

# Thematic assessment of long-term changes in radioactivity in the Baltic Sea, 2007-2010



**Helsinki Commission**

Baltic Marine Environment Protection Commission



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# 1 Introduction

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Radioactive substances occur naturally in the environment - mainly from the substances of which the solar system and the Earth were originally formed and partly from the Earth's atmosphere by the slowing down of particles from the sun. Examples of the former include isotopes of uranium, thorium and potassium which have half-lives comparable to the age of the Earth, i.e. billions of years. Examples of the latter include tritium (<sup>3</sup>H, super heavy hydrogen) and carbon-14 (<sup>14</sup>C) with half-lives of 12 years and 5,700 years, respectively.

The long-lived isotopes <sup>235</sup>U, <sup>238</sup>U and <sup>232</sup>Th are transformed by radioactive decay into a series of decay products that are also radioactive themselves, thus adding to the number of radioactive substances in the environment. Examples include <sup>226</sup>Ra, <sup>210</sup>Pb and <sup>210</sup>Po, with half-lives of 1,600 years, 22 years and 140 days, respectively.

The marine environment thus contains naturally occurring radionuclides. One cubic metre of sea-water typically contains 1,000 Bq <sup>3</sup>H, 4 Bq <sup>14</sup>C, 40 Bq <sup>238</sup>U, 4 Bq <sup>226</sup>Ra, 4 Bq, <sup>210</sup>Pb, 4 Bq <sup>210</sup>Po and 12,000 Bq <sup>40</sup>K (National Academy of Sciences, 1971).

The development and use of nuclear power for military and peaceful purposes have resulted in the production of a number of man-made radioactive substances. Explosions of nuclear weapons in the atmosphere distribute radioactive substances via air in the environment, while underground nuclear explosions release little or no radiation into the environment. While routine operations of nuclear power plants give rise to small controlled discharges of radioactive substances, an accident can cause the release of considerable amounts of radioactivity. Man-made radionuclides of particular concern to man and the environment are <sup>90</sup>Sr and <sup>137</sup>Cs, which are both formed by nuclear fission. As both of these radioisotopes have half-lives of about 30 years, they remain in the environment for many years when released. Furthermore, <sup>90</sup>Sr and <sup>137</sup>Cs are readily transported through food chains since strontium and caesium have chemical similarities to calcium and potassium, which means that they may contaminate food and expose humans

to radioactivity through ingestion. Other man-made radionuclides of concern are <sup>239</sup>Pu and <sup>99</sup>Tc, with half-lives of 24,000 years and 210,000 years, respectively. <sup>14</sup>C is also a man-made nuclide in addition to natural origin.

The occurrence of man-made radioactive substances found in the Baltic Sea is due to four main events:

1. During 1950-1980, the United States and the Soviet Union carried out atmospheric nuclear weapons tests which peaked in the 1960s causing radioactive fallout throughout the northern hemisphere. This pollution is still noticeable in the seas and on land (UNSCEAR, 2000).
2. The accident at the Chernobyl nuclear power plant in 1986 caused heavy pollution in the vicinity of the power plant and also considerable fallout over the Baltic Sea.
3. The two European facilities for reprocessing spent nuclear fuel - Sellafield in the UK and La Hague in France - have both discharged radioactive substances into the sea. Some of this radioactivity has been transported by sea currents to the North Sea, from where a small proportion has entered the Baltic Sea.
4. Authorised discharges of radioactivity into the sea occurring during the routine operation of nuclear installations in the Baltic Sea region (nuclear power plants and nuclear research reactors) have also contributed.

The impact of non-nuclear facilities (e.g. hospitals, industry) on the radioactivity in the Baltic Sea is negligible and very local (Ilus et al, 2000).

Although the nuclear accident that occurred after the March 2011 Great East Japan Earthquake and tsunami in Fukushima, Dai-ichi, Japan, falls outside of the period considered in this assessment, it is of interest to scale its impact on the Baltic Sea area as compared to other sources. The accident resulted in a major contamination of the nearby Pacific coastal environment; however, radioactive fallout over the Baltic Sea from the nuclear accident in Japan was very small and was not detected in sea-water and fish. The corresponding radiological risks are estimated to be negligible.

This report describes work carried out by HELCOM's project on the Monitoring of Radioactive Substances in the Baltic Sea (MORS-PRO) during

the period 2007-2010. Chapter 2 defines the sources of the main man-made radioactivity in the Baltic Sea. Chapter 3 describes the levels of man-made radionuclides in seawater, sediments and biota. Chapter 4 presents work on modelling and evaluations of the risks to man and environment caused by radioactivity in the Baltic Sea. Chapter 5 summarises the project's conclusions. Finally,

Chapter 6 presents the consequent recommendations with the work on data quality presented as an Appendix.

In addition to authors mentioned in the assessment, the following persons from the MORS expert group have also contributed data:

Country/Institute	Person	Institute/department
Estonia	Ms. Eia Jakobson	Environmental Board Radiation Safety Department
Poland	Ms. Maria Suplinska	Central Laboratory for Radiological Protection
Poland	Ms. Tamara Zalewska	Institute of Meteorology and Water Management National Research Institute Maritime Branch
Russia	Ms. Anna Osokina	Khlopin Radium Institute
Russia	Mr. Andrey Stepanov	Khlopin Radium Institute Division for Radioecological and Analytical Research
International Atomic Energy Agency	Ms. Iolanda Osvath	International Atomic Energy Agency, Environment Laboratories, Monaco

## 2 Sources of radioactivity in the Baltic Sea

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<sup>3</sup> BSH, Federal Maritime and Hydrographic Agency, Germany

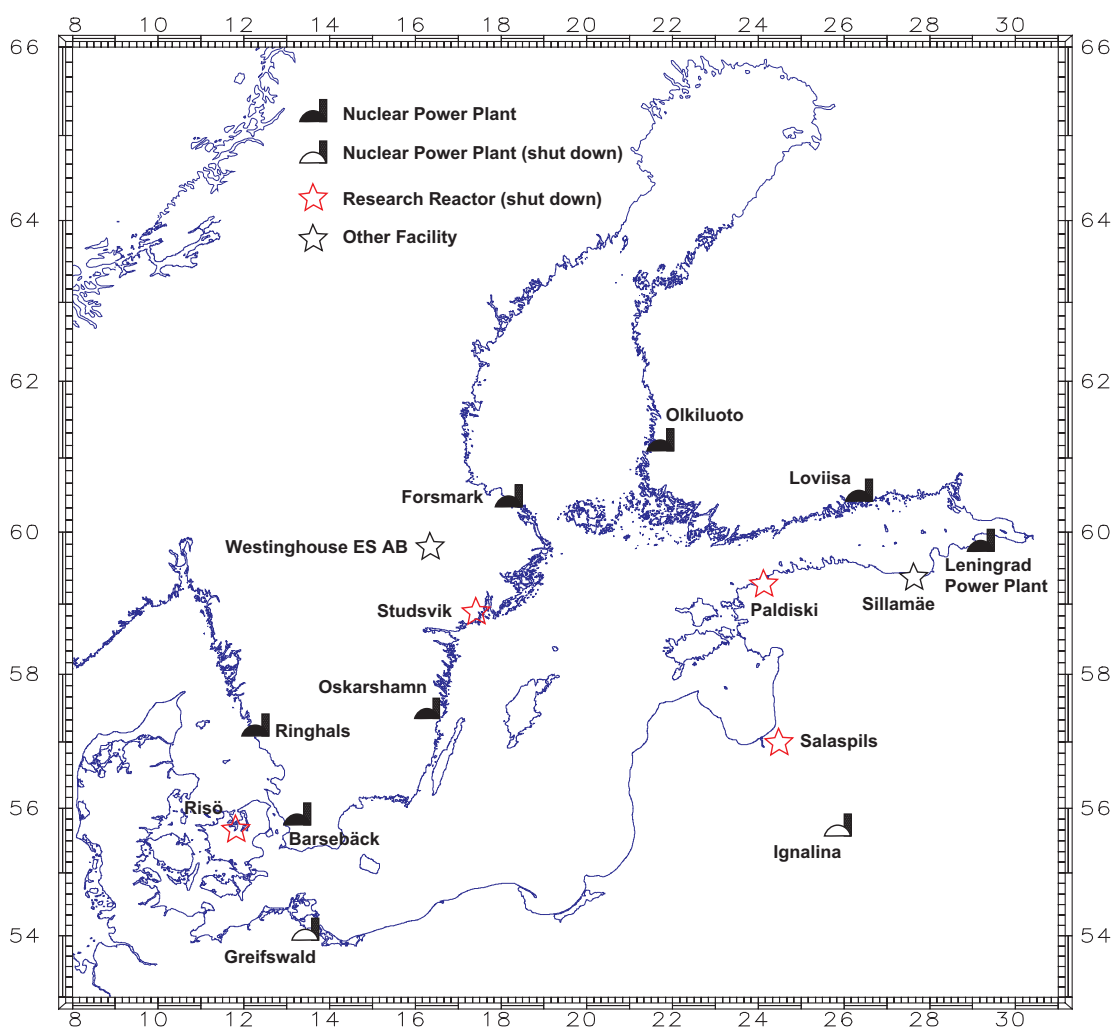
- Non-nuclear facilities in the Baltic Sea drainage area (hospitals, non-nuclear industries, etc.).
- Discharges from facilities located outside the Baltic Sea drainage area.
- The Chernobyl accident.
- Atmospheric nuclear weapons tests.

The injection of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  to the Baltic Sea from the different sources are presented in **Figure 2.2**.

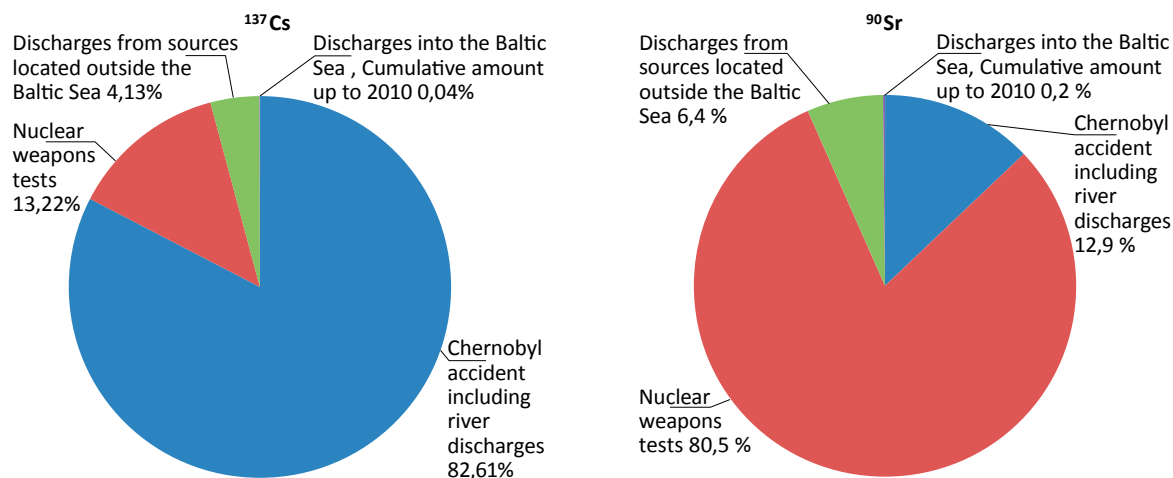
### 2.1 Introduction

This chapter covers the different sources of the current amount of radioactive substances found in the Baltic Sea. The sources are divided as follows:

- Nuclear Facilities in the Baltic Sea drainage area (NPPs, research reactors, waste handling, fuel handling, etc.). The locations of the facilities are shown in **Figure 2.1**.



**Figure 2.1.** Location of nuclear facilities in the Baltic Sea region.



**Figure 2.2** Total injections of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  into the Baltic Sea from different sources up to 2010.

## 2.2 Discharges from facilities in the Baltic Sea drainage area

### Nuclear Facilities (NPPs, research reactors, waste handling, fuel handling, etc.)

This includes all nuclear facilities (power reactors, research reactors, waste handling facilities, fuel

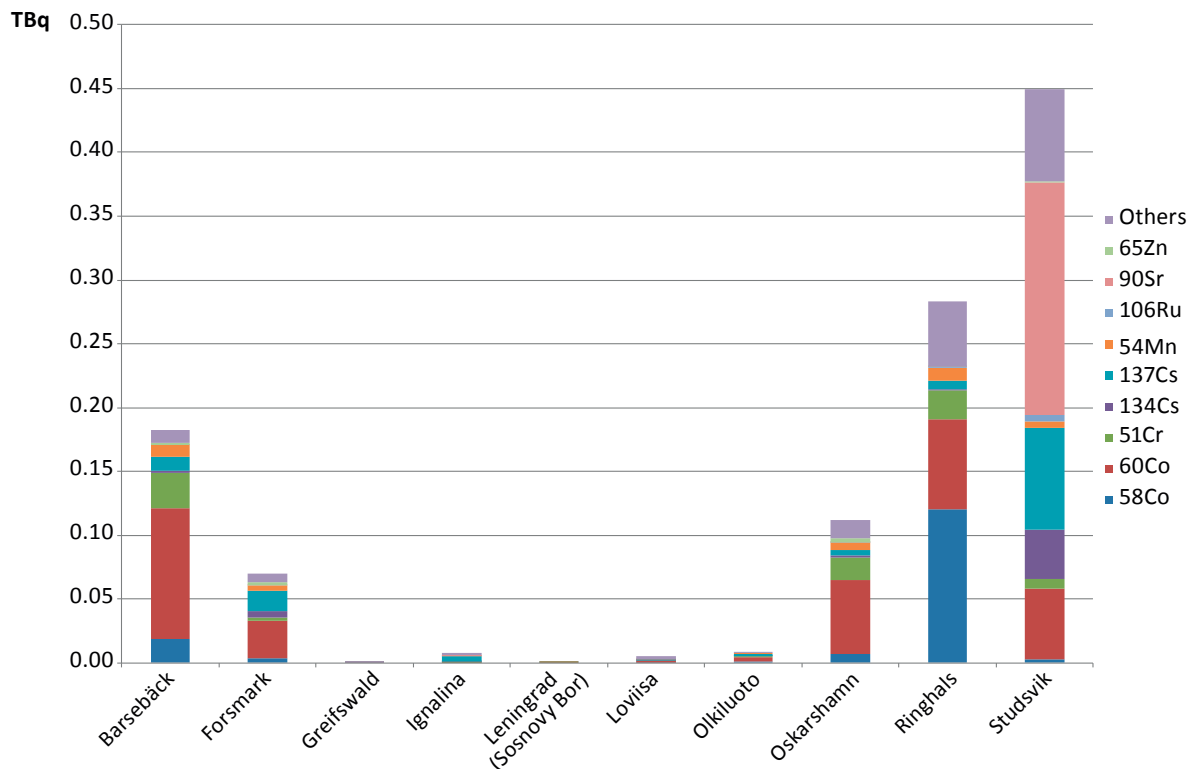
production, etc.) that are located in the drainage area of the Baltic Sea and which discharge directly or indirectly into the sea. The main characteristics of the facilities are summarised in **Table 2.1**.

**Table 2.1.** Nuclear facilities in the drainage area of the Baltic Sea and the main discharge nuclides.

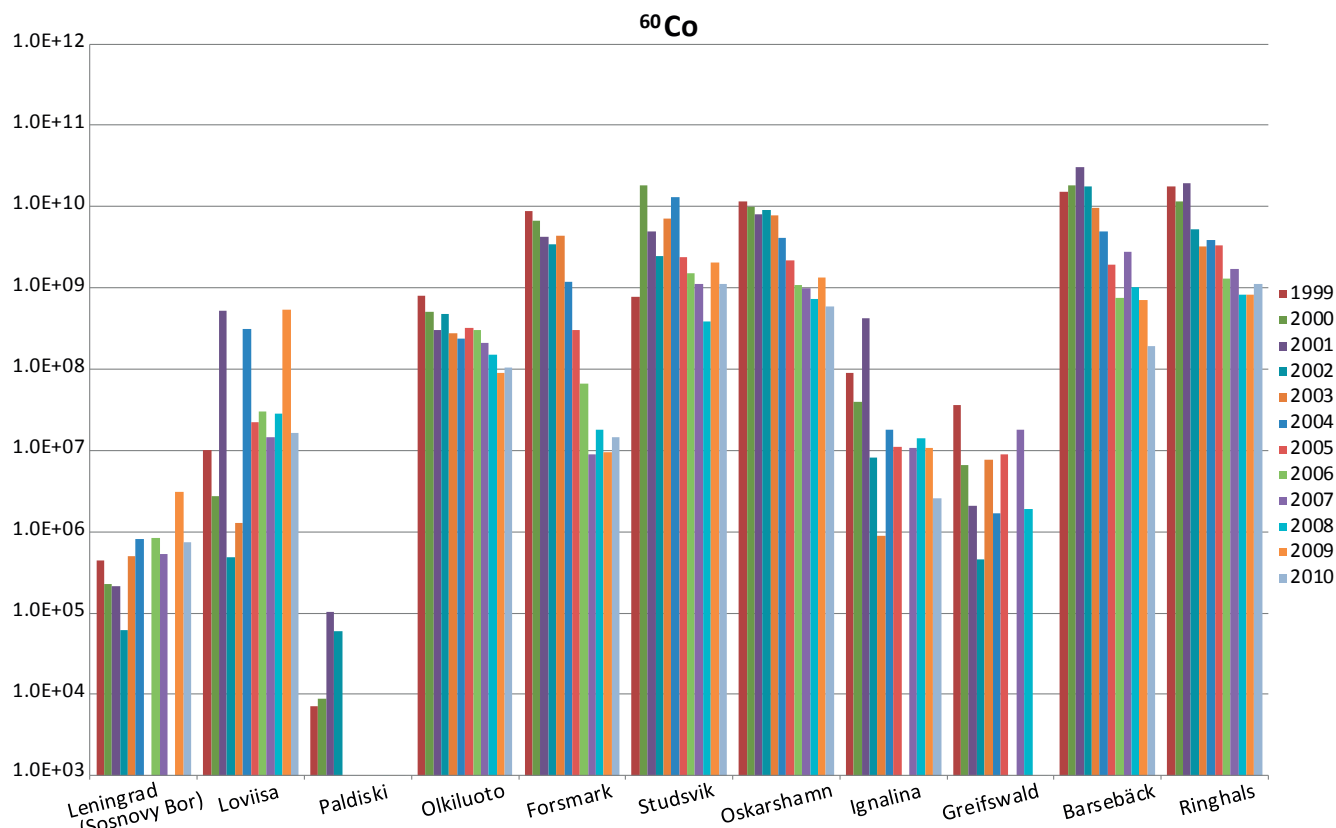
Facility	Country	Type of facility; number of units	Main radionuclides discharged	Remarks
Loviisa	Finland	Power plant; 2 PWR	$^3\text{H}$ , $^{60}\text{Co}$ , $^{137}\text{Cs}$ , $^{110\text{m}}\text{Ag}$ , $^{124}\text{Sb}$ , $^{58}\text{Co}$ , $^{54}\text{Mn}$	
Olkiluoto	Finland	Power plant; 2 BWR	$^3\text{H}$ , $^{60}\text{Co}$ , $^{137}\text{Cs}$ , $^{51}\text{Cr}$ , $^{58}\text{Co}$ , $^{54}\text{Mn}$ , $^{134}\text{Cs}$	
Greifswald	Germany	Power plant; 5 PWR	$^3\text{H}$ , $^{137}\text{Cs}$ , $^{60}\text{Co}$	Shut down in 1990
Ignalina	Lithuania	Power plant; 2 RBMK	$^3\text{H}$ , $^{137}\text{Cs}$ , $^{60}\text{Co}$	First reactor was shut down in 2004 and the second in 2009
Leningrad	Russia	Power plant; 4 RBMK	$^{137}\text{Cs}$ , $^{60}\text{Co}$	H-3 not reported
Barsebäck	Sweden	Power plant; 2 BWR	$^3\text{H}$ , $^{60}\text{Co}$ , $^{51}\text{Cr}$ , $^{58}\text{Co}$ , $^{137}\text{Cs}$ , $^{54}\text{Mn}$	First reactor was shut down in 1999 and the second in 2005
Forsmark	Sweden	Power plant; 3 BWR	$^3\text{H}$ , $^{60}\text{Co}$ , $^{137}\text{Cs}$ , $^{134}\text{Cs}$ , $^{58}\text{Co}$ , $^{54}\text{Mn}$ , $^{51}\text{Cr}$ , $^{65}\text{Zn}$ , $^{124}\text{Sb}$	
Oskarshamn	Sweden	Power plant; 3 BWR	$^3\text{H}$ , $^{60}\text{Co}$ , $^{51}\text{Cr}$ , $^{58}\text{Co}$ , $^{110\text{m}}\text{Ag}$ , $^{54}\text{Mn}$ , $^{65}\text{Zn}$ , $^{125}\text{Sb}$ , $^{137}\text{Cs}$	
Ringhals	Sweden	Power plant; 3 PWR, 1 BWR	$^3\text{H}$ , $^{58}\text{Co}$ , $^{60}\text{Co}$ , $^{124}\text{Sb}$ , $^{51}\text{Cr}$ , $^{54}\text{Mn}$ , $^{125}\text{Sb}$ , $^{137}\text{Cs}$ , $^{95}\text{Nb}$	
Risø	Denmark	Research reactor	$^3\text{H}$	Shut down in 2000
Salaspils	Latvia	Research reactor	$^3\text{H}$ , $^{137}\text{Cs}$ , $^{134}\text{Cs}$	Shut down in 1998
Studsvik	Sweden	Research reactor, Waste handling facility	$^3\text{H}$ , $^{90}\text{Sr}$ , $^{137}\text{Cs}$ , $^{60}\text{Co}$ , $^{134}\text{Cs}$ , $^{192}\text{Ir}$ , $^{144}\text{Ce}$ , $^{54}\text{Mn}$ , $^{106}\text{Ru}$	Research reactor was shut down in 2005
Paldiski	Estonia	Training centre for nuclear submarines	$^3\text{H}$ , $^{137}\text{Cs}$ , $^{90}\text{Sr}$	Shut down in 1989
Sillamäe	Estonia	Chemical metallurgy plant, waste depository	$^{238}\text{U}$ , $^{226}\text{Ra}$ ,	Construction work on the final cover system of the waste depository was completed in October 2008
Westinghouse Electric Sweden AB	Sweden	Fuel fabrication plant	$^{234}\text{U}$ , $^{238}\text{U}$ , $^{60}\text{Co}$	

The discharge pattern for most of the NPPs is similar; the most abundant nuclides present in the discharges are shown in **Table 2.1**. The amounts

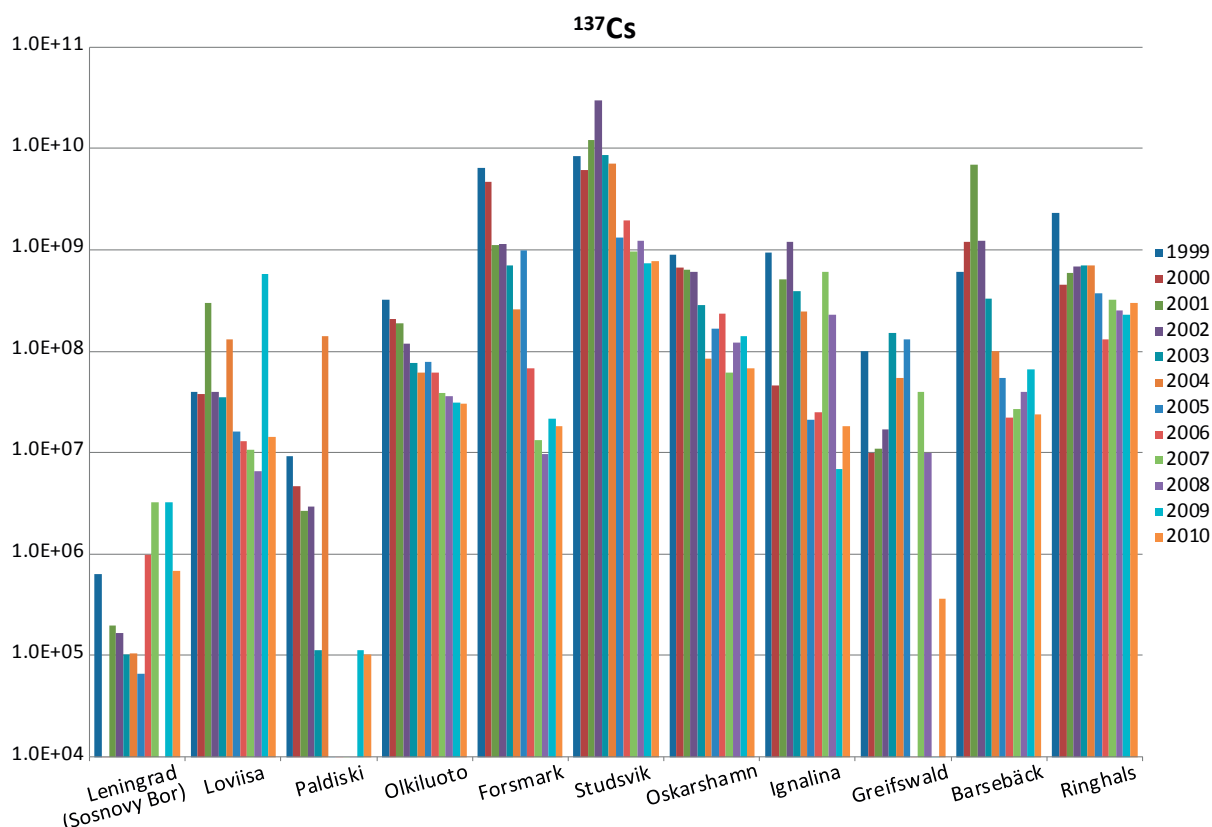
of the most significant radionuclides discharged are shown in **Figures 2.3-25**.



**Figure 2.3.** Total aquatic discharges from local nuclear facilities into the Baltic Sea in 1999-2010 (excluding  $^3\text{H}$ ).



**Figure 2.4.** Annual  $^{60}\text{Co}$  discharges from local nuclear facilities into the Baltic Sea in 1999-2010.



**Figure 2.5.** Annual  $^{137}\text{Cs}$  discharges from local nuclear facilities into the Baltic Sea in 1999-2010.

### Non-nuclear facilities (e.g. hospitals, non-nuclear industries, etc.)

This category includes hospitals, research institutes, non-nuclear industries using radioactive substances and NORM-industries. It is not possible to obtain reliable data on discharges from these sources since information is scarce.

Radionuclides are used for various purposes in the fields of industry, medicine and research, for example, and their use is increasing. However, according to reports of UNSCEAR, their contribution to overall man-made exposures is relatively insignificant. Most radionuclides used in hospitals are short-lived and as their discharges are small, their impact on the radioactivity in the Baltic Sea is negligible and very local (Ilus & Ilus, 2000).

## 2.3 Discharges from facilities located outside the Baltic Sea area

### Nuclear reprocessing plants

A small proportion of the discharges from Sellafield, situated on the west coast of England and discharging into the Irish Sea, and La Hague, situated on the northwest coast of France and discharging into the English Channel are transported by the inflow of saline water through the Danish Straits into the Baltic Sea. The transport time for the radionuclides is about 4-5 years after discharge into the Irish Sea (Sellafield) or about two years after discharge into the English Channel (La Hague) (Nies et al., 1995). Model calculations indicate that only about 4% of the discharges from Sellafield and about 8% of the discharges from La Hague reach the Skagerrak. Due to the efficient mixing of water masses in the Kattegat and the Belt Sea, the main part of the activity returns to the Skagerrak and only about 1% enters the Baltic Sea (Nielsen et al., 1995).



## Chernobyl accident

The accident at the Chernobyl NPP occurred in April 1986 and has since then been the main source of radioactivity in the Baltic Sea (Table 2.2).

**Table 2.2. Total injections of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  into the Baltic Sea from different sources.**

Source	$^{137}\text{Cs}$ TBq	% of total amount	$^{90}\text{Sr}$ TBq	% of total amount
Chernobyl accident <sup>1)4)</sup> / incl. river discharges <sup>5)</sup>	4700 / 300	83	80	13
Nuclear weapons tests <sup>6)</sup>	800	13	500	81
Discharges from sources located outside the Baltic Sea <sup>2)3)</sup>	250	4	40	6
Discharges into the Baltic Sea <sup>1)</sup> , cumulative amount up to 2010	2.4	0.04	1.04	0.2

<sup>1)</sup> based on measurements

<sup>2)</sup> estimated

<sup>3)</sup> according to Nies et al., 1995

<sup>4)</sup> according to Nielsen et al., 1999

<sup>5)</sup> according to Ilus & Ilus, 2000

<sup>6)</sup> according to Nielsen (pers. comm.)

The total input of  $^{137}\text{Cs}$  from the Chernobyl accident into the Baltic Sea area was estimated to 4,700 TBq (Nielsen et al., 1999). 80 TBq of  $^{90}\text{Sr}$  (decay-corrected to 1991) were also injected into the Baltic Sea as a consequence of the accident (Nies et al., 1995). The deposition of the Chernobyl fallout was very unevenly distributed in the drainage area of the Baltic Sea; the most contaminated areas were situated in the land areas surrounding the Bothnian Sea and the eastern Gulf of Finland.

## River discharges

The amount of Chernobyl-derived  $^{137}\text{Cs}$  carried into the Baltic Sea by river runoff has been evaluated in Finland for all Finnish rivers discharging into the Baltic Sea; in Russia for five rivers discharging from the former Soviet Union; and in Poland for the River Vistula. The total river input of  $^{137}\text{Cs}$  was estimated at 300 TBq during 1986-1996 (Ilus & Ilus, 2000).

## Atmospheric nuclear weapons tests

The impact of global fallout caused by the nuclear weapons tests in the 1950s and 1960s as a source of radioactivity in the Baltic Sea has been thoroughly considered in the first Joint Evaluation Report of the HELCOM/MORS Group (Nies et al., 1995). According to recent calculations (Nielsen, pers. comm.), the total injections of weapons-test

$^{90}\text{Sr}$  and  $^{137}\text{Cs}$  into the Baltic Sea were 500 and 800 TBq ( $5.0\text{E}+14$  and  $8.0\text{E}+14$  Bq), respectively (decay-corrected to 1998). Inventories based on measured concentrations of these nuclides in water and sediments of the Baltic Sea resulted in similar values: 490 TBq for  $^{90}\text{Sr}$  and 620 TBq for  $^{137}\text{Cs}$  (calculated to 1981) (Salo et al., 1986).

## Dumping of radioactive waste

Five officially confirmed incidents of dumping of radioactive waste at three different dump sites have been reported in the Baltic Sea region. This small-scale dumping was carried out in the late 1950s or early 1960s. A radiological assessment of the dumping showed that doses to man from these activities were negligible (Nielsen et al., 1999).

## 2.4 Conclusions

The most significant source with respect to the total inventory of artificial radionuclides in the Baltic Sea is the fallout caused by the accident at the Chernobyl NPP in 1986. The most important radionuclides present in the deposition were  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . The total input of  $^{137}\text{Cs}$  from Chernobyl to the Baltic Sea has been estimated at 4,700 TBq.  $^{134}\text{Cs}$  is now no longer detectable due to its short half-life of two years. The post-Chernobyl river dis-

charges of  $^{137}\text{Cs}$  were estimated in the MARINA-BALT Study at 300 TBq, comprising 6-7% of the total injection.

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

The predominant radionuclide in the discharges from the nuclear power plants and research reactors in the Baltic Sea region is  $^3\text{H}$ . The total discharges of  $^3\text{H}$  from these local sources have amounted to 3,500 TBq and those of other beta-

gamma nuclides to about 24 TBq until the end of 2010. The total discharges of alpha nuclides have been 0.005 TBq.

For  $^{137}\text{Cs}$ , the main source was the fallout from Chernobyl (83%) and the nuclear weapons test fallout (13%); the remaining 4% comes from sources outside the Baltic Sea, mainly Sellafield and La Hague. Prior to 1990, the inflow from contaminated water from the North Sea was also a contributor to the inventory of  $^{137}\text{Cs}$  from the discharges from Sellafield. There existed a strong correlation between the  $^{137}\text{Cs}$  activity concentration and the salinity before 1986. For  $^{90}\text{Sr}$ , the main source of contamination was the fallout from the nuclear weapons tests (81%), while the proportion from the Chernobyl fallout was smaller (13%). The remaining 6% comes from sources outside the Baltic Sea, mainly Sellafield and La Hague.

# 3 Radioactivity in the Baltic Sea (trends and regional distribution)

## 3.1 Radionuclides in seawater

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### Introduction

This chapter describes the distribution of artificial radionuclides in the seawater of the Baltic Sea from 2007 – 2010. During this period, nine countries contributed the results of about 850 seawater samples from all sub-regions of the Baltic Sea to the common database. The monitoring programme covered all sub-basins during the report period, with the exception of the Gulf of Riga (see **Figure 3a.1**). As presented in earlier reports (Panteleev *et al.* 1995; Mulsow *et al.* 2003; Herrmann *et al.* 2007), the predominant radionuclide in the Baltic Sea is <sup>137</sup>Cs as it was released by the Chernobyl accident in great amounts in 1986. <sup>134</sup>Cs - the other main contaminant of the Chernobyl event - has vanished to concentrations below detection limit because of its relatively short physical half-life of 2.07 years.

Other artificial radionuclides of relevance in the seawater of the Baltic Sea are <sup>90</sup>Sr, (<sup>239</sup>+<sup>240</sup>)Pu and <sup>99</sup>Tc. The sources of the mentioned radionuclides are described in Chapter 2. Generally, concentrations mentioned in this chapter are understood as activity concentrations. A detailed description of the methods was given in the earlier joint report (HELCOM 1995). The collecting of monitoring data was accompanied by a thorough programme of quality assurance covering both <sup>137</sup>Cs and <sup>90</sup>Sr in seawater in annual exercises; this is presented in the appendix of this report.

### Distribution and temporal evolution of <sup>137</sup>Cs

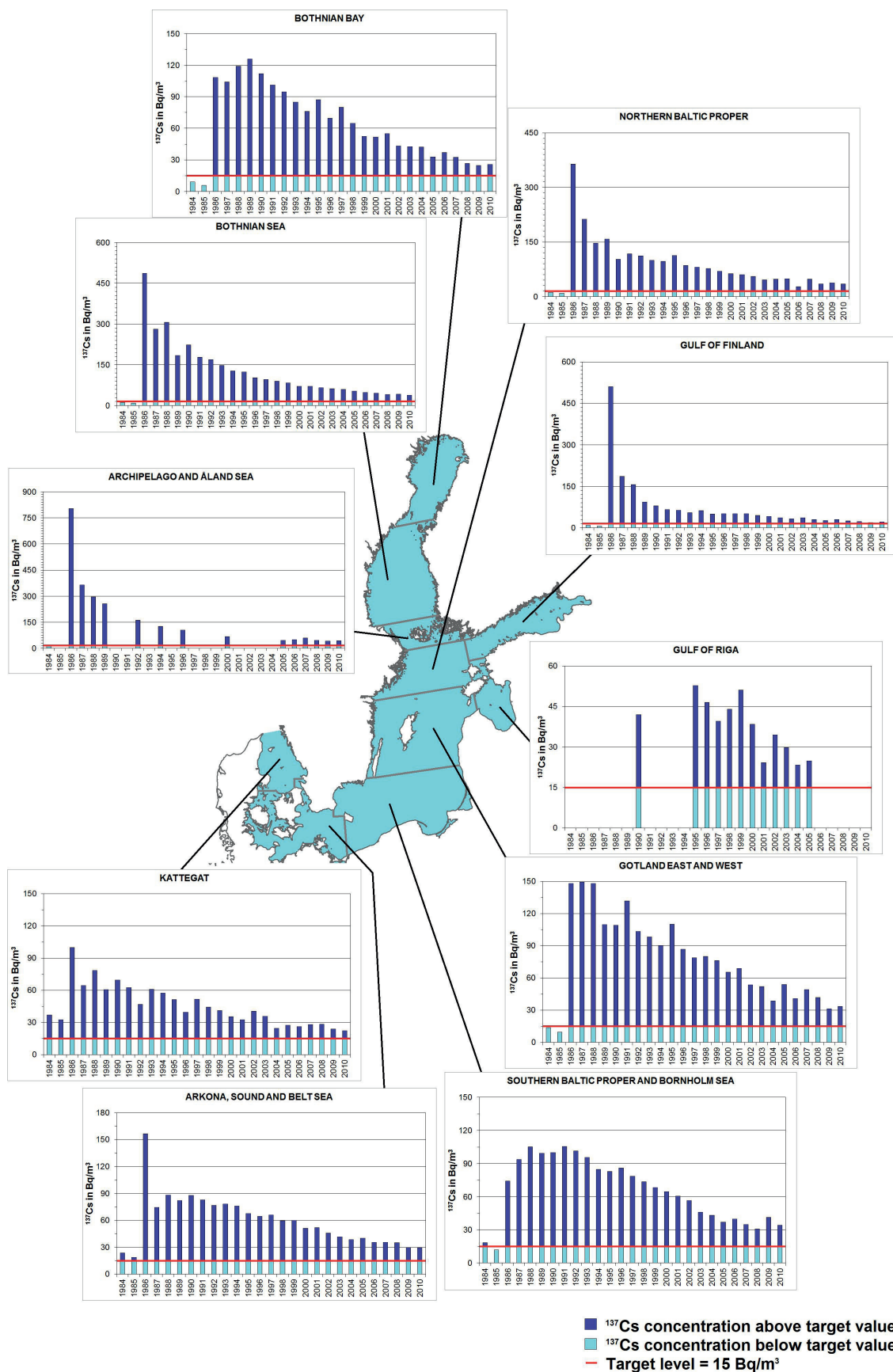
The fate of any pollutant introduced into the sea is determined by its chemical properties as well as by the hydrographic conditions of the sea itself. As a relatively small, semi-enclosed brackish sea - connected to the North Sea and thereby to the North Atlantic only by the narrow Danish Straits - the Baltic Sea suffers possibly more than any other part of the World Ocean from pollution. The Chernobyl

accident made this situation most clear as its legacy is still abundant 24 years after the event.

The Chernobyl accident caused a very uneven <sup>137</sup>Cs deposition in the Baltic Sea region. The Bothnian Sea and the Gulf of Finland were the two most contaminated sea areas. Since 1986, the spatial and vertical distribution of Chernobyl derived <sup>137</sup>Cs has changed as a consequence of river discharges, mixing of water masses, sea currents and sedimentation processes (Illus 2007). In the early phase after Chernobyl, the <sup>137</sup>Cs concentrations decreased rapidly in the Gulf of Finland and in the Bothnian Sea, while at the same time they increased in the Baltic Proper (Ref: 2007; **Figures 3a.1 and 3a.2**). During the period 2007-2010, concentrations of <sup>137</sup>Cs have continued to decrease in all regions of the Baltic Sea. At the beginning of the period, the highest <sup>137</sup>Cs concentrations were reported in the Archipelago and Åland Sea where the concentrations decreased from 58 to 44 Bq/m<sup>3</sup> during the monitoring period. In the Bothnian Sea and the Northern Baltic Proper, the concentrations decreased from about 50 to 42 and 37 Bq/m<sup>3</sup>, respectively. In 2010 in the Gulf of Finland, the concentrations were lower at around 29 Bq/m<sup>3</sup>.

<sup>137</sup>Cs concentrations in the Southern Baltic Proper and the Bornholm started at 46 Bq/m<sup>3</sup> in 2007 and decreased to around 40 Bq/m<sup>3</sup> in 2010. Concentrations in the Western Baltic have been lower and decreased from 42 to 35 Bq/m<sup>3</sup> during the monitoring period. The Kattegat, as the transition area to the North Sea, showed concentrations in surface water decreasing from 35 to 27 Bq/m<sup>3</sup> in 2010. The variation of <sup>137</sup>Cs in surface water between different sea areas of the Baltic Sea is 1.5 between the most contaminated Bothnian Sea and the least contaminated Kattegat.

The vertical distribution of radionuclides in the water column is influenced by physical and biological processes as described above. Whereas in former years the <sup>137</sup>Cs concentrations in surface water was higher compared to those in near-bottom water in the Baltic Proper, this situation has changed during the reporting period. Measured data show evidence that the Chernobyl contamination has finally reached waters deeper than 200 m by vertical transport processes. In other compartments of the Baltic, such as the Bothnian Sea, the Gulf of Finland and the Bothnian Bay,



**Figure 3a.1.**  $^{137}\text{Cs}$  concentrations (Bq/m<sup>3</sup>) in surface water (sampling depth  $\leq 10\text{m}$ ) in 1984-2010, as annual mean values by basin. Target values have been calculated as the average of pre-Chernobyl (1984-1985) concentrations.

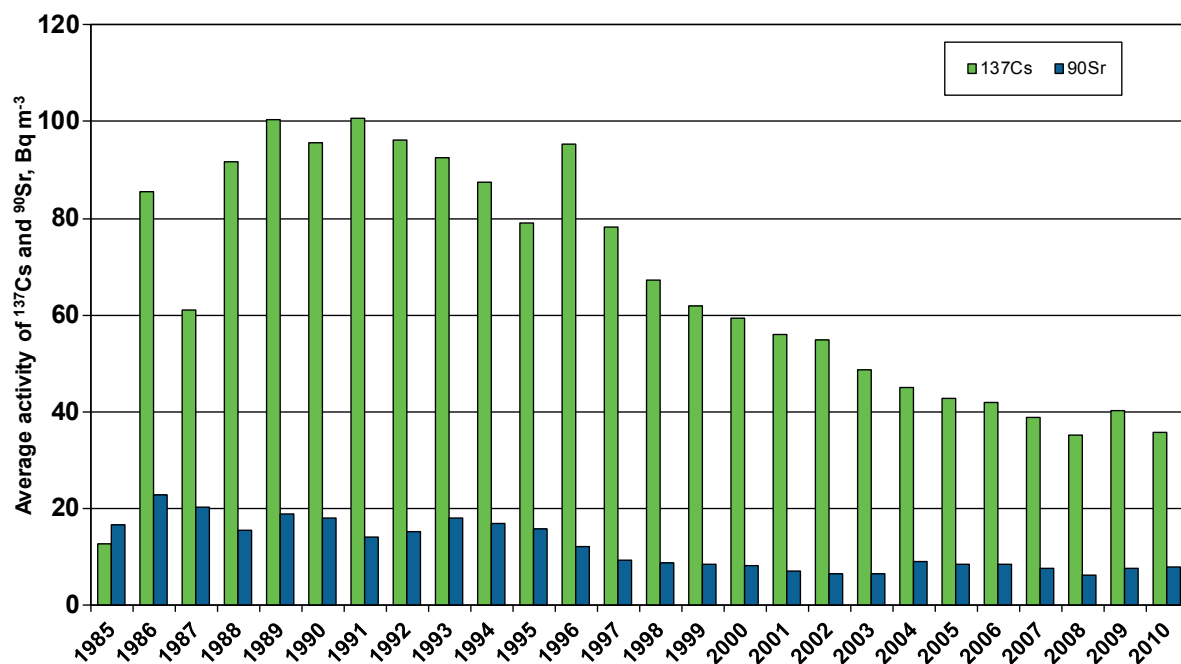


Figure 3a.2. Time series of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  mean concentrations in the Southern Baltic.

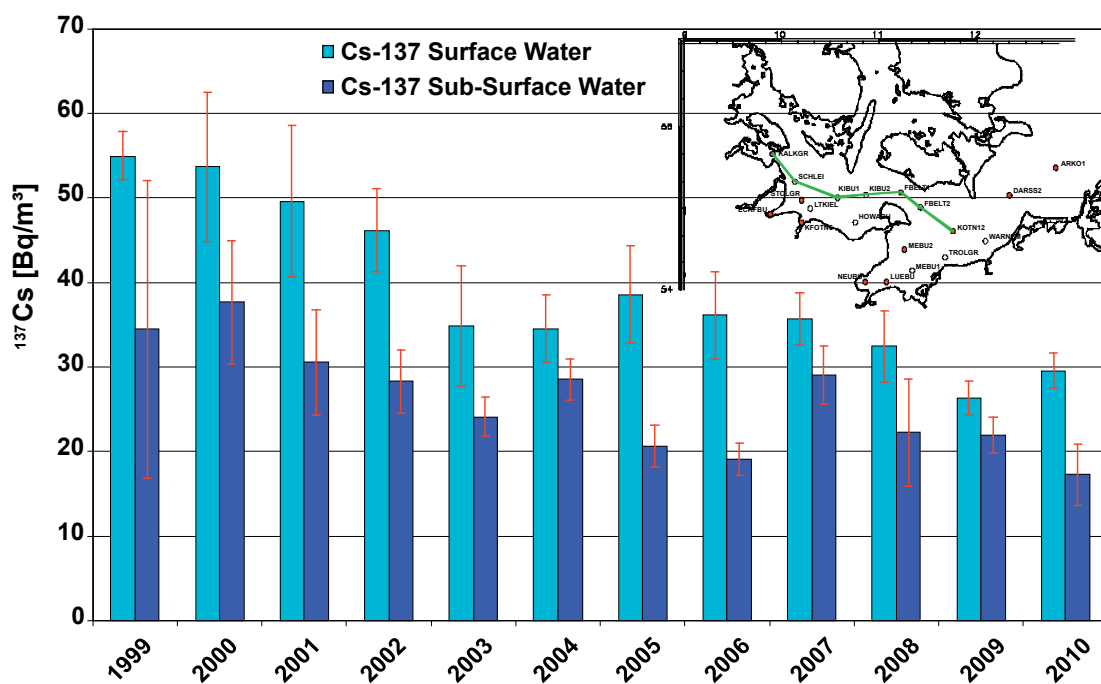


Figure 3a.3. Time series of  $^{137}\text{Cs}$  mean concentrations from stations characterising the inflow of bottom water into the Western Baltic.

this vertical exchange was much more efficient because of a lack of stratification; therefore, a homogeneous distribution with a clear tendency towards higher  $^{137}\text{Cs}$  concentrations in near-bottom water could be observed much earlier. No evidence of remobilization of  $^{137}\text{Cs}$  from bottom sediments has yet been detected, although long-term monitoring may bring more information about remobilization in the future. The circulation

of near-bottom waters in the Baltic Sea can also redistribute the  $^{137}\text{Cs}$  contamination by transferring contaminated near-bottom water from the Bothnian Sea to the Baltic Proper. The  $^{137}\text{Cs}$  concentration in near-bottom water is highest in the Bothnian Sea and decreases towards the Sound and the Kattegat, spanning from 51 to 4 Bq/m<sup>3</sup> during the reporting period.

The Western Baltic as a transition area between the North Sea and the Baltic Sea has special hydrographic conditions different from the rest of the Baltic Sea. Most notably, it is shallow, the average depth being around 20 m. The bottom water bears oxygen and high salinity, and is steadily supplied by currents from the North Sea. The surface water has a net current out of the Baltic because the great catchment area results in a surplus of fresh water into the Baltic Sea. This water exchange is not dominated by tidal currents but by wind forces that result in a current system of high intra-annual and inter-annual variability. As an indicator of the inflow of bottom water, the mean  $^{137}\text{Cs}$  concentration of seven selected stations is given in

**Figure 3a.3.** Besides the general decreasing trend of the  $^{137}\text{Cs}$  concentrations, the figure shows information on the variability of surface and bottom water concentrations, which was much less in recent years compared to 1999. The year with the smallest difference between surface and bottom water was 2008, obviously at a time when bottom water from the Kattegat with low  $^{137}\text{Cs}$  concentration did not reach the indicator stations.

The bottom water from the North Sea supplies the Baltic Sea with contaminants from the La Hague and Sellafield reprocessing plants (see Chapter 2) such as  $^{239}\text{Pu}$ ,  $^{99}\text{Tc}$  and  $^{129}\text{I}$ , whereas the outflowing surface water of the Baltic Sea represents a significant source of  $^{137}\text{Cs}$  to the North Sea. Its effects are detectable along the entire south coast of Norway at least until  $60^\circ\text{N}$ . Today, the Baltic Sea can be regarded as the strongest source of  $^{137}\text{Cs}$  to the North Atlantic, only comparable with the sediments of the Irish Sea. While the quantification of this source is still unclear because of the high variability of the outflow, it is estimated at tens of TBqs per year.

#### Effective half-life of $^{137}\text{Cs}$

Effective half-life is the time required for a given contaminant concentration to decrease 50% as a result of physical, chemical and biological processes. It is specific to the radionuclide and the environment where the radionuclide is present. Effective half-lives for  $^{137}\text{Cs}$  in different parts of the Baltic Sea have been calculated and are shown in **Table 3a.1**. Currently, the effective half-life of  $^{137}\text{Cs}$  in surface water varies from about nine years in the Bothnian Sea to about 11 years in the Baltic

Proper and the Gulf of Finland. The slightly longer residence time of  $^{137}\text{Cs}$  in the Baltic Proper and the Gulf of Finland is due to the inflow of more contaminated water from the northern part of the Baltic Sea and the higher river inflow.

In the time period following Chernobyl (1986-1988), the effective half-lives of  $^{137}\text{Cs}$  were much shorter in most contaminated regions: 0.8 years in the Gulf of Finland and 2.5 years in the Bothnian Sea. The shorter effective half-life of  $^{137}\text{Cs}$  in the Gulf of Finland as compared to the Bothnian Sea during 1986-1988 was probably due to the different water exchange and sedimentation processes in these two regions (Ilus *et al.* 1993). Over time, the effective half-lives increased in both regions. Today, the effective half-lives seem to have turned to decrease (Table 3a.1) since the amount of  $^{137}\text{Cs}$  due to the river inflow to the Baltic Sea has been decreasing.

The target level for  $^{137}\text{Cs}$  concentration in Baltic seawater is defined as  $15 \text{ Bq/m}^3$ , the average pre-Chernobyl concentration. Based on calculated effective half-lives, this level will be reached by 2017-23, which is earlier than previously estimated (Herrmann *et al.* 2007). However, it has to be noticed that these are only rough estimations and are only valid if the effective half-lives remain constant and no substantial remobilization of  $^{137}\text{Cs}$  from the sediment takes place.

**Table 3a.1.** Effective half-lives of  $^{137}\text{Cs}$  in surface water in different basins of the Baltic Sea (1993-2010).

Baltic Sea Region	Time period		
	1986–1988	1993–2006	1993–2010
Bay of Bothnia	-	10	9
Bothnian Sea	2,5	9	9
Gulf of Finland	0,8	13	11
Baltic Proper	-	15	11

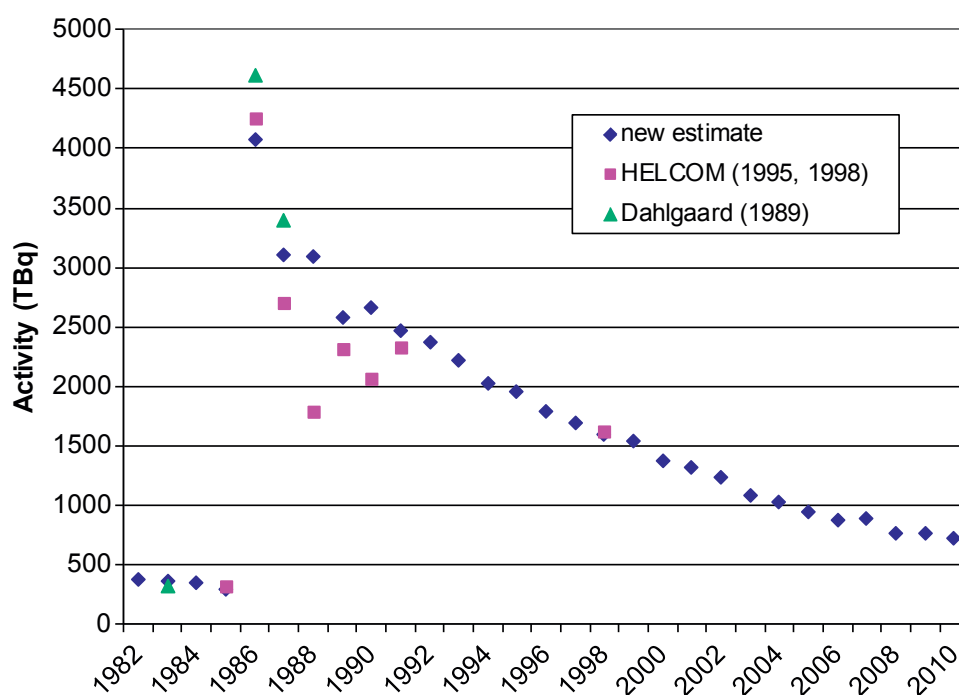
#### Inventories of $^{137}\text{Cs}$ in Seawater

Inventories of  $^{137}\text{Cs}$  in the Baltic seawater are given in **Table 3a.2**. These estimates show that the inventory of  $^{137}\text{Cs}$  in the Baltic water mass was 1540 TBq in 1999, and had decreased to 730 TBq in 2010 due to the sedimentation and physical decay. The inventories were estimated by calculating  $^{137}\text{Cs}$  inventories for seawater in various

regions of the Baltic Sea (Bothnian Bay, Bothnian Sea, Gulf of Finland, Gulf of Riga, Baltic Proper, Kattegat, and Belt Sea) and then combining these estimates. Inventories for different basins were calculated using their volumes (HELCOM, 1996) and their average  $^{137}\text{Cs}$  concentrations, which were calculated from the observation data. The temporal evolution of the  $^{137}\text{Cs}$  inventory in the Baltic seawater is presented in **Figure 3a.4**. Previous estimates for  $^{137}\text{Cs}$  inventories (Panteleev *et al.* 1995, HELCOM 1998; Dahlggaard 1989) are shown together with the new estimates. The HELCOM 1995 and 1996 estimates were calculated with assumption that the estimated mean concentration of  $^{137}\text{Cs}$  was the same for the entire Baltic Sea.

**Table 3a.2. Estimated  $^{137}\text{Cs}$  inventories in the Baltic seawater.**

Year	$^{137}\text{Cs}$ (TBq)
1999	1540
2000	1380
2001	1320
2002	1230
2003	1090
2004	1030
2005	950
2006	870
2007	890
2008	770
2009	770
2010	730



**Figure 3a.4.  $^{137}\text{Cs}$  inventories in the Baltic seawater during 1984-2010.**

## Other radionuclides

### $^{90}\text{Sr}$

$^{90}\text{Sr}$  concentrations in Baltic seawater varied in general from 5 to 15 Bq/m<sup>3</sup> in surface water during 2007-2010. Similar levels were detected in the water column and the near bottom water. The exception is the Kattegat, where the near bottom water originates from the North Sea and has activity concentrations of  $^{90}\text{Sr}$  of only 1-2 Bq/m<sup>3</sup>.  $^{90}\text{Sr}$

inventory in the Baltic Sea was about 165 TBq in 2010. This was less than half of the  $^{90}\text{Sr}$  inventory in 1985, the year before the Chernobyl accident. The  $^{90}\text{Sr}$  concentration decreases slowly with time and its behaviour in seawater is different from  $^{137}\text{Cs}$ .  $^{90}\text{Sr}$  is more soluble in water and its effective half-life in seawater is longer than that of  $^{137}\text{Cs}$ , around 20 years during 1987-2010.



### <sup>(239+240)</sup>Pu

Concentrations of <sup>(239+240)</sup>Pu were very low and varied generally from 1 to 10 mBq/m<sup>3</sup>.

### <sup>99</sup>Tc

For the detection of <sup>99</sup>Tc, seawater was only analysed from the Bornholm Sea, the Arkona Sea, the Kattegat and the Sound. Activity concentrations varied from 0.06 to 0.09 Bq/m<sup>3</sup> with the highest levels detected in the Kattegat. The main source of <sup>99</sup>Tc is the inflow of contaminated waters from the North Sea originating from the Sellafield nuclear reprocessing plant. <sup>99</sup>Tc concentrations have slightly decreased during the study period and the levels were three times lower in comparison to the previous period of 1999 - 2006.

### <sup>3</sup>H

The reported tritium activity concentration in surface water covered the whole reporting period. It varied generally from 1,000 to 2,000 Bq/m<sup>3</sup>, some values up to 12,700 Bq/m<sup>3</sup> were reported from the Gulf of Finland. Although higher than usual in the Baltic Sea, there is no radiological risk from these elevated tritium concentrations.

## Conclusions

<sup>137</sup>Cs concentrations are the main indicator of the radioactive status of the waters of the Baltic Sea. The highest concentrations in the report period were found in the Baltic Proper and the Bothnian Sea. The general trend is steadily decreasing. It is estimated that the pre-Chernobyl target value of 15 Bq/m<sup>3</sup> will be reached between 2017 and 2023, which is earlier than previous estimations. Estimates of effective half-lives for different parts of the Baltic Sea have been updated as between 9 and 11 years. An updated calculation of the inventory of <sup>137</sup>Cs in the seawater of the Baltic Sea resulted in 730 TBq in 2010.

In regard of <sup>137</sup>Cs, the Baltic Sea is still one of the most contaminated areas of the World Ocean, even 24 years after the Chernobyl accident.

## 3.2 Radionuclides of the Baltic Sea sediments

Authors: Kaisa-Leena Hutri, Tarja K. Ikäheimonen and Vesa-Pekka Vartti, STUK, Radiation and Nuclear Safety Authority, Finland

### Introduction

Clay sediments of the Baltic Sea contain valuable information of the radioactivity in the marine environment. Radionuclides, especially <sup>137</sup>Cs, have a tendency to bind into clay and gyttja particles while they settle down. Due to the slow exchange of water between the Baltic Sea and the North Sea, and quite rapid sedimentation rates, the radionuclides have prolonged residence times in the Baltic Sea (Ikäheimonen et al. 2009).

Monitoring radioactivity in the Baltic Sea has been on-going for several decades. With regard to HELCOM, data on the radioactivity of the sea bottom sediments have been gathered since 1984 from some two hundred monitoring stations in order *to compile a baseline* for the existing and changing radioactivity in the Baltic Sea. These results have been reported earlier in HELCOM 1995, 2003 and 2009. The radioactivity data have also been used for several studies, e.g. as a tracer in intercomparison of sediment sampling devices (HELCOM 2000) and in the estimation of sediment accumulation rates and the dating of the sediments (Jensen et al. 2003; Mattila et al. 2006).

This report is a short summary of the radioactivity in the Baltic Sea sediments during the years 1999 – 2010, concentrating on the development of the last four years of the reporting period.

### Material and methods

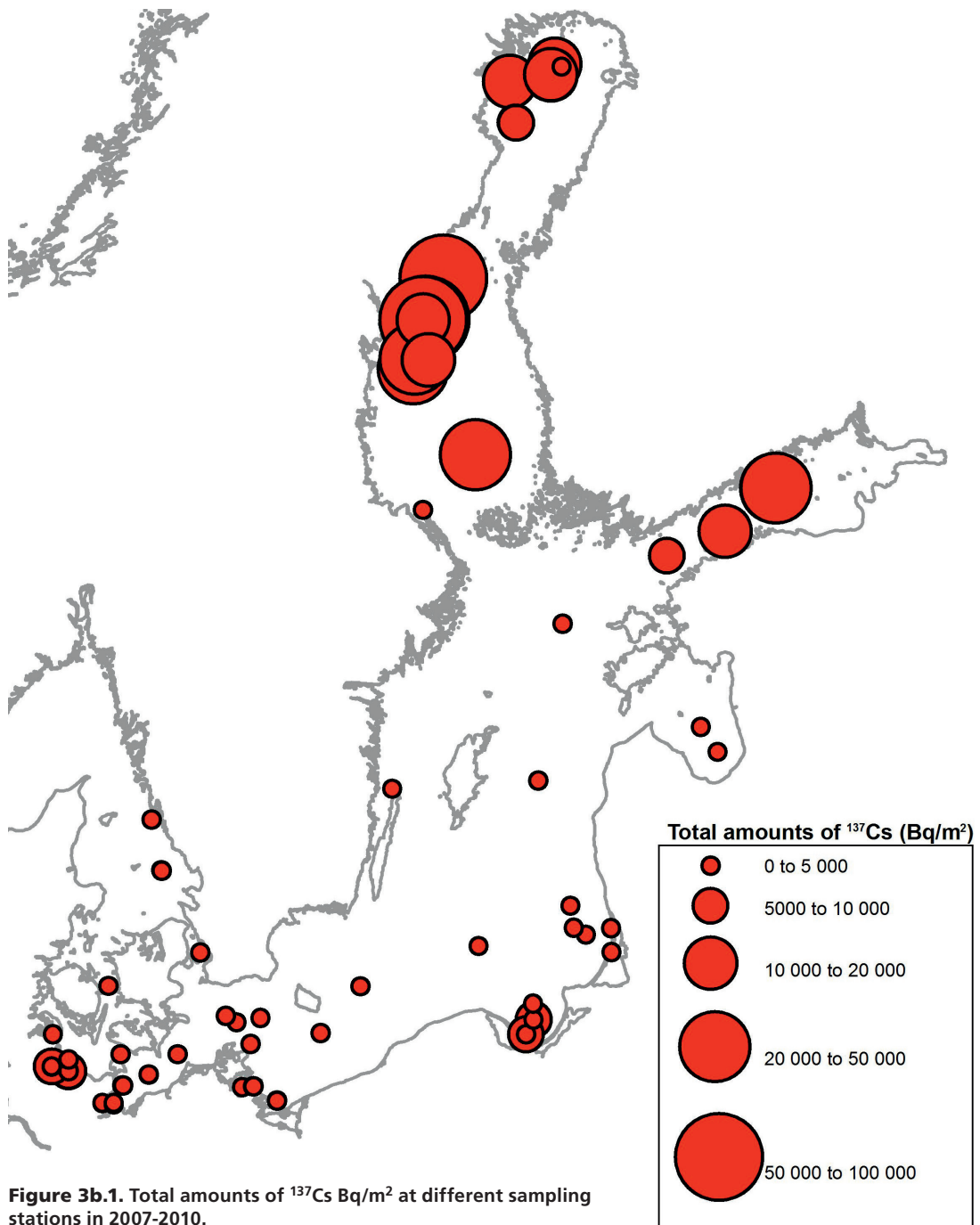
Most of the data are based on the HELCOM database, in which the Contracting Parties have yearly reported recent data. Additional data of STUK's database have been used to assess strontium and plutonium concentrations. During 2007-2010, sampling to measure radioactivity was conducted from 55 monitoring stations. The sampling techniques used by different countries have been described earlier by HELCOM (2003). Sediment types and the bottom morphologies in different parts of the Baltic Sea have also been described in several

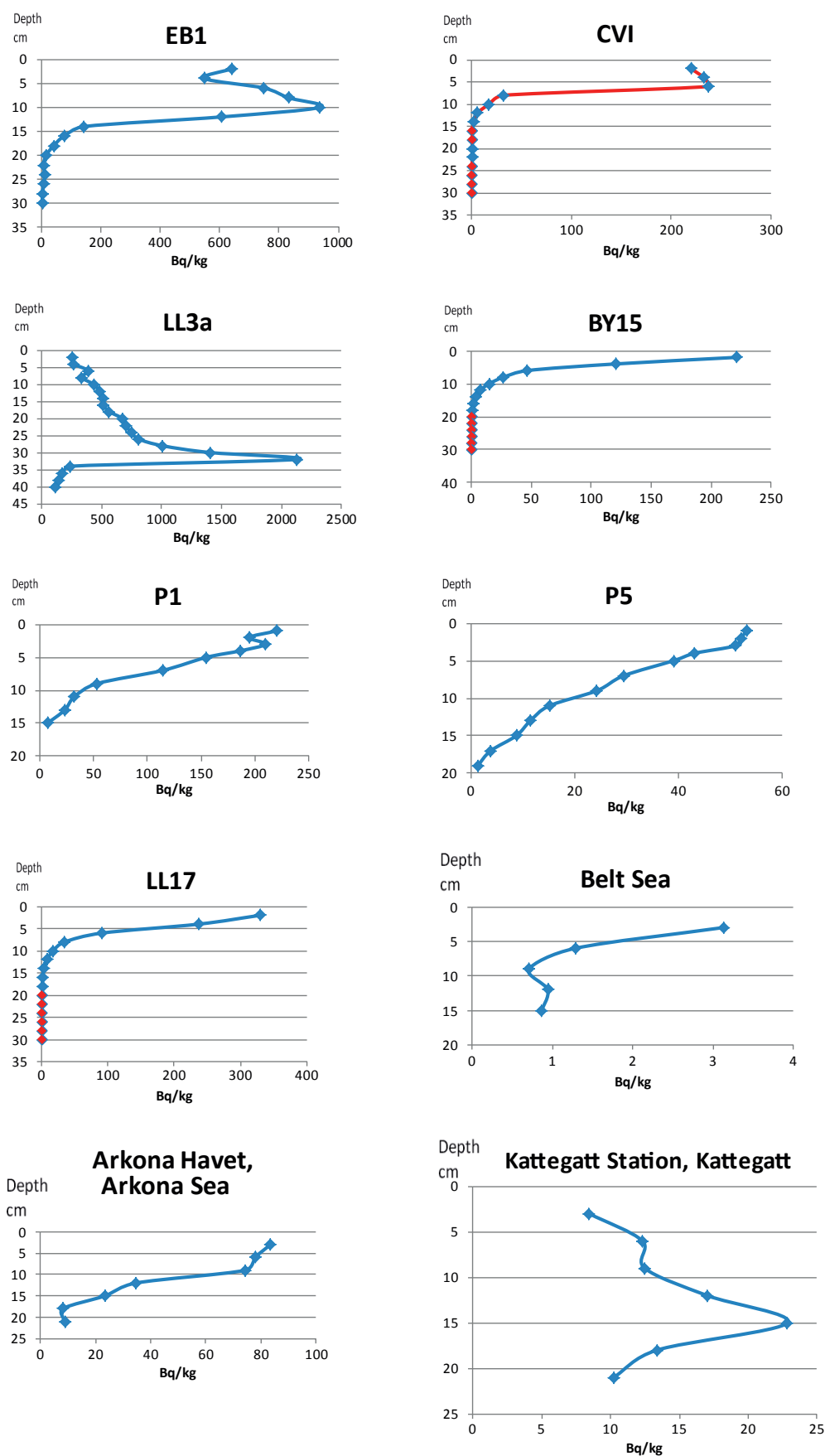
studies (e.g. Winterhalter et al. 1981; Winterhalter 1992, HELCOM 2003; Virtasalo 2006; Hutri 2007).

The methods used in the radionuclide inventories are described in many contexts (Salo et al. 1986, HELCOM 2003; Ilus et al. 2007). In general, the inventories in sediments were based on the mean total amounts of radionuclide activity concentrations in sediments ( $\text{Bq m}^{-2}$ ) and the surface areas of different basins, taking into account the surface areas and the activities in soft and hard bottoms (Ikäheimonen et al. 2009).

## Results and discussion

The amounts of artificial radioactivity have not increased noticeably in the bottom sediments during the years 1999 – 2010, but there is spatial variation in the results. Most of the long lived artificial radioactivity is due to  $^{137}\text{Cs}$  and is mostly found in the bottom sediments in the Bothnian Sea and in the eastern part of the Gulf of Finland (**Figure 3b.1**). Artificial radioactivity from the Chernobyl and global fallouts will partly be buried in sediments in the accumulation bottoms that usually take place in sea basins and depressions.





**Figure 3b.2.** Concentrations of  $^{137}\text{Cs}$  (Bq/kg) as a function of depth at some monitoring stations in the Baltic Sea in 2010. The measured values under the detection limits are marked in red.

The amounts of  $^{137}\text{Cs}$  have been relatively even in different monitoring stations of the Baltic Sea, although showing spatial variation during the reporting period (**Figure 3b.1**). However, due to local or areal accumulation, transportation and erosion rates at some monitoring stations (especially in the Southern Baltic Proper, the Western Baltic and the Gulf of Finland) show a more fluctuant trend of radioactivity concentration of  $^{137}\text{Cs}$ . The sedimentation rate is relatively high in the Baltic Sea and varies widely (between 0.2 and 29 mm/year (HELCOM 2000)) depending on the area and local environmental factors, which is clearly seen by the vertical profiles in **Figure 3b.2**. In addition to the geological conditions, the differences in sampling techniques increase the variability of the results.

Most of the radioactivity in the sediments in the Baltic Sea originates from naturally occurring radionuclides with long half-lives such as  $^{40}\text{K}$ ,  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ . The half-lives and some descriptive statistics of the concentrations of these nuclides in the surface sediments are given in **Table 3b.1**.

( $T_{1/2}$  372.6 h);  $^{110\text{m}}\text{Ag}$  ( $T_{1/2}$  249.8 d); and  $^{125}\text{Sb}$  ( $T_{1/2}$  2.8 a); however, because of their short half-lives, the activities of these radionuclides have decreased considerably or even mostly vanished.

In the recent inventory, we estimated that the total amount of  $^{137}\text{Cs}$  activity in the Baltic Sea sediments was about 2,150-2,480 TBq (Table 2). This amount is about 8 - 9 times higher in comparison to the amounts of pre-Chernobyl level at the beginning of the 1980s (**Table 3b.2**). In recent years, caesium has continued to deposit onto bottom sediments and, at same time, the physical half-life (30.2 a) reduces the activities slowly. Most of the  $^{137}\text{Cs}$  activity is in the bottoms of the Bothnian Sea and in the eastern Gulf of Finland (**Figures 3b.1-3b.2**). The total amounts of  $^{137}\text{Cs}$  activities on so-called hard bottoms varied from 0.3% to nearly 14% of those on soft bottoms, when the average ratio was only about 4% (Ilus et al. 2007).

The reported values during 1999-2010 of  $^{239,240}\text{Pu}$  ( $^{239}\text{Pu}$   $T_{1/2}$   $2.4 \times 10^4$  a,  $^{240}\text{Pu}$   $T_{1/2}$  6,563 a) activities have ranged between and 14.10 Bq  $\text{kg}^{-1}$  dry

**Table 3b.1. Descriptive statistics of the concentrations (Bq  $\text{kg}^{-1}$  dry weight.) of some naturally occurring radionuclides in the surface sediments (0-10 cm) of the Baltic Sea sediments.**

Nuclide	Half-life	Mean	Median	Std	Q1-Q3	N
K-40	$1.3 \times 10^9$ a	777	764	261	624-939	2557
Ra-226	1600 a	39	39	154	28-48	967
Th-232	$1.4 \times 10^{10}$ a	28	28	7	20-32	104

**Table 3b.2. Results of inventories of the total activities of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  in the sediments of the Baltic Sea (Ilus et al. 2007 (before 2007)).**

Years	$^{90}\text{Sr}$ (TBq)	$^{137}\text{Cs}$ (TBq)	$^{239,240}\text{Pu}$ (TBq)	Reference
beginning of the 1980s	12	277	15	Salo et al. 1986
1990 - 1991	-	1,200 - 1,400	18**	HELCOM 1995
1998	-	1,940 - 2,210	-	HELCOM 2003
2000 - 2005	26*	2,100 - 2,400	15.3	HELCOM 2007
2007 - 2010		2,150- 2,480		

\*=rough estimate, \*\* years 1987 - 1988

In the Baltic Sea sediments, there are still considerable amounts of artificial radioactivity due to radionuclides with longer half-lives. Nonetheless, the artificial radioactivity is not expected to cause harmful effects to the Baltic Sea wildlife. After the Chernobyl fallout, there were also elevated concentrations of many short living radionuclides such as  $^{60}\text{Co}$  ( $T_{1/2}$  5.3 a);  $^{103}\text{Ru}$  ( $T_{1/2}$  39.3 d);  $^{106}\text{Ru}$

weight, and values of  $^{238}\text{Pu}$  ( $T_{1/2}$  87.7 a) activities between 0.005 and 0.920 Bq  $\text{kg}^{-1}$ . Most of the plutonium originates from the global fallout; however, there were small amounts of  $^{238}\text{Pu}$  and  $^{241}\text{Pu}$  in the fallout of the Chernobyl accident that could be seen in the activity ratios of  $^{238}\text{Pu}/^{239,240}\text{Pu}$  and  $^{241}\text{Pu}/^{239,240}\text{Pu}$  and in the excess amounts of  $^{241}\text{Pu}$  (Ikäheimonen 2003).  $^{241}\text{Pu}$  increases the amount of

$^{241}\text{Am}$  via radioactive decay. In the surveillance of the marine sediments of the Finnish nuclear power plants, the measured values of  $^{239,240}\text{Pu}$  activities ranged between 0.51 and 3.9 Bq/kg dry weight (STUK database). Due to the limited data, the present total amounts of these activities are difficult to estimate.

The activities of the artificial radionuclides  $^{90}\text{Sr}$  ( $T_{1/2}$  28.5 a) and  $^{241}\text{Am}$  ( $T_{1/2}$  432.7 a) mostly originate from the global fallout. Because of the small share in the Chernobyl fallout and costly analytical methods, the interest on  $^{90}\text{Sr}$  has been reduced and thus only relatively few data are available. In the years 1999 – 2010, the reported  $^{241}\text{Am}$  concentrations ranged from 0.03 to 4.8 Bq kg $^{-1}$  dry weight, and those of  $^{90}\text{Sr}$  from 0.1 to 73.8 Bq kg $^{-1}$  dry weight. In the surveillance of the marine sediments of Finnish nuclear power plants, the measured values of  $^{90}\text{Sr}$  ranged between 0.63 and 8.5 Bq/kg dry weight. (STUK database). Due the limited data, the present total amounts of these activities are difficult to estimate.

### Future work and recommendations

Monitoring radioactivity in the Baltic Sea is an essential part of environmental monitoring and will provide a realistic baseline, e.g. for possible radiation emergency situation modelling purposes. The continuous monitoring work and time trends of the radioactive substances are the bases for understanding the state of the Baltic Sea environment and its radioactivity. However, there are still several gaps in our knowledge of radioactivity in the sediments of the Baltic Sea, e.g. the amounts of  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  as well as natural radioactivity such as  $^{210}\text{Pb}$ . Moreover, an environmental distribution model for any kind of nuclear accident in the Baltic Sea area is needed. In the future, we should attempt to fill these gaps to form a more comprehensive picture of the radioactivity in the sediments of the Baltic Sea. Relevant work in the future should include assessments of the impacts of ionising radiation on the environment of the Baltic Sea.

## 3.3 Radionuclides in biota

Author: Günter Kanisch, TI-FOE, Thünen Institute for Fisheries Ecology, Germany

### Introduction

Levels of radionuclide activity concentrations in marine biota are linked to the corresponding levels in seawater and sediment via accumulation through food chains. The complexity of food chains increases with the number of trophic levels of the species considered. Fish - which represent the Baltic Sea's most important biota group for human seafood consumption - accumulate most of the radionuclides from food, not from water.

Baltic Sea biota received the most important contribution to their levels of radionuclide activity concentrations from the Chernobyl accident in 1986, predominantly from  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . The ratio of activity concentrations  $^{134}\text{Cs}/^{137}\text{Cs}$  determined in biota agreed very well with that of the Chernobyl fallout. The high trophic level species - predators such as cod and pike - showed the highest  $^{137}\text{Cs}$  levels; however, compared to seawater with some delay in reaching the maximum values after 1986. In the long-term, however,  $^{137}\text{Cs}$  time trends in biota will follow the trends in seawater.

Monitoring radionuclides in Baltic Sea biota started in 1984, before the Chernobyl accident. Average activity concentrations in biota from this pre-Chernobyl time have been used later to define target values, which are expected to be reached again post Chernobyl.

In this report, the evaluation of biota data is restricted to fish.

### Material and methods

The number of biota samples collected annually by the MORS-PRO group decreased slightly from about 85 at the beginning of the period 1999 – 2010 to about 70 in 2008, and increased to about 113 in 2009/2010 at the end of this period. The sample numbers over this period were 985, 183 and 140 for fish, *Fucus vesiculosus* and benthic animals, respectively. The main artificial radionuclides that were measured in biota in the assessment period were  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ .

## Results and discussion

### Time trends of activity concentrations

Time trends of activity concentrations had been evaluated in the previous thematic assessment (HELCOM, 2009) for the period up to 2006, where the radionuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in different fish species had been considered. The results from an update of these evaluations to 2010 are discussed below.

$^{137}\text{Cs}$  is continuing to show slowly decreasing trends, which is demonstrated by **Figure 3c.1** showing such trends for herring muscle (*Clupea harengus*). In the western parts of the Baltic Sea, i.e. the Kattegat, the Sound, and the Belt and Arkona Seas, the values already show levels that are slightly below the target value of  $2.5 \text{ Bq kg}^{-1}$  wet weight. In the remaining Baltic Sea basins, the target value is still exceeded in the Bothnian Bay and in the Gotland area by a factor of up to 5.

**Figure 3c.2** shows measured  $^{137}\text{Cs}$  activity concentrations in cod muscle (*Gadus morhua*). At the end of this assessment period, between 4 and  $10 \text{ Bq kg}^{-1}$  wet weight are observed for the means of the measured values.

For the flat fish group consisting of flounder (*Platichthys flesus*) and plaice (*Pleuronectes platessa*), **Figure 3c.3** shows the  $^{137}\text{Cs}$  time series in the western and southern Baltic Sea areas. At the end of the assessment period (2010), the values were below  $5 \text{ Bq kg}^{-1}$  wet weight.

In the coastal areas of Baltic Sea basins around Finland, freshwater fish, i.e. pike (*Esox lucius*), has also been used for monitoring measurements since 1985. Due to larger freshwater fractions in this seawater, their concentration factors are higher resulting in slightly larger  $^{137}\text{Cs}$  values. The trends of  $^{137}\text{Cs}$  in pike are clearly decreasing. By the end of the assessment period, the annual means approached values of around  $15 \text{ Bq kg}^{-1}$  wet weight.

For the radionuclide  $^{90}\text{Sr}$ , mainly originating from nuclear weapons fallout and related runoff from rivers, the values of the specific activity in fish flesh are more than two orders of magnitude lower than those of  $^{137}\text{Cs}$ . The  $^{90}\text{Sr}$  activities in the flesh of flat fish species (flounder, plaice) exhibit slightly larger

values than in round fish species (herring, cod, whiting and mackerel). This is because the analysed samples of the former may contain more amounts of small bones, which accumulate the bone-seeking  $^{90}\text{Sr}$  much more than fish muscle.

The highest measured activity concentration values of  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  are one to two orders of magnitude lower than those of  $^{90}\text{Sr}$ ; consequently, they do not have any significance with respect to the dose accumulated by fish consumption.

The radionuclide which is the most important with respect to dose by fish consumption is the alpha-emitting  $^{210}\text{Po}$  that belongs to naturally-occurring radionuclides. The activity values of 117 samples of fish flesh that have been collected in the Kattegat and the Bornholm Sea since 1990 are summarised in **Table 3c.1**. The average of the three median values is  $0.68 \text{ Bq kg}^{-1}$  wet weight. In a former evaluation of  $^{210}\text{Po}$  in fish from Danish waters including the North Sea (Dahlgard, 1996) average values of 0.35, 0.65 and  $0.96 \text{ Bq kg}^{-1}$  wet weight were observed for cod, herring and plaice fillets, respectively. Mean values of additional few Baltic Sea fish data from Finland (2010) were  $0.4 \text{ Bq kg}^{-1}$  wet weight for pike (flesh;  $N=4$ ) but larger for herring (edible parts;  $N=3$ ) with  $3.4 \text{ Bq kg}^{-1}$  wet weight, respectively, with 20% standard deviation, while the corresponding values of  $^{210}\text{Pb}$  were 0.7 and  $0.07 \text{ Bq kg}^{-1}$ , i.e. about 1/5 of  $^{210}\text{Po}$ .

**Table 3c.1: Activity values of the naturally occurring alpha emitting radionuclide  $^{210}\text{Po}$  in Baltic Sea fish (flesh), in  $\text{Bq kg}^{-1}$  wet weight (1990-2010).**

Species	number	$^{210}\text{Po}$			
		min.	max.	mean	median
Herring	38	0.19	8.5	1.3	0.79
Cod	39	0.043	1.5	0.39	0.30
Flounder	40	0.26	3.4	1.1	0.95

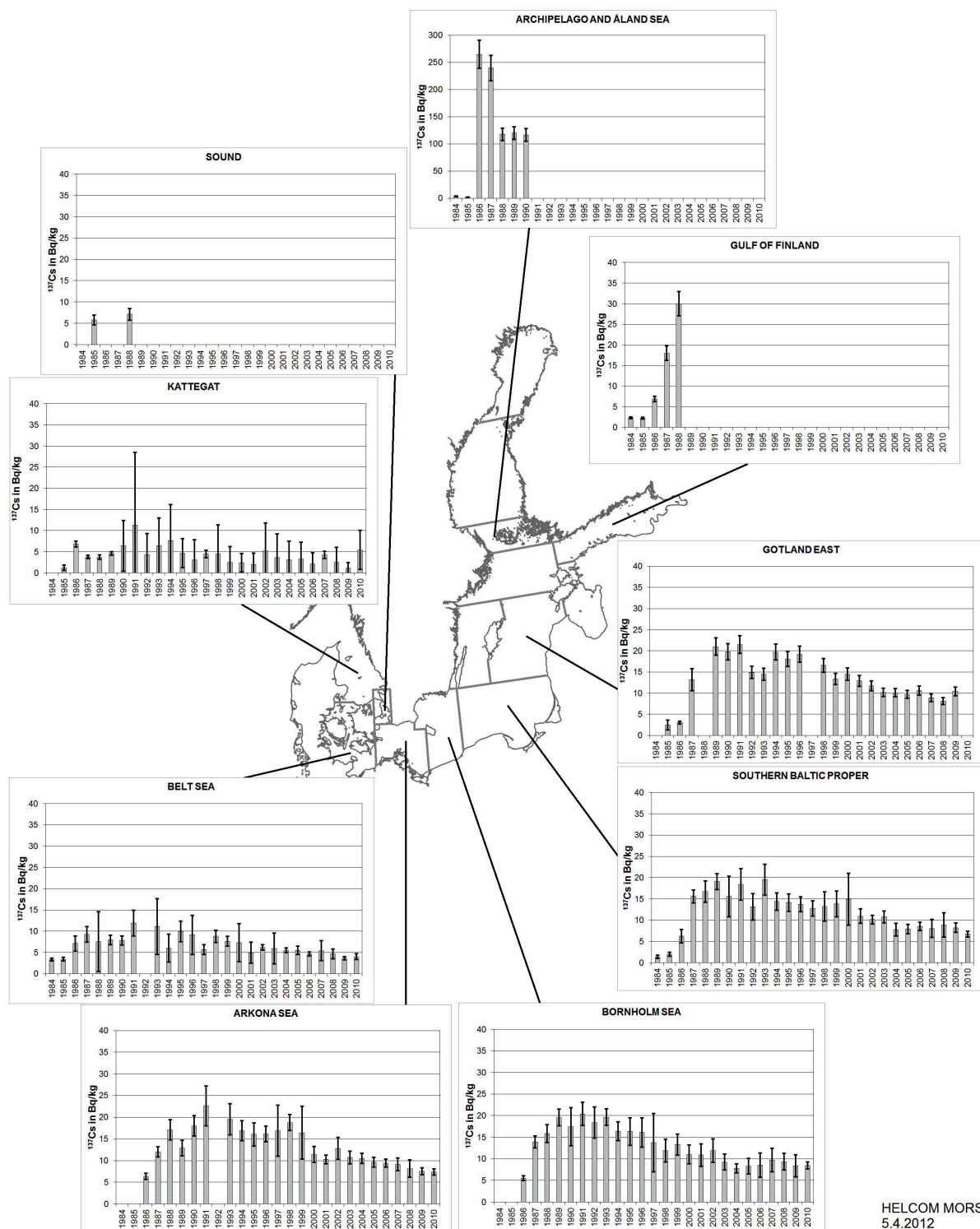
**Figure 3c.4** gives  $^{137}\text{Cs}$  the time series of three fish species from the Polish economic zone (data from Poland), which allowed the effective half-life values of  $^{137}\text{Cs}$  in fish to be estimated at around 14.5 years. This value is comparable with an estimate of about 11 years obtained for  $^{137}\text{Cs}$  in seawater of the Baltic Proper (c.f. chapter 'Seawater' of this report).





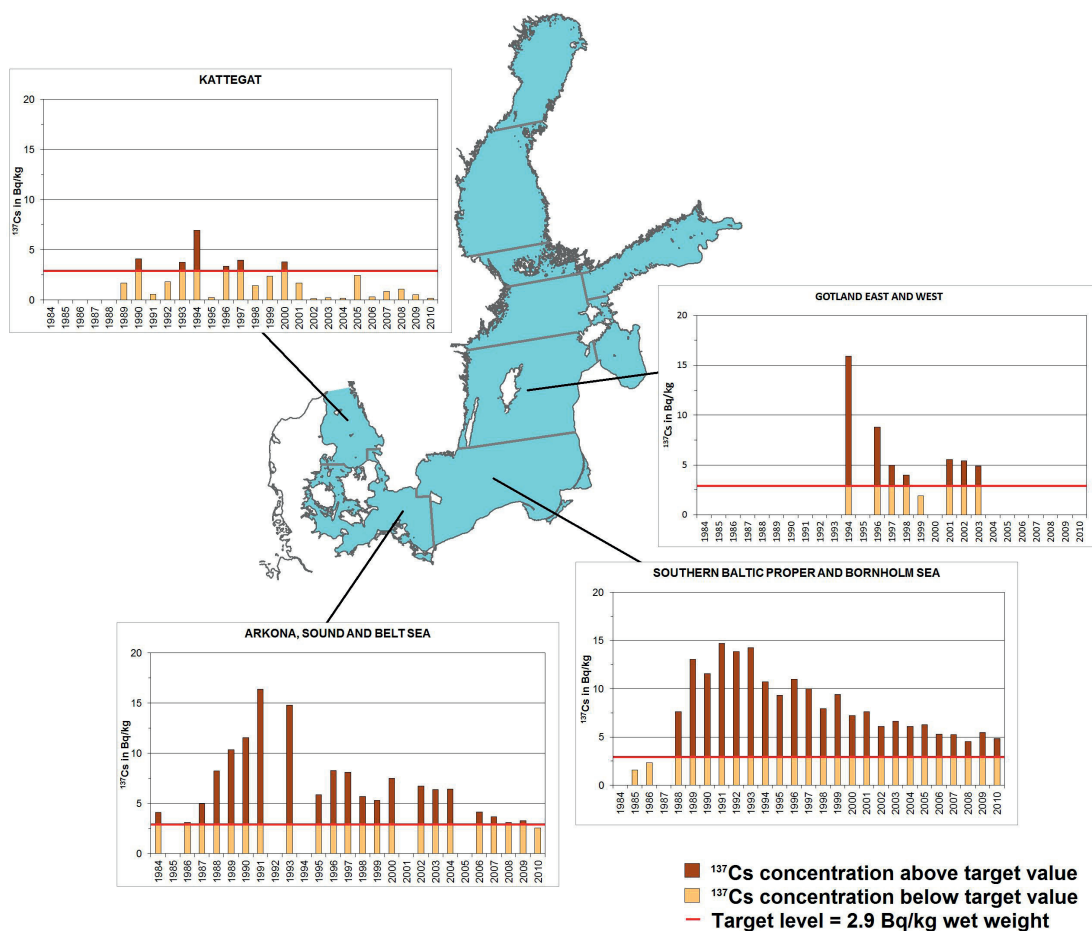
**Figure 3c.1:** Annual average  $^{137}\text{Cs}$  concentrations (Bq/kg wet weight) in herring muscle (fillets) in 1984-2010. Uncertainty has been indicated as bar lines. The target value has been calculated as average of the pre-Chernobyl (1984-1985) activity concentrations.



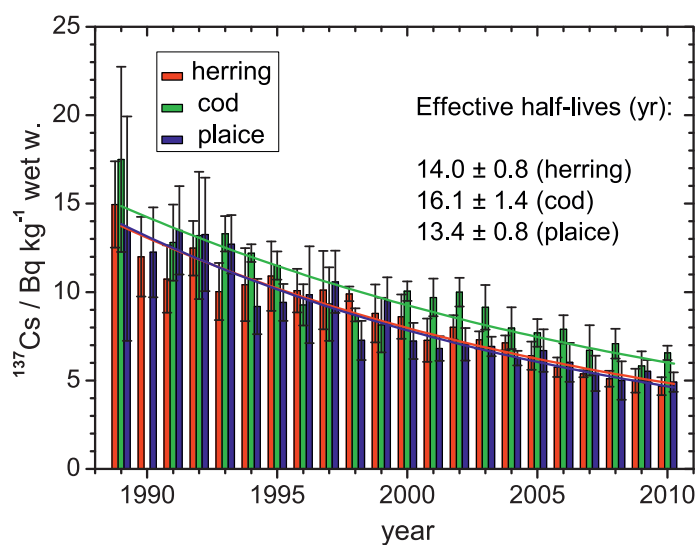


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**Figure 3c.2.** Annual average  $^{137}\text{Cs}$  concentrations (Bq/kg wet weight) in cod muscle (fillets) in 1984-2010. Uncertainty has been indicated as bar lines. NOTE: Variable scale in graphs.



**Figure 3c.3.** Annual average  $^{137}\text{Cs}$  concentrations (Bq/kg wet weight) in plaice and flounder muscle (fillets) in 1984-2010. Uncertainty has been indicated as bar lines.



**Figure 3c.4.** Annual average  $^{137}\text{Cs}$  concentrations ( $\text{Bq kg}^{-1}$  wet weight) in samples of herring, cod and plaice muscle (fillets) collected by Poland in 1989-2010, with 1s uncertainty bars. Effective  $^{137}\text{Cs}$  half-lives (1s uncertainties) were obtained from least-squares fitting exponential decay curves (shown).

## Conclusions

Within the current assessment period, the Chernobyl-derived  $^{137}\text{Cs}$  continued to be the most dominant man-made radionuclide in Baltic Sea fish regarding activity concentrations. By the end of this period (2010), mean values in various Baltic Sea basins were:

- a) between  $0.7 \text{ Bq kg}^{-1}$  wet weight and about  $7 \text{ Bq kg}^{-1}$  wet weight were found in the group of marine round fish (cod, herring, whiting);
- b) between  $1.5$  and  $5 \text{ Bq kg}^{-1}$  wet weight in marine flat fish (plaice, flounder, dab), i.e. slightly lower mean values than in (marine) round fish.
- c) up to  $15 \text{ Bq kg}^{-1}$  wet weight in freshwater fish (pike), in Finnish coastal areas.

# 4 Modelling and Dose Calculations

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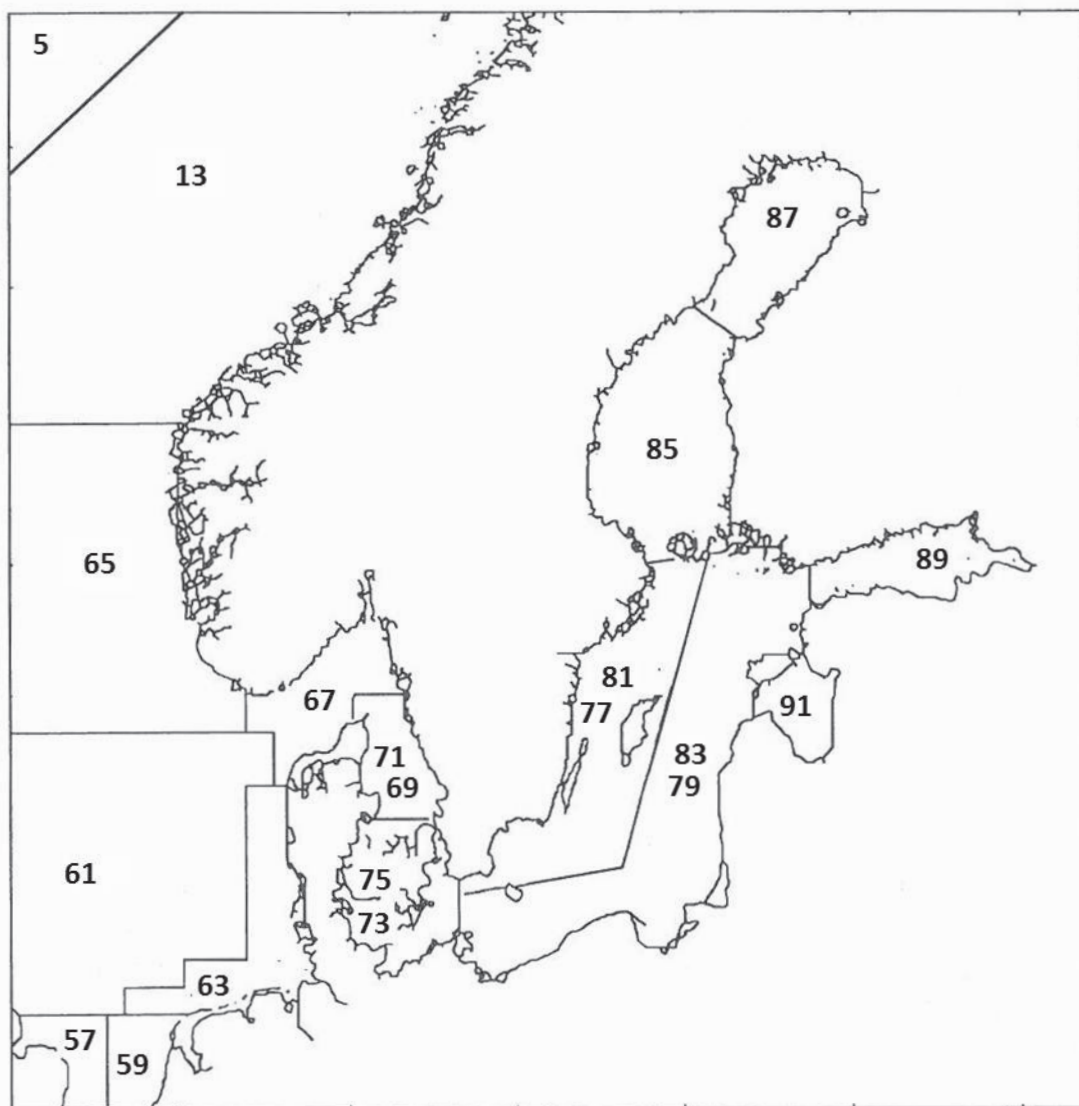
progress of implementing the model in Windows compatible software. The model uses first-order kinetics to simulate the transfer of radioactivity between compartments comprising water regions and underlying sediments.

## 4.1 Model work

Environmental modelling of radioactive substances in the Baltic Sea is used by the MORS Group to support and interpret environmental data and as a tool for radiological assessments.

The Group has used a model (HELCOM, 2003) implemented in software which does not run on contemporary computers. Therefore, work is in

The model is developed for assessing radiological consequences of releases of radioactive material to the marine environment covering European coastal waters including the Baltic Sea. The model simulates the dispersion of radioactive substances in the water due to advective transport, including mixing from wind and tidal forces. Association of radionuclides to suspended sediment material is taken into consideration in addition to subsequent transfer to sediments through particle scavenging.



**Figure 4.1.** Map of the Baltic Sea showing water sub-regions used by the model. Sub-region numbers 69, 73, 77 and 79 refer to bottom waters in sub-regions with stratification. The remaining numbers refer to surface waters and sub-regions without stratification.

From specified inputs of radioactivity to the marine environment, the model calculates time-dependent concentrations in seawater and sediments. These data may be used to calculate doses to man from a range of exposure pathways. The model structure with respect to water boxes is shown in **Figure 4.1**.

A separate implementation of the model (Kanisich et al., 2000) has made it possible to simulate levels of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in Baltic seawater from the dominating sources of input, i.e. atmospheric fallout from nuclear weapons testing; atmospheric fallout from and the Chernobyl accident; and discharges to sea from the European reprocessing facilities Sellafield and La Hague ( $^{99}\text{Tc}$  and  $^{129}\text{I}$ ) show increasing trends in Baltic seawater; however, in terms of radiation dose to man these are insignificant.

## 4.2 Dose calculations

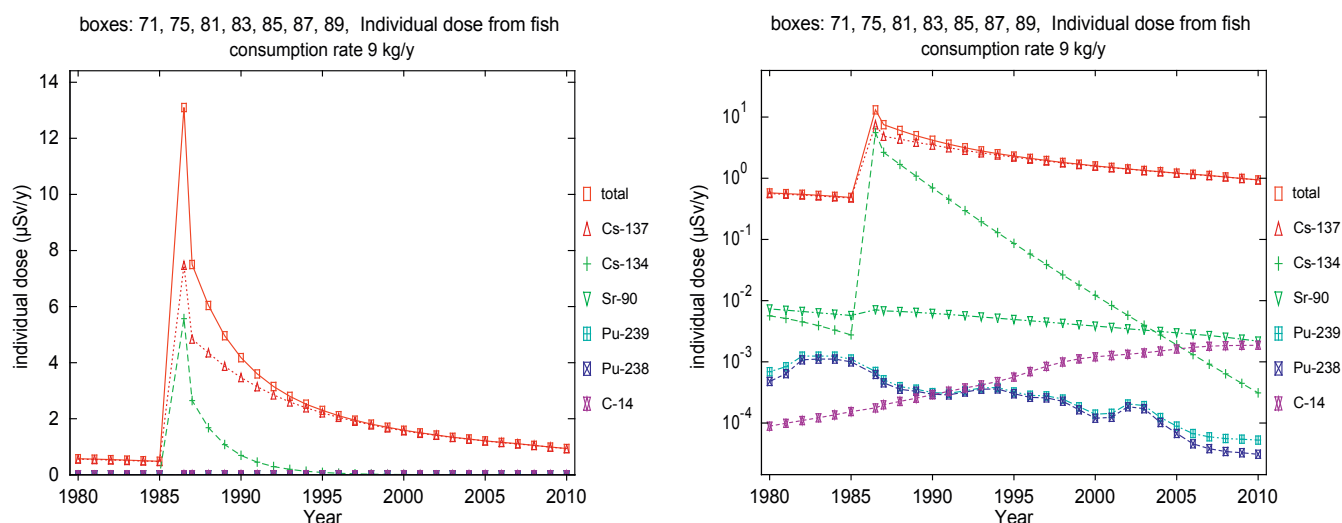
Estimates of radiation doses until the year 2000 to human individuals and populations from radioactivity in the Baltic Sea were made by the MORS Group in HELCOM (2003). The estimates were based on model calculations and included a range of exposure pathways, including ingestion of fish, crustaceans and molluscs, inhalation and external exposure. Doses to individuals were based on human habits assumed to be characteristic for a critical group expected to receive the largest radiation dose.

The dominating exposure was found to be due to  $^{137}\text{Cs}$  and the ingestion of fish.

The concentrations of the dominating man-made radionuclides in the Baltic Sea,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , have been declining since 2000. Only minor amounts of man-made radionuclides discharged from Sellafield and La Hague ( $^{99}\text{Tc}$  and  $^{129}\text{I}$ ) show increasing trends in Baltic seawater; however, in terms of radiation dose to man these are insignificant.

For the reporting period covered by the present report, we may estimate an upper bound for individual doses from man-made radionuclides in the Baltic Sea. During 2007-2010, the concentrations of  $^{137}\text{Cs}$  in marine species of fish from the Baltic Sea have been below 12 Bq/kg. For an individual with a high annual consumption of fish (90 kg), this concentration corresponds to an annual radiation dose of 14  $\mu\text{Sv}$ . This dose is well below the limit of the annual radiation dose to a member of the public of 1,000  $\mu\text{Sv}$  (EC, 1996). The corresponding annual dose from naturally occurring radionuclides in fish is about 100  $\mu\text{Sv}$ , of which the dominating contribution is from  $^{210}\text{Po}$ . The annual dose contribution from tritium ( $^3\text{H}$ ) is insignificant by comparison, less than 0.01  $\mu\text{Sv}$  from natural and man-made sources combined.

Results for the period up to 1980 were shown in HELCOM (2009), including a model comparison of the  $^{137}\text{Cs}$  seawater activity concentrations up



**Figure 4.2.** Calculated annual radiation dose to an individual in the Baltic Sea region consuming 9 kg  $\text{y}^{-1}$  of fish flesh; linear scale left and logarithmic scale right. The six radionuclides shown are those with the largest contributions to the dose; the sum of the radionuclide contributions is shown as 'total'.

to 2005. Calculated annual radiation doses during 1980-2010 to individuals consuming nine kg of fish flesh per year are given in **Figure 4.2**. The calculations show that the annual individual dose from man-made radionuclides peaked in 1986 at 14  $\mu\text{Sv}$  and decreased to about 1  $\mu\text{Sv}$  in 2010. The corresponding dose from naturally occurring radioactivity in fish flesh is about 10  $\mu\text{Sv}$ .

Results from an assessment of the radiological risk to marine biota from radioactive substances in the Baltic Sea are presented in Chapter 4.3.

## 4.3 Assessment of doses to biota

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### Introduction

In recent years, much effort has been directed internationally to the development of a scientific basis for the protection of biota against ionising radiation and the related risk assessment. International bodies essential in the development of radiological criteria or standards for the protection of non-human species have carried out much work in this field (ICRP, 2003, 2007, 2008, 2009; Pröhl et al, 2011; UNSCEAR, 2011). A forthcoming new edition of the EU Basic Safety Standards contains new requirements and recommendations for safety and the protection of non-human species or the total environment (Draft European Basic Safety Standards Directive, 2010).

The potential harmful effects of radiation to biota include increased mortality and the reduced reproductive success of a population. These effects are addressed in environmental risk assessment methods, whereas factors such as variation in radiosensitivity between individuals, interactions between species and adaptation of populations to radiation exposure are omitted due to inadequate scientific knowledge. Furthermore, the possible effects of other contaminants on doses are still excluded in current risk assessment methods.

A variety of tools have been developed to enable the assessment of doses and risks caused by ionising radiation to biota. We have used the ERICA Assessment Tool (Brown et al., 2008) in this study. Intercomparisons of different tools have revealed a wide variability of the results, mainly generated by using default transfer coefficients provided by the tools instead of using site-specific data on activity concentrations (e.g. Beresford et al., 2008; Vives I Batlle et al., 2007, 2011; Yankovich et al., 2010). Therefore, we have used measured activity concentrations in biota and media to obtain more accurate estimations of dose rates to biota.

### Material and methods

The description of data on average activity concentrations of  $^{137}\text{Cs}$ ,  $^{40}\text{K}$ ,  $^{90}\text{Sr}$ ,  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ ,  $^{239,240}\text{Pu}$  in surface water, sediment and biota currently in the Gulf of Finland and in the Bothnian Sea have been given in Outola et al. (2011). The species considered included seaweed (*Fucus vesiculosus*), fish (*Esox lucius*) and bottom living crustacean (*Saduria entomon*). The use of the ERICA Assessment Tool to calculate dose rates to biota has also been described in Outola et al. (2011).

Currently, there are no internationally agreed, legally binding criteria or standards for dose rates to biota that must be met in accordance with regulation. Several dose rate values have been suggested to be used as a screening value, which means that if this value is exceeded further investigation is needed to better understand and quantify the risk. In this assessment, the screening value 10  $\mu\text{Gy/h}$  was used. The derivation of this value is based on the examination of the available data on dose-effect relationships for various organisms obtained in laboratory or field experiments, and has been presented in Andersson et al. (2009) and Garnier-Laplace et al. (2008). The total (internal and external summed) dose rates estimated are compared directly to the selected screening dose rate to enable the assessment of risk to biota.

### Results and discussion

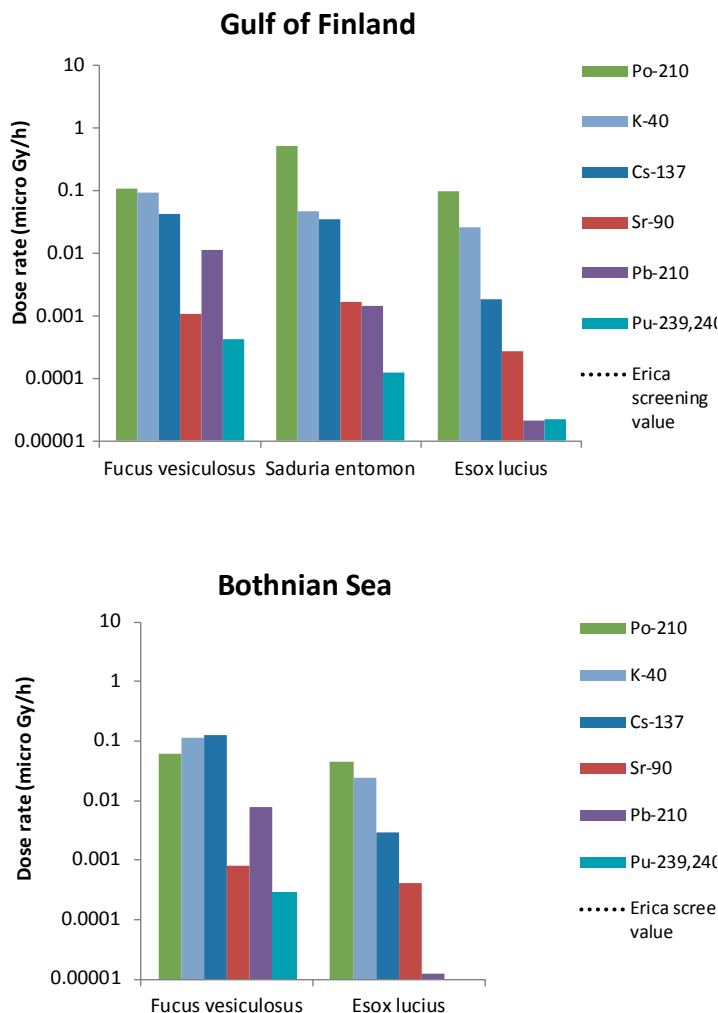
The total dose rate from all studied artificial radionuclides combined ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$ ) is currently 0.002 – 0.1  $\mu\text{Gy/h}$  and is dominated by  $^{137}\text{Cs}$  (**Figure 4.3**). The lowest dose rates resulted to

fish and the highest to sediment-associated organisms (Outola et al., 2011).

The doses to the Baltic Sea biota were mainly dominated by the natural radionuclide  $^{210}\text{Po}$ . The contribution of  $^{137}\text{Cs}$  to the dose was generally less than one tenth of the proportion of  $^{210}\text{Po}$ . The dose rates from  $^{40}\text{K}$  were in the same order as those from  $^{137}\text{Cs}$ , except for fish where the dose rates from  $^{40}\text{K}$  were ten times higher than those from  $^{137}\text{Cs}$ . The contribution from the other radionuclides evaluated ( $^{90}\text{Sr}$ ,  $^{239, 240}\text{Pu}$  and  $^{210}\text{Pb}$ ) to the total dose were of minor importance (Outola et al., 2011).

## Conclusions

The dose rates from the evaluated radionuclides were clearly below the screening level  $10\ \mu\text{Gy/h}$ . Therefore, the biological effects of ionising radiation on biota can be considered negligible in the current radiation situation. According to the present risk assessment methodology, the level of protection of biota against ionising radiation can be considered adequate in the Baltic Sea.



**Figure 4.3.** Average dose rates (microGy/h) from various radionuclides to biota in the Baltic Sea at present.



# 5 Conclusions

The most significant source of man-made radioactivity in the Baltic Sea is fallout from the accident at the Chernobyl nuclear power plant in 1986. The most important radionuclides in the fallout were  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ . The total input of  $^{137}\text{Cs}$  from Chernobyl to the Baltic Sea has been estimated at 4,700 TBq, and the post-Chernobyl river discharges of  $^{137}\text{Cs}$  to the Baltic Sea were estimated at 300 TBq comprising 6-7% of the total fallout.

The second most important source is global fallout from atmospheric nuclear weapons tests carried out during the late 1950s and early 1960s. During the late 1990s, the decay-corrected amounts of weapons-test  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in the Baltic Sea have been evaluated at 800 and 500 TBq, respectively.

The corresponding decay-corrected total inputs to the Baltic Sea of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  originating from nuclear reprocessing plants in Western Europe have been estimated at 250 and 40 TBq, respectively. These sources are now only of minor importance due to significant reductions in discharges in recent years.

The predominant radionuclide in discharges from the nuclear power plants and research reactors in the Baltic Sea region is  $^3\text{H}$ . Total discharges of  $^3\text{H}$  from these local sources have amounted to 3,500 TBq, and those of other beta-gamma emitting radionuclides to about 24 TBq by the end of 2010. The total discharges of alpha emitting radionuclides have been 0.005 TBq.

For  $^{137}\text{Cs}$  in the Baltic Sea, the main source is fallout from Chernobyl (83%), followed by nuclear weapons test fallout (13%). For  $^{90}\text{Sr}$ , the main source of contamination is fallout from nuclear weapons tests (81%), while the proportion from Chernobyl fallout was smaller (13%).

Today,  $^{137}\text{Cs}$  is the main indicator of man-made radioactivity in Baltic seawater. The highest concentrations observed in seawater during the period 2007-2010 were found in the Baltic Proper and the Bothnian Sea. The general trend is steadily decreasing. It is estimated that the target value of 15 Bq/m<sup>3</sup>, corresponding to pre-Chernobyl levels, will be reached between 2017 and 2023, which is earlier than previous estimations. First estimates of effective half-lives for different parts of the Baltic Sea have been calculated at being between 9-11 years.

The inventory of  $^{137}\text{Cs}$  in the Baltic seawater in 2010 is estimated at 730 TBq.

With regard to  $^{137}\text{Cs}$ , the Baltic Sea is still one of the most contaminated areas of the World Ocean, even 24 years after the Chernobyl accident.

The results of the Sediment Baseline Study carried out by the MORS-PRO during the reporting period showed that the concentrations of naturally occurring radionuclides in Baltic Sea sediments remain at background levels. The concentrations of man-made radionuclides in sediments are higher than the target specified in HELCOM's ecological objective of 'radioactivity at the pre-Chernobyl level'. This is particularly true for the Bothnian Sea and the Gulf of Finland, which received the largest amounts of Chernobyl fallout in the Baltic Sea. The total inventory of  $^{137}\text{Cs}$  in the Baltic Sea sediments was estimated at 2,150–2,480 TBq between 2007-2010.

Monitoring radioactivity in the Baltic Sea is an essential part of environmental monitoring and will provide a realistic baseline of possible radiation emergency situations modelling purposes, for example. The continuous monitoring work and time trends of the radioactive substances are the bases for understanding the state of the Baltic Sea environment and its radioactivity. However, there are still several gaps in our knowledge of radioactivity in the sediments of the Baltic Sea, for example the amounts of  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and natural radioactivities such as  $^{210}\text{Pb}$ . In addition, an environmental distribution model for any kind of nuclear accident in the Baltic Sea area is needed. In the future, we should attempt to fill these gaps to form a more comprehensive picture of the radioactivity in the sediments of the Baltic Sea. Moreover, the relevant works in the future should include more assessments of the impacts of ionising radiation on the environment of the Baltic Sea.

Concentrations of man-made radioactivity in fish show generally decreasing trends - in agreement with trends in concentrations in seawater. Chernobyl-derived  $^{137}\text{Cs}$  continued to be the most dominant man-made radionuclide in Baltic Sea fish. By the end of the reporting period, mean values of 0.7-7 Bq kg<sup>-1</sup> wet weight were found in marine round fish (cod, herring, whiting) in various Baltic Sea basins, while the concentrations in pike were

up to 15 Bq kg<sup>-1</sup> wet weight off the Finnish coast. In marine flat fish (plaice, flounder, dab) slightly lower mean values were found than in marine round fish.

Radiation doses to humans from man-made radionuclides in the Baltic Sea are due mainly to ingestion of <sup>137</sup>Cs in fish. Doses from <sup>3</sup>H are lower by several orders of magnitude. During 2007-2010, doses to members of the public from marine pathways have not exceeded an annual value of 0.02 mSv, which is well below the limit of 1 mSv for the general public set in the Basic Safety Standards of the European Council (EC, 1996) and the IAEA (IAEA, 1996).

A detailed assessment of the impact of the nuclear accident that occurred after the 2011 Great East Japan Earthquake and tsunami on the Baltic Sea

area, which occurred outside of the period considered in this assessment, will be presented in the following thematic assessment. In the meantime, the indicator fact sheets are updated regularly with recent data. Radioactive fallout over the Baltic Sea from the nuclear accident in Japan in March 2011 was very small and may not be detectable in seawater and fish. The corresponding radiological risks are estimated to be negligible.

Concentrations of radioactive substances in the Baltic Sea are not expected to cause harmful effects to wildlife in the foreseeable future. However, in line with international developments, the future work of HELCOM will continue to include assessments of the radiological risks to the environment from radioactive substances in the Baltic Sea. The knowledge about the baseline is one of the key factors for the risk assessment.

## 6 Recommendations

The objective of HELCOM MORS-EG is to implement the Helsinki Convention on matters related to the monitoring and assessment of radioactive substances in the Baltic Sea. This work is based on HELCOM Recommendation 26/3 and will support the Monitoring and Assessment Group (HELCOM MONAS).

MORS-EG's main responsibilities are to:

- coordinate basic monitoring programmes on radioactive substances in the Baltic Sea carried out by the Contracting Parties in accordance with HELCOM Recommendation 26/3 and the valid Guidelines;
- annually compile data on discharges of radioactivity from civil nuclear facilities to the Baltic Sea reported by the Contracting Parties;
- annually compile data on discharges and environmental levels of radioactivity in the Baltic Sea submitted to the HELCOM databases in accordance with HELCOM Recommendation 26/3;
- annually validate all data in the HELCOM MORS databases and to make it available on CD-ROMs to MORS-EG Members;
- annually update HELCOM Indicator Fact Sheets on radioactive substances in the Baltic Sea;
- keep the Guidelines on the Monitoring of Radioactive Substances in the Baltic Sea updated;
- coordinate and organise intercomparison exercises on seawater, sediments and biota to assure the high quality of the monitoring data;
- keep under observation trends in the export of radionuclides from the Baltic Sea to the North Sea and vice versa, especially the inflows of radioactivity (e.g.  $^{99}\text{Tc}$  and  $^{129}\text{I}$ ) from Sellafield to the Baltic Sea, and the outflow of Chernobyl radioactivity from the Baltic Sea to the Skagerrak;
- produce thematic reports on man-made and naturally occurring radionuclides in the Baltic Sea, for example, and simple procedures for assessing doses to man from radioactivity in the Baltic Sea;
- finalise in 2013 the periodic assessment on radioactivity in the Baltic Sea covering data up to 2010; this assessment will include levels, inventories and trends for radioactivity in the Baltic Sea and its radiological impact on man and the environment.

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# 9 Appendix: Data Quality

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Ten laboratories from the eight countries bordering the Baltic Sea have contributed to the monitoring programmes during the years 2007 – 2010. Analytical procedures and quality systems of different laboratories have been presented in the earlier assessment by Ikäheimonen and Outola (2009).

Two main intercalibration exercises have been performed during the reporting period: the proficiency test of radionuclides in fish flesh samples organised by IAEA-EL (Pham, 2010) and the Baltic Sea water intercomparison organised by STUK, Finland.

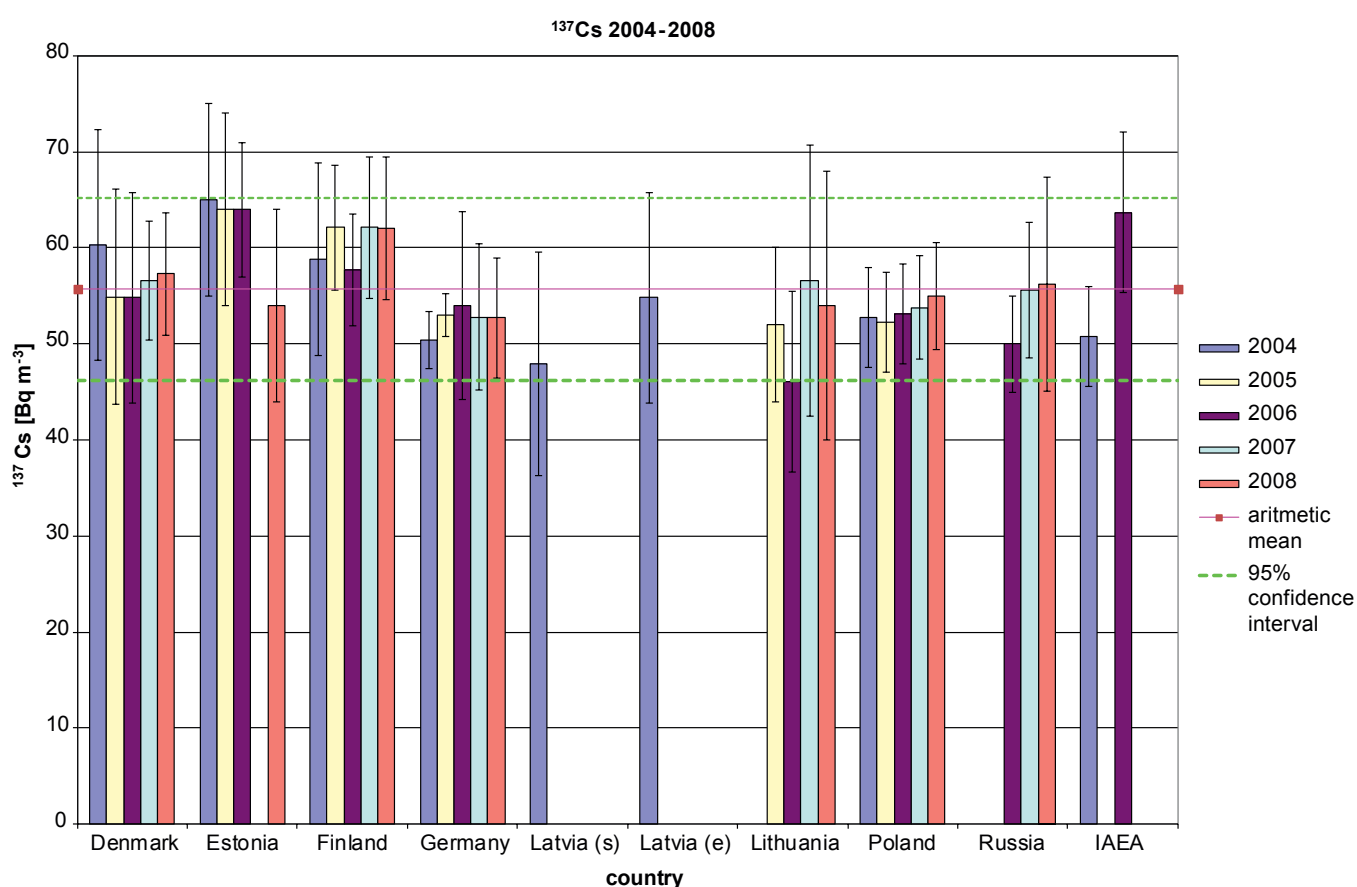
In the proficiency test, 88% of the laboratory means from nine different nuclides were acceptable.

ble. All ten participants reported <sup>137</sup>Cs massic activities, with 90% of the results being acceptable.

This shows a very good performance of the MORS laboratories in the determination of radionuclides.

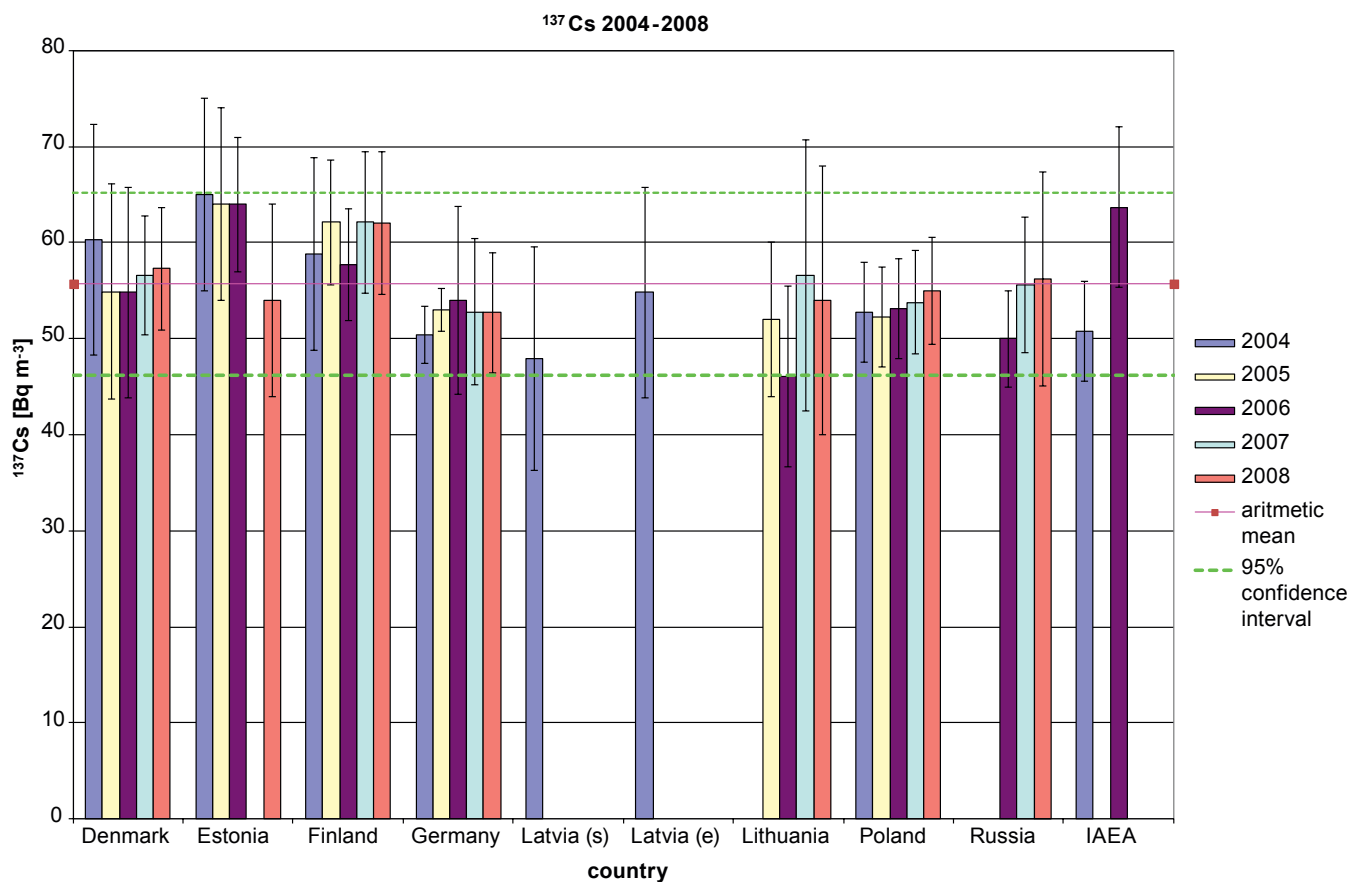
During the years 2004 - 2009, the laboratories were asked to analyse <sup>137</sup>Cs and <sup>90</sup>Sr once a year from a seawater sample collected from the Bothnian Sea (sampling site EB-1); the IAEA-EL participated in the intercalibration. The first results (2004 - 2006) of the intercomparison are reported in the previous report, and the results from the whole period 2004 - 2008 are illustrated in **Figures 9.1** and **9.2**. The results indicate that the measurements are in very good agreement. However, more harmonisation of uncertainty calculations would improve the comparability of the data.

Several laboratories have also regularly participated in the other intercomparisons organised by the IAEA.



**Figure 9.1.** <sup>137</sup>Cs results with two sigma uncertainties in Baltic Sea water intercomparisons, 2004 – 2008.





**Figure 9.2.**  $^{90}\text{Sr}$  results with two sigma uncertainties in Baltic Sea water intercomparisons, 2004 – 2008.



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