr-90 was very small in the Chernobyl fallout. Salo et al. (1986) estimated that the total amount of Sr-90 bound to the bottom sediments of the Baltic Sea was 12 TBq at the beginning of the 1980s.

It should be kept in mind that the calculations presented above are very approximate. The uneven distribution of the Chernobyl fallout causes an additional difficulty in present-day calculations. The global fallout during the 1960s was again more or less evenly deposited in the northern hemisphere.

The preliminary inventories of Cs-137 in Baltic Sea sediments made earlier by the MORS Group resulted in 520-l 466 TBq during 1986-1987. However, at least the higher value may be overestimated, because the calculation was based only on soft-bottom values and the decreasing effect of hard bottoms was not taken into account.

Bojanowski et al. (1993) estimated that the total inventory of Cs-137 in sediments of the Polish Economic Zone was 11 TBq in 1985 and 44 TBq in 1993. This area forms about 8% of the total area of the Baltic Sea. The estimation is in good agreement with our total inventory if we take into account that the Chernobyl fallout was clearly lower in the area surrounding the southern Baltic Proper than in the areas surrounding the Bothnian Sea and the Gulf of Finland (Figure 2.1.1).

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7. RADIONUCLIDES IN BIOTA

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7.1. Introduction

Studies on the transfer of radionuclides between organisms as well as between organisms and their non-living environment are the basis to prepare dose assessments for human exposure to radionuclides due to consumption of different food items. Also, these studies help to get a more profound knowledge of the element transport within ecosystems.

It is well known that organisms form food chains in nature. These are seldom of a simple form, but are linked in very complicated food webs.

In general many radionuclides have a tendency to be enriched while being transferred upwards in the food web, i.e. from primary producers at the lower level of an ecosystem to the consumers (predators). Man is one of the top consumers in marine food webs.

There are many differences between radionuclides concerning their ability to accumulate in aquatic organisms. E.g. the caesium isotopes have a tendency to be enriched in fish flesh, whereas strontium mainly accumulates in bones.

The brackish water character of the Baltic Sea results in special radioecological properties. The salinity of the surface water decreases from around 25 ‰ in the Kattegat to almost fresh water in the inner parts of the Gulfs of Finland and Bothnia. The biota of the Baltic Sea consists of marine and fresh water organisms. The number of original endemic species is very small. The size of many marine organisms decreases considerably when going from higher to lower salinity. At the same time the decreasing salinity results in an increasing accumulation of some radionuclides.

The organisms collected by the different laboratories can be divided into three categories:

1) Organisms directly of importance for human consumption. This category includes a number of species of fish, crab, crayfish and mussels.

2) Organisms of importance in food webs ultimately leading to human consumption. To this category belong species serving as prey, including many kinds of fish and of benthic invertebrates. Other important species to be included here are plankton organisms and macro algae.

3) Organisms useful as bioindicators for radionuclides. Here we have to consider in the first place *Fucus vesiculosus* (bladder wrack), which has proved to be an excellent indicator especially used in monitoring programs at nuclear power plants in the different

areas. Also, many kinds of mussels, e.g. *Macoma baltica* and *Mytilus edulis* might be considered as good indicators, especially for radionuclides in suspended particulate matter.

The areas from where various species of fish were collected are indicated in Figure 3.1 .5. Sampling stations for algae and benthic invertebrates are given in Figure 3.1.4.

7.1.1. Some Important **Biota** in the Baltic Sea

Below a short description is given about the economical and ecological importance of some major species studied within MORS.

Herring is economically the most important fish species in the world. Its main distribution area is the northern part of the Atlantic Ocean and the southern part of the Arctic Ocean. The Baltic herring is a slow-growing and smaller form of herring and has the same economic importance in the Baltic Sea as herring in the oceans. Its importance is greatest in the northern areas of the Baltic Sea. The annual catch of herring in the Baltic Sea is about 400,000 tons and i.e. in Finland the share of herring in the total catch is about 80%. In the Northern Baltic the full-grown herring is only 16 to 19 cm long, but the size increases with increasing salinity. The nutriment consists mainly of planktonic crustaceans.

Cod is the second most important commercial fish species in the world. Its main distribution area ranges from New Foundland to the Lofoten Islands and the French coast. Compared with other important fish species of the Baltic Sea cod requires the most marine conditions. Thus it appears dwarfed in the Baltic Sea and is missing from the innermost parts of the Gulf of Bothnia and Gulf of Finland. In the northern parts of the Baltic Sea the appearance of cod depends on fluctuations of water salinity. The spawning does not succeed north of Gotland, because the salinity should be at least 10 to 12 ‰. During the period 1984 to 1991 the annual catch of cod in the Baltic Sea decreased from 440,000 tons (exceptionally high) to 125,000 tons. *Saduria entomon* forms a marked share of cod's nutriment in the northern Baltic Sea.

Flounder, plaice and dab have the main distribution areas in the northern Atlantic region, as well. From these species flounder has best adapted to the brackish water conditions of the Baltic Sea. It is missing only from the innermost parts of the Gulf of Bothnia and Gulf of Finland, but has significant economical importance only in the southern Baltic Sea. Plaice and dab have a more marine distribution. The annual catch of flounder in the Baltic Sea is about 10,000 tons. Molluscs and crustaceans are main constituents in the nutriment of these three species.

Pike, perch and roach are originally fresh water species, but are able to live in the coastal areas of almost the whole Baltic Sea, at least in shallow bays with low salinity. The premises of spawning are weakening with increasing salinity. Especially pike has economical importance in the Archipelago Sea and the coastal areas of the Gulf of Finland and Gulf of Bothnia. The annual catch of pike from the coastal areas of Finland is about 1,000 to 2,000 tons. Roach is an important prey for full-grown pike and perch. The nutriment of roach consists of small aquatic animals, sometimes merely plants. The young pike and perch eat

small crustaceans and insect larvas. Pike is known as a very local fish living in a small area all its life.

The mussels Mytilus *edulis* and ***Macoma baltica*** live in the North Atlantic region and the Baltic Sea. In the Baltic Sea the size of the mussels decreases with decreasing salinity. ***Macoma*** stands low salinities better and is missing only in the northernmost part of the Bothnian Bay. The distribution of ***Mytilus*** ranges up to the middle parts of the Gulf of Bothnia and Gulf of Finland. In these areas the mussels are very small (l-2 cm). *Mytilus* filters nutriments from water, ***Macoma*** also from the surface layers of sediments. Both species are important prey, especially for flounder. The economical importance of *Mytilus* is restricted in the southernmost areas of the Baltic Sea.

Saduria ***entomon*** is originally an inhabitant of the Arctic Ocean, but lives as a glacial relict in the Baltic Sea and its gulfs down to Bornholm. In the Bothnian Sea it is together with ***Monoporeia affinis*** *the* only constituent of benthic fauna in wide areas. *Saduria* eats dead fish and other organic material existing in the bottom sediments. The full-grown animal is 5-6 cm long. ***Saduria*** is an important prey organism especially for cod, eel and perch.

The bladder-wrack *Fucus* ***vesiculosus*** is a tall brown alga growing in the littoral zones of the Baltic Sea and most of the northern North Atlantic region. *Fucus* communities form a significant part of the ecosystems, especially in the northern Baltic Sea area, where the littoral vegetation in general is poor in species owing to the low salinity of water. *Fucus* belts provide shelter and source of nourishment for fish of different stadia and for other aquatic organisms. The value of ***Fucus*** for human use is negligible. The distribution of *Fucus* corresponds to that of ***Mytilus***. Similarly, the size of the plant decreases with decreasing salinity. In the southern Baltic Sea the thalli can be 70-80 cm high, but only 10 cm in the Quark (Gulf of Bothnia).

7.1.2. **Species Investigated**

The species sampled are listed below. The total number of measured samples (including various tissues from the same organism) was 1138.

A. Invertebrates

	Number of samples:		
	Animal whole	Shells	Soft parts
Arctica islandica		*1*	*5*
Asterias rubens	*3*		
Cardium ***edule***		*2*	*5*
Crangon crangon	*22*		
Macoma baltica	*20*	*2*	*2*
Mya *arenaria*		1	*2*
Mytilus edulis	15	13	46
Saduria entomon	*25*		

B. Algae (whole plant)

	Number of samples:
Fucus vesiculosus	80
Laminaria saccharina	2

C. Fish

	Number of samples:							
	Fish whole	Fish whole without head and entrails	Flesh	Skin	Bones	Entrails	Liver	ovary
Abrumis brama (Bream)			9		6	6		
Clupea hurengus (Herring)	4	40	110		33	10	2	2
Cyprinus carpio (Carp)			3		2	2		
Enchelyopus cimbrius (Four-bearded rockling)	1		1					
Esox lucius (Pike)			44		5	5		
Gadus morhua (Cod)	1		195	14	23	2	101	
Limanda limanda (Dab)			13		3	2		
Merlangius merlangus (whiting)			5					
Myoxocephalus scorpius (Bull rout)			1		1			
Osmerus eperlanus (Smelt)			1		1			
Perca fluviatilis (Perch)			15		11	11		
Platichthys flesus (Flounder)			29		13	2		
Pleuronectes platessa (Plaice)			37		21			
Psettu maxima (Turbot)			1					
Rutilus rutilus (Roach)			11		10	10		
Spruttus sprattus (Sprat)	10		7		4			
Stizostedion lucioperca (Perch-pike)			7		6	6		

D. Others (whole sample)

	Number of samples:
Fish larvae	1
Fry	1
Plankton	24

7.1.3. Sampling Methods

Generally, fish samples were collected by the use of smaller nets, demersal fish by bottom trawls and by utilising commercial fish catches.

For benthic animals usually one of the following sampling methods was used. *Saduria entomon* was caught with bait-nets, e.g. baited with commercial herring, whereas *Mytilus edulis* was collected by divers. For the sampling of *Macoma baltica* Ekman-Birge grabs, and coarse sieves as well as specially designed trawls were used.

Aquatic plants, especially *Fucus vesiculosus* were sampled by diving or collected from the water surface near to the shore.

7.1.4. Radionuclides Studied

Totally, 35 different radionuclides were determined in the biota samples. Also, a number of beta-measurements on Cs-134 + Cs-137 were reported.

The total number of determinations of various nuclides was 3930, including those of Cs-134 + Cs-137. The number of values for radionuclides detected in the four most common organisms can be seen in Table 7.1.1. It may be noted that the number of nuclides detected in *Fucus vesiculosus* is higher than in others. This is mainly due to the fact that most *Fucus* samples were collected at coastal sites at the Archipelago Sea, the Åland Sea, the Gulf of Finland and the Bothnian Sea near to Swedish or Finnish nuclear power plants where discharge of nuclides from the plants is monitored by *Fucus*. Also, these sites received the highest Chernobyl fallout deposition within the Baltic region, thus resulting in an increase of nuclides originating from that accident.

For studying trends in concentrations of nuclides in biota the sub-areas of the Baltic Sea shown in Figure 1.1 (Chapter 1) were used.

Only for a few nuclides of regional occurrence there were enough data to make graphical presentation meaningful, namely Cs-137, Cs-134, Sr-90, Co-60, Ru-106 and Ag-110m. Cs-134 could generally not be detected in the samples before Chernobyl. After that accident its concentration showed an almost constant fraction of that of Cs-137 (after physical decay correction back to the time of the accident). For this reason, the behaviour of Cs-134 in the samples during the period 1984 to 1991 will not be discussed further.

The concentrations of various nuclides in the samples are shown in Figures 7.2.1 to 7.4.1. All of them have been detected in varying quantities in global fallout, normal discharges from the nuclear power and reprocessing plants as well as from the Chernobyl accident. It should be remembered that many sampling sites are situated in the close vicinity of nuclear power plants (marked on Figure 1.1, Chapter 1) from which small amounts of these nuclides are always discharged, also under normal conditions. If data of samples from not more than one or two stations within a Baltic Sea subarea were used, this is indicated in the head line of each single plot within Figures 7.2.1 to 7.3.3 by adding an abbreviated station code (in brackets) to the subarea name.

Table 7.1.1. Number of values for radionuclides detected in the four most common organisms within the MORS biota data 1984 to 1991.

Species	part	Ac-228	Ag-108m	Ag-110m	Ba-140	Be-7	Bi-214	Ce-141	Ce-144
Clupea harengus	fillets	0	7	12	0	0	0	0	0
Fucus vesiculosus	whole	5	1	28	1	13	3	3	5
Gadus morhua	fillets	0	8	10	0	1	0	0	0
Gadus morhua	liver	0	31	100	0	0	0	0	0
		co-57	Co-58	Co-60	cs-134	Cs-134 +137	cs-137	1-131	K-40
Clupea harengus	fillets	0	0	10	*96*	*0*	*108*	*0*	*98*
Fucus vesiculosus	whole	5	29	69	60	0	83	1	44
Gadus morhua	fillets	0	0	11	176	5	191	0	174
Gadus morhua	liver	0	0	26	101	0	101	0	101
		La-140	Mn-54	Nb-95	Pb-212	Pb-214	Pu-238	Pu-239, 240	Ra-224
Clupea harengus	fillets	0	8	1	0	0	0	4	0
Fucus vesiculosus	whole	2	37	5	1	5	2	3	3
Gadus morhua	fillets	0	2	0	0	0	1	7	0
Gadus morhua	liver	0	0	0	0	0	0	1	0
		Ra-226	Ra-228	Ru-103	Ru-106	Sb-124	Sb-125	Sr-89	Sr-90
Clupea harengus	fillets	0	0	0	2	0	0	0	26
Fucus vesiculosus	whole	9	1	6	10	2	10	2	22
Gadus morhua	fillets	2	0	0	1	0	0	1	94
Gadus morhua	liver	0	0	0	0	0	0	0	8
		Tc-99	Te-129m	Tl-228	Zn-65	Zr-95			
Clupea harengus	fillets	0	0	0	0	1			
Fucus vesiculosus	whole	21	2	1	23	3			
Gadus morhua	fillets	0	0	0	1	0			
Gadus morhua	liver	0	0	0	0	0			

At many sampling locations, especially those representing subareas where the main fallout from Chernobyl occurred, only one sample was reported every year. This might lead to doubts concerning the significance of the obtained radioactivity values for a single sample. Certainly in many cases it would be very desirable to have access to a greater number of analyses. However, it should also be kept in mind that most of the measurements, especially from the subareas just mentioned, were made on pooled samples from a number of specimen, the minimum number being generally 10 for Baltic herring, 3 to 5 for pike and cod and around 100 for most invertebrates. In the case of bladder wrack (*Fucus vesiculosus*) a sample is usually made from several plants. Furthermore, it should be kept in mind that in many cases

the values reported to the database are representative for the whole discharge areas of nuclear power plants, where several samples are taken annually, but not reported to the database.

7.2. Fiih

In the following some comments will be made on the occurrence and time-trends for Cs-137 and for some other nuclides.

7.2.1. Cs-137 in Fish

The level of Cs-137 in fish samples at specific sites is largely explained by the varying amounts of initial fallout from Chernobyl in different regions of the Baltic Sea area (Figure 5.2.2, Chapter 5). Thus, the pattern varies considerably if one compares subareas with a high fallout level with those with a lower level. Also, the slow counter clockwise circulation of surface water in the Gulf of Bothnia, as well as in the Baltic Proper and the net southward water transport along the Swedish coast, have a great importance for the time-trends shown by samples from some subareas.

Subareas with a relatively high level of fallout from Chernobyl are the Bothnian Sea, the Gulf of Finland and the Northern Baltic, the Åland and the Archipelago Seas. These subareas are all characterised by maximum Cs-137 levels for most species in 1986 or 1987, followed by a monotonous decrease during the following years. This pattern is recognised e.g. for Baltic herring from the Northern Baltic Proper (Figure 7.2.2g) and cod from the Bothnian Sea (Figure 7.2.1a) as well as in herring from the Bothnian Sea (Figure 7.2.2c) and the Gulf of Finland (Figure 7.2.2f).

The trends for Cs-137 activities in pike from the Archipelago Sea, the Bothnian Sea, and the Gulf of Finland seem to develop differently compared to cod and Baltic herring, showing a more slow increase after Chernobyl and reaching a maximum only in 1988 to 1990 (Figures 7.2.4a, 7.2.4b and 7.2.4d). This observation shows up clearly when comparing pike from the Vaasa region (Bothnian Sea, Figure 7.2.4b) with herring from the same area (Figure 7.2.2c). The trend is interpreted as due to the position of pike as a top predator, thus reaching a maximum for Cs-137 later by consuming fish having reached its maximum of radiocaesium earlier.

Herring from the Bothnian Bay (Figure 7.2.2b) shows a slower increase in caesium levels reaching a maximum around the years 1989-1990. This seems to be the result of riverine inflow of caesium from drainage areas as well as of a slow transport of caesium in more contaminated waters from the south.

In the subareas Gotland East and Southern Baltic Proper the Chernobyl deposition was lower than within the Archipelago Sea and adjacent subareas. A distinct maximum is not observed as a function of time (Figures 7.2. lb, 7.2.2h and 7.2. lc).

Figure 7.2.1. Activities of Cs-137 in cod from different Baltic Sea areas.

Fig. 7.2.1.a: Cs-137 in **GADUS** MORHUA (fillets)
Bothnlan Sea (SWF 22)

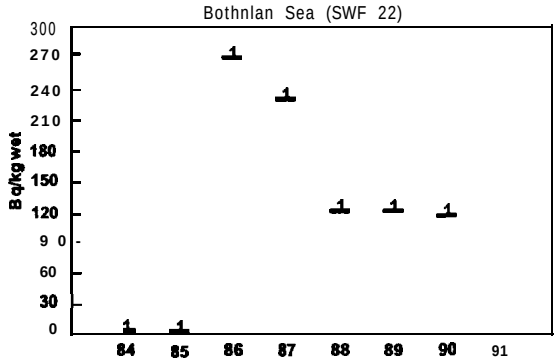


Fig. 7.2.1.b: **Cs-137** in **GADUS** MORHUA (fillets)
Gotland East

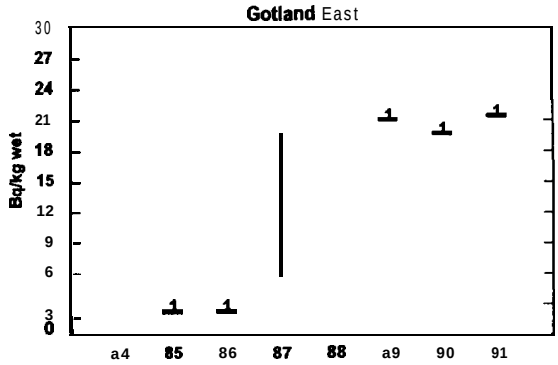


Fig. **7.2.1.c:** Cs-137 in **GADUS** MORHUA (fillets)
Southern Baltic Proper

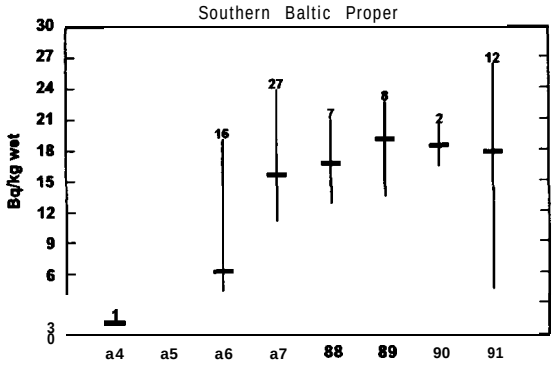
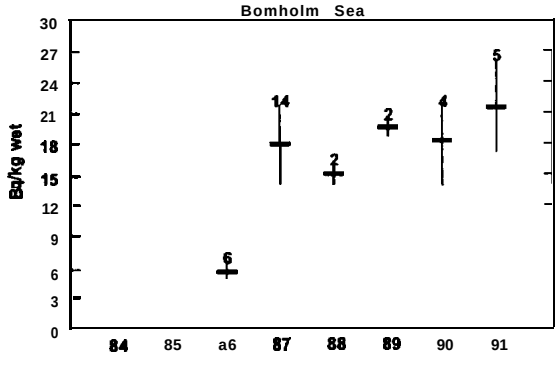


Fig. 7.2.1.d: Cs-137 in **GADUS** MORHUA (fillets)
Bomholm Sea



Fia. 7.2.1.e: Cs-137 in **GADUS** MORHUA **(fillets)**
Arkona Sea

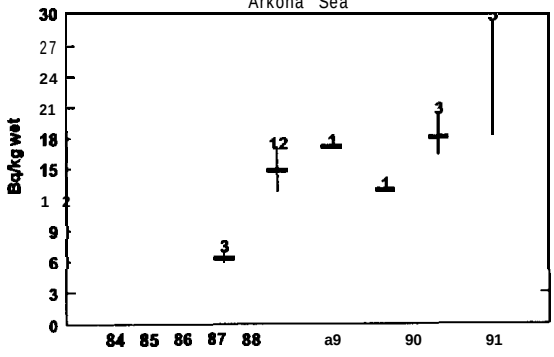


Fig. 7.2.1.f: Cs-137 in **GADUS MORHUA (fillets)**
Belt Sea

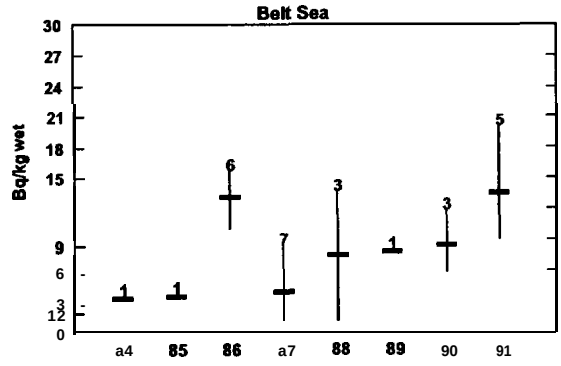
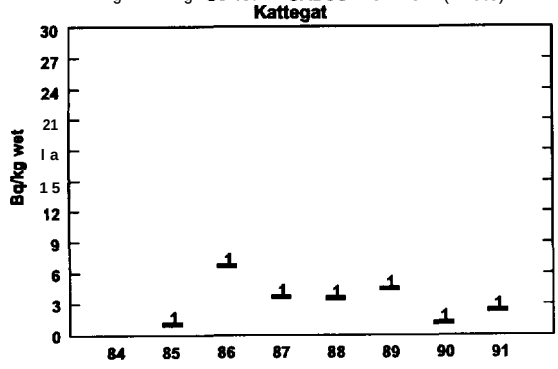


Fig. 7.2.1.g: **Cs-137** In **GADUS** MORHUA (fillets)
Kattegat



Data shown for each year:

arithmetic mean with min-max range and the number of measurements above the min-max range

Figure 7.2.2. Activities of Cs-137 in Baltic herring from different Baltic Sea areas. (continued on next page)

Fig. 7.2.2.a: **Cs-137** in CLUPEA HARENGUS (fillets)
Bothnian Bay (SW **1)**

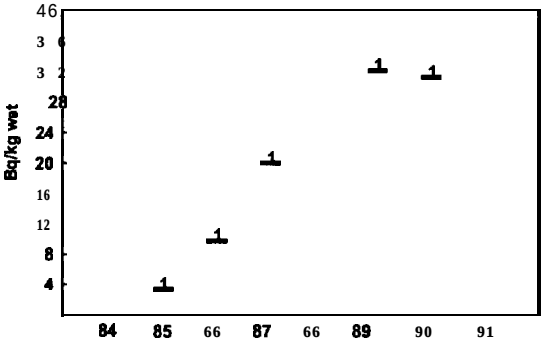


Fig. 7.2.2.b: **Cs-137** in CLUPEA HARENGUS (edible parts)
Bothnlan Bay (HAILUOTO)

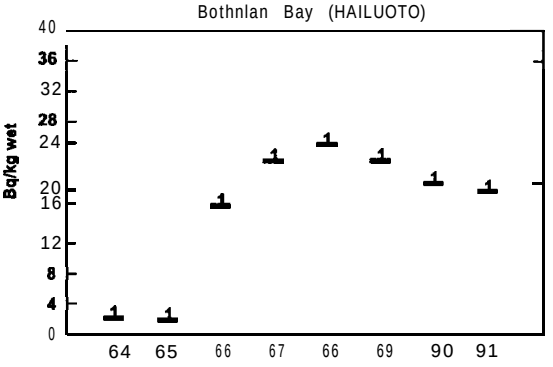


Fig. **7.2.2.c:** Cs-137 in CLUPEA HARENGUS (edible parts)
Bothnian Sea **(VAASA)**

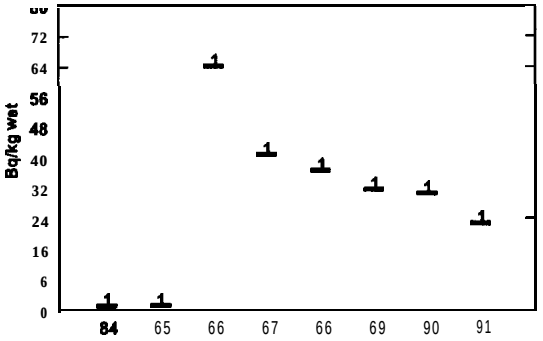


Fig. 7.2.2.d: Cs-137 **in** CLUPEA HARENGUS (fillets)
Bothnian Sea **(SW2)**

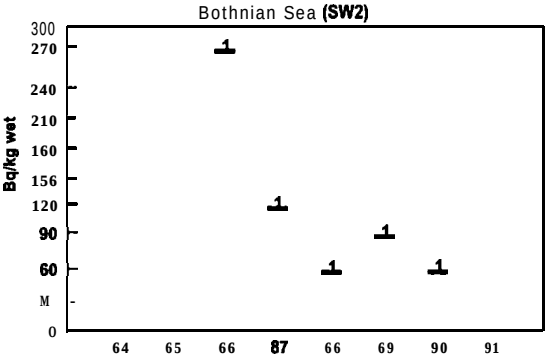


Fig. 7.2.2.e: **Cs-137** in CLUPEA HARENGUS (edible part)
Archipelago Sea **(SEILI)**

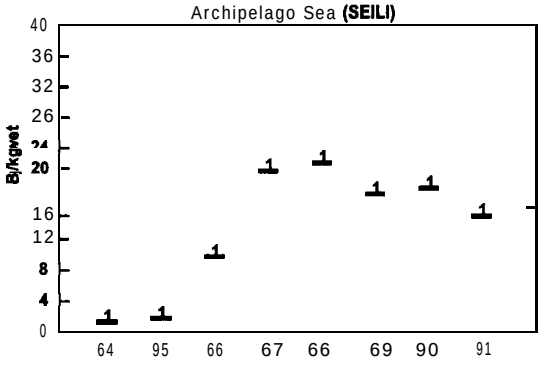


Fig. 7.2.2.f: Cs-137 in CLUPEA HARENGUS (edible parts)
Golf of Finland (LOVIISA + TVAERMINNE)

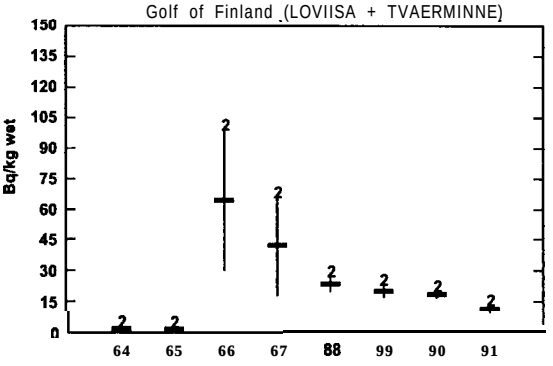


Fig. 7.2.2.g: Cs-137 in CLUPEA HARENGUS **(fillet)**
Northern **Baltic** Proper (SW 3)

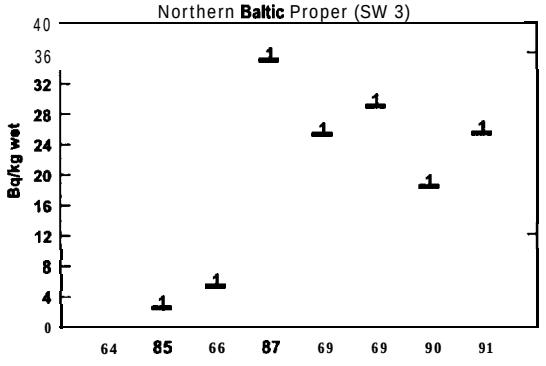


Fig. 7.2.2.h: Cs-137 in CLUPEA HARENGUS (fillet)
Southern Baltic Proper

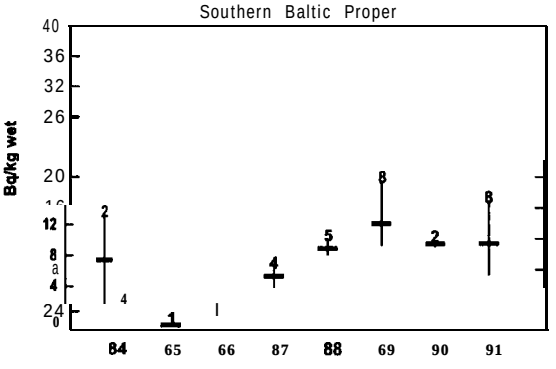


Figure 7.2.2. Activities of Cs-137 in Baltic herring from different Baltic Sea areas. (continued)

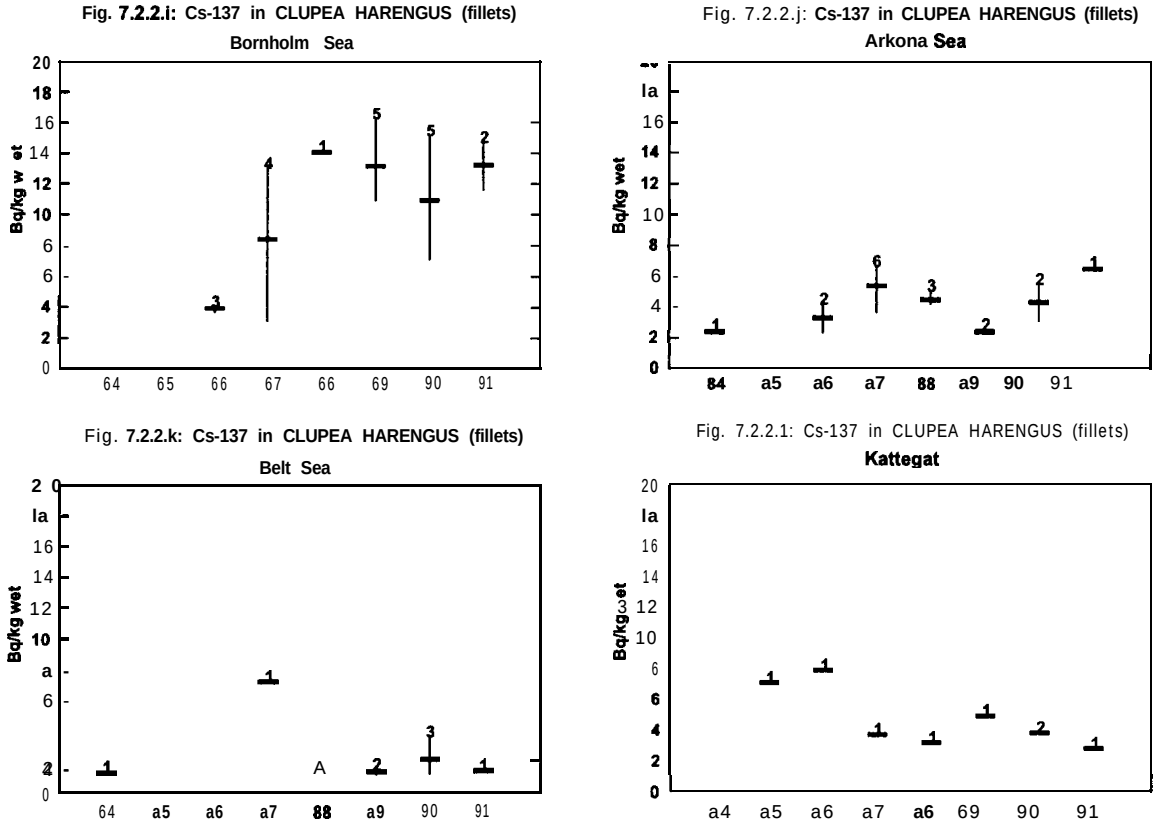
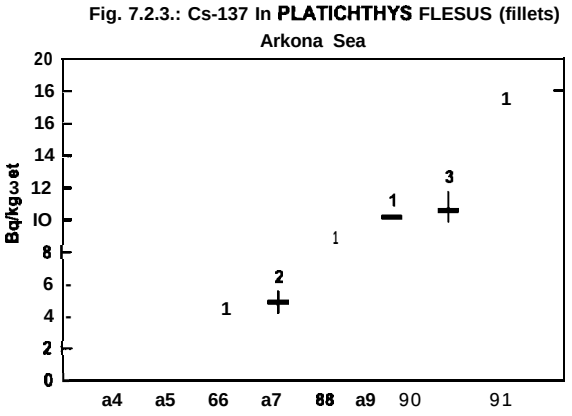
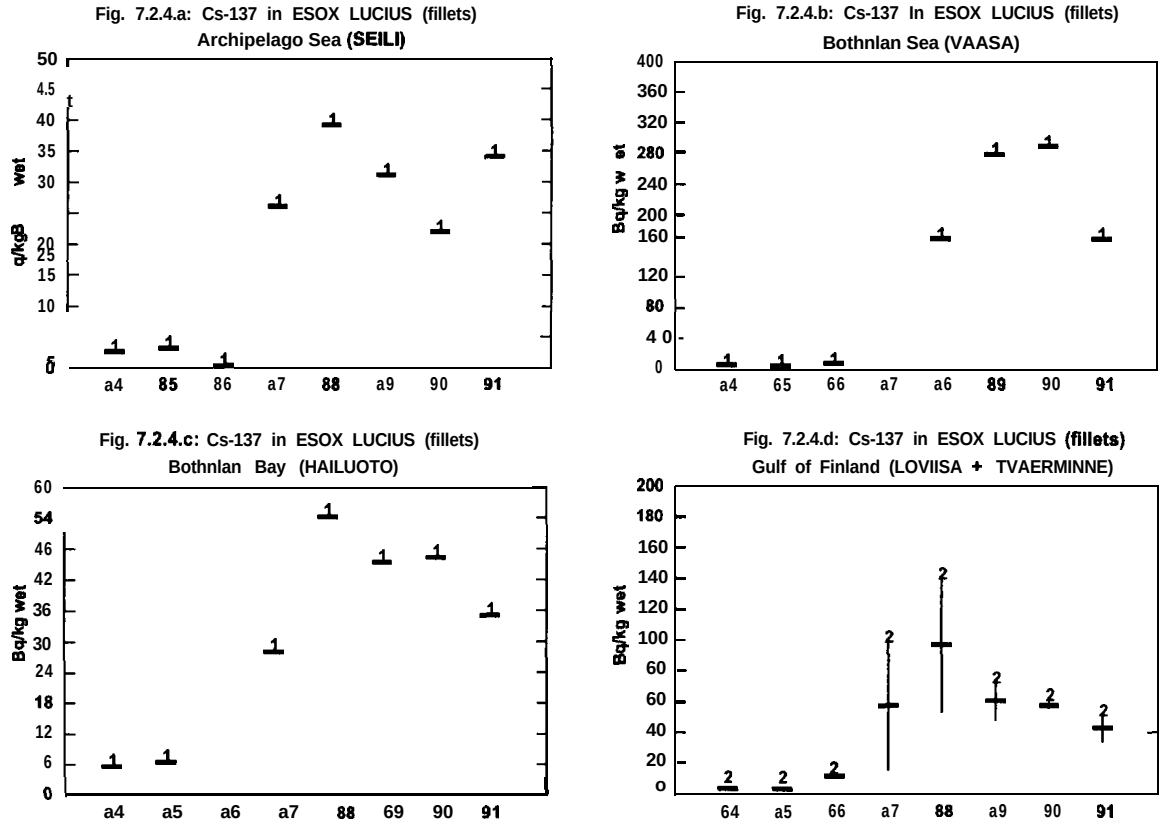


Figure 7.2.3. Activity of Cs-137 in flounder from the Arkona Sea.



The most southern subareas (the Bornholm Sea and west of it) are influenced by the counter clockwise movements of surface water in the Baltic Sea area and the outflow to the North Sea via the Danish straits. This results in a southward directed stream along the Swedish east coast transporting Cs-137 from the areas of initial maximum of Chernobyl disposal to the southern subareas. For this reason the Cs-137 level in fish samples from these subareas generally shows an increasing trend in the period after 1986, the values for that year still being rather low. This is illustrated by Figures 7.2.2j, 7.2. le, 7.2.3 and 7.2. 1f.

Figure 7.2.4. Activities of Cs-137 in pike from different Baltic Sea areas.



In the Kattegat the levels of Cs-137 are, on the average, much lower than in the subareas discussed earlier. In this region a relatively low direct fallout from Chernobyl resulted in an increase during 1986 followed by a decrease thereafter (Figures 7.2.2.1 and 7.2. lg).

7.2.2. **Other Radionuclides in Fish**

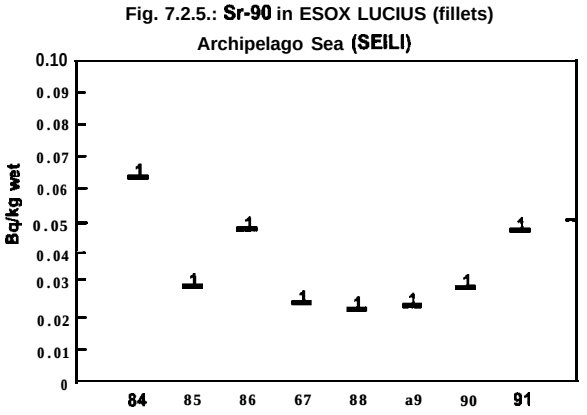
Activity concentrations of Sr-90 in fish do not show any pronounced temporal trends. Indications of a Chernobyl influence, at least in 1986, are in most cases not apparent or only very weak. Figure 7.2.5 gives one representative example of a temporal development with a small enhancement in 1986. Annual averages for three species from different subareas have been used to estimate overall Sr-90 mean values for the period 1984 to 1991. These values as well as minima and maxima are given in Table 7.2.1.

The Sr-90 activities in cod fillets appeared to be lower than in edible parts of Baltic herring, as well as in fillets of pike, by an order of magnitude. For herring, this results from the fact that Sr-90 is enriched mainly in bones and skin. The pike lives in waters with low salinity, for which the concentration factor for Sr-90 in flesh is assumed to be higher than in marine fish.

Table 7.2.1. Sr-90 activities (Bq kg^{-1} wet weight) in fish representing the period 1984 to 1991 in different Baltic Sea subareas

Subarea	fish species	tissue	mean	minimum	maximum
			Bq kg-'	Bq kg-'	Bq kg-'
Belt Sea	Cod	fillets	0.004	0.002	0.005
Bomholm Sea	Cod	fillets	0.005	0.004	0.005
Bothnian Bay	Herring	edible part	0.110	0.084	0.160
Bothnian Sea	Herring	edible part	0.078	0.057	0.120
Archipelago Sea	Herring	edible part	0.073	0.046	0.150
Gulf of Finland	Herring	edible part	0.090	0.038	0.230
Bothnian Bay	Pike	fillets	0.063	0.036	0.250
Bothnian Sea	Pike	fillets	0.051	0.034	0.083
Archipelago Sea	Pike	fillets	0.035	0.022	0.064
Gulf of Finland	Pike	fillets	0.036	0.017	0.059

Figure 7.2.5. Activity of Sr-90 in pike (Archipelago Sea).



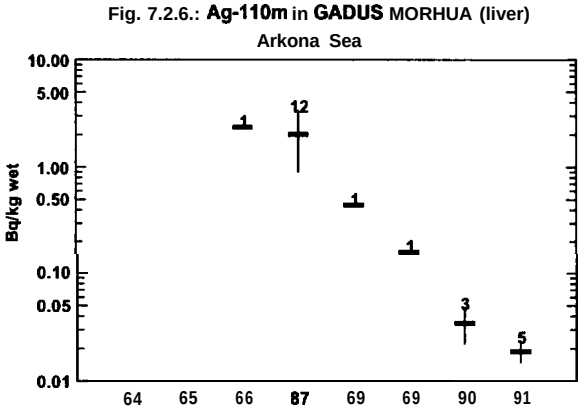
Most of the activity concentrations for plutonium isotopes in fish fillets (mainly cod and herring) were below the detection limit. Although this limit could be lowered substantially 1990 due to methodological improvements, nearly all values of Pu-239,240 obtained since 1990 were also below it. Therefore, it is concluded that the activity content of Pu-239,240 in fish flesh is at least lower than 50 $\mu Bq\,kg^{-1}$ wet weight.

Five Pu analyses of sprat samples (whole fish) from the Belt Sea, the Arkona Sea and the Bornholm Sea, which were collected in 1990 and 1991 yielded Pu-239,240 values between 0.031 and 0.72 mBq kg-' wet weight with a mean of 0.34 mBq kg-' wet weight. These values are at least ten-fold higher than those in cod fillets which can be explained by a higher Pu enrichment in bone and skin compared to that in fillets. The Pu-238 values were below the

detection limit. Two activity ratios of Pu-238/Pu-239,240 were less than 0.050. This indicates global fallout to be the main source for Pu in sprat.

After the Chernobyl accident gamma measurements of cod liver samples revealed activity contents of Ag-110m which were unexpectedly high. By long-term measurements the long-lived silver isotope Ag-108m (half-live 433 years) could also be detected. Over the years 1987 - 1991 an approximately constant and very low Ag-108m activity of about 0.006 Bq kg^{-1} wet weight was estimated. In each of four investigated subareas of the south-western part of the Baltic Sea a regular decrease of the Ag-110m activity was characterised by an effective half-live of about 0.60 years (15% lower than the physical half-live). This value is common to the four areas. Figure 7.2.6 shows the results for one of these areas. Obviously, Ag-110m originated from Chernobyl. The source of Ag-108m is, however, not clearly identified at present (HELCOM, 1989). Meanwhile, both isotopes have been detected also in North Sea cod liver but with different isotope activity ratios (Kanisch and Nagel, 1991).

Figure 7.2.6. Activity of Ag-110m in cod liver (Arkona Sea).



7.3. Invertebrates

Many invertebrates for which time-series were available are included in the regular monitoring programs at the nuclear power plants. At those sites we frequently find elevated levels of nuclides which generally are major constituents in the discharge from the plants. These are mainly activation products like Co-60, Zn-65, Mn-54, Ag-110m and others. Various fission products in small quantities are generally also found.

Samples of invertebrates were regularly collected in the environment of the nuclear power plants at Barsebäck, Forsmark, Greifswald, Loviisa, Olkiluoto, Oskarshamn and Ringhals. The position of these power plants is seen in Figure 1.1.

7.3.1. Cs-137 in Invertebrates

Samples of the mussel *Macoma baltica* (whole animal) collected every year at Forsmark and Olkiluoto (Figures 7.3.2a and 7.3.2b) showed a radiocaesium content which changed with

time analogous to the results from fish. It should be noted that the data on invertebrates are reported on a dry weight basis, whereas fish data are given on a wet weight basis.

A similar analogy to fish is found for Cs-137 in the common mussel *Mytilus edulis* at Forsmark in the Bothnian Sea (Figure 7.3. la) and for the soft bottom isopode *Saduria entomon* at Loviisa in the Gulf of Finland (Figure 7.3.3a).

7.3.2. Other Radionuclides in Invertebrates

For Sr-90 there was not found any temporal trend in the samples. Certainly this nuclide originates mainly from global fallout. Figures 7.3.2c and 7.3.3b illustrate this for *Macoma baltica* from the Bothnian Sea and for *Saduria entomon* from the Gulf of Finland, respectively.

At the nuclear power plant near Loviisa samples of *Saduria entomon* were regularly collected. It has been shown that this species is an efficient accumulator of Ag-110m. A maximum was found in the year after Chernobyl (Figure 7.3.3d). During a cruise with R/V Gauss (FRG) in October/November 1986 Agnedal (1988) collected, inter alia, *Saduria entomon* samples showing significant activity values of Ag-110m. A value of 61 Bq kg^{-1} dry weight for a sample from the Gulf of Finland agrees very well with that shown in Figure 7.3.3d. In agreement with the distribution pattern of the Chernobyl fallout Agnedal found the highest value of 620 Bq kg-' dry weight in a sample from the south western part of the Bothnian Sea.

At sites close to four of the nuclear power plants mussels rather regularly showed small activities of Co-60. Results for *Mytilus edulis* from the Forsmark area are shown in Figure 7.3.1d and those for the Kattegat in Figure 7.3.1e. In the case of *Macoma baltica* and for *Saduria entomon* analogous results are seen in Figures 7.3.2d, 7.3.2e, 7.3.2f and 7.3.3c. Most probably the Co-60 in these samples was of local origin.

The results of a few plutonium analyses of different invertebrate samples are shown in Table 7.3.1. Apart from 4 samples the whole organisms were analysed. Table 7.3.1 includes ratios of wet weight per dry weight to allow conversion of the activities to the wet weight base. Pu-238 could not be detected in these samples.

Figure 7.3.1. Activities of some radionuclides in blue mussels from different Baltic Sea areas.

F5.7.3.1.a: Co-137 in MYTILUS EDULIS (whole body)
Bothnian Sea **(SWF11** 1)

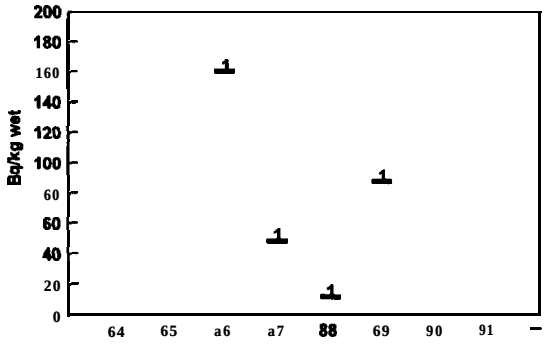


Fig. 7.3.1.b: Cs-137 in MYTILUS EDULIS (soft parts)
Bomholm Sea

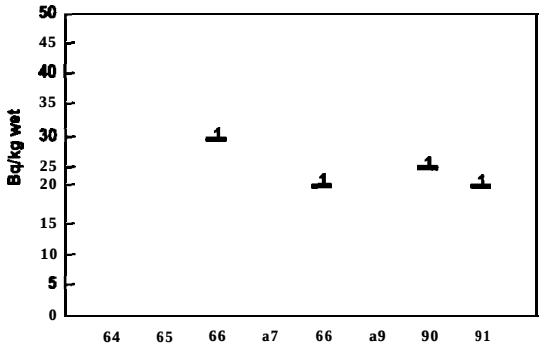


Fig. **7.3.1.c:** Cs-137 in **MYTILUS** EDULIS (soft parts)
Kattegat

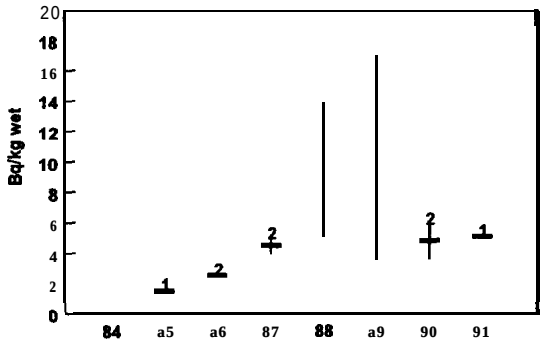


Fig. 7.3.1.d: Co-60 in MYTILUS EDULIS (whole body)
Forsmark Area **(SWF1** 11)

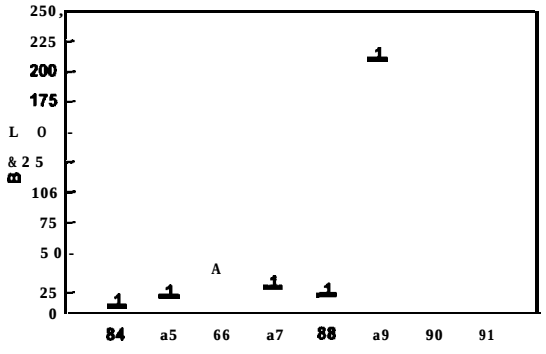
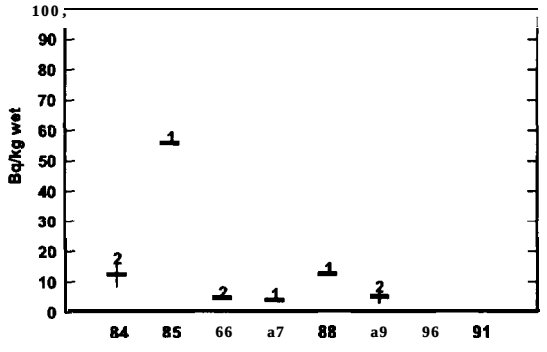


Fig. 7.3.1.e: Co-60 in MYTILUS **EDULIS** (soft parts)
Kattegat



Fii 7.3.2. Activities of some radionuclides in ***Macoma baltica*** from different Baltic Sea areas.

Fig. 7.3.2.a: Cs-137 in **MACOMA BALTICA** (whole body) Bothnian Sea **(FORSMARK)**

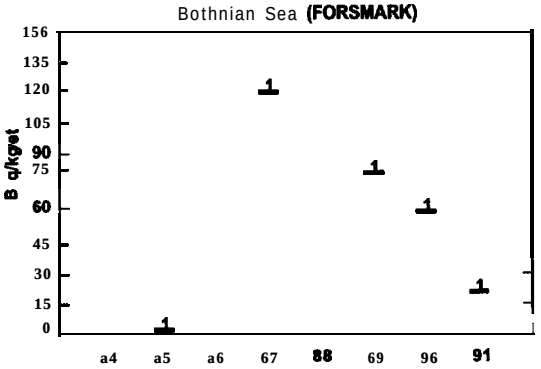


Fig. 7.3.2.b: Cs-137 in **MACOMA BALTICA** (whole body) Bothnian Sea **(OLKILUOTO)**

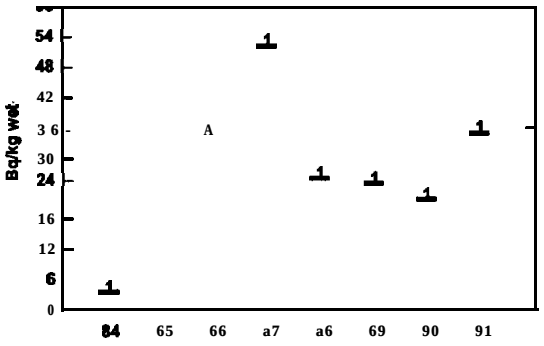


Fig. **7.3.2.c:** Sr-90 in **MACOMA BALTICA** (whole body) Bothnian **Sea** (OLKILUOTO)

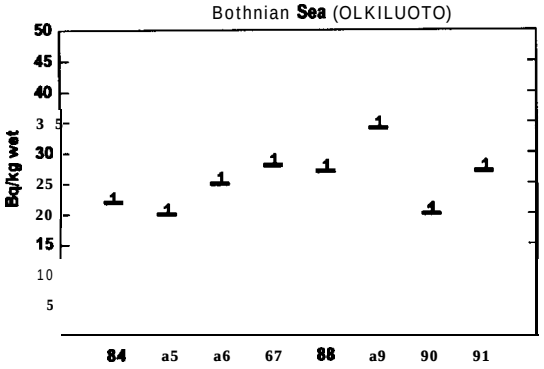


Fig. 7.3.2.d: **Co-60** in **MACOMA BALTICA** (whole body) Bothnian **Sea** (OLKILUOTO)

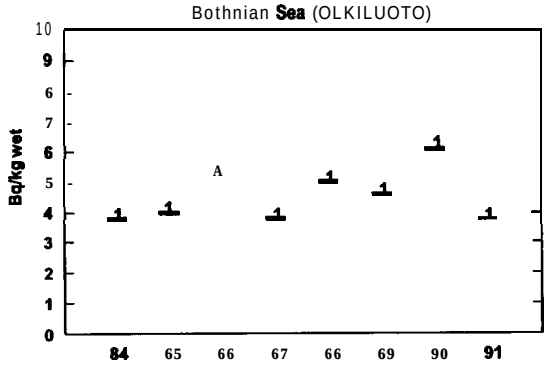


Fig. 7.3.2.e: Co-60 in **MACOMA BALTICA** (whole body) Bothnlan Sea (FORSMARK)

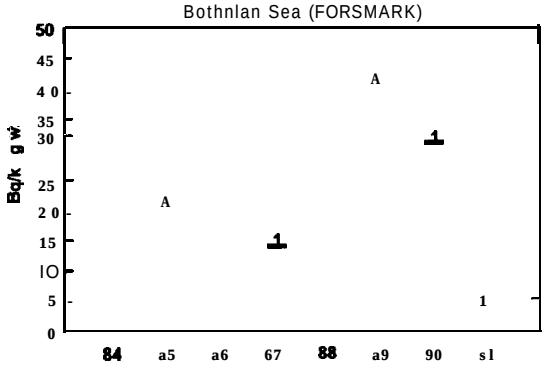
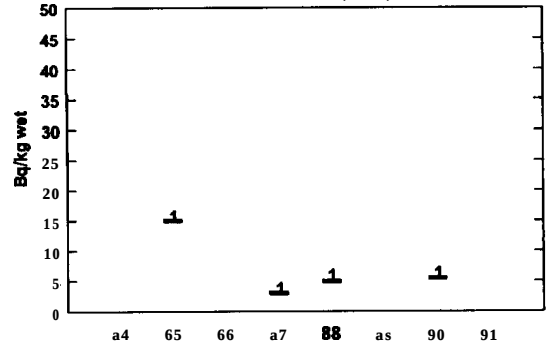
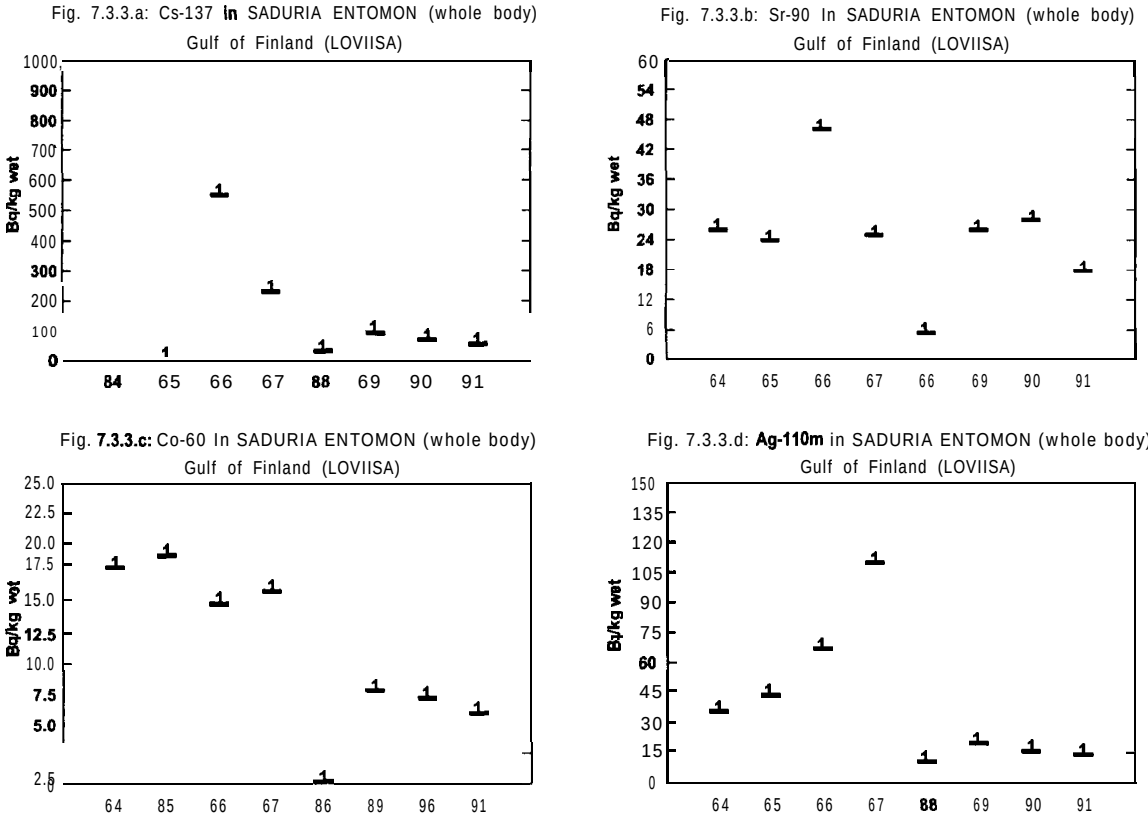


Fig. 7.3.2.f: Co-60 in **MACOMA BALTICA** (whole body) **Gotland** West (OKG)



These data can be compared with results obtained by investigations within the Polish Economic Zone after the Chernobyl accident (Skwarzec and Bojanowski, 1992). Their **Pu**-239,240 mean of 51 **mBq kg^{-1}** dry weight for ***Saduria entomon*** confirms very well our mean of 55 **mBq** kg-'. The Pu-239,240 activities in soft parts of ***Mytilus edulis,*** given in Table **7.3.1,** are within the lower part of the range given by Skwarzec and Bojanowski. However, their mean of about 92 **mBq kg^{-1}** dry weight was nearly four times higher than that of the two samples of soft parts in Table 7.3.1. The only result for ***Macoma baltica***, measured here as whole animals, is lower than their **mean** of 49 **mBq kg^{-1}** dry weight (range **31**- 86 **mBq** kg-').

Figure 7.3.3. Activities of some radionuclides in *Saduria entomon* from the Loviisa area (Gulf of Finland).



Some of the activity ratios of Pu-238/Pu-239,240 from Table 7.3.1 are lower than 0.06 or even 0.04. Neglecting the other “less than values” being significantly higher, we conclude that the Pu activity ratios seem to be consistent with values of about 0.04 being representative for the global fallout (Pentreath, 1988). Skwarzec and Bojanowski (1992) found for their samples from 1986 to 1988 slightly higher activity ratios (between 0.08 and 0.125) which they attributed to an influence by the Chernobyl deposition.

Table 7.3.1. Results for plutonium isotopes (mBq kg^{-1} dry weight, single measurements) in invertebrates from various subareas

Subarea	year	tissue	WD [a]	F'u-239,240 mBq kg" dw	Pu-238/ Pu-239,240
Saduria entomon:					
Southern Baltic Proper	1989	whole	2.9	110	< 0.072
	1989	whole	4.0	25	< 0.10
	1989	whole	4.3	39	< 0.059
	1989	whole	2.9	30	< 0.19
	1990	whole	5.9	41	< 0.078
Gulf of Finland	1989	whole	4.8	84	—
Mytilus edulis:					
Arkona Sea	1989	soft part	7.7	31	< 0.035
Belt Sea	1991	soft part	10.	19	< 0.12
	1991	whole	3.2	30	< 0.11
Macoma baltica:					
Bothnian Sea	1989	whole	2.9	14	---
Arctica islandica:					
Belt Sea	1988	soft part	11.	76	< 0.16
Arkona Sea	1991	soft part	11.	36	< 0.028
Asterias rubens:					
Belt Sea	1989	whole	4.5	22	< 0.18
	1990	whole	3.8	28	< 0.054

[a] ratio wet per dry weight

7.4. Algae

The only species regularly sampled in the area was bladder wrack, ***Fucus vesiculosus***. A relatively large number of samples was collected especially in the close environs of the Swedish and Finnish power plants. A list of the nuclides most frequently detected in ***Fucus*** is found in Table 7.1.1.

The concentration of these nuclides varies depending on the distance and direction of the sampling place in relation to the point of discharge. Some models have rather successfully described this relation, which gives a possibility to use ***Fucus*** as a semi-quantitative indicator for the discharge (Dahlgaard and Boelskifte, **1992).**

The amounts of some radionuclides in the samples were also strongly related to the deposition of the Chernobyl fallout at the different sampling sites, generally being highest at Forsmark and Olkiluoto at the Bothnian Sea. This is well illustrated in Figures 7.4. la, 7.4. lb and 7.4. lc showing Cs-137, Ag-110m and Ru-106 at the different sites.

The results of few plutonium analyses of aquatic plants are shown in Table 7.4.1. Two significant activity ratios Pu-238/Pu-239,240 of *Fucus* are, considering their measurement uncertainties, consistent with global fallout, whereas one ratio of *Laminaria saccharina (0.19)* could indicate an influence of North Sea derived plutonium.

Table 7.4.1. Results for plutonium isotopes (mBq kg^{-1} dry weight, single measurements) in aquatic plants from the Belt Sea

Subarea	year	tissue	WD [a]	Pu-239,240 mBq kg-' dw	Pu-238/ Pu-239,240
Fucus vesiculosus:					
Belt Sea	1988	whole	5.3	190	0.050
	1989	whole	7.7	300	0.057
	1990	whole	6.3	31	< 0.15
Laminaria saccharina:					
Belt Sea	1987	whole	10.	63	0.19
	1988	whole	7.1	31	< 0.077

[a] ratio wet per dry weight

Figure 7.4.1. Some radionuclides in *Fucus vesiculosus*.

Fig. 7.4.1 .a: Cs-137 in Fucus vesiculosus

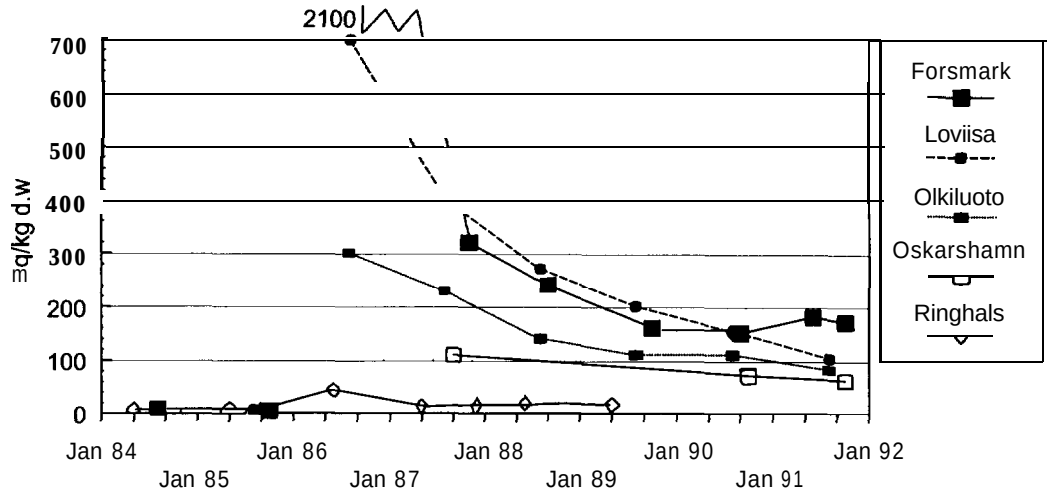


Fig. 7.4.1.b: Ag-110m in Fucus vesiculosus

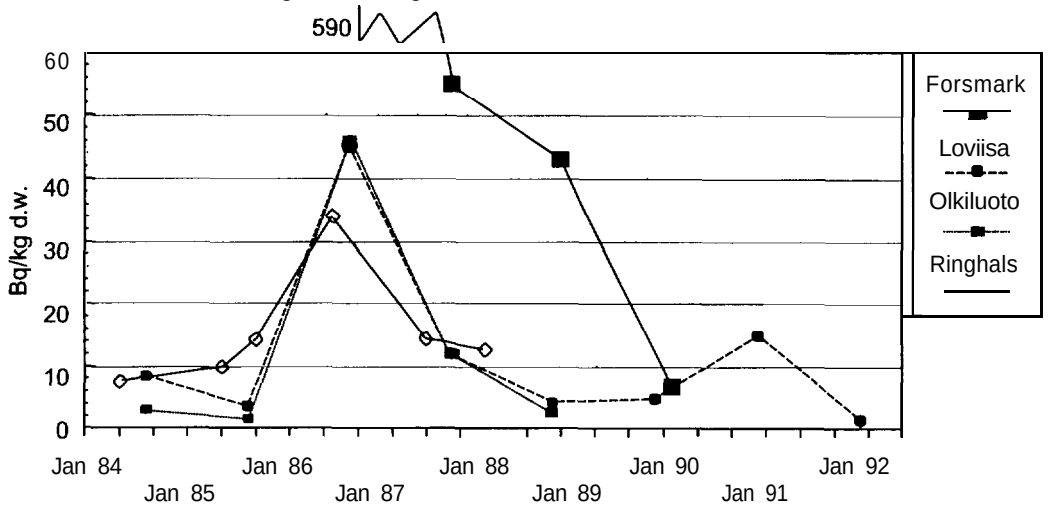
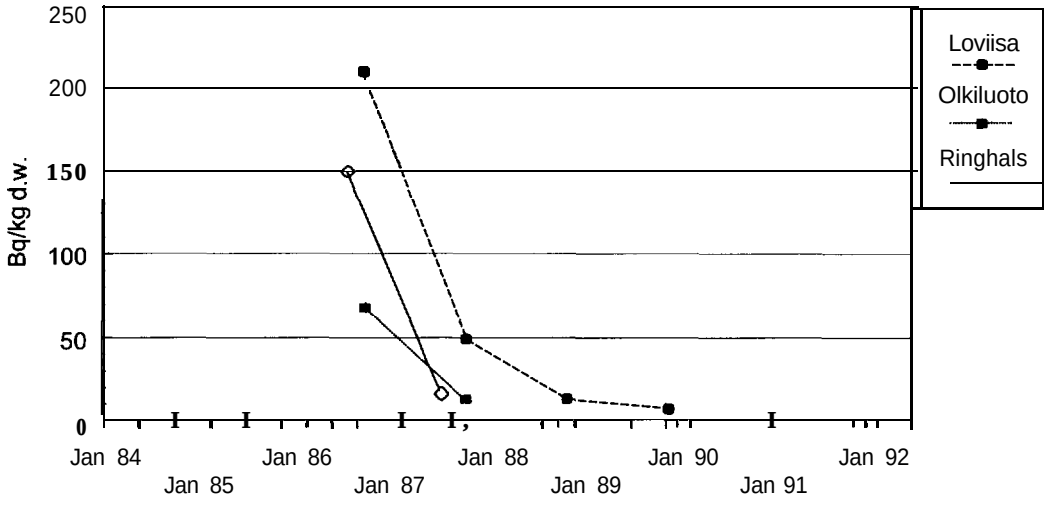


Fig. 7.4.1.c: Ru-106 in Fucus vesiculosus



7.5. Concentration Factors

The discussion of concentration factors for the radionuclides Cs-137 and Sr-90 is confined to the main marine fish, Baltic herring, cod, plaice and flounder, and the fresh water fish pike.

For the different areas of the Baltic Sea average annual concentrations of the two radionuclides in unfiltered seawater from the years 1988 to 1991 were calculated. In general, the averaging was performed using available concentrations from the whole water column. For practically all fish samples considered for the calculation of concentration factors the sampling depths were less than 80 m. Only for very few cod samples sampling depths of more than 80 m were reported. Therefore, the maximum water depth for averaging the radionuclide concentrations in water was restricted to not more than 80 m. Due to incomplete mixing after the Chernobyl deposition, especially the Cs-137 concentrations were lower in deep water than in the surface layer. This implies that an averaging over the whole water column in the three subareas 4, 6 and 9 (Southern Baltic Proper, Bornholm Sea and Gotland East, respectively) would not have been correct.

Usually, the concentration factors of biota are based on filtered seawater concentrations. For Cs-137 and Sr-90 the small error introduced by the use of unfiltered seawater is negligible for the Baltic Sea conditions. The concentration factors were obtained by division of annual means of the radionuclides in fish (Bq kg-' wet weight) by the annual average concentrations of the same radionuclides in water (Bq 1"). The results obtained are presented in the Tables 7.5.1 to 7.5.6. Besides statistical parameters for the concentration factors, the corresponding water concentrations of each box are characterised by mean values which were averaged over the years 1988 to 1991 (4 years in total) as far as measured values were available. The standard deviation of this mean serves as a measure of the variation of the annual averages of the seawater concentrations within the 4 years. It should be emphasised, however, that the concentration factors were calculated from the individual annual averages of the seawater concentrations. Graphical presentations of the concentration factors are given in Figure 7.5.1 for different Baltic Sea areas, which have been ordered by going from west to east and then to north, thereby following the decreasing salinity in the Baltic Sea.

Undoubtedly, the increase of the concentration factors observed by going from west to east and then to north (see Figure 75.1) can be due to several factors, but first of all the well-documented increase when salinity decreases.

A mean Cs-137 concentration factor of 181 was obtained for cod in the areas 2, 6, 4 and 9 (southern part of the Baltic Sea) with a very low standard deviation of 3 between these areas (Table 7.5.1, Figure 7.5. la). In the same areas the averaged Cs-137 concentration factor for fillets of Baltic herring (Table 7.5.2, Figure 7.5.1c) was about half of that of cod with a larger standard deviation. However, in the northern part of the Baltic Sea (Gulf of Bothnia and Gulf of Finland, areas 7, 8 and 11) the averaged concentration factor was 169 $\pm$ 19 ($\pm 1\sigma$ SD) in edible parts of Baltic herring (Table 7.5.3). In the southern part of the Baltic Sea (areas 2, 6 and 4) the averaged Cs-137 concentration factors were 122 $\pm$ 14 and 137 $\pm$ 18 for plaice and flounder, respectively (Tables 7.5.4 and 7.5.5, Figures 7.5. le and 7.5. 1f).

Figure 7.5.1. Concentration factors of Cs-137 and Sr-90 in fish from different Baltic Sea areas (see Figure 7.1.1 for the method of data presentation; number of years indicated above min-max range).

Fig. 7.5.1.a: **Cs-137** concentration factors of cod (fillets)

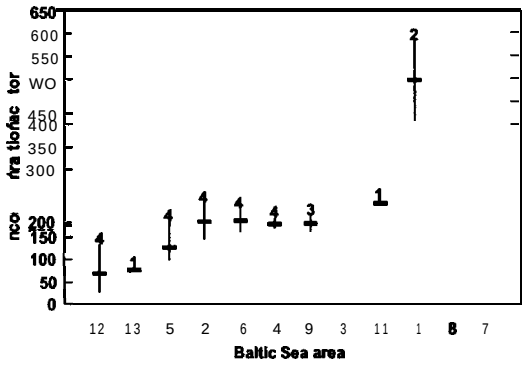


Fig. **7.5.1.b: Sr-90** concentration factors of cod (fillets)

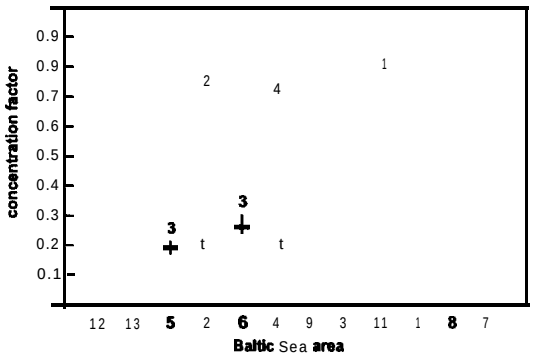


Fig. **7.5.1.c: Cs-137 concentration** factors of herring **(fillets)**

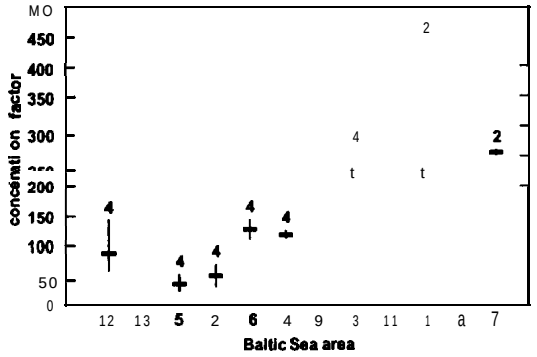


Fig. 7.5.1 **.d: Sr-90** concentration factors of herring (fillets)

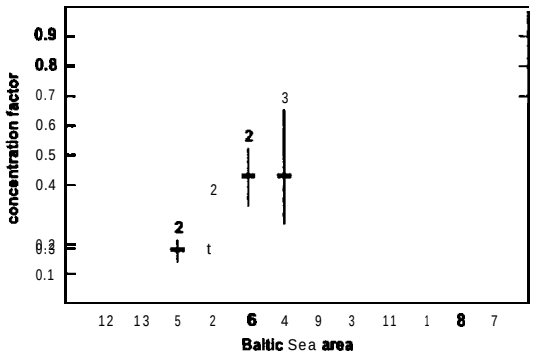


Fig. 7.5.1.e: **Cs-137** concentration factors of plaice (fillets)

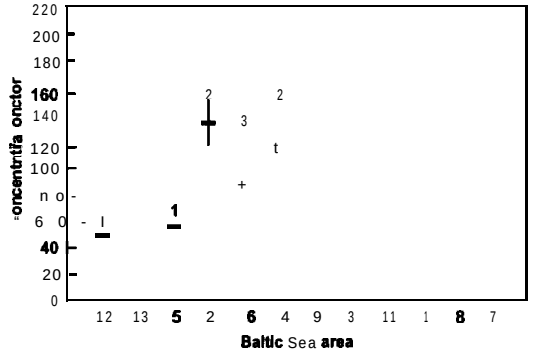


Fig. **7.5.1.f:** Cs-137 concentration factors of flounder (fillets)

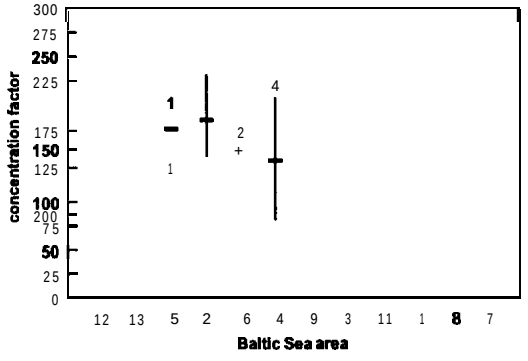


Fig. 7.5.1.g: **Cs-137** concentration factors of pike (fillets)

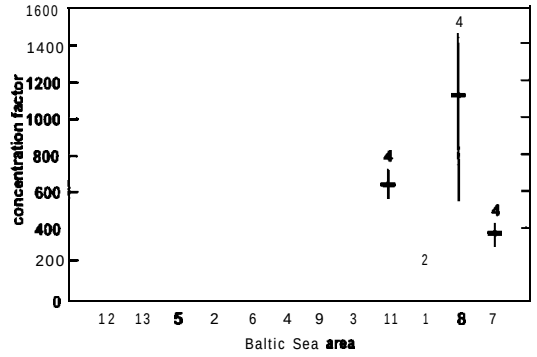
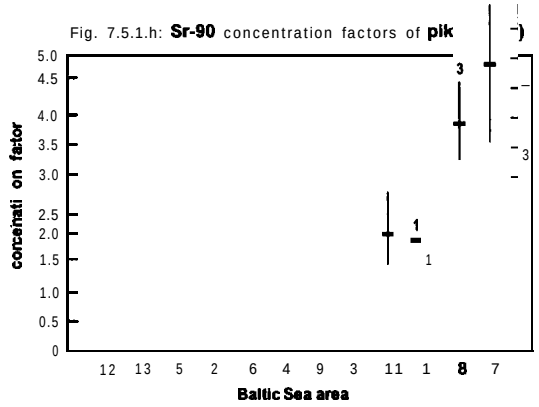


Fig. 7.5.1.h: **Sr-90** concentration factors of **pik**



Sr-90 concentration factors of cod fillets and herring fillets in the western and southern parts of the Baltic Sea were around 0.4 (Tables 75.1 and 7.5.2, Figures 7.5. lb and 7.5. 1d). The corresponding concentration factors measured in edible parts of Baltic herring from the northern parts are an order of magnitude higher, around 4 (Table 7.5.3).

High Cs-137 concentration factors were observed in fillets of pike (Table 7.5.6), which lives in waters with low salinity. In the Bothnian Sea values around 1100 were found. This is supported by results of Swedish investigations in the Biotest Basin (southern Swedish coast of the Bothnian Sea, near to the Forsmark power plant) after the Chernobyl accident. For adult pike steady state concentrations of Cs-137 resulted in a Cs-137 concentration factor of about 1000 (Evans, 1991). In fresh water systems this concentration factor was much higher. In Sweden Sundblad et al. (1991) investigated e.g. the Lake Hillesjön, which was heavily contaminated by Chernobyl fallout with about 100 $kBq\ m^{-2}$ Cs-137. Their results for pike suggest a Cs-137 concentration factor of about 4000.

The Sr-90 concentration factors in pike fillets were around 2 (Table 7.5.6).

The concentration factors for Cs-137 and Sr-90 determined in the Baltic Sea before the Chernobyl accident agree quite well with those presented here. Cs-137 concentration factors for cod fillet estimated by Grimås and Holm (1983) were as follows:

	cs-137
Gotland (area 9 and 10):	183
Öresund (area 13):	172
Smogen (area 12):	135

From investigations at the nuclear power plants of Olkiluoto and Loviisa in the areas 8 and 11 (Salo, 1980) the following concentration factors were estimated:

	cs-137	Sr-90
Herring (edible parts):	200	4
Cod (fillets):	200	1

Studies in the "Greifswalder Bodden" (Arkona Sea, area 2) before 1983 lead to a Cs-137 concentration factor of 170 for cod fillet (IAEA, 1986).

For marine fish an IAEA report (IAEA, 1985) gives a Cs-137 concentration factor of 100 (range 10-300). The Sr-90 concentration factor for the whole fish as far as it is edible is given as 2 (range 0.3-10), whereas the factor for fish flesh is stated to be less than 1.

For the central and southern parts of the North Sea (ICES areas IVB and IVC) Cs-137 concentration factors were derived among other species for fillets of cod and plaice (Steele, 1990). The mean concentration factors and the range of annual means between 1978 and 1985 were as follows:

	Mean	Range
Cod	92	66 to 120
Plaice	39	22 to 45

Our Cs-137 concentration factors of cod from the Kattegat and the Sound are similar to the mean North Sea value. For few samples of plaice from the Kattegat and the Belt Sea we found as to be expected only slightly higher concentration factors than that of 39 in the North Sea, however, they may be biased by not using local near-bottom Cs-137 concentrations in water in our calculations.

Table 7.51. Concentration factors of cod (*Gadus morhua*; fillets)

Subarea	number of years	mean water concentration	concentration factor Bq kg^{-1} per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
cs-137 :						
1 Åland/Archipel.	2	0.251 ± 0.063	496	407	582	124
2 Arkona Sea	4	0.096 ± 0.005	183	144	225	34
4 South. B. Proper	4	0.101 ± 0.008	178	169	190	9
5 Belt Sea	4	0.076 ± 0.003	125	97	184	40
6 Bornholm Sea	4	0.102 ± 0.003	185	161	209	20
9 Gotland East	3	0.117 ± 0.013	179	162	195	17
11 Gulf of Finland	1	0.135	223			
12 Kattegat	4	0.046 ± 0.009	70	28	133	45
13 Sound	1	0.094	76			
Sr-90 :						
2 Arkona Sea	2	0.017 ± 0.0001	0.46	0.21	0.71	0.35
4 South. B. Proper	4	0.020 ± 0.004	0.46	0.19	0.68	0.21
5 Belt Sea	3	0.016 ± 0.002	0.19	0.17	0.21	0.02
6 Bomholm Sea	3	0.016 ± 0.001	0.26	0.30 0.24		0.03
11 Gulf of Finland	1	0.02	0.76			

Table 7.5.2. Concentration factors in Baltic herring (*Clupea harengus*, fillets)

Subarea	number of years	mean water concentration	concentration factor Bq kg^{-1} per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
Cs-137 :						
1 Åland/Archipel.	2	0.251 ± 0.063	330	214	445	165
2 Arkona Sea	4	0.096 ± 0.005	46	28	65	15
3 North. B. Proper	4	0.101 ± 0.006	241	200	276	32
4 South. B. Proper	4	0.101 ± 0.008	117	112	124	5
5 Belt Sea	4	0.076 ± 0.003	32	20	48	12
6 Bornholm Sea	4	0.102 ± 0.003	126	110	142	13
7 Bothnian Bay	2	0.123 ± 0.001	256	252	260	5
12 Kattegat	4	0.046 ± 0.009	85	56	142	39
Sr-90 :						
2 Arkona Sea	2	0.017 ± 0.0001	0.26	0.17	0.34	0.12
4 South. B. Proper	3	0.021 ± 0.004	0.43	0.27	0.65	0.20
5 Belt Sea	2	0.016 ± 0.001	0.18	0.14	0.21	0.05
6 Bornholm Sea	2	0.020 ± 0.008	0.43	0.33	0.52	0.13

Table 7.5.3. Concentration factors in Baltic herring (*Clupea harengus*, edible parts)

Subarea	number of years	mean water concentration	concentration factor Bq kg^{-1} per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
Cs-137 :						
1 Åland/Archipel.	2	0.251 ± 0.063	81	75	87	9
7 Bothnian Bay	4	0.123 ± 0.001	176	161	193	17
8 Bothnian Sea	4	0.213 ± 0.055	147	127	166	17
11 Gulf of Finland	4	0.100 ± 0.025	183	149	207	27
Sr-90 :						
1 Åland/Archipel.	1	0.017	3.8			
7 Bothnian Bay	4	0.016 ± 0.002	6.8	5.3	7.8	1.0
8 Bothnian Sea	3	0.019 ± 0.001	3.2	3.0	3.5	0.3
11 Gulf of Finland	4	0.021 ± 0.001	2.4	1.8	3.1	0.6

Table 7.5.4. Concentration factors of plaice (***Pleuronectes platessa***, fillets)

Subarea	number of years	mean water concentration	concentration factor Bq kg^{-1} per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
Cs-137 :						
2 Arkona Sea	2	0.093± 0.005	133	117	150	23
4 South. B. Proper	2	0.107± 0.007	127	111	144	24
5 Belt Sea	1	0.073	56			
6 Bornholm Sea	3	0.103± 0.003	106	84	125	21
12 Kattegat	1	0.035	49			
Sr-90 :						
2 Arkona Sea	1	0.018	0.79			
6 Bornholm Sea	1	0.026	0.34			

Table 7.5.5. Concentration factors of flounder (***Platichthys flesus***, *fillets)*

Subarea	number of years	mean water concentration	concentration factor Bq kg-' per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
Cs-137 :						
2 Arkona Sea	4	0.096± 0.005	117	80	163	34
4 South. B. Proper	4	0.101± 0.008	142	81	206	51
5 Belt Sea	1	0.078	108			
6 Bornholm Sea	2	0.101 ± 0.002	153	148	158	7
sr-90 :						
2 Arkona Sea	3	0.017± 0.0001	0.39	0.25	0.6	0.19

Table 7.5.6. Concentration factors of pike (*Esox Lucius,* fillets)

Subarea	number of years	mean water concentration	concentration factor Bq kg^{-1} per Bq l^{-1}			
		Bq l^{-1} ± 1σSD	mean	minimum	maximum	1σSD
cs-137 :						
1 Åland/Archipel.	2	0.251 ± 0.063	141	132	150	13
7 Bothnian Bay	4	0.123 ± 0.005	366	297	418	51
8 Bothnian Sea	4	0.213 ± 0.055	1119	549	1450	429
11 Gulf of Finland	4	0.100 + 0.025	635	561	716	65
Sr-90 :						
1 Åland/Archipel.	1	0.017	1.3			
7 Bothnian Bay	4	0.016 ± 0.001	3.4	2.1	4.4	1.1
8 Bothnian Sea	3	0.019 ± 0.003	2.4	1.8	3.1	0.71
11 Gulf of Finland	4	0.021 ± 0.001	1.4	0.9	2.1	0.52

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8. MODELLING THE TRANSFER OF RADIONUCLIDES IN THE BALTIC SEA

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The assessment model used for the present investigation includes a description of the physical dispersion of radionuclides in the marine environment, a description of the transfer of radionuclides to biota and a calculation of doses to individuals and populations exposed to radionuclides in seafood. The dispersion model (Nielsen, 1994) covers the North-East Atlantic coastal waters including the Baltic Sea. The whole model rather than just the part covering the Baltic Sea is used, in order to take into account the transfer of radionuclides into the Baltic Sea from the two European nuclear reprocessing plants, Sellafield and La Hague. Due to limited time and resources for the present work, it was decided to restrict the assessment to cover only the radionuclides Cs-137 and Sr-90 and only the ingestion pathway. Other radionuclides and pathways contribute to the doses to man in the Baltic Sea, but it is expected that Cs-137 is the single dominating radionuclide and that the ingestion pathway is the single dominating pathway as was found from the CEC Marina Study (CEC, 1990). Furthermore, observed data for the two radionuclides selected (available from the HELCOM database described in Section 3.3) permit comparisons with model predictions to be made in most areas of the Baltic Sea, which allow quantitative estimates of model reliability to be made.

8.1. Description of Models

Compartmental or box-model analysis is used to simulate the movement of radionuclides between parts of the marine environment. Box-model analysis assumes instantaneous uniform mixing within each box with rates of transfer across the boundaries of the box being proportional to the inventories of material in the source boxes. The model includes water advection and mixing between adjacent boxes, sedimentation, bioturbation, and bioconcentration. The overall model is used to calculate doses to man from ingestion of seafood.

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

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

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

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

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

where V_i is the volume of water represented by box I. The present model was modified somewhat from the previous version (Nielsen, 1994) by adjusting the mixing conditions (water exchange rates) for some adjacent water boxes of the Baltic Sea while keeping the net advective water fluxes of the model unaltered. The modification was carried out by minimising the difference between the model predictions and observed levels of Cs-137 in the Baltic seawater.

Figure 8.1.1 shows the regions used in the marine box model near the Baltic Sea, and Figure 8.1.2 shows a diagrammetric representation of the positions, and interconnections among individual water boxes for the entire model. Each of the water compartments has associated suspended sediment and the water compartments having seabed boundaries have underlying sediment compartments. The water compartments have odd numbers and the surface sediment compartments have even numbers. The latter are not shown in Figure 8.1.2.

Figure 8.1.1. Regions around the Baltic Sea covered by the marine box model. The numbers of the water boxes are indicated.

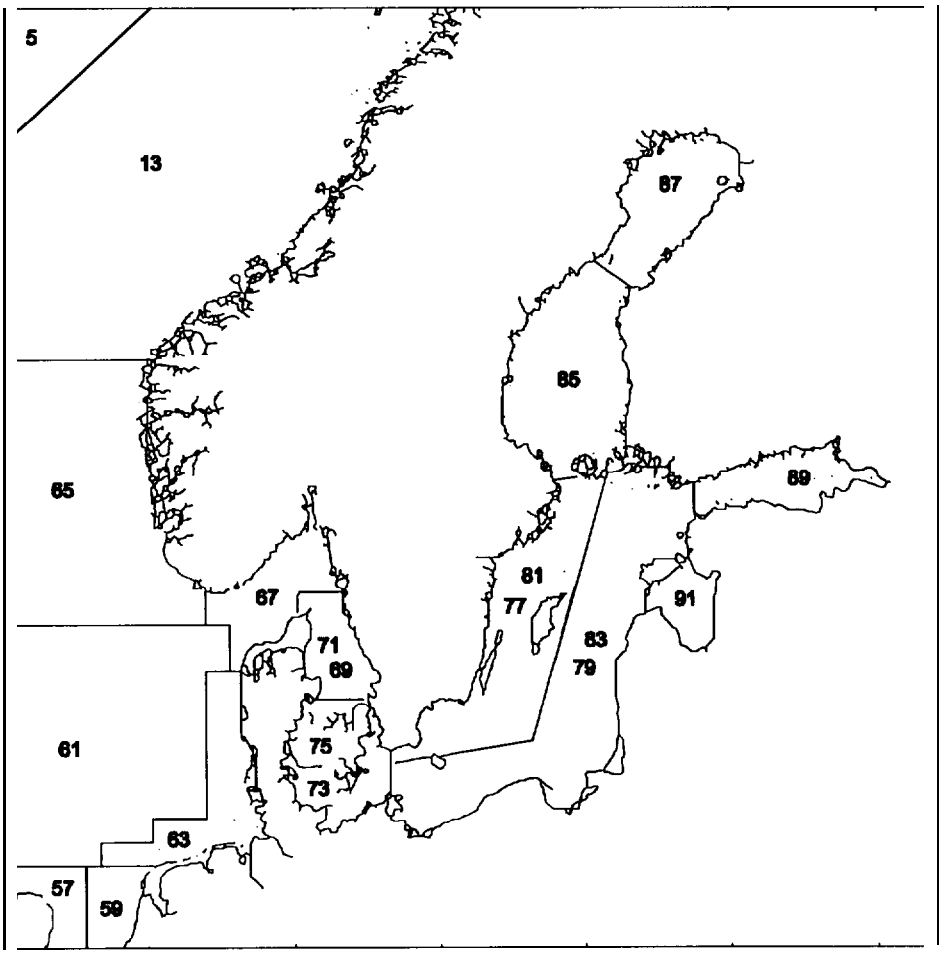
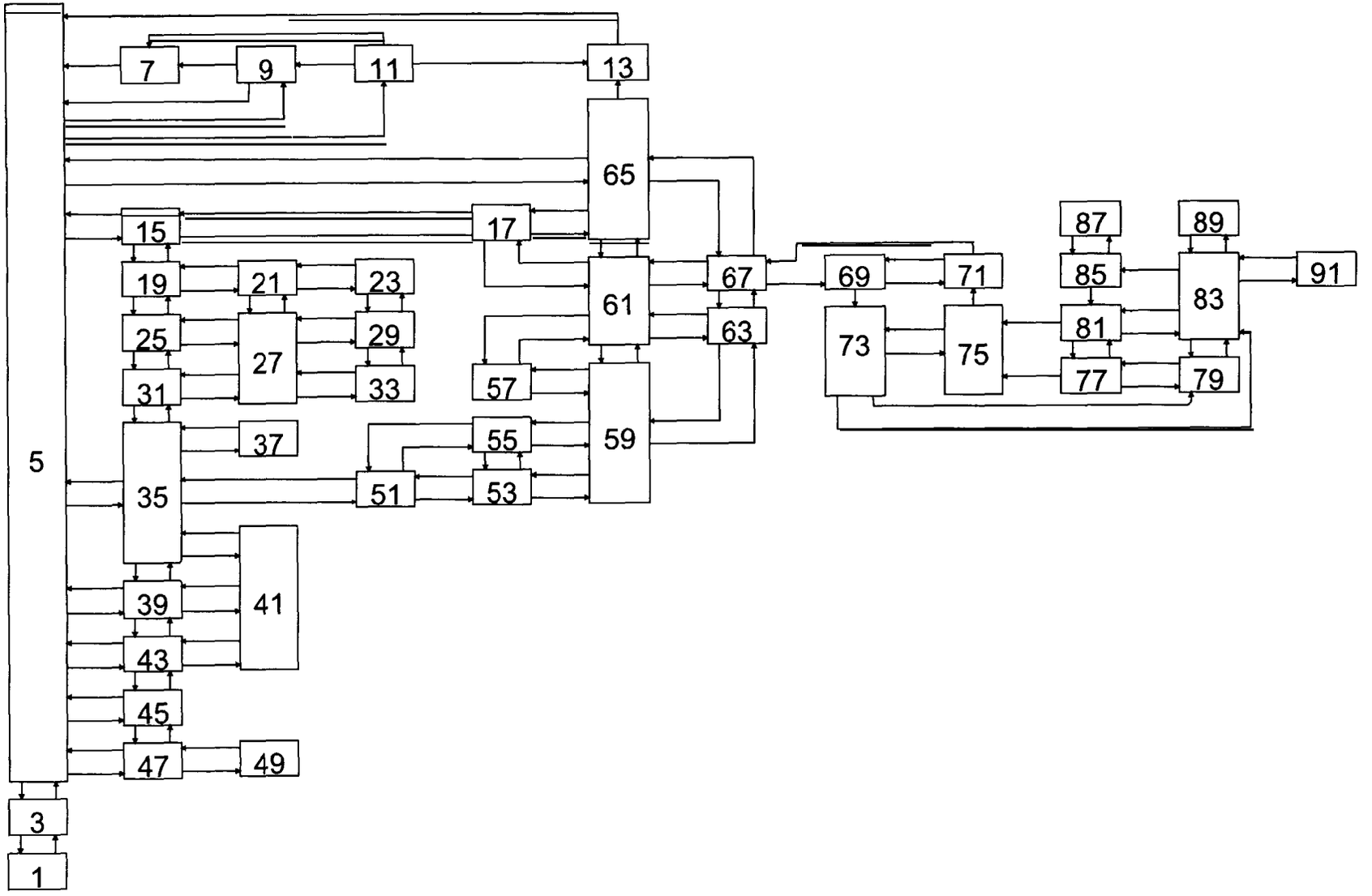


Figure 8.1.2. Schematic structure of the water boxes of the entire model showing routes of water exchange (arrows) between adjacent boxes.



The compartment names, volumes, mean depths, suspended sediment loads and sedimentation rates are given in Table 8.1.1. The volume exchange rates used by the model are given in Table 8.1.2.

Table 8.1.1. Compartment names, volumes, mean depths, suspended sediment loads (SSL) and sedimentation rates (SR).

Box	Name	Volume (m^3)	Depth (m)	SSL ($t\ m^{-3}$)	SR ($t\ m^2\ y^{-1}$)
1	Other Oceans	1.0E+18	4.0E+03	1.0E-07	5.2E-05
3	Atlantic Ocean	3.0E+17	3.5E+03	1.0E-07	1.0E-04
5	North-East Atlantic	5.0E+16	3.5E+03	1.0E-07	1.0E-05
7	Arctic Ocean	1.7E+16	1.2E+03	1.0E-07	1.0E-05
9	Spitsbergen Waters	1.0E+14	1.2E+03	1.0E-07	1.0E-05
11	Barents Sea	3.0E+14	2.0E+02	1.0E-07	1.0E-05
13	Norwegian Coastal Waters	1.0E+15	1.2E+03	1.0E-07	1.0E-05
15	Scottish Waters West	1.0E+13	1.1E+02	1.0E-06	1.0E-04
17	Scottish Waters East	3.0E+12	1.1E+02	1.0E-06	1.0E-04
19	Irish Sea North West	4.1E+11	9.3E+01	3.0E-06	3.0E-03
21	Irish Sea North	6.0E+10	3.4E+01	3.0E-06	5.1E-03
23	Irish Sea North East	5.2E+10	2.4E+01	3.0E-06	3.6E-03
25	Irish Sea West	6.6E+11	6.3E+01	3.0E-06	2.0E-03
27	Irish Sea South East	1.6E+11	3.1E+01	3.0E-06	4.7E-03
29	Cumbrian Waters	3.8E+10	2.8E+01	3.0E-06	4.2E-03
31	Irish Sea South	1.1E+12	5.7E+01	1.0E-06	1.0E-04
33	Liverpool and Morcambre Bays	3.2E+10	1.3E+01	3.0E-06	2.0E-03
35	Celtic Sea	2.0E+13	1.5E+02	1.0E-06	1.0E-04
37	Bristol Channel	1.0E+12	5.0E+01	1.0E-06	1.0E-04
39	Bay of Biscay	6.5E+14	4.0E+03	1.0E-06	1.0E-05
41	French Continental Shelf	3.5E+13	3.5E+02	5.0E-07	1.0E-04
43	Cantabrian Sea	3.0E+13	7.6E+02	1.0E-06	2.0E-04
45	Portuguese Continental Shelf	1.5E+13	4.9E+02	1.0E-06	2.0E-04
47	Gulf of Cadiz	2.3E+14	1.7E+03	2.0E-07	5.0E-05
49	Mediterranean Sea	4.0E+15	1.3E+03	1.0E-06	1.0E-04
51	English Channel West	3.2E+12	6.0E+01	1.0E-06	1.0E-04

Table 8.1.1. continued.

Box	Name	Volume (m^3)	Depth (m)	SSL ($t\ m^{-3}$)	SR ($t\ m^{-2}\ y^{-1}$)
53	English Channel South East	6.5E+11	4.0E+01	1.0E-06	1.0E-04
55	English Channel North East	6.5E+11	4.0E+01	1.0E-06	1.0E-04
57	North Sea South West	4.5E+11	3.1E+01	6.0E-06	1.6E-04
59	North Sea South East	9.5E+11	3.7E+01	6.0E-06	1.9E-04
61	North Sea Central	1.3E+13	5.0E+01	6.0E-06	1.0E-04
63	North Sea East	1.2E+12	2.2E+01	6.0E-06	4.4E-05
65	North Sea North	5.6E+13	2.4E+02	6.0E-06	1.0E-04
67	Skagerrak	6.8E+12	2.1E+02	1.0E-06	7.5E-04
69	Kattegat, deep	2.0E+11	1.0E+02	1.0E-06	7.5E-04
71	Kattegat, surface	3.2E+11	2.0E+01	1.0E-06	7.5E-04
73	Belt Sea, deep	1.4E+11	3.0E+01	1.0E-06	7.5E-04
75	Belt Sea, surface	1.5E+11	1.4E+01	1.0E-06	7.5E-04
77	Baltic Sea West, deep	7.7E+11	1.1E+02	3.0E-07	7.5E-04
79	Baltic Sea East, deep	1.5E+12	1.1E+02	3.0E-07	7.5E-04
81	Baltic Sea West, surface	3.8E+12	4.9E+01	3.0E-07	7.5E-04
83	Baltic Sea East, surface	7.0E+12	5.3E+01	3.0E-07	7.5E-04
85	Bothnian Sea	4.9E+12	6.2E+01	3.0E-07	1.5E-03
87	Bothnian Bay	1.5E+12	4.1E+01	3.0E-07	1.0E-03
89	Gulf of Finland	1.1E+12	3.7E+01	3.0E-07	1.5E-03
91	Gulf of Riga	4.1E+11	2.3E+01	3.0E-07	7.5E-04

Table 8.1.2. Exchange rates between boxes ($km^3\ y^{-1}$).

From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)
1	3	1000000	5	11	100000	5	43	109000
3	1	1000000	5	13	14500	5	45	460000
3	5	500000	5	15	8300	5	47	510000
5	3	500000	5	35	10000	5	65	34000
5	9	100000	5	39	670000	7	5	240000

Table 8.1.2. continued.

From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)
9	5	20000	31	27	75	51	55	3500
9	7	220000	31	35	600	53	51	1000
11	7	20000	33	27	109	53	55	300
11	9	140000	33	29	55	53	59	6000
13	11	60000	35	5	4600	55	51	1000
15	5	500	35	31	3000	55	53	1300
15	17	10700	35	37	2000	55	59	50
15	19	5000	35	39	150000	57	59	609
17	15	500	35	41	140000	57	61	381
17	61	8000	35	51	7000	59	53	50
17	65	2400	37	35	2000	59	55	1000
19	15	7400	39	5	670000	59	57	294
19	21	333	39	35	150000	59	61	458
19	25	514	39	41	580000	59	63	7274
21	19	833	39	43	390000	61	17	0
21	23	183	41	35	140000	61	57	696
21	27	173	41	39	580000	61	59	0
23	21	238	41	43	74000	61	63	9735
23	29	100	43	5	109000	61	65	6908
25	19	2414	43	39	390000	61	67	5600
25	27	933	43	41	75000	63	59	2417
25	31	600	43	45	15000	63	61	8700
27	21	568	45	5	460000	63	67	5892
27	25	433	45	43	13000	65	5	1730
27	29	230	45	47	60000	65	13	45400
27	31	75	47	5	510000	65	17	0
27	33	129	47	45	58000	65	61	0
29	23	205	47	49	52890	65	67	4983
29	27	145	49	47	50580	67	61	5500
29	33	35	51	35	2000	67	63	0
31	25	3000	51	53	3500	67	65	11505

Table 8.1.2. continued.

From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)	From box	To box	Exchange rate ($km^3\ y^{-1}$)
67	69	1545	77	75	220	83	85	525
69	71	930	77	79	220	83	89	8
69	73	720	77	81	198	83	91	312
71	67	2020	79	77	440	85	81	970
71	69	100	79	83	208	85	87	372
73	75	930	81	77	198	87	85	472
73	79	220	81	75	720	89	83	133
73	83	270	81	83	4124	91	83	344
75	71	1170	83	79	208			
75	73	700	83	81	4118			

At any given time the activity in the water column is partitioned between the water phase and the suspended sediment material. The fraction of the total activity (F_w) in the water column that is in aqueous solution is given by:

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

where K_d is the sediment distribution coefficient ($m^3\ t^{-1}$) and SSL the suspended sediment load (t m"). Values for k_d of 3000 and 1000 are used for Cs-137 and Sr-90, respectively (IAEA, 1985). Activity on suspended sediments is lost to the underlying boxes when particulates settle out. The fractional transfer from a water column (box i) to the sediments (box j) due to sedimentation is given by:

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

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

The model includes transfers of radioactivity between the surface sediment layer and the bottom boundary layer. This transfer is represented by diffusivity through the pore water, using a diffusion coefficient of $1 \cdot 10^{-9}\ m^2\ s^{-1}$, and mixing due to bioturbation, modelled as a diffusive process using a diffusion coefficient of $1 \cdot 10^{-12}\ m^2\ s^{-1}$ (NRPB et al., 1994). Removal of activity from the top surface sediment to lower sediment layers is taken into account by assuming that the burial rate is equal to the flux of particles settling from the overlying waters. Radioactive decay is accounted for in all boxes.

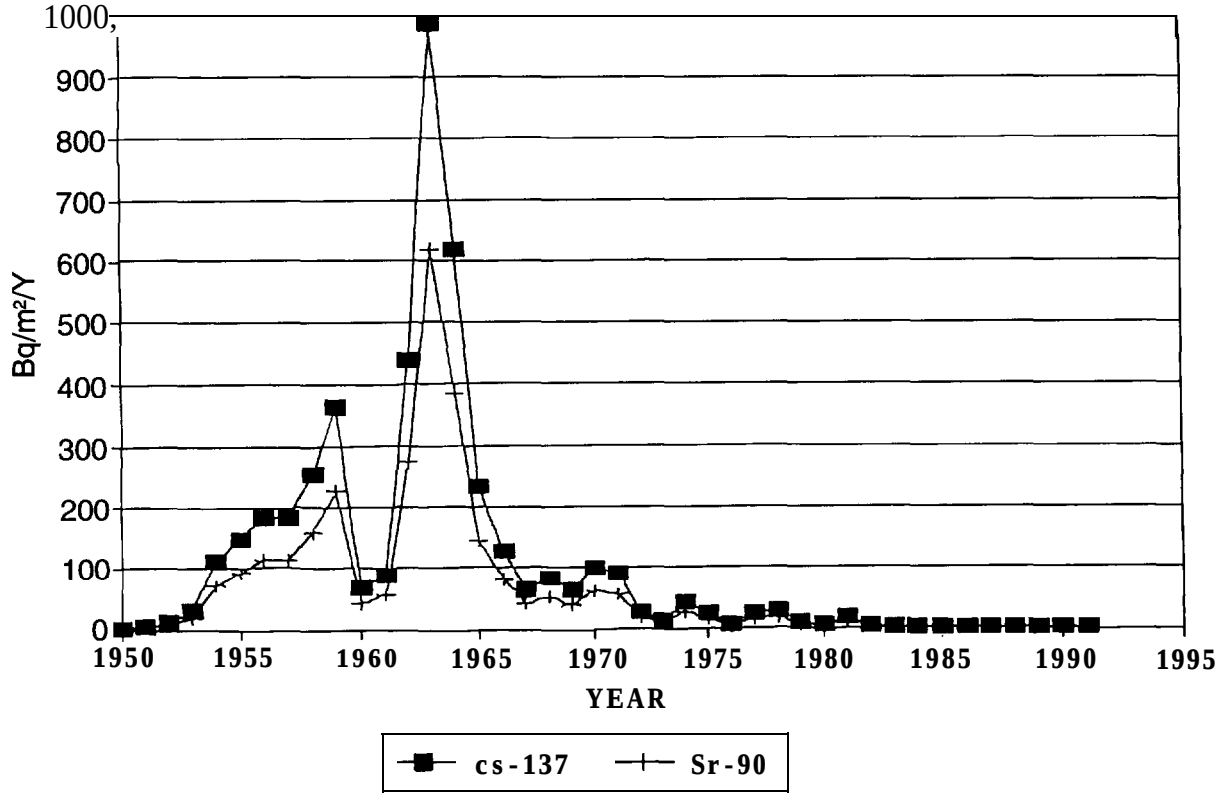
The model is implemented in the TIME ZERO modelling environment (Kirchner, 1989) on a personal computer.

8.2. Source **Terms**

In order to compare environmental levels of Cs-137 and Sr-90 predicted by the model with observations, the following four source terms have been considered as input to the model: nuclear weapons fallout, Chernobyl fallout, liquid discharges from the European reprocessing facilities Sellafield and La Hague, and liquid discharges from nuclear installations bordering the Baltic Sea.

Fallout of Cs- 137 and Sr-90 from the atmospheric nuclear tests has been included based on measurements made in Denmark. These data are considered representative for the Baltic Sea area. The data cover the period from 1955 to 1992 (Aarkrog et al., 1992) as shown in Figure 8.2.1. Each model surface compartment thus every year receives an input of Cs-137 and Sr-90 according to the corresponding surface area. The influence of the Chernobyl accident was eliminated from this source by extrapolating the nuclear weapons fallout for the period 1986-1992 based on exponential regression of the data observed in the period 1970-1985.

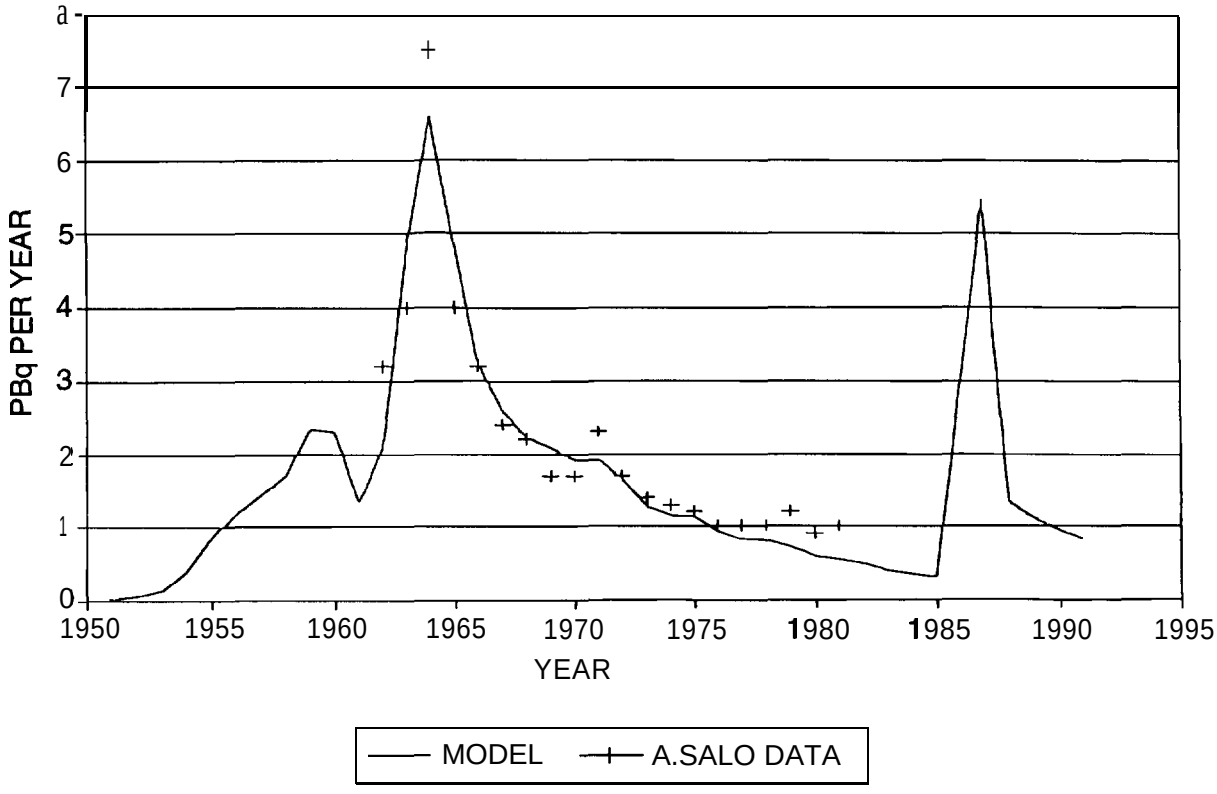
Figure 8.2.1. Atmospheric fallout of Cs-137 and Sr-90 in Denmark (Bq m^{-2}) from nuclear weapon tests.



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

contribution to consider. Salo et al. (1985) investigated the inventories of Cs-137 and Sr-90 in seawater and sediments of the Baltic Sea for the time period 1961 to 198 1 and give data for the total runoff of Cs-137 and Sr-90 with the rivers. Their data show that the river runoff of Sr-90 for the period 1961-1981 constitutes an important contribution to the input of Sr-90 into the Baltic seawater and since 1972 the river runoff has dominated over the direct fallout from the atmosphere to the water. For Cs- 137 the river runoff is less important - of the order of a few percent compared to the atmospheric fallout into the water. We have constructed two simple sub-models and fitted them to these data allowing extrapolations to be made for the runoff of Cs-137 and Sr-90 into the Baltic seawater. The predictions and the data are shown in Figures 8.2.2 and 8.2.3 which give for the time period 1950 to 2000 the estimated input used for the model calculations of Cs-137 and Sr-90 via rivers to the Baltic seawater.

Figure 8.2.2. Predicted and observed runoff **(TBq y^{-1})** of Cs-137 via rivers to the Baltic Sea shown as a function of time.

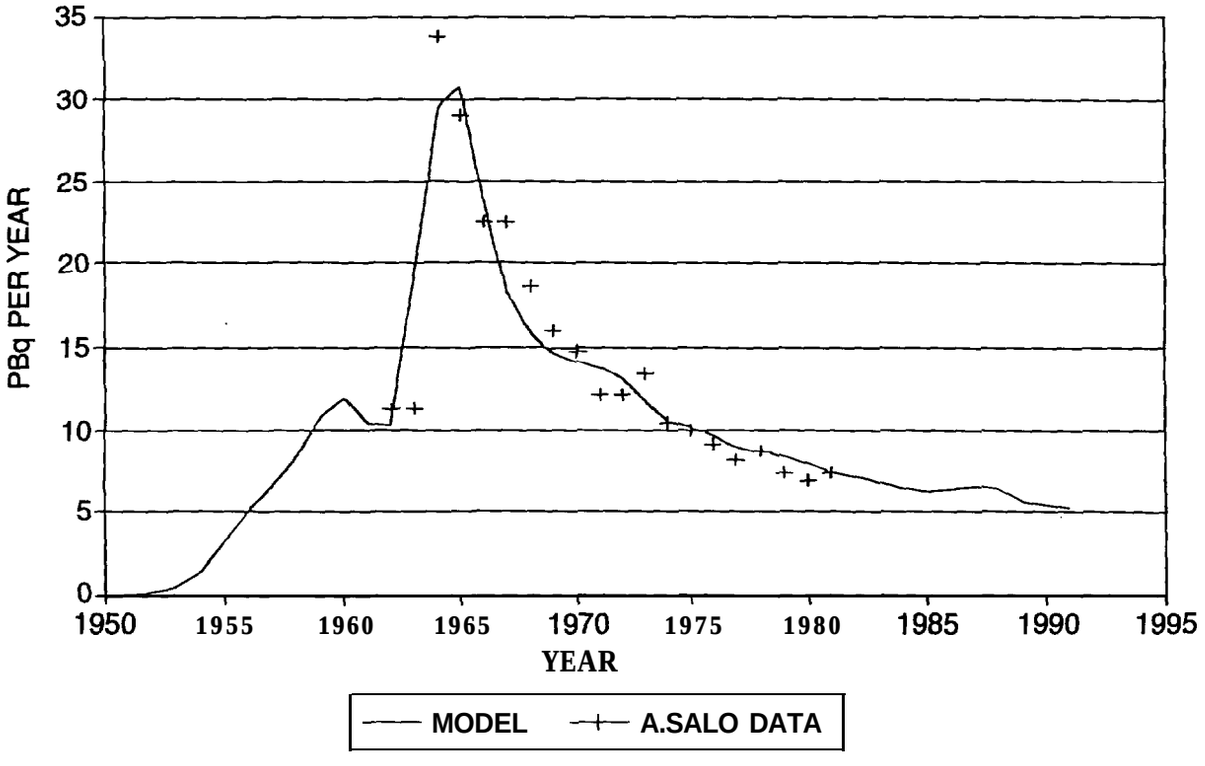


The sub-models are based on the classical UNSCEAR technique (eg. UNSCEAR 1972) using multiple regression of the data from 1961 to 1981 against the annual fallout rates and the accumulated fallout concentrations. The following sub-models were determined:

$$Sr\text{-}90: \quad R_i = 13\, d_{i-1} + 14\, d_{i-2} + 3\, A_{,,} + 8\, A_{,,}$$

$$Cs\text{-}137: \quad R_i = 1.1\, d_i + 3.4\, d_{i-1} + 1.1\, A_{,.}$$

Figure 8.2.3. Predicted and observed runoff (TBq y“) of Sr-90 via rivers to the Baltic Sea shown as a function of time.



The total river runoff R_i (TBq a^{-1}) in year I is a function of the fallout rate d_i (Bq m^{-2} a^{-1}) in the same year, and/or the previous years d_{i-1}, d_{i-2}, and the accumulated fallout concentrations A, (Bq m^{-2}), where H designates the effective half-life. Both regressions give a R^2-value (coefficient of determination) of 0.97. The two sub-models illustrate the well-known radioecological characteristics for the transfer from soil (to vegetation) of Sr-90 and Cs-137. The transfer of Sr-90 is influenced by a time lag of one to two years and the accumulated fallout based on the physical half-life and a much shorter half-life. The transfer of Cs-137 depends on the recent fallout and the accumulated fallout based on a short effective half-life. The values of the coefficients for the two expressions additionally demonstrate the significantly higher transfer of Sr-90 via river runoff compared to that for Cs-137; for the equilibrium situation with a constant fallout rate the river runoff of Sr-90 is about a factor of 20 higher than that of Cs-137. For the model calculations the total runoff for each radionuclide was split between the different Baltic Sea sub-regions according to the sixes of the corresponding catchment areas (based on HELCOM 1993). The catchment areas used are shown in Table 8.2.1.

Table 8.2.1. Catchment areas for the sub-regions of the Baltic Sea used to calculate runoff.

Box No.	Baltic Sea Regions	Catchment areas (km^2)	(%)
71	Kattegat	87400	5.1
75	Belt Sea	4300	0.3
81	Baltic Sea West	108250	6.3
83	Baltic Sea East	469600	27.3
85	Bothnian Sea	228100	13.3
87	Bothnian Bay	277000	16.1
89	Gulf of Finland	413300	24.0
91	Gulf of Riga	132100	7.7
	Total	1720050	100

The input of Cs-137 from the Chernobyl accident into the Baltic Sea area was estimated to 4.5 PBq (CEC, 1990). The amount and distribution of radiocaesium over the area is based on the information given in Chapter 2. Observed data from Denmark, Finland and Russia show that the fallout of Sr-90 from the Chernobyl accident was about 2% relative to that of Cs-137. Table 8.2.2 shows the estimated inputs of Cs-137 into the various regions used for the model calculations.

Table 8.2.2. Direct fallout Cs-137 and Sr-90 to Baltic seawater regions from the Chernobyl accident used for the model calculations.

BOX No.	Baltic Sea Regions	cs-137 (TBq)	Sr-90 (TBq)
75	Belt Sea	60	1.2
81	Baltic Sea West	980	19.6
83	Baltic Sea East	490	9.8
85	Bothnian Sea	2400	48
87	Bothnian Bay	210	4.2
89	Gulf of Finland	320	6.4
91	Gulf of Riga	40	0.8
	Total	4500	90

The reported discharges of Cs-137 and Sr-90 shown in Figures 8.2.4 and 8.2.5 from Sellafield (into box 29) and La Hague (into box 53) have been used (BNFL, 1993; CEC, 1990) as input to the model. However, only a small fraction of these discharges reach the Baltic Sea.

Figure 8.2.4. Discharges of Cs-137 to sea (PBq y^{-1}) from the nuclear reprocessing plants Sellafield in the UK and La Hague in France.

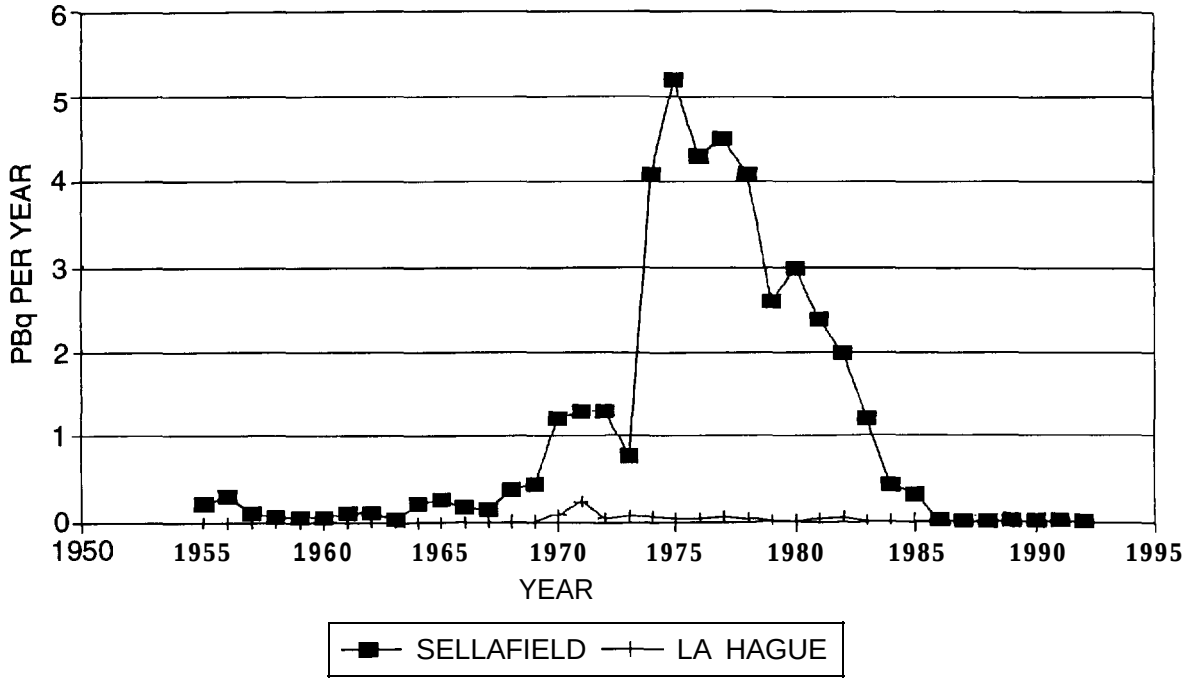
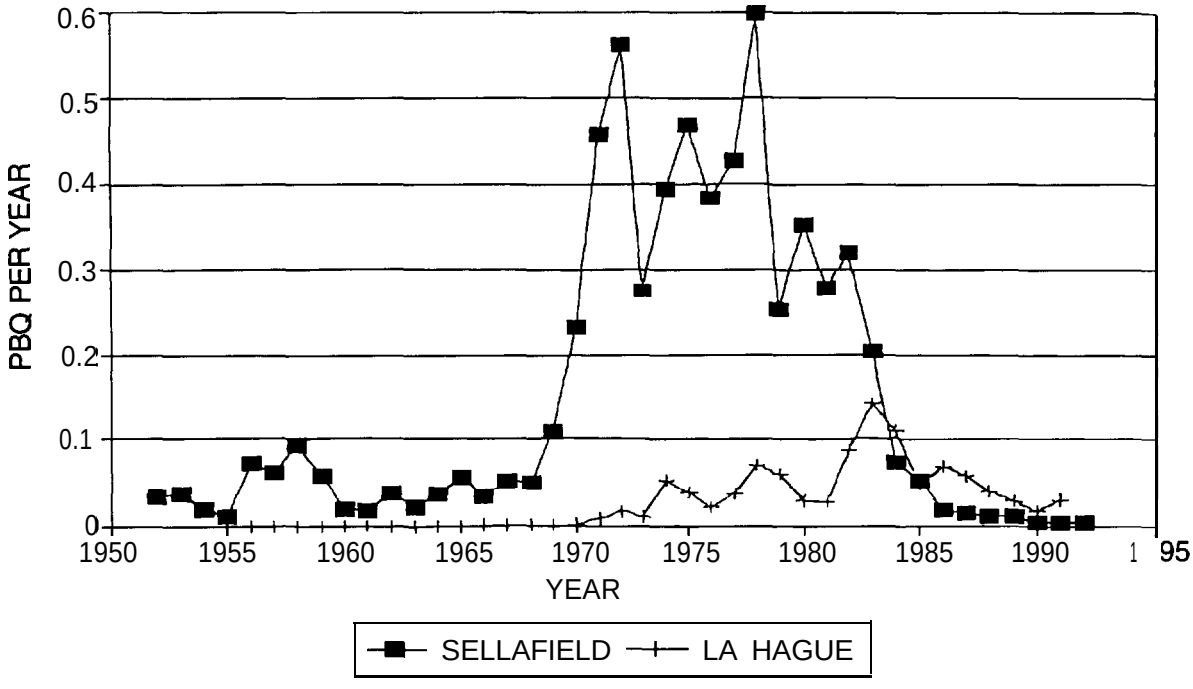


Figure 8.2.5. Discharges of Sr-90 to sea (PBq y^{-1}) from the nuclear reprocessing plants Sellafield in the UK and La Hague in France.



The input to the Baltic Sea has been estimated from the model calculations which indicate that about 4% of the discharges from Sellafield are transferred to the Kattegat compared to about 8% of the discharges from La Hague. The relative transfer of Cs-137 is lower (about 10% relative difference) than that of Sr-90 because of the chemical differences that result in a higher transfer to sediments of Cs-137 than of Sr-90. Due to the efficient mixing of the upper and lower waters in the Kattegat and the Belt Sea, the main part of the activity of these radionuclides from the two reprocessing plants return to the Skagerrak and only about 1% of the discharges of Cs-137 and Sr-90 are estimated to be transferred to the Baltic Proper.

Furthermore, discharges of Cs-137 and Sr-90 from the nuclear installations bordering the Baltic Sea as specified in Section 2.3 have been included in the model calculations.

Table 8.2.3 presents an overview of the sources considered for the present calculations.

Table 8.2.3. Discharges of Cs-137 and Sr-90 to the Baltic Sea in 1950 to 1994 used for the model calculations.

Source	cs-137 (TBq)	Sr-90 (TBq)
Weapons fallout, direct deposition to sea	1780	1130
Weapons fallout, river runoff	70	410
Chernobyl fallout	4500	90
European reprocessing	380	70
Nuclear installations	0.65	0.4
Total	6732	1700

8.3. Model Results for Cs-137

Calculations have been carried out with the model for the time period 1950 to 2000 using the sources specified in the previous section. The calculated concentrations of Cs-137 in seawater, sediments and biota are shown in the Figures 83.1, 8.3.3 and 8.3.5, respectively. For the water concentrations in Figure 8.3.1 it is noted that all regions reflect the culmination of the nuclear weapons fallout in the 1960's. In the 1970's and early 1980's the large Sellafield discharges determine the levels in the Kattegat and Belt Sea, but not further into the Baltic Sea. Nearly all regions reflect the impact of the significant Chernobyl fallout in 1986 which is the reason that the Baltic Sea at present has the highest Cs-137 concentrations found in seawater anywhere. The observed data are included in the graphs where circles show the annual mean concentrations. It is noted that there generally is a good agreement between the observations and the model predictions. However, in the deep waters of the Eastern Baltic (box 79) the predicted levels differ significantly from the observed values after 1986. The comparison is further illustrated in Figure 8.3.2 showing a scatterplot between all (n= 130) observed and predicted concentrations of Cs-137 in seawater. The full line gives the ideal 1: 1

relationship, and the points are seen to scatter on both sides of the line. The geometric mean of the predicted-to-observed (P/O) ratio is 1.1 with a geometric standard deviation of 1.4.

Figure 8.3.1. Calculated and observed concentrations of Cs-137 in seawater (Bq m^{-3}) in the water regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

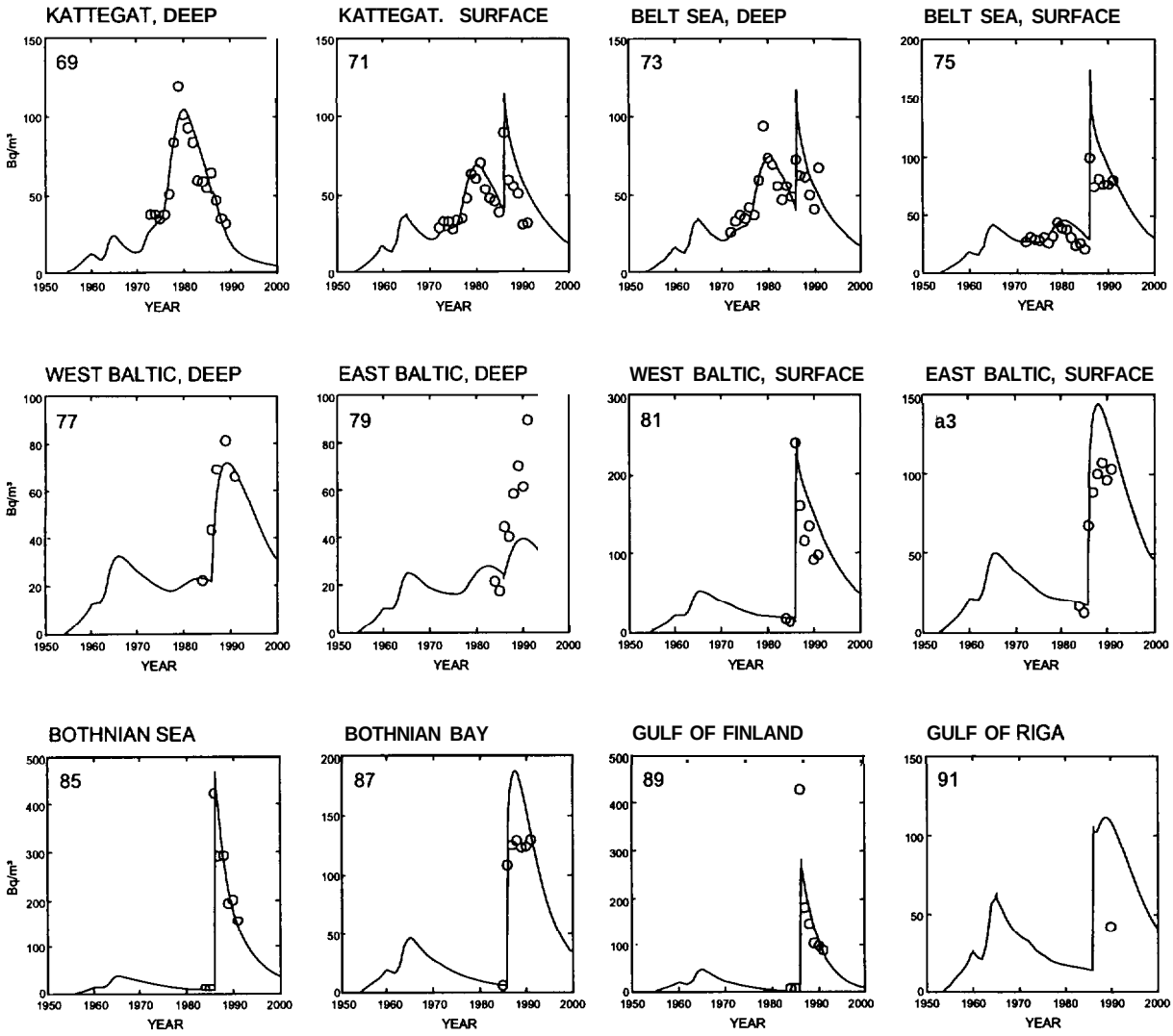
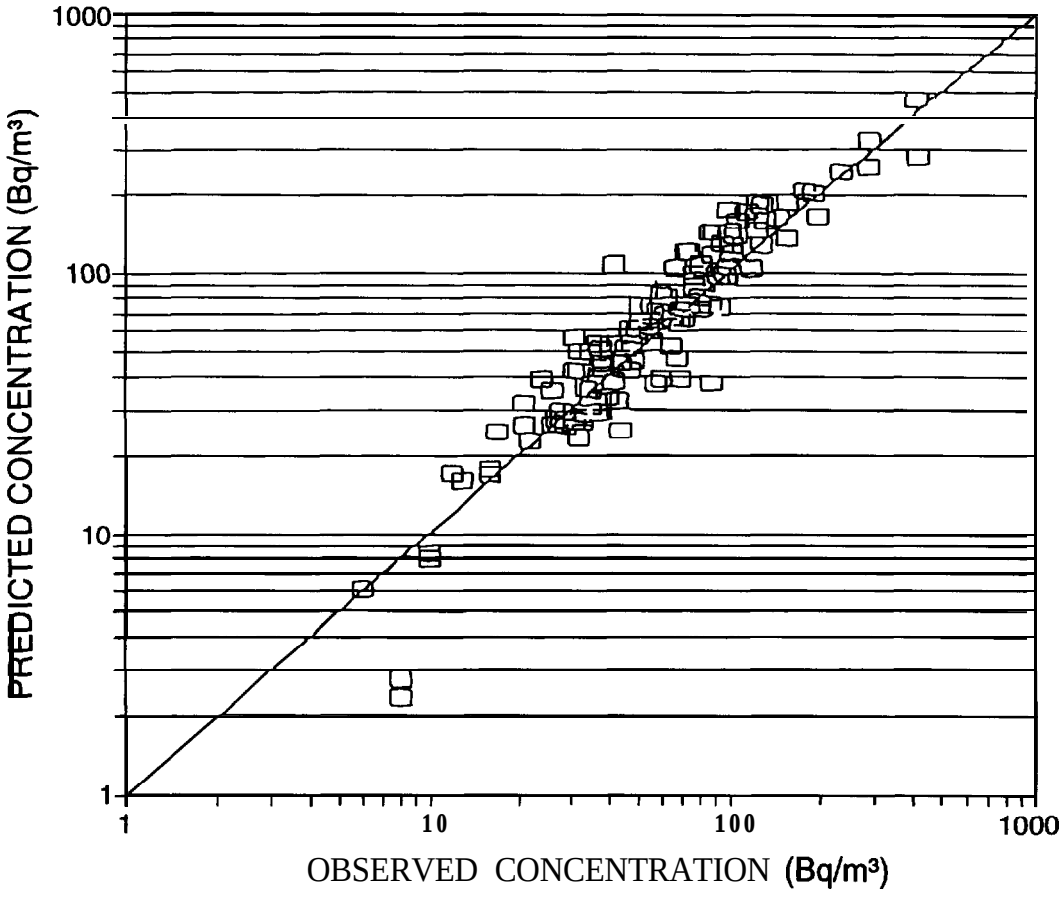


Figure 8.3.2. Scatter-plot of predicted and observed annual mean concentrations of Cs-137 in Baltic seawater. The line indicates 1: 1 relationship.



The calculated concentrations of Cs-137 in surface sediments are shown in Figure 8.3.3. The sediment concentrations are seen to respond more slowly to the source terms than the water concentrations. The observed data are much more scarce than for seawater. This reflects the fact that sediment sampling is much more demanding in terms of time and resources compared to water sampling. Furthermore, the variability of marine sediment data is very much larger within a small area than seawater data due mainly to inhomogeneities in sediments and very different mixing processes in the two media. However, the comparison between observed and predicted values shows a distinct tendency to under predict the concentrations of Cs-137 in surface sediments after the Chernobyl accident. Figure 8.3.4 shows a scatter-plot of the observed and predicted sediment concentrations (n = 35). The geometric mean of the P/O ratio is 0.3 with a geometric standard deviation of 2.4.

The calculated concentrations of Cs-137 in biota are shown in Figure 8.3.5. For Kattegat (box 71) calculations are done for both molluscs and fish, and for the remaining regions for fish only. The biota concentrations are calculated from concentrations of Cs-137 in filtered seawater using recommended bioconcentration factors (200 Bq t^{-1} fish per Bq m^{-3} water, and 30 Bq t^{-1} molluscs per Bq m^{-3} water; Koivulehto and Saxén, 1981; IAEA, 1985). Figure 8.3.6 shows a scatterplot of the observed and the predicted values (n=89). The geometric mean of the P/O-ratios is 1.7 with a geometric standard deviation of 2.3.

Figure 8.3.3. Calculated and observed concentrations of Cs-137 in top sediment (Bq kg^{-1} dw) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

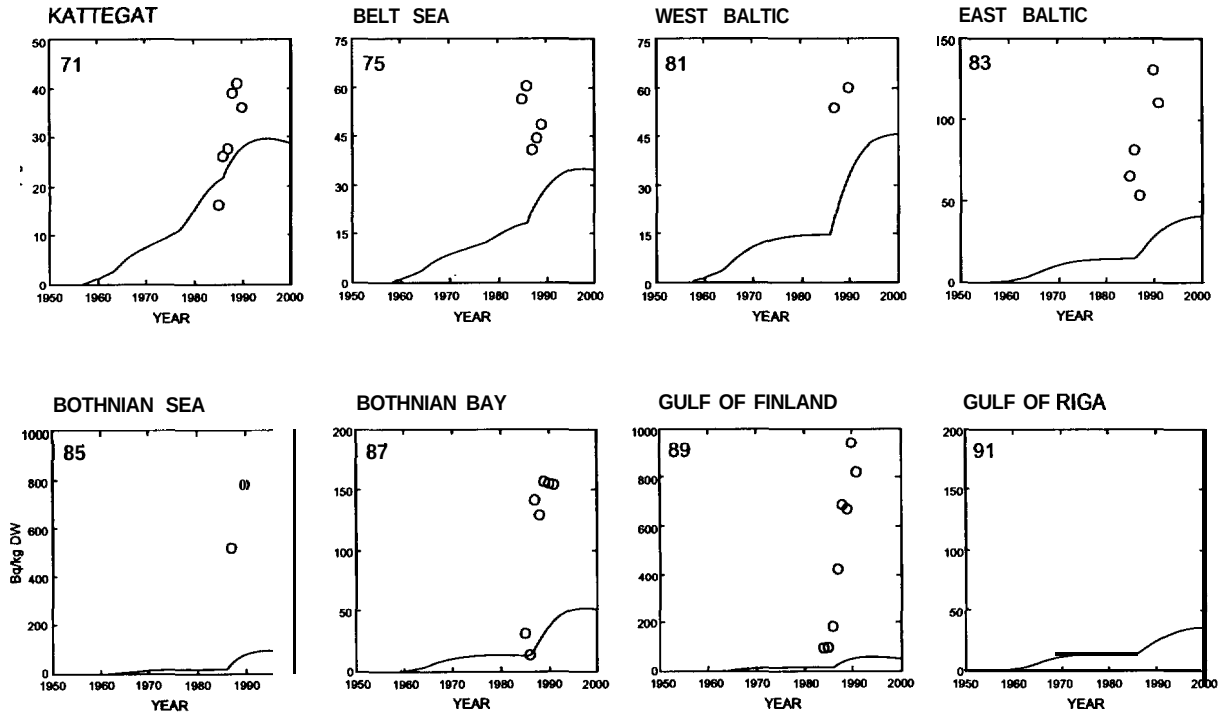


Figure 8.3.4. Scatterplot of predicted and observed annual mean concentrations of Cs-137 in surface sediments. The line indicates 1: 1 relationship.

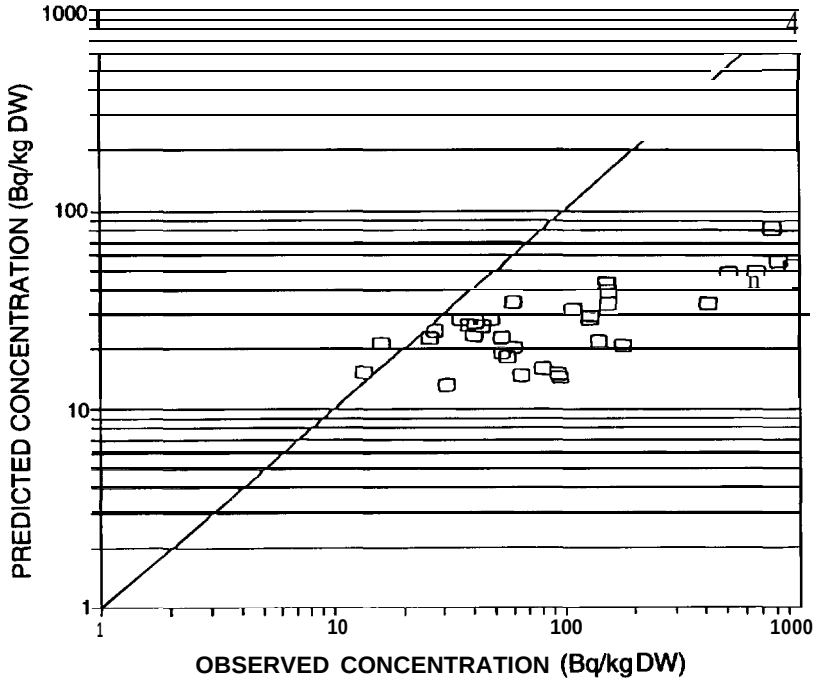


Figure 8.3.5. Calculated and observed concentrations of Cs-137 in biota (Bq kg^{-1} fw) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles and triangles show the annual arithmetic mean values of the observed levels in molluscs and fish, respectively.

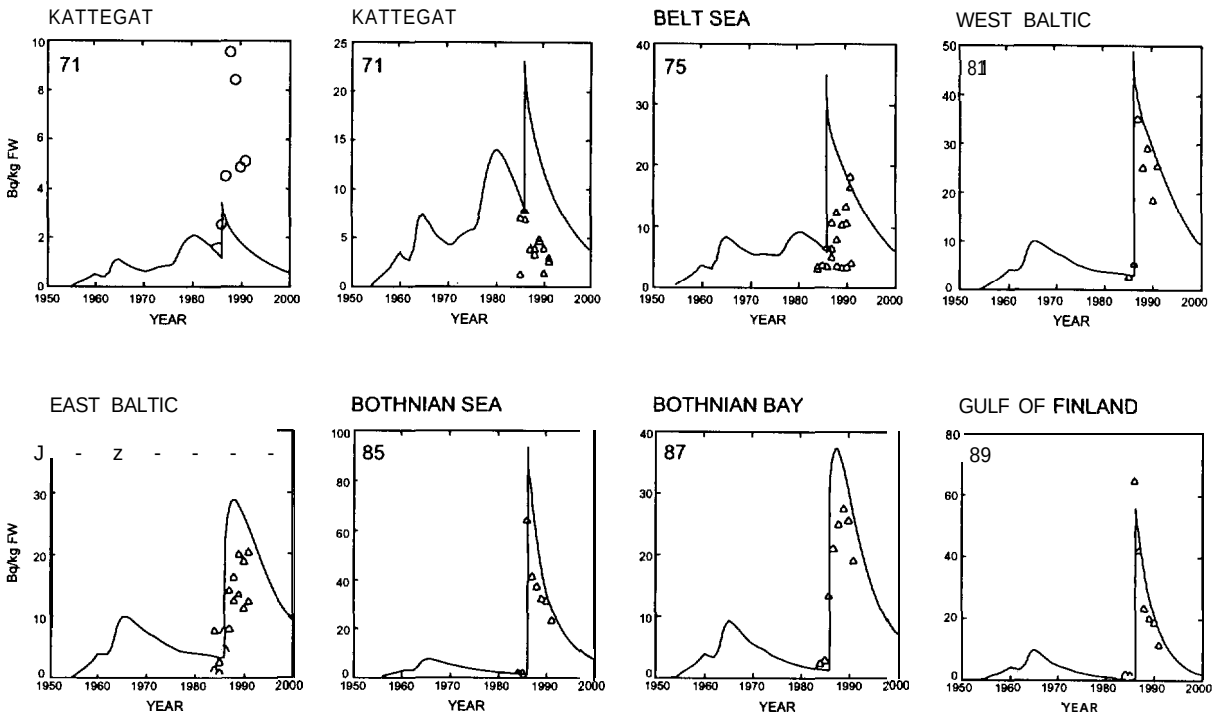
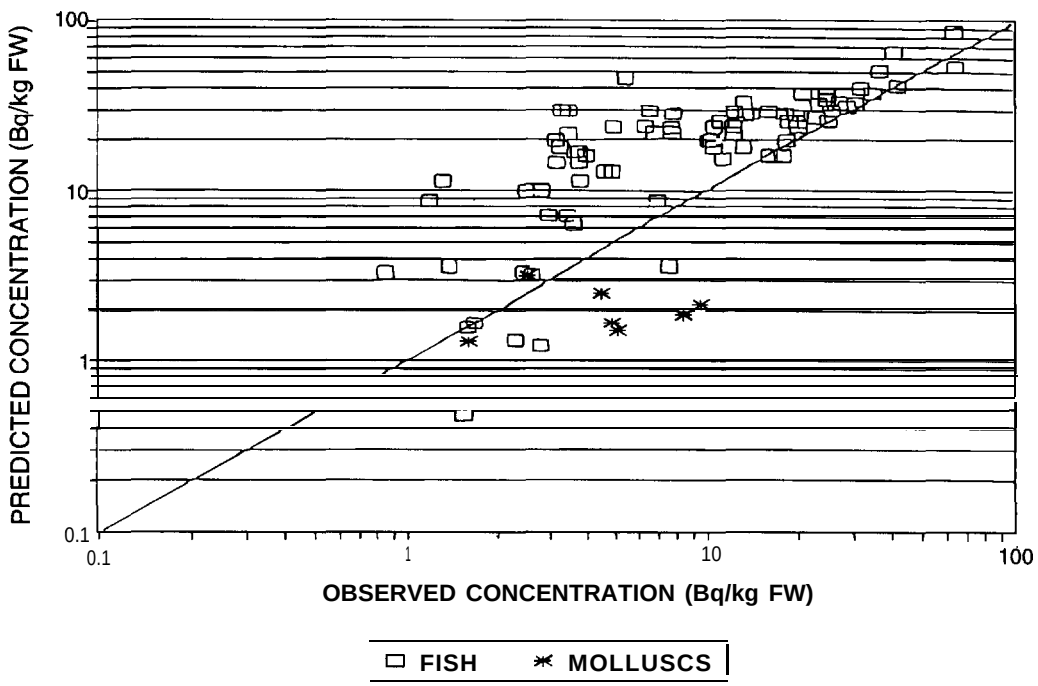
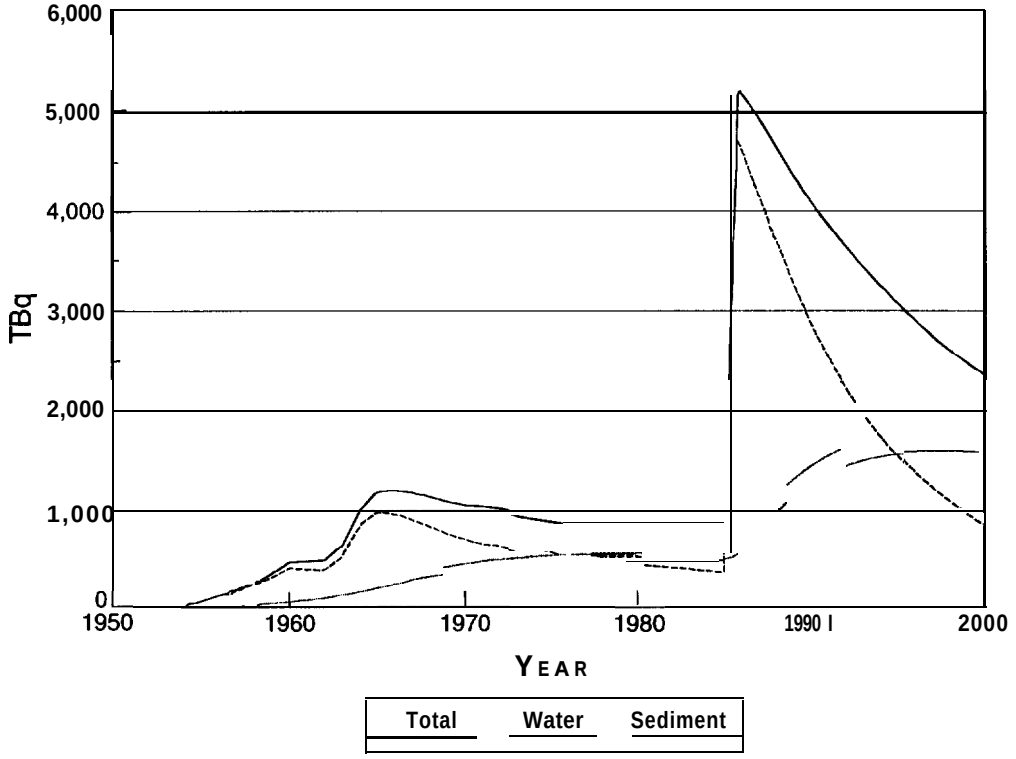


Figure 8.3.6. Scatter-plot of predicted and observed annual mean concentrations of Cs-137 in biota. The line indicates 1: 1 relationship.



The inventory of Cs-137 in the Baltic Sea has been calculated with the model as a function of time. Figure 8.3.7 shows the inventory broken down according to the two compartments: water and sediments. The transfer of Cs-137 from water to the sediments with time is illustrated. The results show that the inventory is approximately equally divided between water and sediments before the Chernobyl accident in 1986 after which most of the activity is found in the water. These predicted data are shown in Figure 8.38 together with observed data from Salo et al. (1985) and Sections 5 and 6 of the present report.

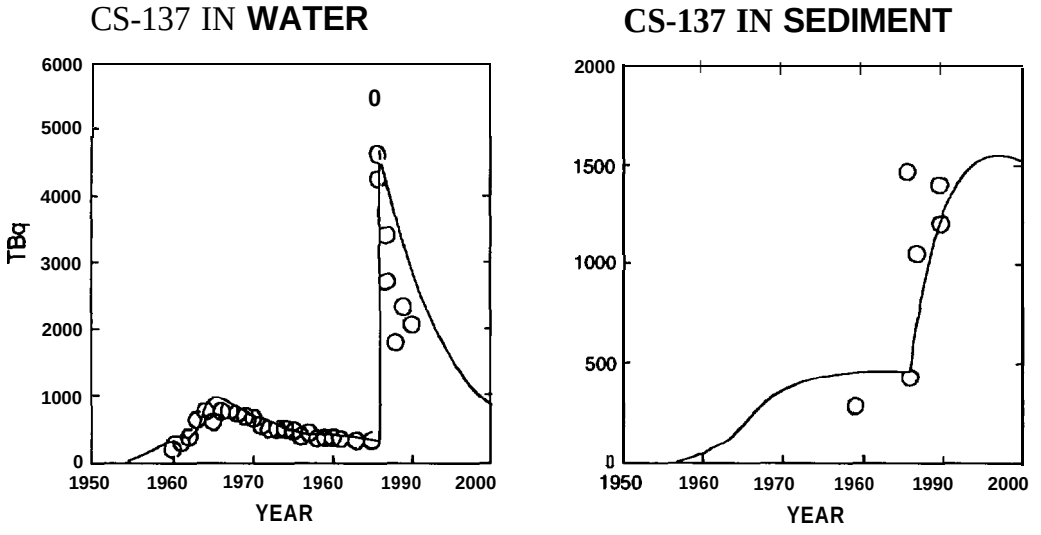
Figure 8.3.7. Predicted inventory of Cs-137 (TBq) in the Baltic Sea in water and sediments shown as a function of time.



It is noted that there is a good agreement for the water data prior to 1986 after which the model overestimates the water levels. This could indicate a different (faster) rate of transfer from the water to the sediments of the Cs-137 from the Chernobyl fallout compared to that from the atmospheric fallout from the nuclear weapons testing. This is in qualitative agreement with the comparison between the observed and predicted sediment concentrations after the Chernobyl accident. For all the water inventory data $(n=32)$ the geometric mean P/O ratio is 1.2 with a geometric standard deviation of 1.2. For the sediment data there is a reasonable agreement between observations and predictions, but the number of observations are few $(n=6)$. The geometric mean of the P/O ratios for the sediment inventory data is 0.9 with a geometric standard deviation of 1.7.

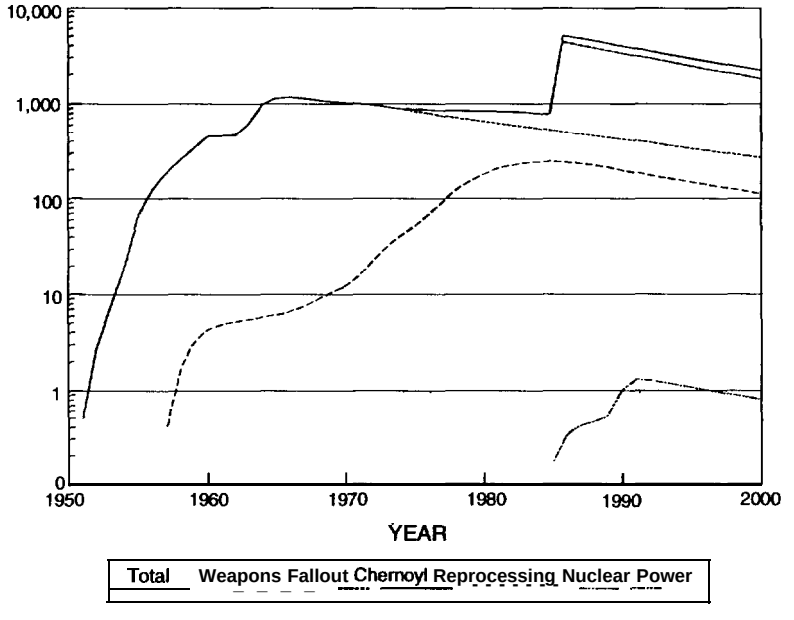
Figure 8.3.9 shows the Cs-137 inventory broken down according to the four sources: nuclear weapons fallout, Chernobyl fallout, discharges from reprocessing facilities and discharges from nuclear installations at the Baltic Sea.

Figure 8.3.8. Comparison of predicted and observed inventories of Cs-137 in Baltic seawater and sediments. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.



The graph is shown with a logarithmic vertical scale in order to cover the range from the total inventory to that from the nuclear installations which is about four orders of magnitude lower than the former. The two major sources to the inventory are the nuclear weapons fallout and the Chernobyl fallout in agreement with the inputs shown in Table 8.2.4. The Cs-137 inventory is estimated in 1994 at a value of about 3200 TBq of which 11% originate from nuclear weapons fallout, 84% from the Chernobyl accident, 5% from European reprocessing and 0.01% from nuclear installations located in the Baltic Sea area.

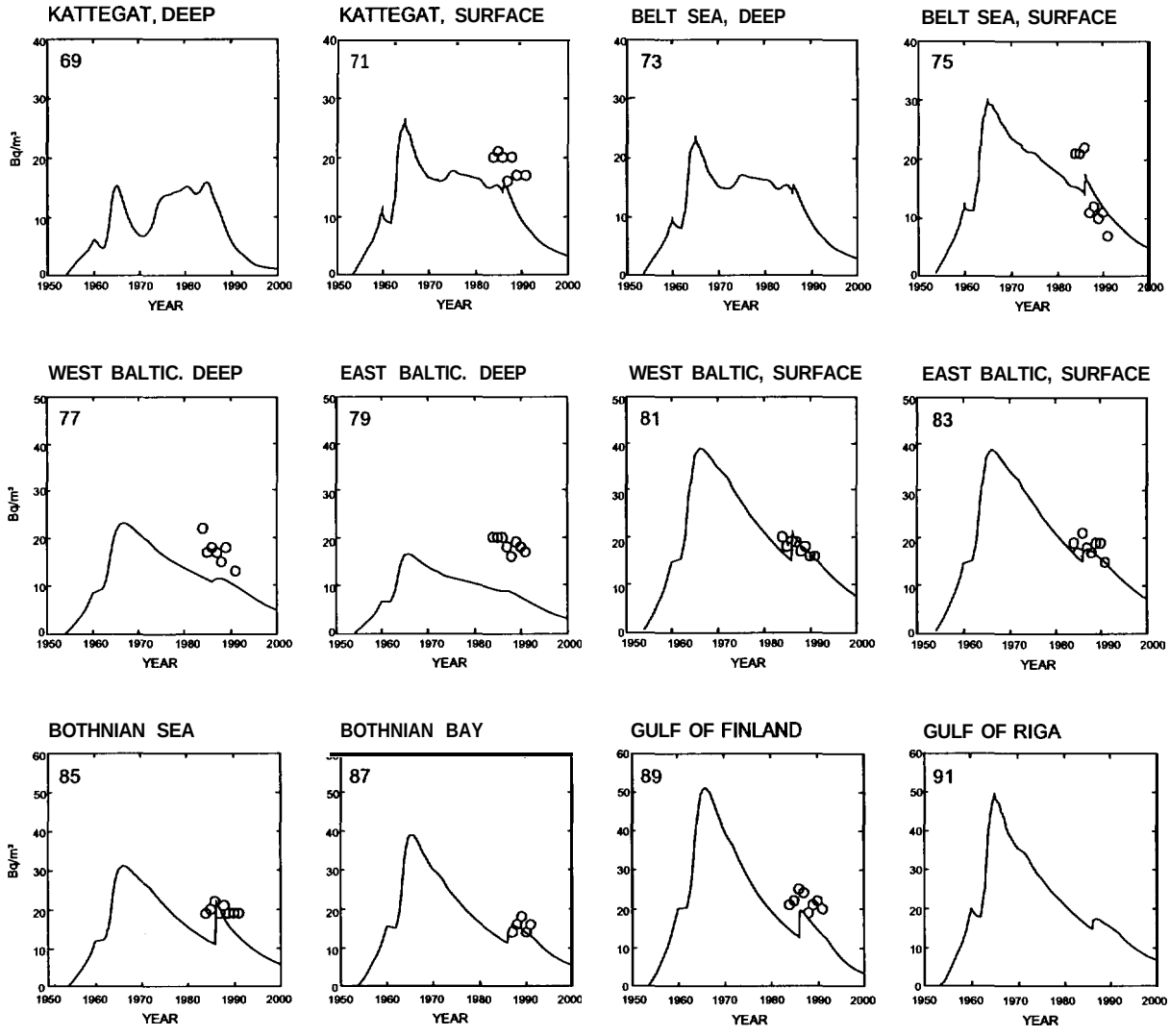
Figure 8.3.9. Predicted inventory of Cs-137 (TBq) in the Baltic Sea broken down according to the sources shown as a function of time.



8.4. Model Results for Sr-90

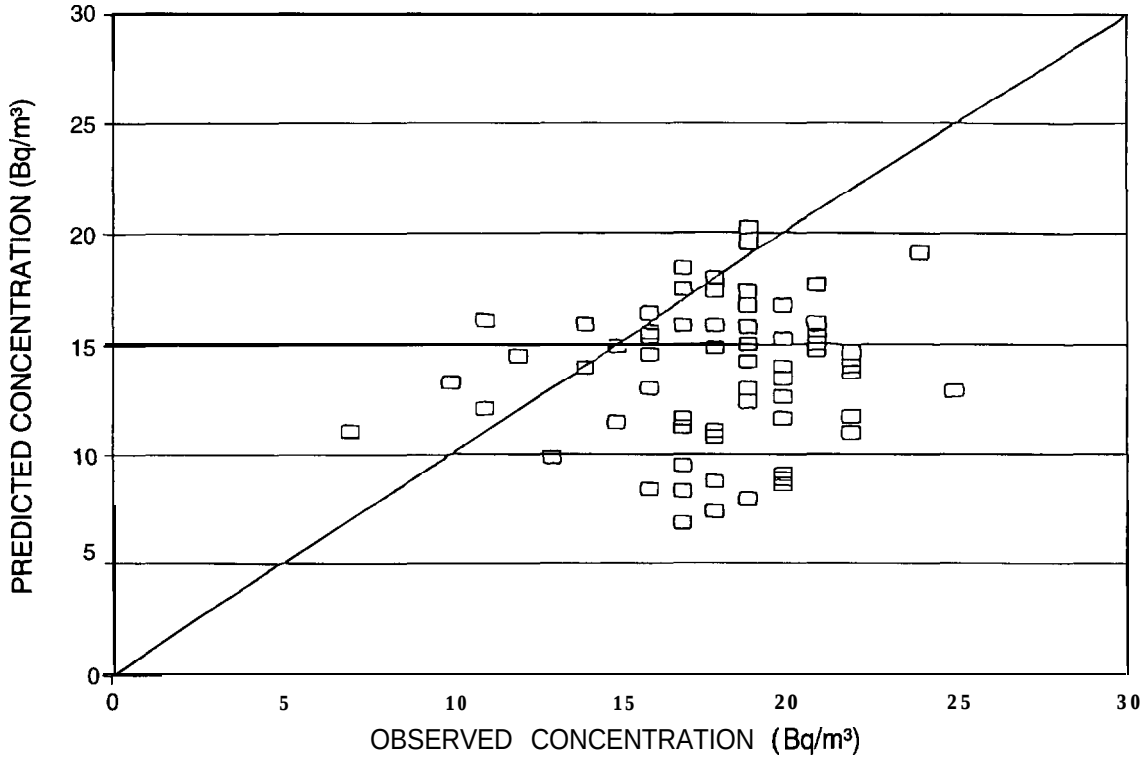
Similar calculations have been carried out for Sr-90 as for Cs-137. The calculated water concentrations of Sr-90 are shown in Figure 8.4.1 together with the observed values. The same tendencies are seen as for Cs-137: the fallout from the nuclear weapons testing is dominating the levels that peak in the early 1960's, the influence of discharges from the European reprocessing plants are only discernible in the Kattegat and the Belt Sea, and the contribution from the Chernobyl accident in 1986 is apparent but relatively much lower than for Cs-137. The observed data generally compare well with the predicted values. It is noted that the predicted values for the deep waters of the Eastern Baltic (box 79) are low compared to the observed values in agreement with the comparison for the Cs-137 data for that box. This indicates a need for improving the model by adjusting the water exchanges between that and the adjacent water regions.

Figure 8.4.1. Calculated and observed concentrations of Sr-90 in seawater (Bq m^{-3}) in the water regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.



The influence of the Chernobyl fallout on the concentrations of Sr-90 in the water is obvious from the predicted values but not equally so from the observed values. However, the comparison between observed and predicted values may be interpreted as a support of the assumptions concerning the input of Chernobyl Sr-90 fallout into the Baltic Sea. Figure 8.4.2 shows a scatterplot of the observed and predicted levels of Sr-90 in seawater. The geometric mean of the P/O values (n=67) is 0.8 with a geometric standard deviation of 1.4.

Figure 8.4.2. Scatter-plot of predicted and observed annual mean concentrations of Sr-90 in Baltic seawater. The line indicates 1: 1 relationship.



The calculated concentrations of Sr-90 in surface sediments in the various regions of the Baltic are shown in Figure 8.4.3. The observed values are few (n = 10) but indicate a tendency by the model to over predict the concentrations. The geometric mean P/O ratio is 2.6 with a standard deviation of 2.5 .

The calculated concentrations of Sr-90 in fish are shown in Figure 8.4.4. The calculated concentrations are based on a bioconcentration factor of 2 Bq t^{-1} fish per Bq m^{-3} filtered seawater (IAEA, 1985). It is noted that the predicted levels fall in two groups that either significantly overestimates or underestimates the observed levels. This is also apparent in Figure 8.4.5 showing a scatter-plot between the observed and predicted values. This indicates that the single bioconcentration factor used for the calculations in this case may be too simplistic a concept (cf. Section 7 on Biota). However, the model may be acceptable for the average conditions since the geometric mean of the P/O ratios is 0.9 which is close to unity; but the geometric standard deviation is large - 4.4.

Figure 8.4.3. Calculated and observed concentrations of Sr-90 in top sediment (Bq kg^{-1} dw) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

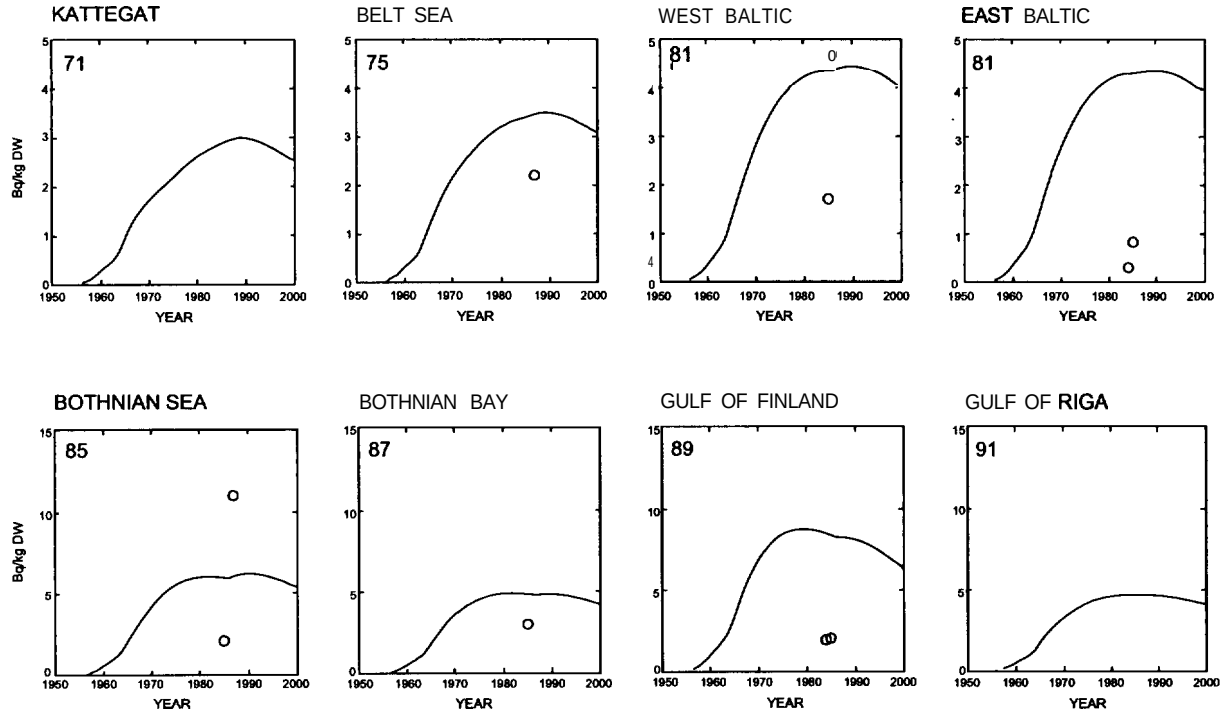


Figure 8.4.4. Calculated and observed concentrations of Sr-90 in fish (Bq kg^{-1} fw) in the regions of the Baltic Sea. The full lines show the predicted levels and the triangles show the annual arithmetic mean values of the observed levels.

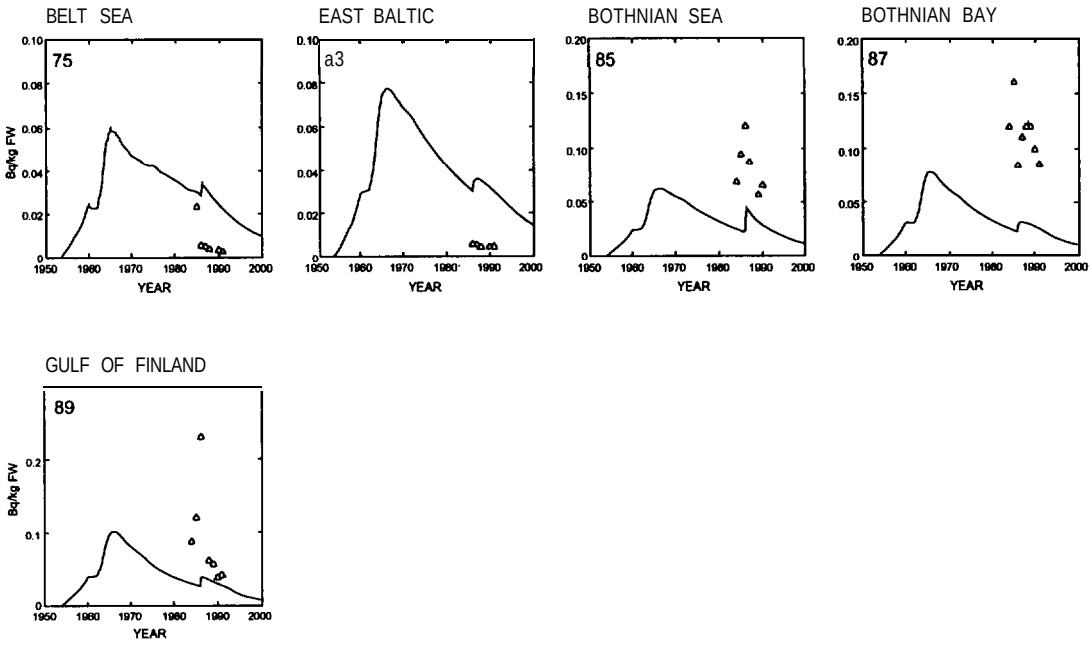
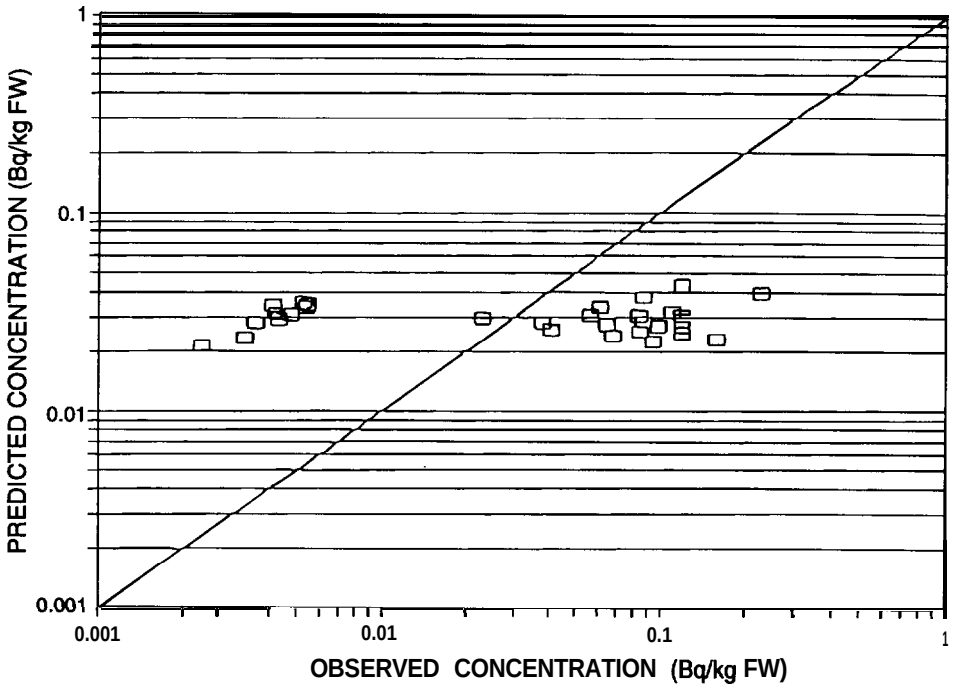


Figure 8.4.5. Scatterplot of predicted and observed annual mean concentrations of Sr-90 in fish. The line indicates 1:1 relationship.



The inventory of Sr-90 in the Baltic Sea is shown in Figure 8.4.6 which gives the inventories in water and sediment as a function of time. It is noted that the relative proportion of Sr-90 in the sediments is estimated to be smaller than for Cs-137.

Figure 8.4.6. Predicted inventory of Sr-90 (TBq) in the Baltic Sea in water and sediments shown as a function of time.

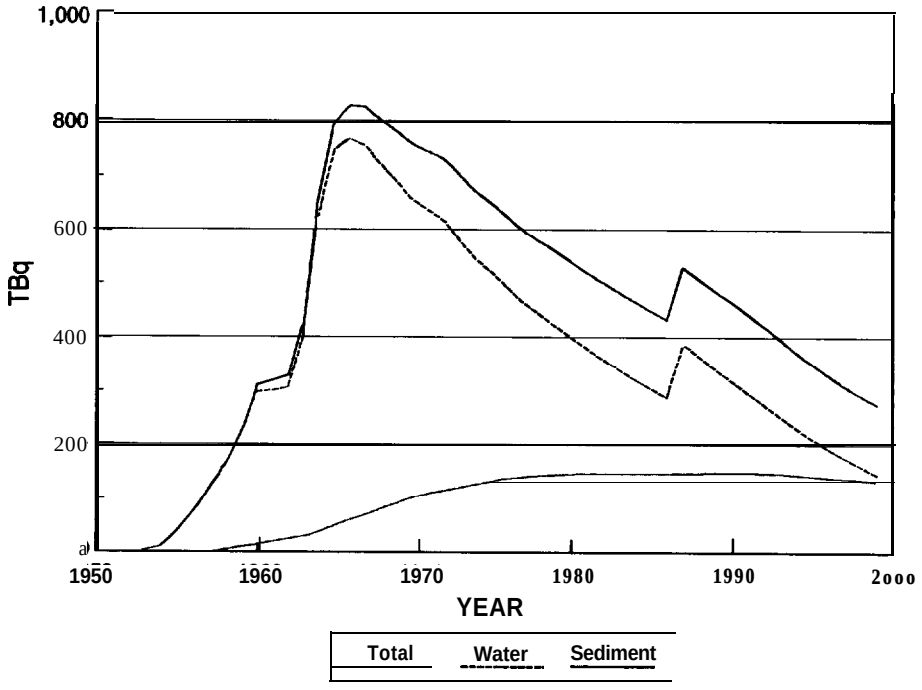
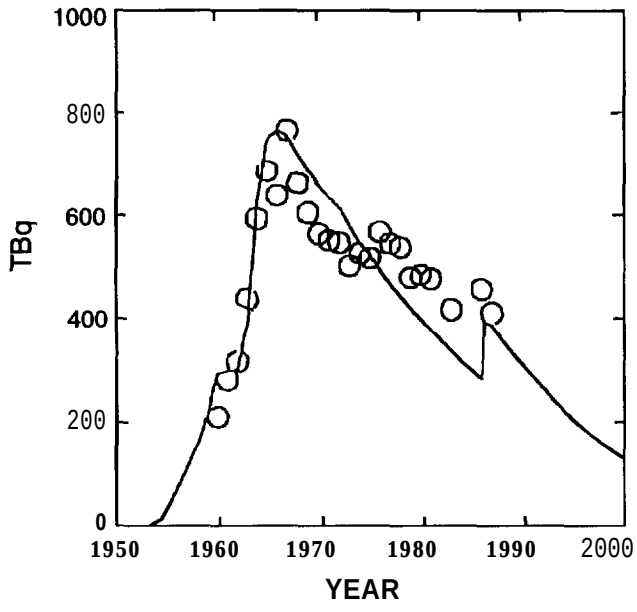


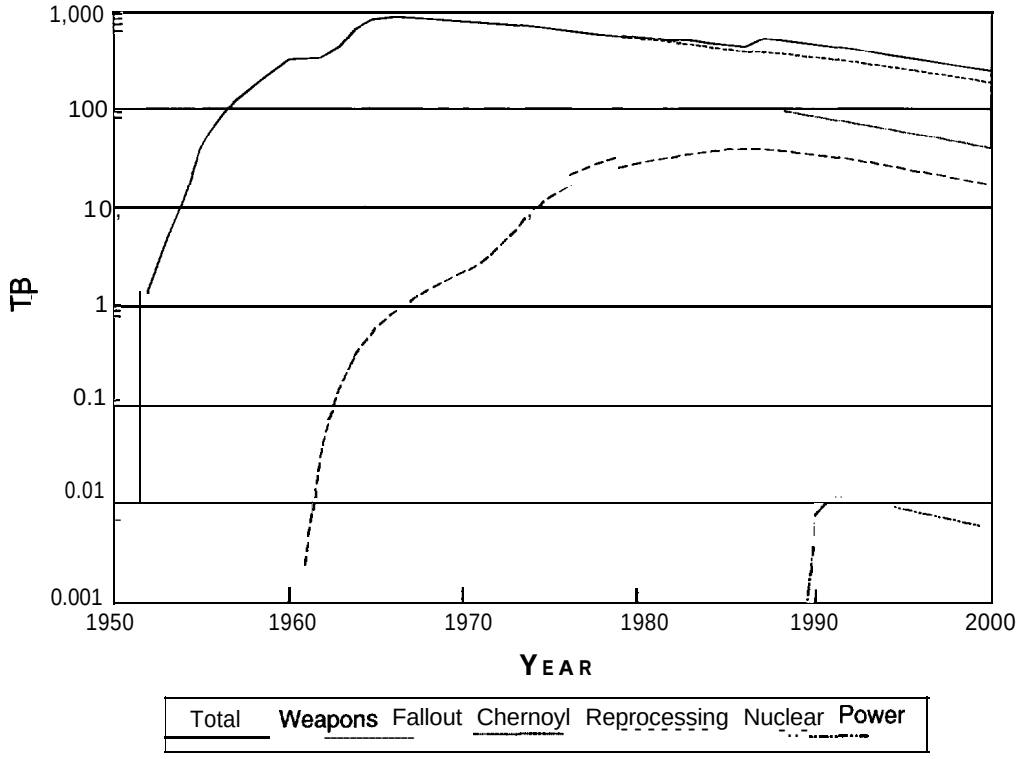
Figure 8.4.7 shows a comparison between the observed (data from Salo et al., 1985, and the present report) and predicted inventory of Sr-90 in the Baltic seawater. The agreement is good and the geometric mean of the P/O ratios is 1.0 with a geometric standard deviation of 1.2.

Figure 8.4.7. Comparison of predicted and observed inventories of Cs-137 in Baltic seawater. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.



The total inventory is shown in Figure 8.4.8 broken down according to the four sources: weapons fallout, Chernobyl fallout, reprocessing and nuclear installations. The dominating source is fallout from nuclear weapons testing. The inventory is estimated at a level of 370 TBq Sr-90 in 1994 of which 75 % originate from weapons fallout, 18 % from the Chernobyl accident, 7% from European reprocessing facilities, and 0.06% from nuclear facilities located in the Baltic Sea area.

Figure 8.4.8. Predicted inventory of Sr-90 (TBq) in the Baltic Sea broken down according to the sources shown as a function of time.



8.5. **Conclusions**

A model has been developed that enables assessments of the radiological consequences of releases of radioactivity into the Baltic Sea to be carried out. The 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 dispersion model is based on box-model analysis and includes 12 water and 12 sediment boxes for the Baltic Sea area. The physical processes covered by the model are net advection and mixing of water between adjacent boxes, sedimentation of particulate material from the water column to the top sediment, and 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 are calculated by the model from radionuclide concentrations in filtered seawater using recommended concentration factors.

The quality of the model predictions has been investigated by comparing predicted levels of Cs-137 and Sr-90 in water, top sediment and biota with observed levels. The observed data originate partly from the HELCOM database (Section 3.3) and partly from published material and cover the time period from 1960 to 1992. The sources of radioactivity into the Baltic region that have been considered are fallout from atmospheric nuclear weapons testing, fallout

from the Chernobyl accident in 1986, discharges of radionuclides from the two European reprocessing plants (Sellafield and La Hague) transported into the Baltic Sea, and discharges of radionuclides from nuclear installations located in the Baltic Sea area.

The comparisons have been made in terms of predicted-to-observed (P/O) ratios for which geometric means and geometric standard deviations have been calculated. The geometric mean of the P/O-ratio is an overall factor that indicates the predictive accuracy by how much the model overestimates the observed data (P/O-ratio higher than unity) or underestimates the observed data (P/O-ratio lower than unity). Similarly, the geometric standard deviation indicates the predictive precision by a single factor within which 68% of the values vary around the geometric mean value. From the geometric standard deviation another factor may be calculated that corresponds to the variability of 95 % of the values. The comparisons of the levels of Cs-137 and Sr-90 in the Baltic Sea are summarised in Table 8.5.1, which gives for the two radionuclides the factor of predictive accuracy and the factor of predictive precision at a 95 % confidence level.

Table 8.51. Summary of comparisons between model predictions and observations of Cs-137 and Sr-90 in different environmental compartments of the Baltic Sea expressed in terms of a factor of predictive accuracy (geometric mean of predicted-to-observed ratios) and factor of predictive precision corresponding to a 95% confidence level.

Environmental compartment	cs-137		Sr-90	
	Factor of predictive accuracy	Factor of predictive precision	Factor of predictive accuracy	Factor of predictive precision
Water	1.1	1.8	0.8	1.9
Sediments	0.3	5.9	2.6	6.4
Biota	1.7	5.3	0.9	20
Inventory Water Sediment	 1.2 0.9	 1.5 2.9	1.0	1.4

The factor of predictive precision thus indicates the overall agreement between individual values, but gives no information of possible bias. The factor of predictive accuracy indicates the possible bias expressed as the overall agreement between the model predictions and observations.

The predicted water concentrations are generally in good agreement with the observations for both radionuclides; the factor of predictive accuracy is close to unity and the factor of predictive precision is below 2. For the top sediment concentrations the agreement is less favourable; for Cs-137 the model underestimates the observed levels with about a factor of 3 and for Sr-90 the model overestimates with about a factor of 3, and the factor of predictive precision is about 6 for both isotopes. For the biota data the comparison shows that the model

overestimates the Cs-137 levels with about a factor of 1.7 combined with a factor of predictive precision of about 5, whereas the predictive accuracy for Sr-90 is fine but with a very large factor of predictive precision of 20. The predicted water inventory data compare well with the observed data for the two radionuclides both in terms of predictive accuracy and predictive precision. For the Cs-137 sediment inventory data the predictive precision is also good, but not as good as for the water data.

The inventories of Cs-137 and Sr-90 in the Baltic Sea in 1994 have been estimated at 3200 TBq and 370 TBq, respectively. For both radionuclides most of the activity is in the water while the rest is in the bottom sediments. For Cs-137 the inventory is due mainly to fallout from the Chernobyl accident (84%), while the rest originates from nuclear weapons fallout (11%), discharges from European reprocessing plants (5 %) and discharges from nuclear facilities located in the Baltic Sea area (0.01 %). For Sr-90 the main source of contamination is fallout from nuclear weapons testing (75%), while the rest is due to fallout from the Chernobyl accident (18%), discharges from the European reprocessing plants (7%) and discharges from nuclear facilities located in the Baltic Sea area (0.06%)

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9. DOSE CALCULATIONS

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The model described in Chapter 8 has been used for an assessment of doses to man via the ingestion pathway from the radionuclides Cs-137 and Sr-90 present in the Baltic Sea covering the time period from 1950 to 1992. The assessment is not complete since other radionuclides and other exposure pathways contribute to the exposure. But for the present purpose it has been necessary to restrict the assessment to the items mentioned. However, the dominating dose contribution to man from a single radionuclide and a single exposure pathway is believed to be due to the consumption of seafood contaminated with Cs-137. This was found to be the case in the Marina study (CEC, 1990) that evaluated the radiological impact of radioactivity in North European marine waters. The results from the present assessment are thus expected to approximate the doses from all radionuclides and all exposure pathways.

The assessment concerns dose rates (sieverts per year, Sv y^{-1}) to individual members of the public and collective dose rates (manSv y^{-1}) and committed collective doses (manSv) to members of the public in the Baltic Sea area for the time period 1950-1992 from the consumption of seafood contaminated with Cs-137 and Sr-90. Doses to man from radiation exposure are indicators of the associated health risk. At low dose rates the health risk is that of fatal cancer and the risk factor is $5 \cdot 10^{-2}$ Sv" (ICRP, 1991). The dose calculations are based on dose-per-unit-intake factors from Phipps et al. (1991).

9.1. Assumptions Concerning Seafood Consumption

For the calculation of doses to individual members of the public the rates of intake have been based on the assumptions made by the IAEA (1986) concerning the prevention of marine pollution by dumping of radioactive wastes and other radioactive matter. The individuals from a critical group are assumed to consume fish, crustacea and molluscs at rates of 0.3 kg d^{-1}, 0.1 kg d^{-1} and 0.1 kg d^{-1}, respectively.

With respect to doses to populations (collective doses), the total catches of fish, crustacea and molluscs were taken into account and assumed to be used for human consumption. The fishery statistics from 1991 were obtained from the International Council for the Explorations of the Seas (ICES, 1993) and are reproduced in Table 9.1.1. The fish catch is dominated by the species herring (60%), cod (20%), sprat (16%), and the remainder concerns flounder, salmon, dab, trout, turbot and plaice. For the calculations no distinction has been made between fish species. The total landings shown in Table 9.1.1 have been used for the calculations combined with relative fractions of the total landings assumed to be actually eaten by man. For these fractions the following values were used: one half for fish, one third for crustacea and one sixth for molluscs. It has furthermore been assumed that the annual harvest of marine produce has remained unaltered equal to that shown in Table 9.1.1 throughout the time period considered.

Table 9.1.1. Catches of fish, crustacea and molluscs (metric tonnes) in the Baltic Sea areas in 1991 (ICES 1993).

Area	Fish (t)	Crustacea (t)	Molluscs (t)
Kattegat	66000	988	375
Belt Sea	40989	143	11034
Baltic Sea West	215151		
Baltic Sea East	232638		
Bothnian Sea	30797		
Bothnian Bay	7739		
Gulf of Finland	39941		
Total	633255	1131	11409

9.2. Doses

9.2.1. Individual Dose Rates

Estimates of radiation doses to individuals of a critical group near sites of nuclear installations are required by national authorities. However, such local evaluations are beyond the scope of the present investigation that is concerned with consequences on a regional scale. But in order to illustrate consequences to individuals from the radionuclides and sources considered, dose rates to individuals have been calculated for each of the regions covered by the model based on rates of intake of seafood described in the previous section. Other exposure pathways than ingestion of seafood contribute (eg. external radiation from beach exposure, inhalation of resuspended sediment), but they have not been considered here.

Results of the calculations are shown in Figures 9.2.1 and 9.2.2 showing for the Bothnian Sea the dose rates by source and by radionuclide, respectively. The total dose rates were lower in all other regions of the Baltic Sea than those in the Bothnian Sea. The dose rate is noted to be dominated by Cs-137 throughout the period; the contribution from Sr-90 is estimated to range from one to two orders of magnitude below that from Cs-137. Until 1985 the dose rate to individuals is determined by atmospheric fallout from nuclear weapons testing and peaks in the 1960's at about 13 $\mu Sv\ y^{-1}$. The Chernobyl accident in 1986 is predicted to have further raised the dose rate to individuals about one order of magnitude to about 150 $\mu Sv\ y^{-1}$, and this source of contamination is predicted to dominate the future doses. From 1990 and onwards the dose rate from weapons fallout is estimated to approximate that from the European reprocessing plants Sellafield and La Hague at about one order of magnitude below the dose rate from the Chernobyl accident. Furthermore, the dose rate from nuclear installations in the Baltic Sea area is estimated to range about four orders of magnitude below that from the Chernobyl accident. It should be noted that liquid discharges of Cs-137 and Sr-90 from nuclear installations have only been considered since 1984 although some installations were in operation earlier. However, such discharge data prior to 1984 were not available for this work.

Figure 9.2.1. **Dose rates (Sv y^{-1})** to an individual member of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Bothnian Sea according to sources.

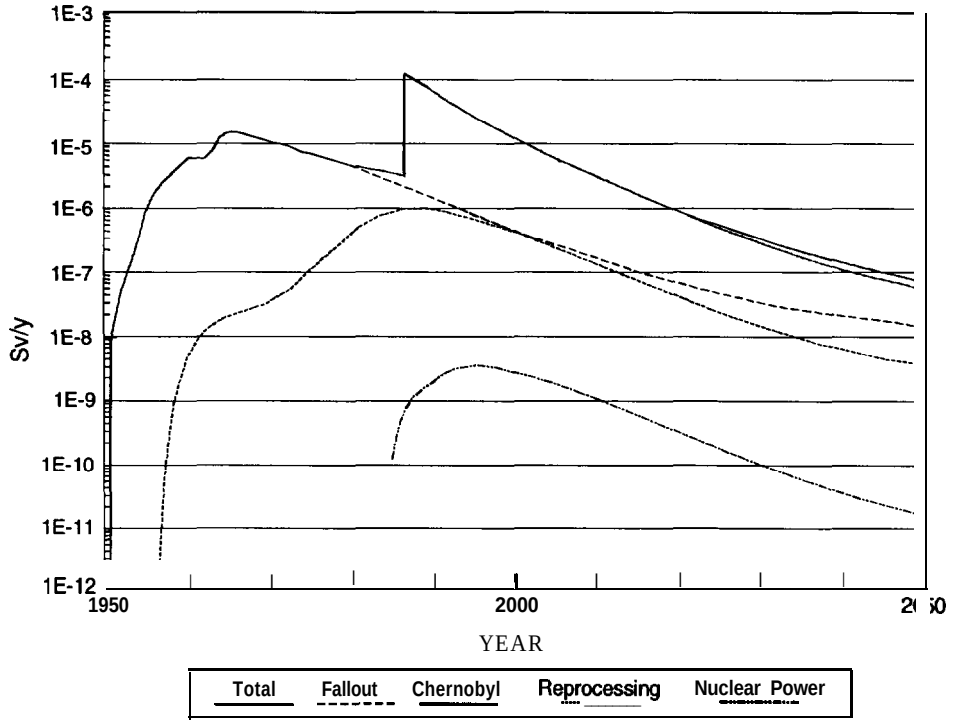
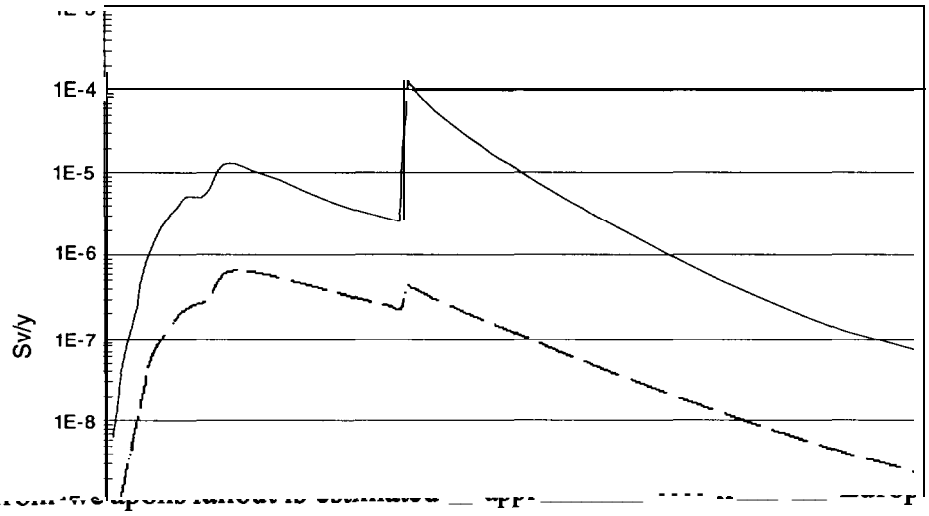


Figure 9.2.2. Dose rates (Sv y^{-1}) to an individual member of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Bothnian Sea.



plants Sellafield and La Hague at about one order of magnitude below the dose rate from the Chernobyl accident. Furthermore, the dose rate from nuclear installations in the Baltic Sea area is estimated to range about four orders of magnitude below that from the Chernobyl accident. It should be noted that liquid discharges of Cs-137 and Sr-90 from nuclear installations have only been considered since 1984 although some installations were in operation earlier. However, such discharge data prior to 1984 were not available for this work.

9.2.2. Collective Dose Rates and Collective Doses

The present study does not include an identification or grouping of the human populations exposed to radioactivity from ingestion of seafood from the Baltic Sea area, and assumes that all marine produce is consumed by man. However, it may be assumed that the main part of the seafood is consumed by members of the public in the countries bordering the Baltic Sea and that only a small fraction is exported to other countries.

Calculations of collective dose rates and doses to man from consumption of seafood from the Baltic Sea area were made based on calculations with the model and source terms outlined in Chapter 8 and the assumptions mentioned in Section 9.1. The source terms comprise releases of Cs-137 and Sr-90 to the environment from the Chernobyl accident, nuclear weapons testing, the two European reprocessing plants Sellafield and La Hague, and nuclear installations bordering the Baltic Sea. Releases have been considered for the time period from 1950 up to the end of 1992.

The results for the collective dose rates are shown in Figures 9.2.3 and 9.2.4 giving the collective dose rates broken down in components according to sources and nuclides. The graphs show the same qualitative relationship between the different components of the collective dose rates as for the individual dose rates.

Figure 9.2.3. Collective dose rates **(manSv y^{-1})** to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area according to sources.

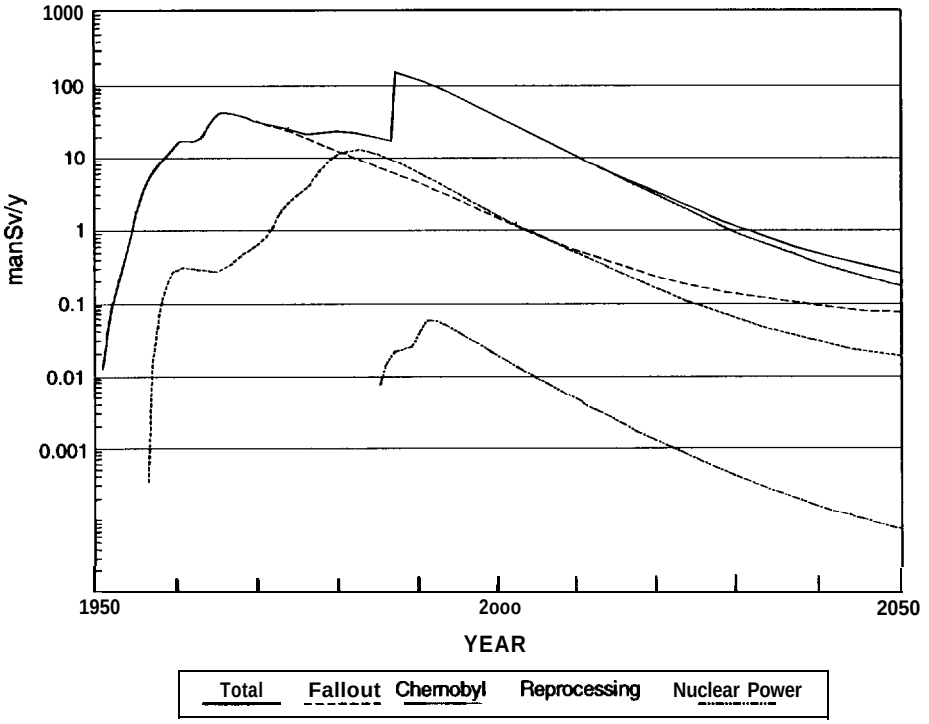
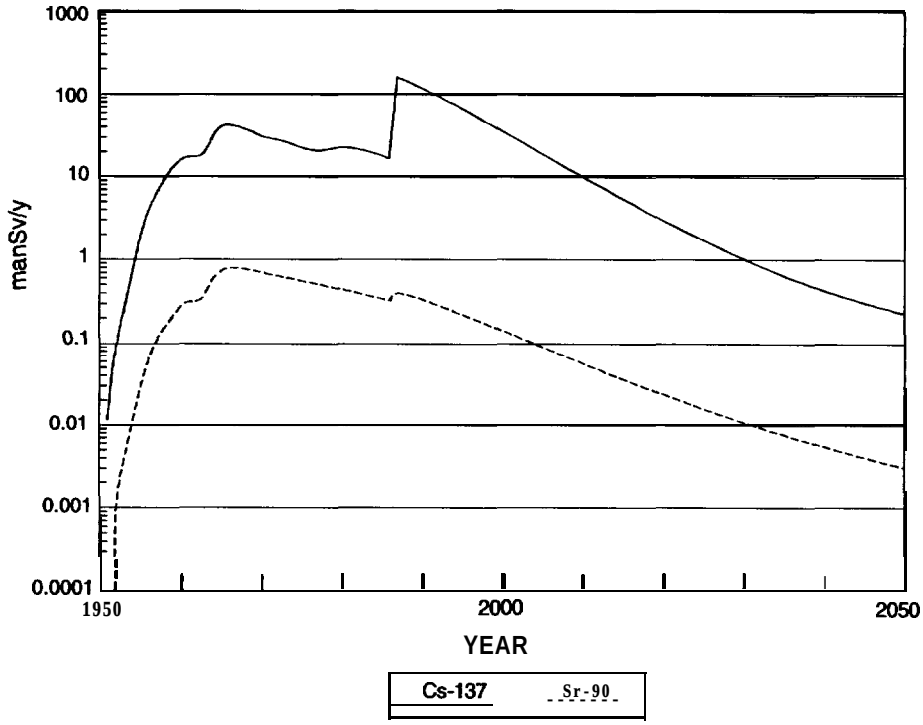


Figure 9.2.4. Collective dose rates **(manSv y^{-1})** to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area.



The collective dose rate is estimated to reach a maximum of about 42 **manSv y^{-1}** in the **1960's** before 1986 where the collective dose rate peaks at about 160 **manSv y^{-1}** due to the Chernobyl accident. The time course of the dose rate from weapons fallout is predicted to be reduced at a rate slower than that from the other sources after year 2000 due to inflow of radionuclides to the Baltic Sea from the North Atlantic via the North Sea.

The collective committed doses which are the time-integrated collective dose rates are shown up to the year 2050 in graphical form in Figures 9.2.5 and 9.2.6. The collective committed doses are given in numerical form in Table **9.2.1.** The collective committed dose from Cs-137 and Sr-90 is thus estimated at 2300 **manSv** of which about 1% is estimated to be due to **Sr-90** and the rest to Cs-137. The main contribution of the dose from Sr-90 is due to fallout from nuclear weapons where transfer from the catchment area to the sea via river runoff gives a significant contribution (cf. Chapter 8).

Figure 9.2.5. Committed collective doses (manSv) to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area according to sources.

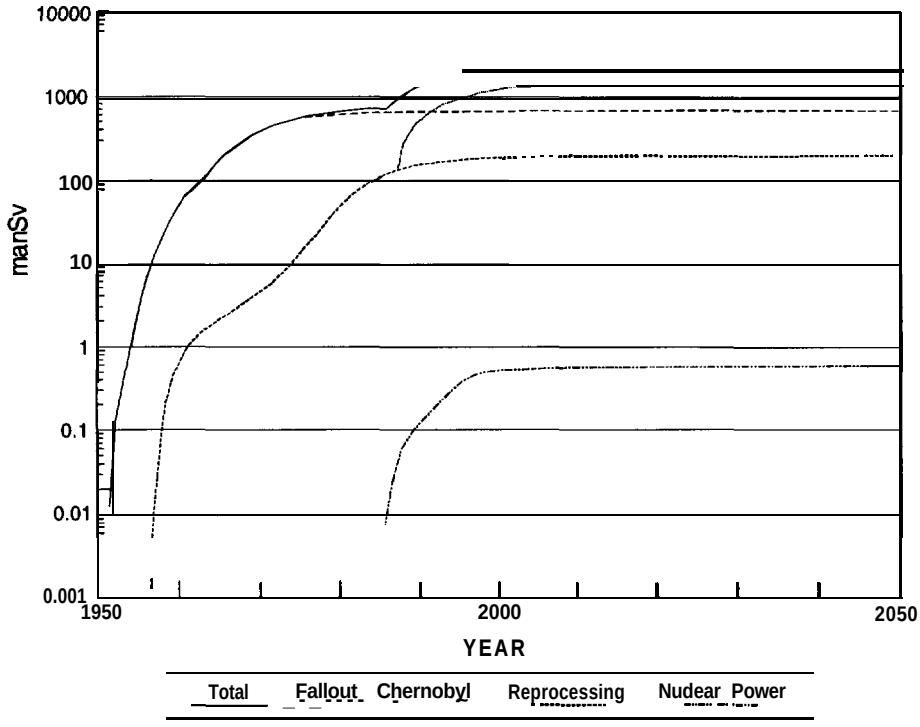


Figure 9.2.6. Committed collective doses (manSv) to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area.

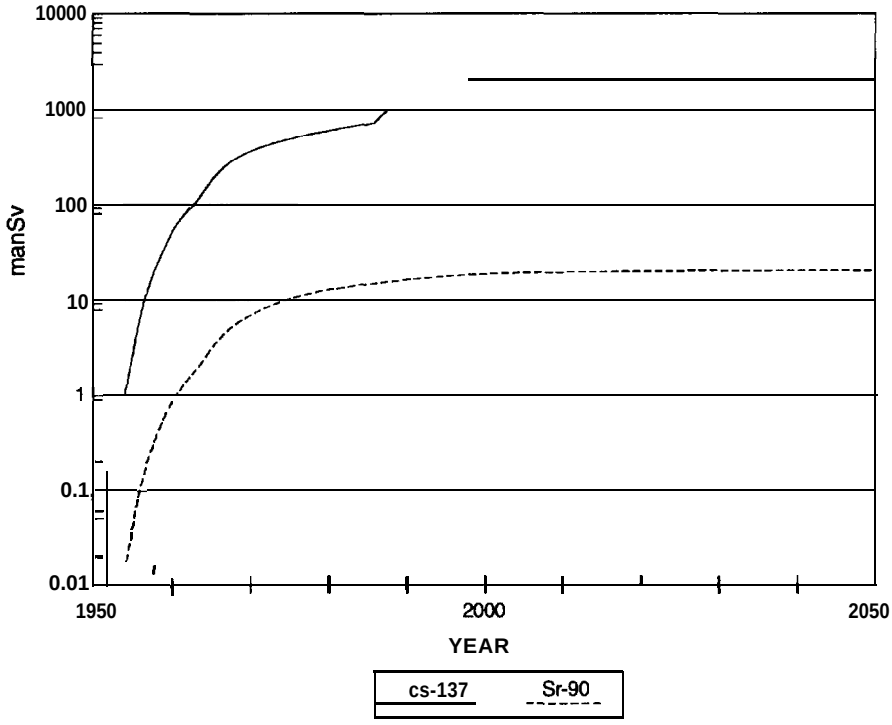


Table 9.2.1. Collective committed doses to members of the public from ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area calculated for up to the year 2050.

Source	Cs-137 (manSv)	Sr-90 (manSv)	Total (manSv)
Chernobyl	1387	1	1388
Weapons fallout	650	18	668
Reprocessing plants	194	1	195
Nuclear installations	0.2	0.005	0.2
Total	2232	20	2252

The dominating source to the collective committed dose is the fallout from the Chernobyl accident which is estimated to contribute 1400 manSv corresponding to about 60% of the total. Then follows the atmospheric fallout from nuclear weapons testing estimated to give rise to a collective dose of about 700 manSv corresponding to about 30% of the total. Discharges from European nuclear reprocessing plants are estimated to cause a committed collective dose of 200 manSv in the Baltic Sea area corresponding to about 10% of the total, and discharges from nuclear installations bordering the Baltic Sea area are estimated to cause 0.2 manSv corresponding to about 0.01% of the total.

In the Marina project (CEC, 1990) estimates were made of collective doses from ingestion of Cs-137 in seafood from Chernobyl fallout into the Baltic Sea. Two methods were applied, one based on mean residence times of conservative substances and radiocaesium inventories in the seawater and another using a box model similar to the present one, but without considering sedimentation. The committed doses were estimated at 2400 manSv and 3800 manSv by the two methods, respectively. These values are significantly higher than the present estimate of 1400 manSv. But if one considers the differences between the assumptions made (source term, mean residence time and fishery statistics), the first method agrees well (within 15%) with the present estimate. The second method still gives an estimate well (about a factor of two) above the present one, but this difference is explained by lack of sedimentation in that box model.

Furthermore, it should be noticed that the present collective dose estimate from the Chernobyl accident is probably on the high side. The dose depends mainly on the time course from 1986 and onwards of the concentration of Cs-137 in the Baltic seawater. From the comparison made in Chapter 8 of the measured and calculated inventories of Cs-137 in the Baltic seawater it is noted, that the model underestimates the rate of reduction after the Chernobyl accident in 1986. The rate of reduction of the model corresponds to a mean residence time of 8 years whereas the rate of reduction from the observed data corresponds to a mean residence time of 3 years ($\pm$ 25 % , 1 sd). However, the observed data cover five years only, and a longer time of observation is needed for a more reliable estimate of the rate of reduction which additionally may not be adequately described by a single exponential function (a mean residence time) but rather by two exponential terms. So in order to improve the estimate of the collective doses from seafood ingestion of Cs-137 from the Chernobyl accident it is important that the observed data for Cs-137 in Baltic seawater are updated and followed for a longer time period for a more reliable estimate of the rate of reduction.

9.3. Comparison with Doses from Natural Radionuclides

Naturally-occurring radionuclides in seawater are incorporated into marine organisms and give rise to radiation exposure to humans from ingestion of seafood. Polonium-210 is the major contributor in this context due to the high bio-accumulation in marine organisms and the relatively high dose factor. Typical PO-210 concentrations in fish from the Baltic Sea are 0.8 Bq kg^{-1} (Bojanowsky, 1994; Dahlgaard, 1994; Holm, 1994), in crustacea 20 Bq kg-' (Swift et al., 1994) and in molluscs 30 Bq kg-' (Dahlgaard, 1994). From the same assumptions of seafood intake as in the preceding sections and using a dose factor of $6.2 \cdot 10^{-7}$ Sv Bq^{-1} (Phipps et al., 1991) one may calculate a dose rate of 1.2 $mSv\ y^{-1}$ to the individual member of the public from PO-210. This value is about an order of magnitude higher than the peak dose rate from Cs-137 and Sr-90 in 1986 calculated for an individual member of the public eating seafood from the Bothnian Sea. Similarly, the collective dose rate from PO-210 may be calculated using the data on fishery statistics in Table 9.1.1 resulting in a value of about 200 $manSv\ y^{-1}$. This is slightly higher than the peak value of the collective dose rate from Cs-137 and Sr-90 in 1986 (160 $manSv\ y^{-1}$).

9.4. Conclusions

Doses have been calculated to members of the public from the ingestion of the radionuclides Cs-137 and Sr-90 in seafood produced in the Baltic Sea area. The calculations cover the time period from 1950 to 1992 and include source contributions from nuclear weapons testing, the Chernobyl accident, the two European reprocessing plants Sellafield and La Hague, and nuclear installations bordering the Baltic Sea area. The results are based on calculations with the box model described in Chapter 8.

Dose rates from Cs-137 and Sr-90 to individual members of the public (a critical group) have been calculated based on rates of daily intake (0.3 kg fish, 0.1 kg crustacea and 0.1 kg molluscs). The dose rates to individuals from the ingestion of seafood from the Bothnian Sea are predicted to be larger than from any other area in the Baltic Sea due to the pattern of deposition of fallout from the Chernobyl accident. The dose rate is predicted to peak in 1986 at a value of 0.1 $mSv\ y^{-1}$.

Collective dose rates and doses to members of the public have been calculated based on predicted concentrations of Cs-137 and Sr-90 in biota and fishery statistics. The collective dose rate peaks in 1986 at a level of 160 $manSv\ y^{-1}$ due to fallout from the Chernobyl accident. The total collective dose from Cs-137 and Sr-90 in the Baltic Sea is estimated at 2300 manSv of which about 60% originates from Chernobyl fallout, about 30% from fallout from nuclear weapons testing, about 10% from European reprocessing facilities, and about 0.01% from nuclear installations bordering the Baltic Sea area.

Doses from naturally-occurring radioactivity in seafood (polonium-210) have been calculated on a similar basis and compared with the doses from Cs-137 and Sr-90. The results of this comparison are shown in the Tables 9.4.1 and 9.4.2 which summarise the calculated dose rates and doses. It is noted that dose rates and doses from natural radioactivity dominate except for the year 1986 where the collective dose rate from Chernobyl fallout approached that from natural radioactivity.

Table 9.4.1. Summary of maximum dose rates from Cs-137, Sr-90 and Po-210 to the population around the Baltic Sea.

Sources	Individual dose rate peak (year) (mSv y^{-1})	Collective dose rate peak (year) (manSv y^{-1})
Chernobyl fallout	0.15 (1986)	160 (1986)
Nuclear weapons fallout	0.013 (1965)	42 (1966)
European reprocessing plants	0.0009 (1988)	12 (1982)
Baltic nuclear installations	0.000004 (1991)	0.02 (1987)
Natural radioactivity (Po-210)	1.2	200

Table 9.4.2. Collective exposure of the population around the Baltic Sea for the period from 1950 to 2050 from Cs-137, Sr-90 and Po-210 in Baltic seawater.

Sources	Collective dose to year 2050 (manSv)
Chernobyl fallout	1400
Nuclear weapons fallout	700
European reprocessing plants	200
Baltic nuclear installations	0.2
Natural radioactivity (Po-210)	20 000

The reliability of the model calculations of Cs-137 in biota, which are important for the dose calculations, was tested in Chapter 8 which showed a predictive accuracy slightly less than 2. No formal uncertainty analysis has been carried out, but it is estimated that the predictive accuracies of the calculated dose rates and doses are about a factor of 3.

The present work represents a first step towards a full assessment of the exposure of members of the public to radionuclides in the Baltic Sea. A full assessment should include a more comprehensive list of radionuclides and exposure pathways.

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10. CONCLUSIONS

This report is based on the information and data submitted from the Contracting Parties and comprises the most improtant aspects of artificial radionuclides in the Baltic Sea.

The main source with respect to the total inventory of artificial radionuclides in the Baltic Sea was the fallout after the Chernobyl accident in April 1986. The two major processes affecting the distribution of the fallout were the direct deposition onto the sea surface and the subsequent input from river run-off. The river run-off from 1987 onwards was less significant.

The second important source of radionuclides was the fallout from atmospheric weapon tests during the sixties. This source led to a more homogeneous distribution in the Baltic Sea. The dominating nuclides were Cs-137 and Sr-90 in an activity ratio of about 1.6.

Discharges from the reprocessing plants at Sellafield and La Hague influenced the concentration in the Baltic Sea by the inflow of contaminated saline water through the Danish Straits. The transport of contaminated waters from Sellafield at the Irish Sea coast to the Western Baltic takes about five years, the one from La Hague at the Channel takes between two to three years. However, due to the significant reductions of discharges from Sellafield and La Hague in recent years this source became of minor importance. Model calculations have shown that about 4% of the soluble radionuclides from Sellafield and about 8 % from La Hague were transferred to the Kattegat and only about 1% of the Sellafield discharge will reach the Baltic Proper.

The least significant source of artificial radionuclides are the discharges from the operation of nuclear installations on the drainage area of the Baltic Sea. Most of the nuclides from these sources are detectable only in the vicinity of the point of release.

Comparing these four sources of radionuclides for Cs-137 and Sr-90, one comes to the following quantitative total inputs into the Baltic Sea, decay corrected to the year 1991:

	cs-137 TBq	Sr-90 TBq
Global fallout	900	600
Chernobyl fallout	3700 - 4600	80
Western European Reprocessing	250	40
Nuclear Installations	0.6	0.3

In order to monitor the radioactive substances in the Baltic Sea the Contracting Parties to the Helsinki Convention set up a monitoring programme which covers water, sediment, suspended particulate matter, fish, aquatic plants and benthic animals in different sub-areas of the Baltic Sea.

The main emphasis has been on the analyses of gamma-emitters in seawater, sediment and fish. After the Chernobyl fallout there was a substantial increase in the number of analyses of samples from all compartments.

The environmental data on radionuclides in seawater and sediments submitted to the HELCOM database cover the whole area of the Baltic Sea. However, most of the sampling locations of fish are concentrated to the Southern regions. The present monitoring programme is considered to be adequate to fulfil those objectives of the HELCOM/MORS work that deal with the collection of data on radionuclides in the Baltic Sea, the evaluation of the results and assessment of radiation doses to the population living around the Baltic Sea. However, future changes to the monitoring programme should take into consideration that the number of sampling locations for radionuclides in sediments is much smaller than that for water and thus put a stronger focus on samples of sediments. The transfer of radionuclides from the water column to the sediments and the subsequent resuspension of activity back to the water column are processes of importance for the evaluation of radionuclide inventories and the prediction of long-term radiation doses to man.

Due to the monitoring programme any significant release of radioactivity from RTG's (Radionuclide Thermoelectric Generators) used in lighthouses in the former USSR or releases from dumping of radioactive waste would have been detected. There is no evidence of releases from any of these RTG's during the period of this report.

In addition to the environmental data on radionuclides, data are collected on releases from nuclear installations. The monitoring programme will detect any radioactive release resulting in a significant impact to the Baltic Sea.

Within the monitoring work special emphasis has been given to the reliability of data. During 1984-1991, nine intercomparison exercises were organized by the International Atomic Energy Agency - Marine Environment Laboratory (IAEA-MEL). One Baltic seawater exercise (IAEA-299) was specially designed for the laboratories of the HELCOM-MORS group while others were organized on a worldwide scale. The data reported by the laboratories of the Contracting Parties compared with the reference values established through the worldwide intercomparison exercises are very satisfactory, especially for Cs-137 and Pu-239,240. This shows that the data produced by laboratories of the MORS group are of high quality.

Comprehensive studies on seawater were made during the evaluation period, 1984- 1991. After the Chernobyl accident the concentration of Cs-137 increased by several orders of magnitude in some areas. The highest concentrations of Cs-137 were detected in the Gulf of Finland and the Southern Bothnian Sea. The initial heterogeneous distribution of the deposition of Cs-137 in water was levelled during the following years up to 1991 due to mixing of water masses. However, in 1991 levels about 200 Bq m^{-3} were still detected in the southern part of the Bothnian Sea compared to about 100 Bq m^{-3} in the Baltic Proper.

The Cs-137 inventory in Baltic seawater increased in 1986 by more than a factor of 10, and decreased by sedimentation and outflow to about half that value by 1991. In 1991 the Cs-137 concentrations in the Baltic Sea were significantly higher compared to those of the other shelf seas in the world, e.g. the central North Sea, where the levels were about 20 Bq m^{-3}.

Bottom sediments play an important role in radioecological studies of the marine environment, because a large proportion of radioactive substances entering the water is adsorbed on suspended particulate matter and deposited to the sediments.

As early as in June 1986 a clear increase of Chernobyl derived Cs-137 was observed in the sediments at some sampling stations. During the whole period the largest sedimentation of Cs-137 took place in the eastern part of the Gulf of Finland and in the Bothnian Sea. This was in good agreement with the distribution pattern of Cs-137 deposition in the drainage area of the Baltic Sea. In 1990 the highest total depositions of Cs-137 in sediments were 43 $kBq\,m^{-2}$ in the Eastern Gulf of Finland and 21 $kBq\,m^{-2}$ in the Bothnian Sea.

The total inventory of Cs-137 in the Baltic Sea sediments in 1991 was estimated to 1400 TBq and that of Pu-239,240 to 18 TBq. The corresponding values estimated at the beginning of the 1980's were 277 and 15 TBq, respectively.

Generally, the main pathway of radiation exposure to humans from contamination of the marine environment is given by consumption of marine fish. Therefore, special emphasis was given to follow the contamination in Baltic Sea fish. Consequently, after the Chernobyl accident the most important radionuclides measured in biota samples were Cs-137 and Cs-134. The results for fish, mainly cod, herring, flat fish and pike, clearly showed that they reflected the distribution pattern of the Chernobyl fallout. The temporal evolution of Cs-137 especially in the south western sub-areas of the Baltic clearly demonstrated the influence of slow circulation of surface water from the Gulf of Bothnia as well as in the Baltic Proper and the water transport southward along the Swedish coast.

Benthic animals showed radionuclides from aquatic discharges from nuclear power plants, e.g. Co-60 and Ag-110m. This could also be demonstrated for the bladder wrack, *Fucus vesiculosus*.

For use in model calculations, concentration factors were calculated for the main fish species for the radionuclides Cs-137 and Sr-90. The concentration factors varied between different areas of the Baltic Sea as expected according to different salinities. Average Cs-137 concentration factors (fish flesh) varied between 140 and 1120 for pike (fresh water fish) and between 30 and 500 for marine species. Average Sr-90 concentration factors (fish flesh) ranged from 1.3 to 3.4 for pike and between 0.2 and 0.8 for marine fish. The concentration factors for marine fish from higher salinity waters are in reasonable agreement with the concentration factors given by IAEA.

A box model covering the North-East Atlantic waters including the Baltic Sea has been used to calculate the dispersion of the radionuclides Cs-137 and Sr-90 in the Baltic Sea considering the four sources previously mentioned. The model was used to predict concentrations of Cs-137 and Sr-90 in seawater, sediments and biota in addition to radionuclide inventories for the time period 1950-2000. The model includes simulation of hydrodynamical transport, partitioning of radioactivity between water and particulate matter, interaction with sediments, and transfer of radioactivity to the marine fauna and to man.

Comparisons were made between predicted and observed values of radionuclide concentrations

in seawater, sediments and biota in addition to the inventories in water and sediments. Overall it may be concluded from this comparison that the reliability of the model predictions is good considering that the predictive accuracy of the model is better than a factor of 2.

The model has been used to show the temporal trend of the inventories of Cs-137 and Sr-90 in the Baltic Sea separated into the components from the four above mentioned sources. The calculations show that the Cs-137 inventory in the Baltic Sea until 1985, where it amounted to about 1 PBq, was dominated by atmospheric fallout from nuclear weapons testing. After the Chernobyl accident in 1986 the inventory rose to a value of about 5 PBq, and this input has dominated the inventory since that time. The contribution to the inventory of Cs-137 in the Baltic Sea from the nuclear reprocessing plants Sellafield and La Hague is calculated to peak in 1985 at a value of about 0.3 PBq after which this component has been decreasing. The 0.3 PBq of Cs-137 corresponds to 1% of the total discharge of Cs-137 from the Sellafield facility for the time period considered. Furthermore, the calculations show that the inventory of Cs-137 from nuclear facilities bordering the Baltic Sea remains below about 1 TBq or more than three orders of magnitude below the levels from nuclear weapons fallout and Chernobyl fallout.

For Sr-90 the calculated inventories show a corresponding picture with the exception that the Sr-90 fallout from the Chernobyl accident contributed little to the total inventory compared to Cs-137. The inventory of Sr-90 in the Baltic Sea is thus dominated by atmospheric fallout from nuclear weapons testing for the whole time period considered, presently at a level of about 400 TBq. The inventory of Sr-90 in the Baltic Sea from the nuclear reprocessing plants Sellafield and La Hague peaks in 1986 at a value of about 40 TBq which again corresponds to about 1% of the total Sellafield discharges for the time period covered. The inventory from the nuclear facilities bordering the Baltic Sea contribute very little to the total inventory. The peak value is reached in 1991 at a value of about 0.01 TBq which is more than four orders of magnitude below the inventory from weapons fallout.

Radiation doses to man from consumption of seafood contaminated with Cs-137 and Sr-90 in the Baltic Sea have been calculated. The doses were calculated 1) in terms of dose rates to individuals from a critical group assumed to consume seafood from the area of the Baltic Sea with a high level of radioactive contamination and at a high rate of intake and 2) in terms of collective committed doses (and dose rates) to populations at an unspecified number of people assumed to consume the total seafood catch from the Baltic Sea (about 0.6 mill. tons fish per year).

The calculated dose rate to an individual from the critical group is analysed in terms of the relative contributions from the two radionuclides and from the four sources considered. The dose rate is clearly dominated by the contribution from Cs-137 which is one to two orders of magnitude higher than that from Sr-90. This is mainly due to the difference between the biological uptakes in fish of the two radionuclides from the water. For the four sources considered it is found that the dose rate from nuclear weapons fallout dominates until the year of the Chernobyl accident in 1986 after which the latter dominates. The maximum dose rate occurs in 1986 at a value of about 0.2 $mSv\ y^{-1}$. The corresponding annual dose from natural radioactivity in seafood (polonium-210) is about 1.2 mSv, or 6 times higher. Comparing the doses received by marine contamination with those from terrestrial pathways which range

from two to five mSv y^{-1} in the countries bordering the Baltic Sea, one can conclude that the additional radiation exposure to the critical group due to the consumption of Baltic fish was less than 10% in 1986. At present the marine pathway is only in the order of 1% of the terrestrial pathway of the average population and should, therefore, be of no concern. The dose rate from radionuclides discharged from the nuclear reprocessing plants Sellafield and La Hague reaches a maximum in 1990 at a value similar to that from the weapons fallout. The dose rate contribution from the nuclear facilities bordering the Baltic Sea is about two orders of magnitude lower than the contribution from nuclear weapons fallout.

The total collective committed dose to man from intake of seafood from the Baltic Sea contaminated with Cs-137 and Sr-90 is estimated at 2300 mansieverts (manSv). About 60% of this dose originates from the Chernobyl fallout, about 30% from nuclear weapons fallout, about 10% from European reprocessing facilities, and about 0.03% from nuclear installations bordering the Baltic Sea. Releases of radioactivity to the Baltic Sea area were considered for the time period 1950-1991, and doses were calculated for 100 years until the year 2050. For the same period of time the collective doses to man from intake of natural radioactivity in seafood is estimated at 20,000 manSv or about 10 times greater than the doses from Cs-137 and Sr-90. The maximum annual collective dose from Cs-137 and Sr-90 occurred in 1986 at about 160 manSv which is close to the annual collective dose from natural radioactivity in seafood of 200 manSv.

The present work represents a first step towards a full assessment of the exposure of members of the public to radionuclides in the Baltic Sea. Future work should include the full range of radionuclides discharged from the nuclear facilities bordering the Baltic Sea (e.g. also H-3, Co-60, Zn-65, Ag-110m, Sb-125) and other exposure pathways than that of ingestion (e.g. external exposure on beaches and inhalation). Furthermore, the modelling would benefit from improvement with respect to hydrodynamic water transport and transfer of radioactivity to the sediments.

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ANNEX

QUESTIONNAIRE REGARDING MONITORING OF RELEASES AND CORRESI'ONDING QUALITY ASSURANCE FOR NUCLEAR INSTALLATIONS IN THE BALTIC SEA AREA

INTRODUCTION

Among the subjects to be studied by the EC MORS-group is the compiling and subsequent reporting of releases of radioactive substances from nuclear installations in the Baltic region. It has been recognized by the Group that additional information on how these releases have been measured would be useful for further evaluation of these data. It has therefore been agreed that this questionnaire should be sent out in order to facilitate such a compilation of information. It has also been recognized that by adding some extra questions to monitoring practices about release limits and organizational structures this compilation could provide a basis to the Contracting Parties to the Helsinki Convention, if so desired, to evaluate whether or not further improvement of applied practices could be worthwhile.

INSTALLATIONS

Give a list of all nuclear installations in the catchment area of the Baltic Sea in your country that releases significant amounts of radioactive substances to the Baltic Sea region.

REGULAR ASPECTS

a) Name of responsible authority

b) Principles for release limit (i.e. given i Bq/nuclide or nuclide group or dose to a critical group etc.) and specify release limits for airborne and liquid releases. Indicate if limits is given per installation or per unit.

c) Describe the reporting procedure (to whom, frequency and by whom)

d) Describe the authorities role for control of releases (i.e. measurement of its own and/or supervised by authority.

f) Describe the procedure for the archives of data and samples.

MONITORING SYSTEMS

The following information should be given for each unit at all installations indicated in the Installations section.

Releases to air

a) Give name(s) of installation(s) and unit(s) for which the description below is valid

b) Give a general description of ventilation systems and the number of release points.

c) Give a general description of the monitoring systems (if necessary, specified for each release point).

d) Describe separately how the following groups of nuclides are sampled, including sampling periods, and describe the corresponding analyzing procedure:

1. gamma emitting noble gases
2. gamma emitting halogens
3. gamma emitting particulate
4. alpha emitting nuclides
5. strontium-90
6. tritium
7. carbon-14

e) Indicate if other nuclides with complex analyzing procedures like I-129, Fe-55 and Cs-135 are considered and if so how (by extrapolation from other nuclides etc.)

f) Describe the calibration procedures for the monitoring system itself (i.e if based on injection experiments or based on flow relations only) and for the different analyzing procedures for the above mentioned nuclide groups.

g) Give average detection limits for the following nuclides in Bq/year assuming typical release rates:

Kr-88
Xe-133
I-131
co-60

Releases to water

a) Give name(s) of installation(s) and unit(s) for which the description below is valid

b) Give a general and brief description of the waste water treatment systems.

c) Describe the procedures for the discharges and corresponding sampling, including frequencies and quantities.

d) Describe the sample analyzing procedures for the following groups of nuclides, specified for particulate and soluble fractions, if separated.

1. gamma emitting nuclides
2. alpha emitting nuclides
3. strontium-90
4. tritium
5. carbon-14

e) Indicate if other nuclides with complex analyzing procedures like I-129, Fe-55 and Cs-135 are considered and if so how (by extrapolation from other nuclides etc.)

f) Describe the calibration procedures for the different analyzing procedures for the above mentioned nuclide groups.

g) Give average detection limits for the following nuclides in Bq/l and in Bq/year with typical waste water release quantities.

I-131
Co-60
Zn-65
Mn-54

QUALITY ASSURANCE

Describe all major quality assurance activities within the installations, within the country and international ones, in form of education of staff, intercalibration exercises etc.

Compilation of Questionnaire Regarding Release Monitoring and Quality Assurance at Nuclear Installations in the Baltic Catchment Area

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Foreword

Answers to the questionnaire have been given by Denmark, Finland, Germany, Latvia, Poland and Sweden. Poland has no installations in the area and is not included in the compilation. Germany's only installation, Greifswald, has been shut down since 1990, but answers to the questionnaire were given by the German authorities anyhow and Germany is consequently included in the compilation.

Neither Russia nor Lithuania have replied and information from Sosnovy Bor and Ignalina NPP are thus missing. Some informal information of the conditions at these installations are included by the author, based on information achieved in the bilateral Swedish-Lithuanian and Swedish-Russian co-operation programme.

The compilation does not reflect the level of details and ambition on the replies. For detailed information, the replies as well as the questionannaire are given in the appendix. Detailed information are not repeated in the compilation, instead focus is given more to principles and general features.

This questionnaire has focused on the technical aspects of release monitoring. The philosophy for release reduction, i.e. whether you use the ALARA (As Low As Reasonably Achievable) principle, the BAT (Best Available Technology) principle or any other principle was not included in the questionnaire but could be evaluated in another study.

Results and Conclusions

The compilation shows that similar principles for release limits are used in the different countries, although the implementation procedures are slightly different. The limits are based on dose to a critical group with derived activity limits. The latter limits could either correspond to single nuclides or groups of nuclides.

The release monitoring is normally carried out by the licensee, except in Germany where in addition a remote monitoring system operated by the authorities exists. The role of the authorities in general is to set up guidelines for the quality of the systems and to carry out some control measurements.

The principles of monitoring are quite similar in the different countries. The airborne release monitoring systems normally involve two ways of function, one on-line monitoring function

with relatively simple instrumentation and a more sophisticated system for sampling and averaging during a longer time period (typically a week) with spectroscopic analysing equipment. Similar principles go for the aquatic releases, with a sample collection prior to the release to determine if more cleaning is necessary and then collection of a large sample during release or prior to the release for detailed activity assessment. The reports to authorities are normally based on the latter type of analysis.

The degree of sophistication naturally varies with the type of facility. The power production units normally have more expensive equipment, with a higher degree of automatic functionality. The quality assurance procedures are also more developed at these installations.

As an overall conclusion one can say that similar principles for protection of the public are used and that the quality of the equipment and procedures for release monitoring corresponds to the actual and potential environmental impact that these releases could give rise to.

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
INSTALLATIONS	Research laboratory (Risø, 1 reactor)	2 BWR, 2 PWR Loviisa, TVO	4 PwR (shut down 1990)	Research laboratory	9 BWR, 3 PWR, research laboratory (Studsvik, 1 reactor)	
REGULAR ASPECTS						
a. Authorities involved	2	1	2 national and 1 regional (Mecklenburg-Vorpommern)	2	2	VATESI
b. Principles for release limits	Dose limit 0.1 mSv/a per installation (related to safety rather than routine releases). Derived laboratory and nuclide specific activity limits.	Dose limit 0.1 mSv/a per installation and 5 manSv/GW. Derived site and nuclide group (noble gas, iodine, tritium, particulates) specific activity limits.	Dose limit (overall not given, 0.3 mSv/a for liquid discharges). Derived site and nuclide group (noble gas, iodine, tritium and particulates) specific activity limits.	Dose limit (not given). Derived nuclide group (noble gas, Sr-90 and particulates(beta)) specific activity limits.	Dose limit 0.1 mSv/a per installation and 5 manSv/GW. Derived site and nuclide specific activity limits.	Dose limit 0.2 mSv/a for airborne releases and 0.05 mSv/a for aquatic releases. Derived nuclide group specific activity limits.
c. Reporting	Annual	Quarterly and annually	Quarterly	Annual	Monthly	
d. Role of authority	Release monitoring carried out by licensee. No control measurements by authority.	Release monitoring carried out by licensee. Control measurement and approval of methodology carried out by authority.	Release monitoring primarily carried out by operator. A remote monitoring system operated by licensee and authority, are also in operation. Control measurement and approval of methodology by authority.	Release monitoring carried out by licensee. Control measurements carried out by authority.	Release monitoring carried out by licensee. Control measurement and approval of methodology carried out by authority.	

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
MONITORING SYSTEMS	**Based on reactor DR3**	**LOVIISA and TVO unless specified**	**Based on KTA SAFETY STANDARD**	**Based on research reactor in Salaspils**	**Based on all units and research reactor in Studsvik unless specified**	**Igualina**
Releases to air						
a. Ventilation system	One exhaust point	Common stack with separate channels for each unit with separate vent. systems for chemistry lab. and turbine hall (LOVIISA) or one common stack per unit (TVO). Offgas delay system (35-90 h Krypton and 25-45 d Xenon).	Not given.	Two separate systems, one for reactor basin with experimental devices and one for laboratories. Both systems are filtrated.	Main stack with ventilation from offgas, steam leakage condenser flow, reactor and turbine hall. Recombine, filters and delay systems used in combination. Auxiliary buildings may have separate stacks.	Main stack with 3 pipe and a flow of 700 m3/s (100 in Swedish units). Recombine, aerosol- and iodine filter and delay system for off gases.
b. Monitoring design	In-stack on-line monitoring of a iodine filter with scintillation counter and ionisation chamber.	Double circuit sampling system with one particulate filter and two activated charcoal filters. Beta-sensitive scintillation counter for continuous monitoring of noble gases.	Single circuit sampling system with aerosol and charcoal filters with continuous beta monitoring. Double beta-sensitive counters for continuous monitoring of noble gases.	Single circuit sampling system with aerosol filter. Beta-sensitive scintillation counter in 40 1 chamber for continuous monitoring of noble gases (Ar-41).	Two level single circuit sampling system with aerosol and iodine filters and continuous noble gas measurement chamber.	Double on-line monitoring systems for noble gases, long-life and short-life aerosols and iodine with use of gamma compensated betacounters.

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
Releases to air						
:. Sampling and analyses 1. noble gases 2. halogens 3. aerosols 4. aslpha 5. Sr-90 6. tritium 7. c-14	1. Continuously with ionisation chamber (Ar-41) 2. Continuously with scint. counter on filter 3. Weekly with sample from gold trap	1. One grab sample per week. Gamma spectrometric meas. 2. one week sample. Gamma spectrometric meas. 3. Similar as 2. 4. One month sample. Gross alpha counting. 5. Quarterly combined week sample. Chemical separation and ingrowth of Y-90. Low bkg gas flow beta counting. 6. One month sample. Liquid scint. of silicagel sample. 7. Estimations based on experimental data.	1. One representative sample per week. Gamma spectrometric meas. 2. One week sample. Gamma spectrometric meas. 3. Similar as 2. 4. One quarterly sample. Alpha spectroscopy. 5. One quarterly sample. 6. One quarterly sample. 7. One quarterly sample.	1. No gamma spectrometric meas. Half life analysis to determine Ar-4 1. 3. Beta measurements 4. Gross alpha meas.	1. In-stack continuous gamma spectroscopic meas. l-2 hours meas. BARSEBACK: Two grab samples per week and gamma spectr. STUDSVIK: 1 1 grab samples per year. 2. One week sample.Gamma spectrometric meas. 3. Similar as 2. 4. Half-year samples. Alpha spectr. 5. Half-year samples. 6. No routine meas. 7. Estimation based on meas. campaigns.	1. One 6 l grab sample/day. Gamma spectroscopy. 2. One week sample. Gamma spectroscopy. 3. Two samples/week. Gamma spectroscopy. 7. Estimated to 1 TBq/GW.

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
Releases to air						
d. Detection limits (Bq/a)	Ar-41: 4E13 I-131: 5E7a	LOVIISA: Kr-85: 1E10 Xe-133: 1E10 TVO: Kr-85: 2E12 Xe-133: 3E12 BOTH: I-131: 5E6 Co-60: 5E6	Not given directly, but assuming an air flow of 100 m3/s: Xe-133: 1E12 1-131: 5E6 Co-60: 1E7	Not given	Kr-85: 1-6E12 Xe-133: 6-6E12 I-131: 2-50E6 Co-60: 4-60E6	Kr-85: 4E13 Xe-133: 8E13 I-131: 5E8 Co-60: 5E7
Releases to water						
a. Waste water system	Centralised system with evaporation and ion exchange as cleaning methods.	Centralised system. Cleaning methods not specified.	Centralised system with evaporation and ion-exchange as cleaning methods.	Waste water system und reconstruction. Ion exchange is used for cleaning.	Centralised waste water system for system drainage, chemistry water and cleaning water. Treatment methods used are filtering, ion exchange and evaporation.	Reactor coolant, condensate and waste water cleaning systems. Ion exchange and evaporation used as treatment methods.

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
Releases to water						
b. Discharge procedure	Beta measurement is carried out before discharge. Gamma spectroscopy on quarterly pooled samples.	Sample measured before discharge.	Sample measured before discharge. Continuous monitoring during discharge.	Sample measured before discharge. Discharge is mixed with sanitary water from Salaspils and treated in a waste water treatment facility and then released to the river Daugava.	Sample measured before discharge. During discharge a proportional sample is taken, called juridical sample, which is used for analysis.	Sample measured before discharge (total beta). During discharge continuous monitoring (NaI).
c. Analysis procedures 1. gamma 2. alpha 3. Sr-90 4. Tritium 5. c-14	1. Gamma spectroscopy 4. Liquid scintillation	1. Gamma spectroscopy on weekly samples. 2. Gross alpha counting with scintillator on monthly pooled sample. 3. Quarterly combined sample. Chemical separation and ingrowth of Y-90. Low bkgr gas flow beta counting. 4. Analysis on monthly pooled samples with liquid scint.	1. Gamma spectroscopy on weekly samples. 2. Gross alpha on quarterly pooled samples. 3. Analysis on monthly pooled samples. 4. Similar as 3.	1. Gamma spectroscopy, frequency not given. 3. Radiochemical analysis. Monthly pooled samples.	1. Gamma spectroscopy on monthly pooled samples. 2. Gross alpha (additionally spectroscopic on some units) on monthly pooled samples. 3. Analysis on half year pooled samples. 4. Analysis on monthly pooled samples.	1. Gamma spectroscopy on 20 l sample.

	Denmark	Finland	Germany	Latvia	Sweden	Ignalina (Sosnovy Bor) Unofficial Information
Releases to water						
1. Detection limits (Bq/l)	General beta and gamma: E-3	I-131: 5 Co-60: 5 Zn-65: 10 Mn-54: 5	I-131: 30 Co-60: 40 Zn-65: 75 Mn-54: 40	All nuclides: 0.5	I-131: 5 Co-60: 5 Zn-65 10 Mn-54 5	Gamma: 0.01
QUALITY ASSURANCE						
	Yearly calibrations with standard sources. International intercalibration exercises.	Comprehensive QA programmes organised by licensee. National intercalibrations.	Detailed instructions of QA procedures. National intercalibration excercises.	Calibration against standard sources.	Comprehensive QA-programmes organised by licensee. National inter-calibrations. Control measurements by authority. QA meetings with authority twice a year.	

No. 18 ACTIVITIES OF THE COMMISSION 1985
- Report of the activities of the Baltic Marine Environment Protection Commission during 1985 including the Seventh Meeting of the Commission held in Helsinki 11-14 February 1986
- HELCOM Recommendations passed during 1986
(1986)*

No. 19 BALTIC SEA MONITORING SYMPOSIUM
Tallinn, USSR, 10-15 March 1986
(1986)

No. 20 FIRST BALTIC SEA POLLUTION LOAD COMPILATION
(1987)

No. 21 SEMINAR ON REGULATIONS CONTAINED IN ANNEX II OF MARPOL 73/78 AND REGULATION 5 OF ANNEX IV OF THE HELSINKI CONVENTION
National Swedish Administration of Shipping and Navigation;
17-18 November 1986, Norrkiiping, Sweden
(1987)

No. 22 SEMINAR ON OIL POLLUTION QUESTIONS
19-20 November 1986, Norrkiiping, Sweden
(1987)

No. 23 ACTIVITIES OF THE COMMISSION 1986
- Report on the activities of the Baltic Marine Environment Protection Commission during 1986 including the Eighth Meeting of the Commission held in Helsinki 24-27 February 1987
- HELCOM Recommendations passed during 1987
(1987)*

No. 24 PROGRESS REPORTS ON CADMIUM, MERCURY, COPPER AND ZINC
(1987)

No. 25 SEMINAR ON WASTEWATER TREATMENT IN URBAN AREAS
7-9 September 1986, Visby, Sweden
(1987)

No. 26 ACTIVITIES OF THE COMMISSION 1987
- Report on the activities of the Baltic Marine Environment Protection Commission during 1987 including the Ninth Meeting of the Commission held in Helsinki 15-19 February 1988
HELCOM Recommendations passed during 1988
(1988)

No. 27A GUIDELINES FOR THE BALTIC MONITORING PROGRAMME FOR THE THIRD STAGE; PART A. INTRODUCTORY CHAPTERS
(1988)

No. 27B GUIDELINES FOR THE BALTIC MONITORING PROGRAMME FOR THE THIRD STAGE; PART B. PHYSICAL AND CHEMICAL DETERMINANDS IN SEA WATER
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No. 9 SECOND BIOLOGICAL INTERCALIBRATION WORKSHOP
Marine Pollution Laboratory and Marine Division of the National Agency of Environmental Protection, Denmark, August 17-20, 1982, Rønne, Denmark
(1983)

No. 10 TEN YEARS AFTER THE SIGNING OF THE HELSINKI CONVENTION
National Statements by the Contracting Parties on the Achievements in Implementing the Goals of the Convention on the Protection of the Marine Environment of the Baltic Sea Area
(1984)

No. 11 STUDIES ON SHIP CASUALTIES IN THE BALTIC SEA 1979- 198 1
Helsinki University of Technology, Ship Hydrodynamics Laboratory, Otaniemi, Finland
P. Tuovinen, V. Kostilainen and A. Hämäläinen
(1984)

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(1984)*

No. 13 ACTIVITIES OF THE COMMISSION 1983
- Report of the activities of the Baltic Marine Environment Protection Commission during 1983 including the Fifth Meeting of the Commission held in Helsinki 13-16 March 1984
- HELCOM Recommendations passed during 1983 and 1984
(1984)

No. 14 SEMINAR ON REVIEW OF PROGRESS MADE IN WATER PROTECTION MEASURES
17-2 1 October 1983, Espoo, Finland
(1985)

No. 15 ACTIVITIES OF THE COMMISSION 1984
- Report of the activities of the Baltic Marine Environment Protection Commission during 1984 including the Sixth Meeting of the Commission held in Helsinki 12-15 March 1985
- HELCOM Recommendations passed during 1984 and 1985
(1985)

No. 16 WATER BALANCE OF THE BALTIC SEA
A Regional Cooperation Project of the Baltic Sea States;
International Summary Report
(1986)

No. 17A FIRST PERIODIC ASSESSMENT OF THE STATE OF THE MARINE ENVIRONMENT OF THE BALTIC SEA AREA, 1980-1985; GENERAL CONCLUSIONS
(1986)

No. 17B FIRST PERIODIC ASSESSMENT OF THE STATE OF THE MARINE ENVIRONMENT OF THE BALTIC SEA AREA, 1980-1985; BACKGROUND DOCUMENT
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BALTIC SEA ENVIRONMENT PROCEEDINGS

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(1979)*

No. 2 REPORT OF THE INTERIM COMMISSION (IC) TO THE BALTIC MARINE ENVIRONMENT PROTECTION COMMISSION
(1981)"

No. 3 ACTIVITIES OF THE COMMISSION 1980
- Report on the activities of the Baltic Marine Environment Protection Commission during 1980
- HELCOM Recommendations passed during 1980
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No. 4 BALTIC MARINE ENVIRONMENT BIBLIOGRAPHY 1970-1979
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PART A-l : OVERALL CONCLUSIONS
(1981)*

No. 5B ASSESSMENT OF THE EFFECTS OF POLLUTION ON THE NATURAL RESOURCES OF THE BALTIC SEA, 1980
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PART A-2: SUMMARY OF RESULTS
PART B: SCIENTIFIC MATERIAL
(1981)

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Institut für Meereskunde an der Universität Kiel, Department of Marine Chemistry, March 23 -April 3, 1981
(1982)

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- Report of the activities of the Baltic Marine Environment Protection Commission during 1981 including the Third Meeting of the Commission held in Helsinki 16-19 February 1982
- HELCOM Recommendations passed during 1981 and 1982
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- Report on the activities of the Baltic Marine Environment Protection Commission during 1988 including the Tenth Meeting of the Commission held in Helsinki 14-17 February 1989
- HELCOM Recommendations passed during 1989
(1989)

No. 30 SECOND SEMINAR ON WASTEWATER TREATMENT IN URBAN AREAS
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(1989)

No. 31 THREE YEARS OBSERVATIONS OF THE LEVELS OF SOME RADIONUCLIDES IN THE BALTIC SEA AFTER THE CHERNOBYL ACCIDENT
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No. 32 DEPOSITION OF AIRBORNE POLLUTANTS TO THE BALTIC SEA AREA 1983-1985 AND 1986
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No. 33 ACTIVITIES OF THE COMMISSION 1989
- Report on the activities of the Baltic Marine Environment Protection Commission during 1989 including the Eleventh Meeting of the Commission held in Helsinki 13-16 February 1990
- HELCOM Recommendations passed during 1990
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No. 34 STUDY OF THE RISK FOR ACCIDENTS AND THE RELATED ENVIRONMENTAL HAZARDS FROM THE TRANSPORTATION OF CHEMICALS BY TANKERS IN THE BALTIC SEA AREA
(1990)

No. 35A SECOND PERIODIC ASSESSMENT OF THE STATE OF THE MARINE ENVIRONMENT OF THE BALTIC SEA, 1984-1988; GENERAL CONCLUSIONS
(1990)

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(1990)

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(1990)

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- Report on the activities of the Baltic Marine Environment Protection Commission during 1990 including the Twelfth Meeting of the Commission held in Helsinki 19-22 February 199 1
- HELCOM Recommendations passed during 1991
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- Report of the activities of the Baltic Marine Environment Protection Commission during 199 1 including the 13th meeting of the Commission held in Helsinki 3-7 February 1992
- HELCOM Recommendations passed during 1992
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No. 44 NITROGEN AND AGRICULTURE, INTERNATIONAL WORKSHOP
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No. 45 SECOND BALTIC SEA POLLUTION LOAD COMPILATION
(1993)

No. 46 SUMMARIES OF THE PRE-FEASIBILITY STUDIES
Prepared for the Baltic Sea Joint Comprehensive Environmental Action Programme
(1993)

No. 47 HIGH LEVEL CONFERENCE ON RESOURCE MOBILIZATION
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- Report on the activities of the Baltic Marine Environment Protection Commission during 1992 including the 14th meeting of the Commission held in Helsinki 2-5 February 1993
- HELCOM Recommendations passed during 1993
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- Report on the activities of the Baltic Marine Environment Protection Commission during 1993 including the 15th meeting of the Commission held in Helsinki 8-11 March 1994
- HELCOM Recommendations passed during 1994
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(1994)

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(1994)

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