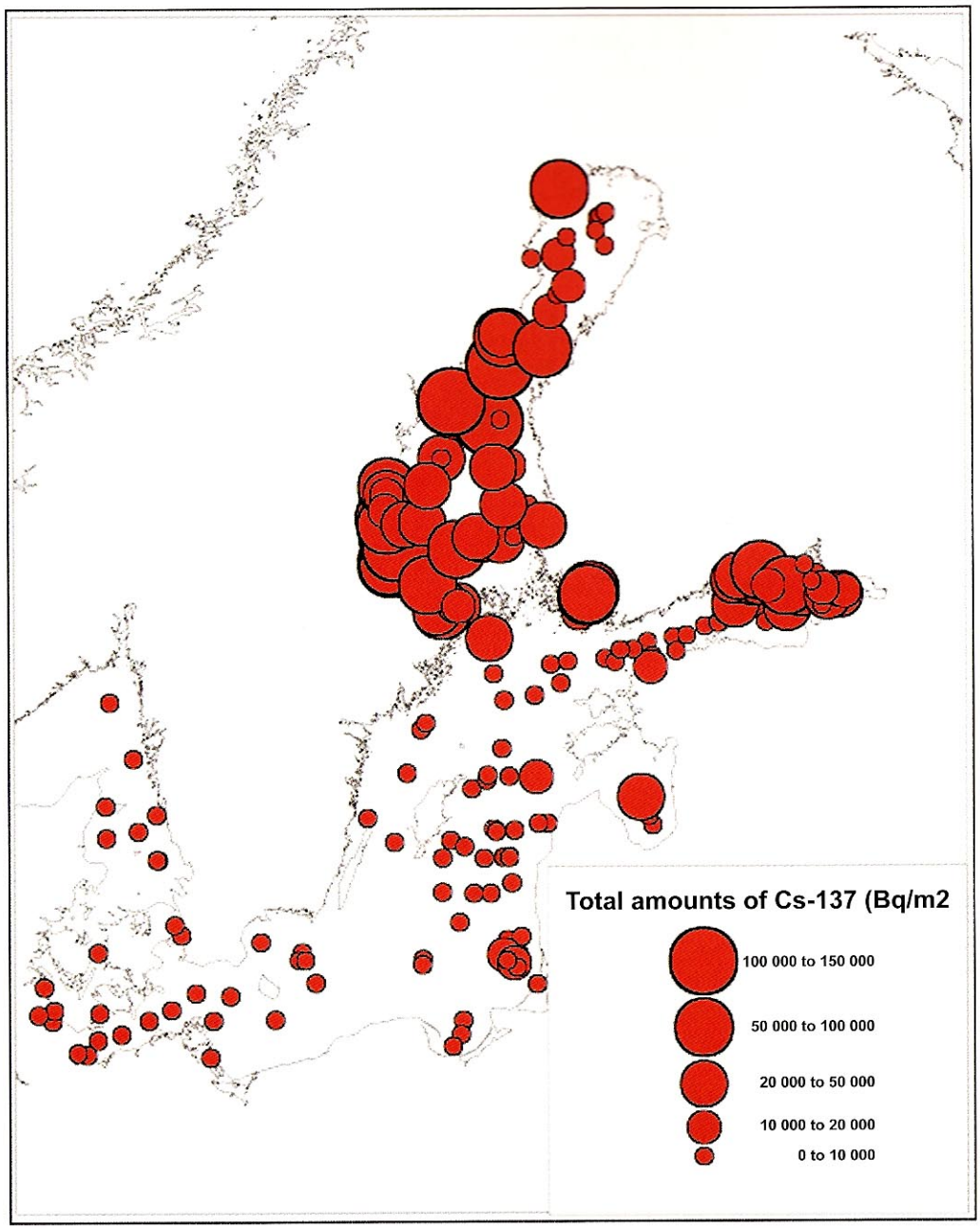


Figure 1:
Total amounts of ^{137}Cs (Bq/m^2) at different sampling stations in the Baltic Sea in the late 1990s and the beginning of the 2000s (Ilus et al. 2007). Most of ^{137}Cs is in the sediments of the Bothnian Sea and in the eastern Gulf of Finland, due the high deposition of the Chernobyl fallout in these areas, the high tendency of ^{137}Cs to bind in sediment particles in brackish water environments, and the high sediment accumulation rates in these areas.

sediment / Bq kg^{-1} in water) in brackish water environments, and because it is relatively easy to measure. In the recent inventory, we estimated that the total amount of ^{137}Cs activity in the Baltic Sea sediments was about 2,100-2,400 TBq at the beginning of the 2000s (Ilus et al. 2007). This amount was about 8-9 times higher compared to the pre-Chernobyl level in the beginning of the 1980s (***Table 2***). In recent years, caesium has continued to be deposited on the sea bottom and, at the same time its physical half-life has reduced its activity slowly. Most of the ^{137}Cs activity is in the sediments of the Bothnian Sea and in the eastern Gulf of Finland (***Figure 1***). The newly compiled data has increased our knowledge of the distribution of ^{137}Cs activity between the so-called hard and soft bottoms in open sea areas. The total amounts of ^{137}Cs activity on hard bottoms varied from 0.3% to nearly 14% of those recorded on soft bottoms, while the average ratio was only about 4% (Ilus et al. 2007).

Additional data on transuranic elements was also compiled during the study period. The reported values of $^{239,240}Pu$ activities varied between 0.01 and 14.1 Bq kg^{-1} d.w. and the values of ^{238}Pu activities between 0.006 and 0.48 Bq kg^{-1}. The total amount of $^{239,240}Pu$ was estimated to be about 15.3 TBq (***Table 2***). Most of the plutonium originated from the

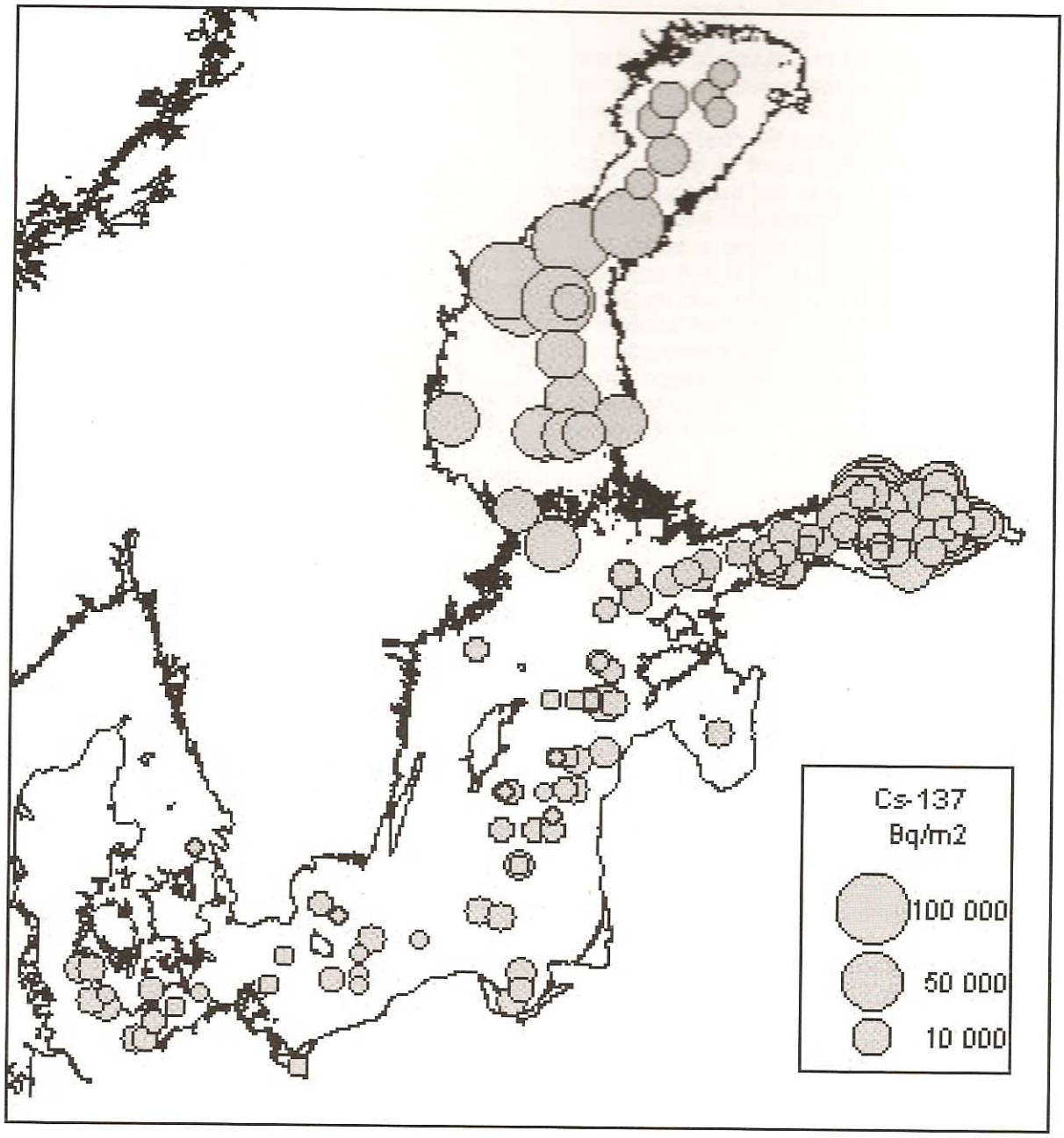


Figure 6.5.1 *Total amounts of ^{137}Cs [Bq m^{-2}] at different sampling stations in 1998.*

Sediment samples were usually taken from soft bottoms, i.e. from the sedimentation bottoms of deep basins. Soft bottoms very often act as "sinks" for radionuclides, whereas hard bottoms are regarded as transport bottoms with very low accumulation rates for sinking matter. However, erosion bottoms are very seldom uncontaminated, because bioturbation caused by benthic fauna may transfer contaminants and organic material into deeper sediment layers. Studies carried out on the Polish coast have shown that ^{137}Cs penetrates effectively into near-shore sandy sediments, and that rapidly accumulating sediments affected by river discharges have much higher contents of exchangeable radiocaesium than slowly accumulating marine sediments (Knapinska-Skiba et al., 1994, 1995, 1997).

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

sediment layers (0–30 cm) ranged between 1 900 Bq m^{-2} and 3 900 Bq m^{-2}.

5.7 Np-237

Neptunium-237 is an artificial radionuclide produced in nuclear reactors as a result of the irradiation of uranium-235 and uranium-238. Large amounts are found in spent nuclear fuel. The activity concentrations of Np-237 in surface sediments (0–20 cm) of the Baltic Sea were very low: 0.2–6.5 millibequerels per kg dry weight. The total amounts of Np-237 (mBq m^{-2}) at 26 sampling stations are shown in Figure 7. The total inventory of Np-237 in the seabed of the Baltic Sea was estimated at 0.02 TBq.

5.8 Pu-238

Results on plutonium-238 were reported from 11 stations in the southern Baltic Proper and four stations in the northern areas of the Baltic Sea. In the southern Baltic Proper, the activity concentrations of Pu-238 ranged from 6 mBq kg^{-1} to 250 mBq kg^{-1} dry weight in the surface sediments (0–15 cm) and the total amounts were generally from 0.5 Bq m^{-2} to 4.1 Bq m^{-2}. Only at one station in the southern Baltic Proper was the total amount of Pu-238 regularly higher (4.8–9.1 Bq m^{-2}). In the

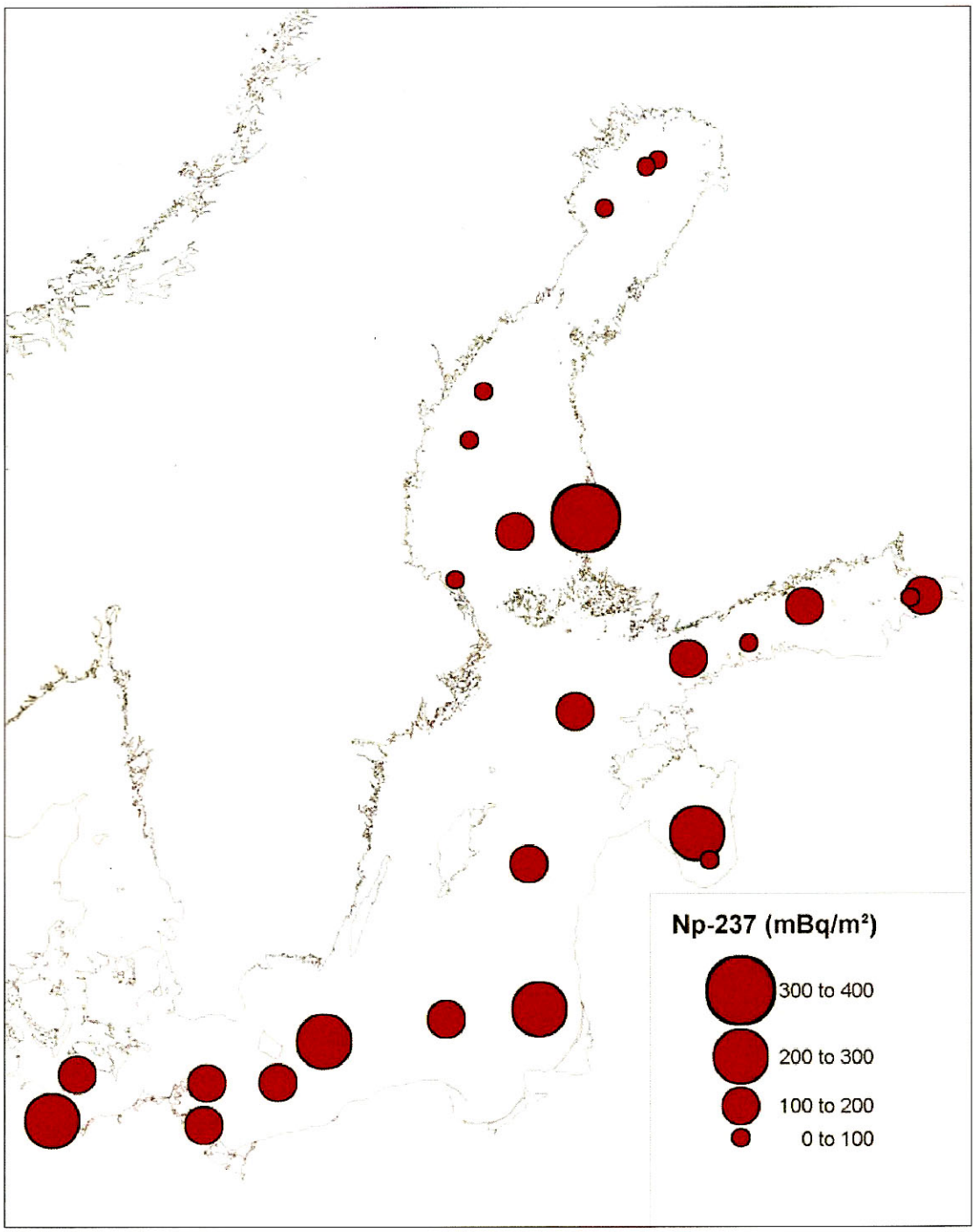
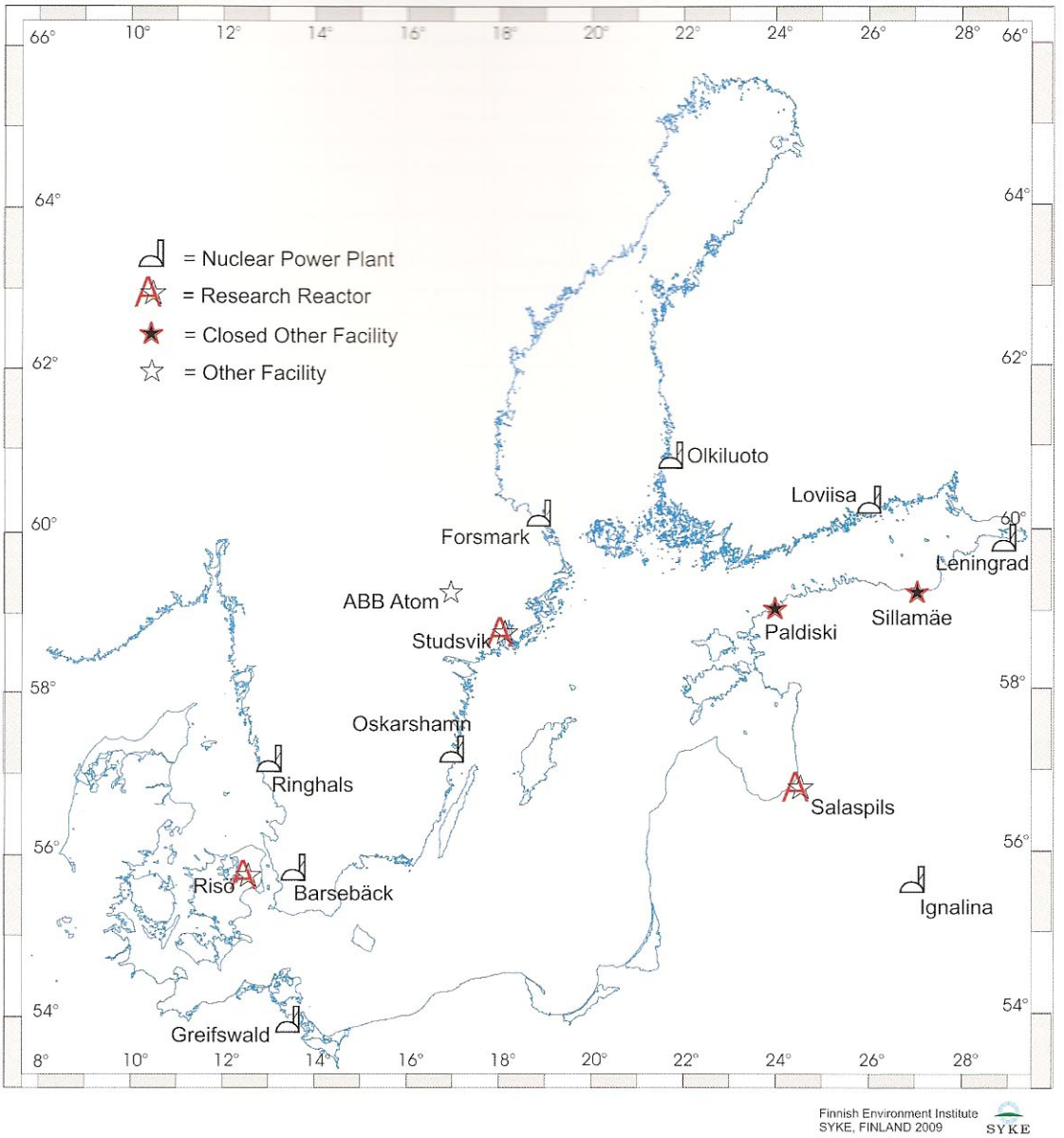


Figure 7. Total amounts of Np-237 (mBq m^{-2}) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

Figure 1:
Location of nuclear facilities in the Baltic Sea region.



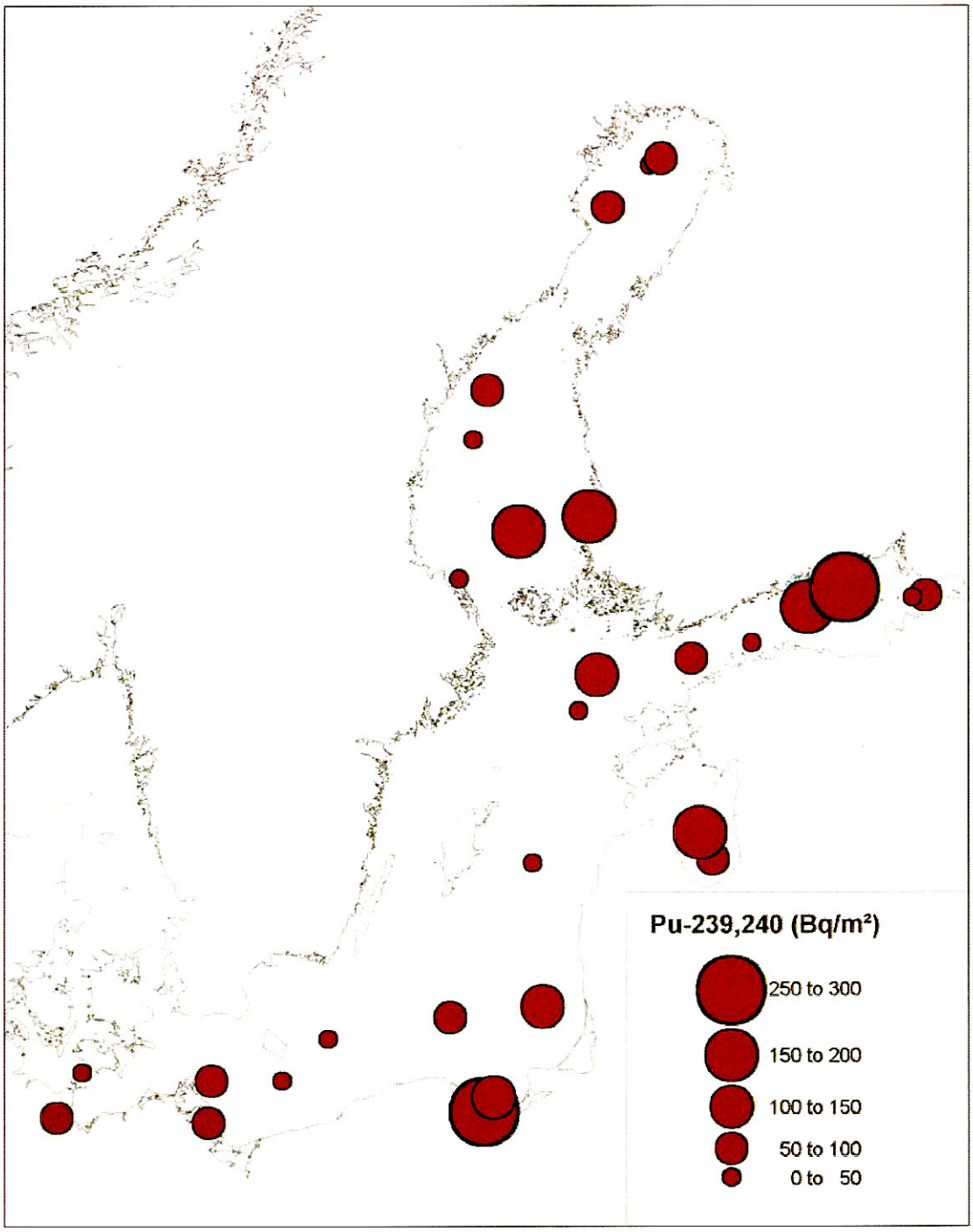


Figure 10.
Total amounts of Pu-239,240 (Bq m^{-2}) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

Sub-region	Number of sediment cores	Maximum (Bq m^{-2})	Median (Bq m^{-2})	Total inventory (TBq)
Bothnian Bay	4	94	69	1.25
Bothnian Sea	8	178	156	6.86
Gulf of Finland	8	299	92	1.60
Baltic Proper + Gulf of Riga	29	265	47	5.60
Belt Sea	2	51		
Total	51			15.3

Table 3.
Total inventories and maximum and median values of Pu-239,240 in different sub-regions of the Baltic Sea.

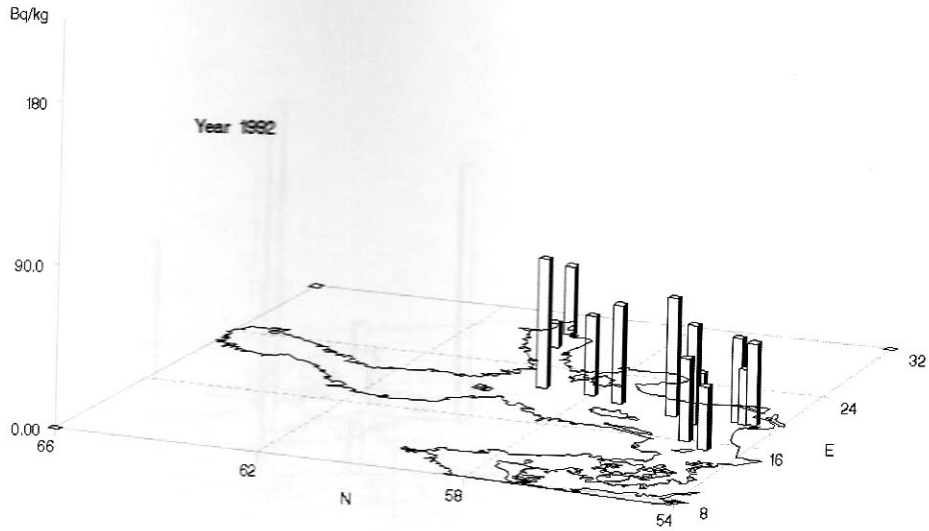
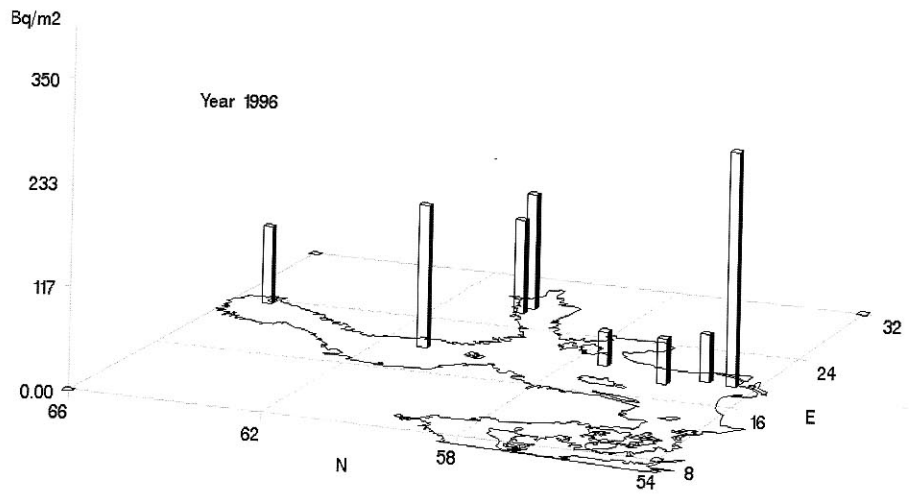


Figure 6.4.2.5 *Radium-226 in surface sediment layers at different sampling stations in 1992.*



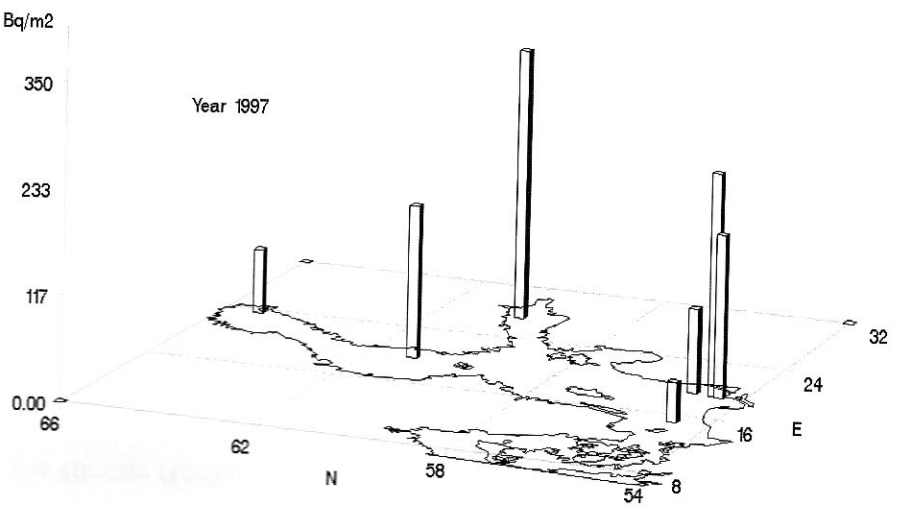


Figure 6.4.2.6 *Total amounts of plutonium-239,240 at different sampling stations in 1996 and 1997.*

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Radioactivity in the Baltic Sea: inventories and temporal trends of ^{137}Cs and ^{90}Sr in water and sediments

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Abstract The Baltic Sea is ecologically unique as one of the world's largest brackish water basins. It was significantly contaminated by radioactivity following the Chernobyl accident in 1986, the major contaminant being long-lived ^{137}Cs. Due to the slow exchange of water between the Baltic Sea and the North Sea and the relatively rapid sedimentation rates, radionuclides have prolonged residence times in the Baltic Sea. ^{137}Cs levels are consequently still clearly higher than in other water bodies around the world. In addition to the Chernobyl accident, artificial radionuclides in the Baltic Sea originate from the global fallout following nuclear weapons testing in the 1950s and 1960s, while discharges into the Baltic Sea from nuclear power plants and other facilities are of minor importance. Here, inventories and the temporal evolution of radionuclides both in seawater and sediments of the Baltic Sea are presented and discussed.

Keywords Baltic Sea · Inventory · Radioactivity

Introduction

The Baltic Sea (Fig. 1) is neither an ocean nor a lake, but a large brackish water basin that is only connected to the world's oceans by the narrow and shallow Danish straits. The Baltic Sea consists of a series of sub-basins that are mostly separated by shallow sills. It has a surface area of 390,000 km^2 and volume of 21,205 km^3, while the average depth is only 54 m [1, 2]. The water is a mixture of seawater from the North Sea and fresh water from rivers and rainfall. The water exchange between the Baltic Sea and North Sea is limited and the renewal time of the entire water mass is around 50 years [1]. The drainage basin covers an area that is 4.3 times larger than the area of the Baltic Sea itself, bringing large amounts of freshwater to the sea through rivers [2]. The salinity of the surface water varies regionally and the mean salinity is about 7‰ [1]. The Baltic Sea has very strong salinity stratification that prevails throughout the year. The saltier water flowing from the North Sea does not easily mix with the less dense water already in the Baltic Sea but sinks into the deeper basins.

^{137}Cs and ^{90}Sr in the Baltic Sea mainly originate from the Chernobyl accident in 1986 and from the fallout following nuclear weapons testing that was mostly conducted in the 1950s and 1960s. The contribution from Chernobyl amounts to 82% of ^{137}Cs and 13% of ^{90}Sr present in the Baltic Sea, while the nuclear weapons testing accounts for 14 and 81% of these radionuclides, respectively [3]. Radioactivity from nuclear weapons testing was evenly distributed in the Baltic Sea area, whereas the deposition from the Chernobyl accident was very uneven. Small amounts of these radionuclides are discharged from local nuclear facilities and also from facilities outside the Baltic Sea. However, local discharges from the 11 facilities situated in the Baltic Sea discharge area only account for 0.1% of the total activity injected. Discharges from sources outside the Baltic region are mainly associated with the Sellafield reprocessing plant in the UK and account for 4 and 6% of ^{137}Cs and ^{90}Sr inputs, respectively [3].

Nearly 85 million people live in the Baltic Sea catchment area. Excessive inputs of nutrients and hazardous

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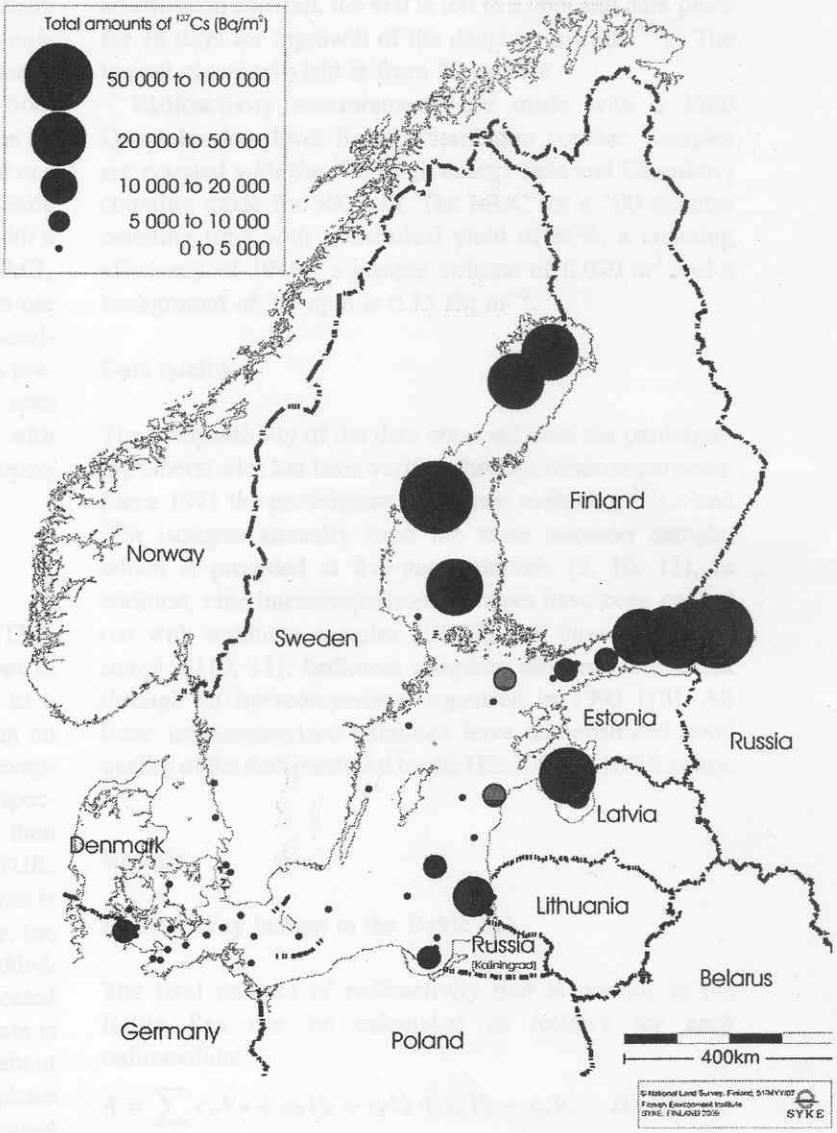

Fig. 1 Areal distribution of ^{137}Cs in sediments of the Baltic Sea in 2004–2006

substances originating from agriculture, urban areas, industry and transport have caused major environmental problems in the Baltic Sea. The Helsinki Commission (HELCOM) works to protect the marine environment of the Baltic Sea from all sources of pollution through intergovernmental co-operation between Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Poland, Russia, Sweden, and the European Community. In 1984 the HELCOM-MORS group (The Helsinki Commission-Monitoring of Radioactive Substances) was established to monitor radioactive substances in the Baltic Sea, but the monitoring of radioactivity in the Baltic Sea had been going on for much longer. In Finland, for example, such studies were started in the early 1960s.

Of all the seas in the world, the Baltic Sea is considered to be one of the most contaminated with artificial radioactivity and it has been subject for many research projects [4–9]. This article aims to provide a general overview of radioactivity in the Baltic Sea. Inventories of radionuclides are evaluated for both seawater and sediments using the data available from the HELCOM-MORS database complemented with measurements performed by the Finnish Radiation and Nuclear Safety Authority (STUK).

Materials and methods

Sampling and radioanalytical methods used by HELCOM-MORS laboratories

Monitoring of radionuclides in the Baltic Sea is carried out by individual countries in the monitoring network under HELCOM. Samples of seawater, sediment, fish, aquatic plants and benthic animals are collected annually. The

ults of the monitoring programme are submitted annually o the HELCOM-MORS database. Sampling and the analytical methods used by the HELCOM-MORS laboratories have been described earlier [3]. ^{137}Cs is determined from water by first concentrating it by evaporation, adsorption on AMP or with hexacyanoferrates. All laboratories except one then use HPGe or Ge(Li) gamma spectrometry to measure the activity. The single laboratory measuring ^{137}Cs with a gas-flow beta counter performs ion exchange and Cs_2PtCl_6 precipitation prior to the measurement. All laboratories use gamma spectrometry for sediment samples. ^{90}Sr is chemically separated in most laboratories either with various precipitations or using HDEHP extraction. One laboratory uses Sr resin (Eichrom). ^{90}Sr activity is determined either with liquid scintillation or with a beta counter (gas flow proportional counter).

Methods used at STUK

The following accredited in-house method is used at STUK to determine ^{137}Cs and ^{90}Sr radioactivity concentrations in seawater samples. An inactive Sr carrier is added to a seawater sample of about 30 L, the amount depending on the salinity of the water sample. The sample is first evaporated to about 0.5 L, measured with HPGe gamma spectrometry in Marinelli geometry, and the spectrum is then analysed with GAMMA-99 software developed by STUK. The MDC for ^{137}Cs with a 1000 min measurement time is typically under 2 Bq m^{-3}. After gamma spectrometry, the sample is diluted to about 4 L and an Fe carrier is added. Iron scavenging is performed with NH_4OH for a heated solution. A cooled solution is filtered and the precipitate is discarded. The solution is heated to near boiling and about 10 g of ammonium carbamate is added to precipitate strontium as carbonate at pH 9–10. The solution is allowed to cool and the precipitate to settle. After decanting, the rest of the solution is centrifuged and the supernatant is discarded. The carbonate precipitate is dissolved in 20 mL of 8 M nitric acid for extraction chromatography. The extraction chromatographic column (diameter 0.8–1 cm) contains 3 g of strontium-specific Sr Resin (Triskem International), which is pretreated with 3 M nitric acid. The sample is introduced into the column, which is then washed twice with 20 mL of 8 M nitric acid and once with 3 M nitric acid. Strontium is eluted with 70 mL of 0.05 M nitric acid. The eluate is warmed and the pH is adjusted to 8–9. Strontium is precipitated as carbonate with 2 g of ammonium carbamate and filtered onto filter paper. The precipitate is transferred to a tared 20 mL plastic scintillation vial with 1.6 mL of 1 M hydrochloric acid and the vial is weighed. A 0.1 mL aliquot is taken from the vial for yield determination of strontium by atomic absorption spectrometry. After adding 6 mL of UltimaGold AB scintillation cocktail, the vial is left in a cool and dark place for 18 days for ingrowth of the daughter nuclide ^{90}Y. The typical chemical yield is from 30 to 70%.

Radioactivity measurements are made with a 1220 Quantulus low level liquid scintillation counter. Samples are counted with the ^{14}C (high energy beta and Cerenkov) counting mode for 300 min. The MDC for a 300 minutes counting time with a chemical yield of 50%, a counting efficiency of 194%, a sample volume of 0.030 m^3 and a background of 2.7 cpm is 0.13 Bq m^{-3}.

Data quality

The comparability of the data obtained from the participating laboratories has been verified through intercomparisons. Since 1991 the participants have been analyzing ^{137}Cs and ^{90}Sr isotopes annually from the same seawater sample, which is provided at five-year intervals [3, 10, 11]. In addition, nine intercomparison exercises have been carried out with sediment samples [10–12] and three with biota samples [10, 11]. Sediment sampling devices were tested through an intercomparison organised in 1992 [13]. All these intercomparison exercises have demonstrated good quality of the data produced by the HELCOM-MORS group.

Results

Radioactivity budget in the Baltic Sea

The total amount of radioactivity that is present in the Baltic Sea can be calculated as follows for each radionuclide:

$$A = \sum c_P V_P + c_R V_R + c_I V_I + c_L V_L - c_o V_o - D,$$

where A, the total amount of a single radionuclide in the Baltic sea, is the sum of the activity transferred in the following processes on a yearly basis: precipitation (P), runoff (R), inflow through the Danish straits (I), local discharges (L), outflow through the Danish straits (O) and physical decay (D). In the equation, c is the concentration of the radionuclide and V the water volume in the above-mentioned processes. The amount of ^{137}Cs and ^{90}Sr activity annually transferred in these processes is presented in Figs. 2, 3, and 4. Results prior to 1982 were obtained from the studies of Salo et al. [14] Since 1984 the concentrations of radionuclides in the water flowing into and out of the Baltic Sea through the Danish straits have been obtained from the HELCOM database, and the volumes of inflowing and outflowing water used in calculations have been 471 and 947 km^3/year, respectively [15]. Annual river runoff following the Chernobyl accident was estimated based on the detailed, measured runoff of ^{137}Cs and ^{90}Sr

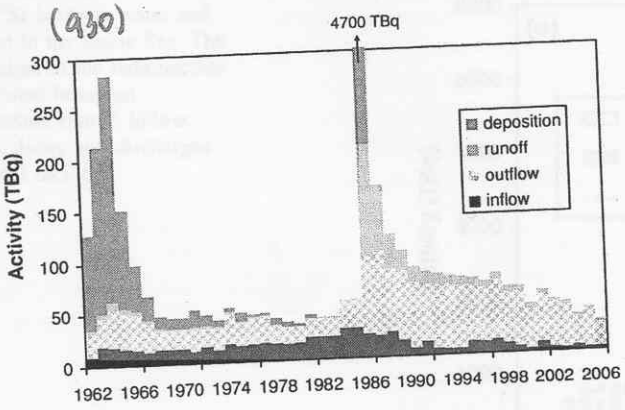

Fig. 2 Inputs (deposition, runoff, inflow) and output (outflow) of ^{137}Cs in the Baltic Sea. Deposition of ^{137}Cs from Chernobyl accident was 4700 TBq (off the scale)

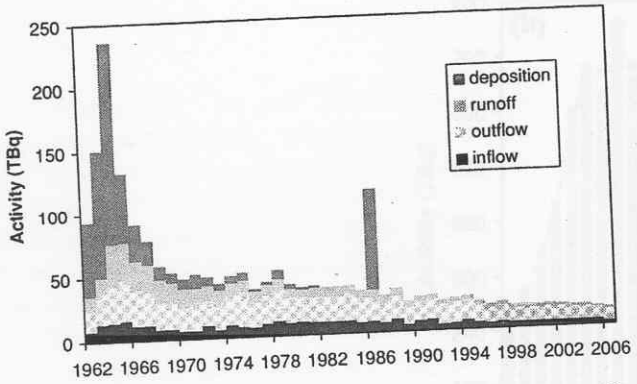

Fig. 3 Inputs (deposition, runoff, inflow) and output (outflow) of ^{90}Sr in the Baltic Sea

from Finland into the Baltic Sea [16] and by scaling these runoff values for the whole Baltic Sea using the information that the river runoff from Finland is about one-fifth of the total runoff of these radionuclides into the Baltic Sea, as determined earlier for ^{137}Cs [17]. The amounts of ^{137}Cs and ^{90}Sr from the Chernobyl accident deposited as precipitation are 4700 and 80 TBq, respectively [10]. While the amounts of these two nuclides were relatively even in the global fallout, much more ^{137}Cs than ^{90}Sr was released into the Baltic Sea following the Chernobyl accident in 1986. The input of radioactivity from local discharges was obtained from the HELCOM database and is presented in Fig. 4, separately from other input terms due to its lower activity contribution. Since the Chernobyl accident in 1986, the amount of ^{137}Cs transferred through the Danish straits has been much larger in outflowing than inflowing waters, reflecting the fact that the Baltic Sea is one of the most contaminated seas in the world. In the era of nuclear weapons testing (1950s–1960s), the significance of runoff as a source of radioactivity was much greater for ^{90}Sr than ^{137}Cs, and it also lasted longer in the case of ^{90}Sr. The proportion of ^{90}Sr in the Chernobyl deposition was much lower than that of ^{137}Cs, and no increase was detected in ^{90}Sr runoff. Deposition from Chernobyl was unevenly distributed and a significant portion of the drainage area was contaminated to a lesser degree than the Baltic Sea itself.

Inventories of ^{137}Cs and ^{90}Sr in the Baltic Sea water and their effective half-lives

The inventories of ^{137}Cs and ^{90}Sr in seawater (Fig. 5) were derived from the observed data (obtained from the HELCOM database) by calculating the average ^{137}Cs and ^{90}Sr concentration in water from different basins of the Baltic Sea and taking into consideration the volumes of these basins [15]. Data prior to 1982 were obtained from the

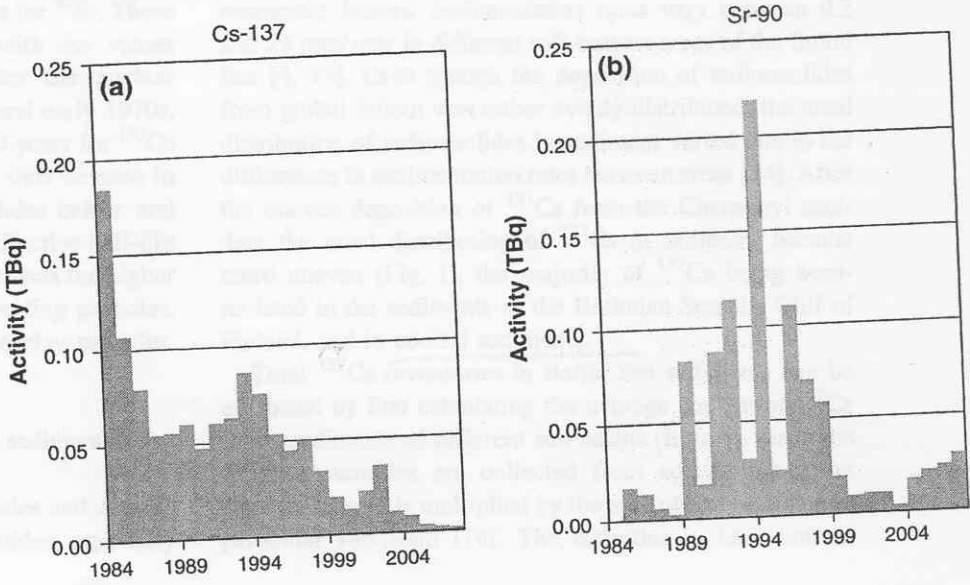

Fig. 4 Discharges of: **a** ^{137}Cs and **b** ^{90}Sr into the Baltic Sea from local nuclear facilities

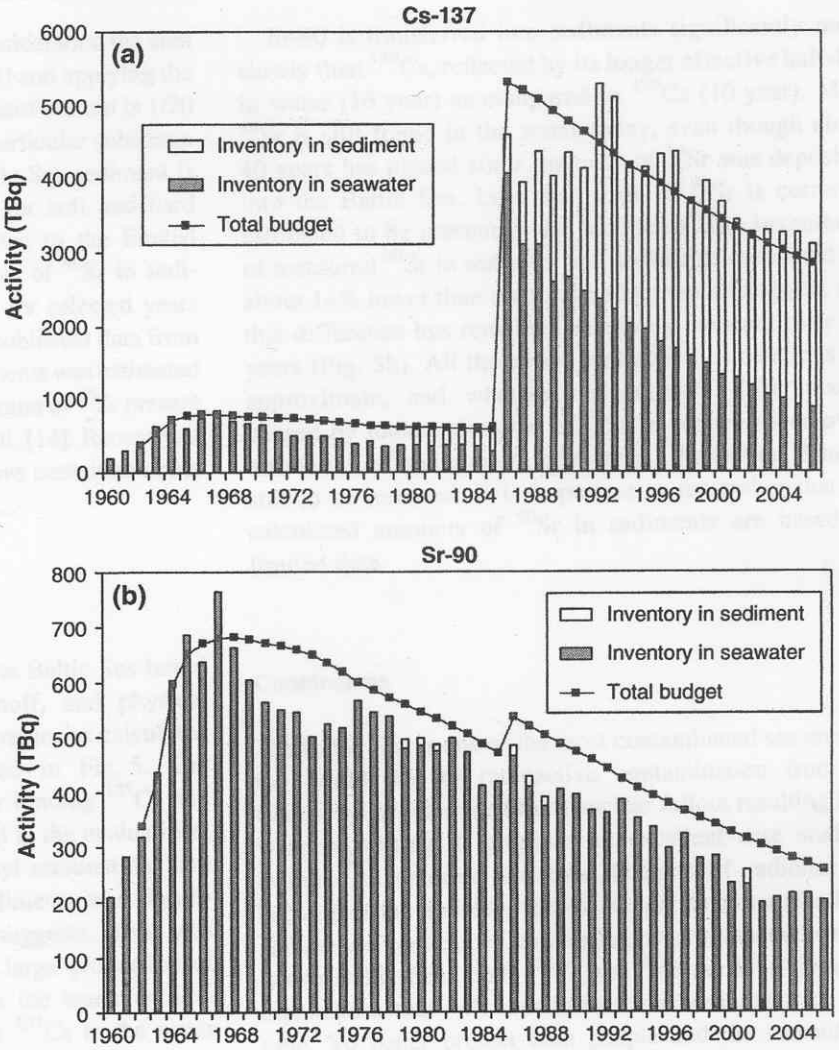

Fig. 5 Distribution of: **a** ^{137}Cs and **b** ^{90}Sr between water and sediment in the Baltic Sea. The total budget of the radionuclide is calculated based on precipitation, runoff, inflow, outflow, decay and discharges from local facilities

article of Salo and Tuomainen [14]. Effective half-lives of ^{137}Cs and ^{90}Sr in seawater were calculated from the inventories in seawater for the period 1987–2006 and were found to be 10 years for ^{137}Cs and 16 years for ^{90}Sr. These half-lives are in very good agreement with the values measured by Salo and Voipio [18] after the nuclear weapons tests ceased at the end of 1960s and early 1970s, when they reported effective half-lives of 9 years for ^{137}Cs and 15 years for ^{90}Sr. No differences can thus be seen in the effective half-lives of these radionuclides before and after the Chernobyl accident. The shorter effective half-life of ^{137}Cs in seawater as compared to ^{90}Sr reflects the higher ability of ^{137}Cs to be absorbed on sedimenting particles. The Baltic Sea sediments are very rich in clay particles, which effectively retain ^{137}Cs.

Inventories of ^{137}C and ^{90}Sr in Baltic Sea sediments

Sediments are the final sink for most particles and organic material present in the seawater. Radionuclides, especially ^{137}Cs, have a tendency to bind to these particles as they settle. The sedimentation rate is quite high in the Baltic Sea and varies widely depending on the area and local environmental factors. Sedimentation rates vary between 0.2 and 29 mm/year in different soft-bottom areas of the Baltic Sea [4, 13]. Even though the deposition of radionuclides from global fallout was rather evenly distributed, the areal distribution of radionuclides in sediment varied due to the differences in sedimentation rates between areas [14]. After the uneven deposition of ^{137}Cs from the Chernobyl accident the areal distribution of ^{137}Cs in sediment became more uneven (Fig. 1), the majority of ^{137}Cs being accumulated in the sediments of the Bothnian Sea, the Gulf of Finland, and in coastal sediments.

Total ^{137}Cs inventories in Baltic Sea sediments can be estimated by first calculating the average content of ^{137}Cs in the sediments of different sub-basins (Bq/m^2). Since the sediment samples are collected from soft bottoms, the average content is multiplied by the area of soft bottom in a particular sub-basin [14]. The activities in hard bottom

sediments are calculated by taking into consideration the area of hard bottoms in different sub-basins [14] and applying the assumption that the activity of ^{137}Cs in a hard bottom is 1/20 of that observed in the soft bottom of a particular sub-basin [12]. The total inventory of ^{137}Cs in Baltic Sea sediment is then obtained by combining the values for soft and hard bottoms, and is presented in Fig. 5a. Due to the limited number of ^{90}Sr measurements, inventories of ^{90}Sr in sediment (Fig. 5b) could only be calculated for selected years using the data from Salo et al. [8] and unpublished data from Finland. The activity in hard bottom sediments was estimated by assuming that it contains 1/5 of the amount of ^{90}Sr present in soft bottoms, as determined by Salo et al. [14]. Recently, it has been estimated that the sediments have contained about 26 TBq of ^{90}Sr in the 2000s [12].

Discussion

The total budget of ^{137}Cs and ^{90}Sr in the Baltic Sea based on precipitation, outflow, inflow, runoff, and physical decay values can be compared to the inventories calculated for water and sediment as illustrated in Fig. 5. The importance of the biota in removing or binding ^{137}Cs and ^{90}Sr is so low (<1%) that it was omitted in the evaluations. In the first few years after the Chernobyl accident the sum of ^{137}Cs in the water phase and in sediments was significantly lower than the total budget suggests. This was probably due to two reasons. First, a large proportion of ^{137}Cs descended with particles within the water column and the calculated inventory value for ^{137}Cs in the water phase has a relatively large uncertainty. Secondly, the newly deposited, soft sediment layer is often lost during sampling [7], which can lead to underestimated values for ^{137}Cs in sediments within the first few years after the Chernobyl accident. Overall, the calculation of the ^{137}Cs inventory in the sediments of some sub-basins in the Baltic Sea has sometimes been based on a single measurement, which has lead to large uncertainty in the estimated sediment values. This is reflected in the data for 1993 and 1994 (Fig. 5a), when the higher than usual ^{137}Cs inventory in sediment was caused by a single value obtained for the Bothnian Sea, the major sink for radioactivity in the Baltic Sea, where unusually high measurements were reported for 1993 and 1994. Inventories of seawater are generally based on a larger amount of data than those for sediments and the estimations are thus more reliable. Currently, 70% of the ^{137}Cs in the Baltic Sea is found in sediments. During the last 10 years the measured inventories in seawater and sediments combined have been about 7% larger than the total budget suggests. This agreement is relatively good taking into consideration the large uncertainties associated with these calculations.

Sr-90 is transferred into sediments significantly more slowly than ^{137}Cs, reflected by its longer effective half-life in water (16 year) as compared to ^{137}Cs (10 year). Most ^{90}Sr is still found in the water today, even though about 40 years has passed since majority of ^{90}Sr was deposited into the Baltic Sea. Less than 15% of ^{90}Sr is currently estimated to be present in the sediments. The inventories of measured ^{90}Sr in seawater and sediments combined are about 14% lower than the total budget would suggest, and this difference has remained relatively constant over the years (Fig. 5b). All the above-mentioned calculations are approximate, and whether the detected difference is caused by underestimation of ^{90}Sr in sediments or overestimation of the source terms (e.g., deposition data) is still to be resolved. It is important to remember that the calculated amounts of ^{90}Sr in sediments are based on limited data.

Conclusions

The Baltic Sea is one of the most contaminated sea areas in the world due to radioactive contamination from the Chernobyl accident and from nuclear fallout resulting from weapons testing. It provides an excellent case study to evaluate inventories and total budgets of radionuclides because of the limited water exchange, its rather small size and the well developed monitoring program that covers the whole Baltic Sea. Both ^{137}Cs and ^{90}Sr are long-living radionuclides that will remain in the environment for long time. To better protect both people and the surrounding environment from the harmful effects of radiation, it is important to understand the long-term behaviour of these radionuclides. Even though the radiation dose to people is low, it is also important to consider the other biota that may be more exposed (e.g., those in close contact with sediments). Due to its particle-reactive nature, ^{137}Cs is transferred into the sediments more rapidly than ^{90}Sr. The effective half-lives of these nuclides in seawater were found to be 10 and 16 years, respectively. Sediments are becoming an important depository of these radionuclides in the Baltic Sea, but not necessarily the final depository, as the radionuclides may be released from the sediments under different environmental conditions or through human activity.

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