

Baltic Sea Environment Proceedings No. 117

Radioactivity in the Baltic Sea, 1999-2006 HELCOM thematic assessment



Helsinki Commission

Baltic Marine Environment Protection Commission

Authors:

Jürgen Herrmann, Tarja K. Ikäheimonen, Erkki Ilus, Günter Kanisch, Maria Lüning, Jukka Mattila, Sven P. Nielsen, Iolanda Osvath and Iisa Outola.

For bibliographic purposes this document should be cited as:

HELCOM, 2009
Radioactivity in the Baltic Sea, 1999-2006
HELCOM thematic assessment

Balt. Sea Environ. Proc. No. 117

Number of pages: 64

Information included in this publication or extracts thereof are free for citation on the condition that the complete reference of the publication is given as stated above

Copyright 2009 by the Baltic Marine Environment Protection Commission
- Helsinki Commission -

ISSN 0357-2994

Language check: Fran Weaver

Cover photo: Elena Bulycheva

Design and layout: Asa Butcher (asa@chameleonproject.net)

Printed by: Erweko Painotuote Oy, Finland

Contents

1	Introduction	7
2	Sources of Radioactive Substances in the Baltic Sea	9
2.1	Introduction	9
2.2	Discharges from facilities in the Baltic Sea drainage area	9
2.2.1	Nuclear facilities (NPPs, research reactors, waste handling, fuel handling, etc.)	9
2.2.2	Non-nuclear facilities (e.g. hospitals, non-nuclear industries, etc.)	10
2.3	Discharges from facilities located outside the Baltic Sea	11
2.3.1	Nuclear reprocessing plants	11
2.3.2	Chernobyl accident	11
2.3.3	Atmospheric nuclear weapons tests	12
2.3.4	Dumping of radioactive waste	13
2.4	Conclusions	13
3A	Radionuclides in Seawater	18
3A.1	Introduction	18
3A.2	Distribution and temporal evolution of ¹³⁷ Cs	18
3A.3	Effective half-life and target levels of ¹³⁷ Cs	22
3A.4	Inventories of ¹³⁷ Cs in seawater	23
3A.5	Other radionuclides	23
3A.6	Conclusions	24
3B	Radionuclides in Sediments	25
3B.1	Introduction	25
3B.2	Material and methods	25
3B.3	Sources of artificial radioactivity	25
3B.4	Results and discussion	25
3B.5	Recommendations and future work	29
3C	Radionuclides in Biota	
3C.1	Introduction	30
3C.2	Material	30
3C.3	Use of a box model for comparison with measured data	30
3C.4	Results and discussion	30
3C.4.1	Concentration factors	30
3C.4.2	Activity concentrations	31
3C.4.3	Activity ratios	32
3C.4.4	Trends in activity concentrations	33
3C.4.5	Conclusions	35

4 Modelling and Dose Calculations	41
4.1 Model work	41
4.2 Dose calculations	42
5 Radioactivity in the Baltic Sea Compared to Other Sea Regions	47
6 Conclusions	50
7 Recommendations	52
8 Acknowledgements	53
Appendix: Data Quality	54
1. Introduction	54
2. Analytical procedures used by MORS laboratories	54
3. Quality assurance, internal and external checking	54
4. Intercalibration excercises (1999-2006) organized by the MORS group	54
4.1 Sediment	54
4.2 Seawater	55
5. Conclusions	55

1 Introduction

Sven Nielsen

Risø DTU, National Laboratory for Sustainable Energy, Denmark

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 (^3H , super heavy hydrogen) and carbon-14 (^{14}C), with half lives of 12 years and 5,700 years, respectively.

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

Therefore, the marine environment contains naturally occurring radionuclides. One cubic metre of seawater typically contains 1000 Bq ^3H , 4 Bq ^{14}C , 40 Bq ^{238}U , 4 Bq ^{226}Ra , 4 Bq ^{210}Po and 12,000 Bq ^{40}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 in the environment, while underground nuclear explosions release little or no radiation into the environment. The routine operations of nuclear power plants give rise to small controlled discharges of radioactive substances, but accidents at nuclear power plants can cause releases of considerable amounts of radioactivity into the environment. Man-made radionuclides of particular concern to man and the environment are ^{90}Sr and ^{137}Cs , which are both formed by nuclear fission. Both of these radioisotopes have half lives of about 30 years, so when released into the environment they remain there for

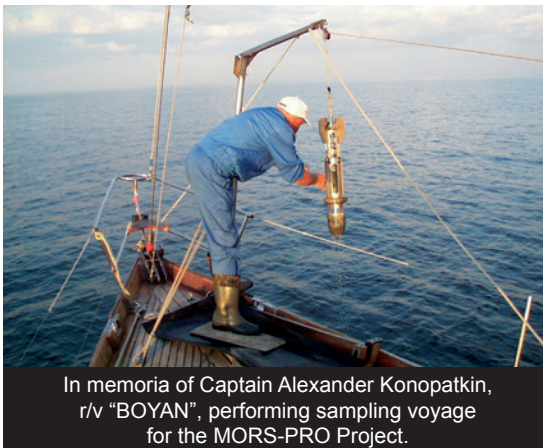


Photo by Visvaldis GRAVERIS

In memoria of Captain Alexander Konopatkin, r/v "BOYAN", performing sampling voyage for the MORS-PRO Project.

many years. Furthermore, ^{90}Sr and ^{137}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 ^{239}Pu and ^{99}Tc , with half lives of 24,000 years and 210,000 years, respectively.

The occurrence of man-made radioactive substances in the Baltic Sea has four main causes:

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 of spent nuclear fuel, at 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.

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 1999-2006. Chapter 2 describes the sources of man-made radioactivity in the Baltic Sea. Chapter 3 describes the levels of man-made radionuclides in seawater, sediments and biota. Chapter 4 describes work on modelling and evaluations of the risks to man caused by radioactivity in the Baltic Sea. Chapter 5 compares the levels of man-made radionuclides in the Baltic Sea with levels in other sea regions. Chapter 6 summarises the project's conclusions. Chapter 7 presents the consequent recommendations, and work on data quality is presented in the Appendix.

References

- National Academy of Sciences (1971):
Radioactivity in the marine environment (RIME), National Academy of Sciences, Washington DC, USA.
- UNSCEAR (2000):
Sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) Report to the General Assembly of the United Nations, New York.



2 Sources of Radioactive Substances in the Baltic Sea

Maria Lünig¹, Erkki Ilus², Jürgen Herrmann³

¹ Swedish Radiation Safety Authority, Sweden

² STUK, Radiation and Nuclear Safety Authority, Finland

³ BSH, Federal Maritime and Hydrographic Agency, Germany

2.1 Introduction

This chapter examines the different sources of the current amount of man-made radioactive substances found in the Baltic Sea. The sources are divided as:

- Nuclear facilities in the Baltic Sea drainage area (nuclear power plants (NPPs), research reactors, waste handling, fuel handling, etc.)
- Non-nuclear facilities in the Baltic Sea drainage area (e.g. hospitals, non-nuclear industries, etc.)
- Discharges from nuclear facilities located outside the Baltic Sea drainage area
- The Chernobyl accident
- Atmospheric nuclear weapons tests

2.2 Discharges from facilities in the Baltic Sea drainage area

2.2.1 Nuclear facilities (NPPs, research reactors, waste handling, fuel handling, etc.)

This category includes all the nuclear facilities (power reactors, research reactors, waste handling facilities, fuel production, etc.) located in the drainage area of the Baltic Sea which discharge directly or indirectly into the Baltic Sea. The main characteristics of these facilities are summarised in **Table 1**. The locations of the facilities are shown in **Figure 1**.

The discharge pattern for most of the NPPs is similar, and the most abundant nuclides present in the discharges are shown in **Table 1**. The amounts of the most significant radionuclides discharged are shown in **Figures 2-10**.

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

Table 1: Nuclear facilities in the drainage area of the Baltic Sea, and their main discharge nuclides.

Table 2:
List of NORM industries (using naturally occurring radioactive materials) in the Baltic Sea area.

Type of industry	Denmark	Estonia	Finland	Germany	Latvia	Lithuania	Poland	Russia	Sweden
Fossil fuel power stations	X	X	X	X		X	X		
Discharge point in the Baltic Sea							No		
Discharge data or produced amount given in tonnes									
Oil and gas extraction	X			X		X	X		
Discharge point in the Baltic Sea							No		
Discharge data or produced amount given in tonnes									
Metal processing - particularly iron and steel production, but also tantalum and niobium	X	X	X	X			X		X
Discharge point in the Baltic Sea							No		X
Discharge data or produced amount given in tonnes									
Phosphate industry	X		X	X		X	X		X
Discharge point in the Baltic Sea							No		X
Discharge data or produced amount given in tonnes									
Titanium oxide pigment production			X	X					
Discharge point in the Baltic Sea									
Discharge data or produced amount given in tonnes									
Zirconium and rare earth processes - refractory products and brick manufacture	X		X	X					
Discharge point in the Baltic Sea									
Discharge data or produced amount given in tonnes									
Cement production	X	X	X	X		X	X		X
Discharge point in the Baltic Sea							No		X
Discharge data or produced amount given in tonnes									
Water purification plant		X	X			X	X		X
Discharge point in the Baltic Sea							No		X
Discharge data or produced amount given in tonnes									X
Paper mill		X	X			X	X		X
Discharge point in the Baltic Sea							No		X
Discharge data or produced amount given in tonnes									
Others?									
Discharge point in the Baltic Sea									
Discharge data or produced amount given in tonnes									

2.2.2 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. Different types of industries have been listed according to EC (2003) concerning the state of NORM (Naturally Occurring Radioactive Material) in Europe. Information about discharges from non-nuclear facilities in the Baltic Sea area was collected from the HELCOM Contracting Parties in a questionnaire. Only a limited amount of data could be compiled in this way, however, and it was not possible to obtain reliable information about discharges from these sources.

Radionuclides are used for various purposes in industry, medicine and research, and their

usage is increasing. However, according to reports of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2000), their contribution to overall man-made exposures is relatively insignificant. Most of the radionuclides used in hospitals are short-lived, so their discharges are small, and their impact on radioactivity in the Baltic Sea is negligible and very local (Ilus & Ilus 2000). **Tables 2 and 3** give an overview of the different types of non-nuclear industries in the countries surrounding the Baltic Sea. Some information about hospitals and laboratories working with radionuclides is also given. The list of industries is based on EC (2003).

	Denmark	Estonia	Finland	Germany	Latvia	Lithuania	Poland	Russia	Sweden
Hospitals,						3	118		18
Discharge point directly in the Baltic Sea							No		Yes
Discharge data or produced amount given in tonnes							-		Yes
Hospitals,									14
Discharge point in rivers or lakes draining into the Baltic Sea									Yes
Discharge data or produced amount given in tonnes									Yes
Laboratories, open sources						6	343		
Discharge point in the Baltic Sea							No		
Discharge data or produced amount given in tonnes							-		
Laboratories, closed sources							141		
Discharge point in the Baltic Sea							No		
Discharge data or produced amount given in tonnes							-		
Industries,									
Discharge point in the Baltic Sea									
Discharge data or produced amount given in tonnes									
Industries,									
Discharge point in rivers or lakes draining into the Baltic Sea									
Discharge data or produced amount given in tonnes									

Table 3: Table summarising other non-nuclear facilities (e.g. hospitals, laboratories or industries producing or working with radioactive materials).

Source	¹³⁷ Cs TBq	Percentage of total input	⁹⁰ Sr TBq	Percentage of total input
1. Chernobyl accident ¹⁾⁴⁾ / incl. river discharges ⁵⁾	4,700 / 300	82	80	13
2. Nuclear weapons tests ⁶⁾	800	14	500	81
3. Discharges from sources located outside the Baltic Sea ²⁾³⁾	250	4	40	6
4. Discharges into the Baltic Sea ¹⁾ , Cumulative amount up to 2006	1.5	0.03	0.73	0.1

Table 4: Total inputs of ¹³⁷Cs and ⁹⁰Sr into the Baltic Sea from different sources.

¹⁾ based on measurements

²⁾ estimated

³⁾ according to Nies et al. 1995

⁴⁾ according to Nielsen et al. 1999

⁵⁾ according to Illus & Illus, 2000

⁶⁾ according to Nielsen (pers. comm.)

2.3 Discharges from facilities located outside the Baltic Sea

2.3.1 Nuclear reprocessing plants

Small proportions 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 times for these radionuclides are about 4-5 years for discharges into the Irish Sea from Sellafield, and about 2 years for discharges into the English Channel from 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

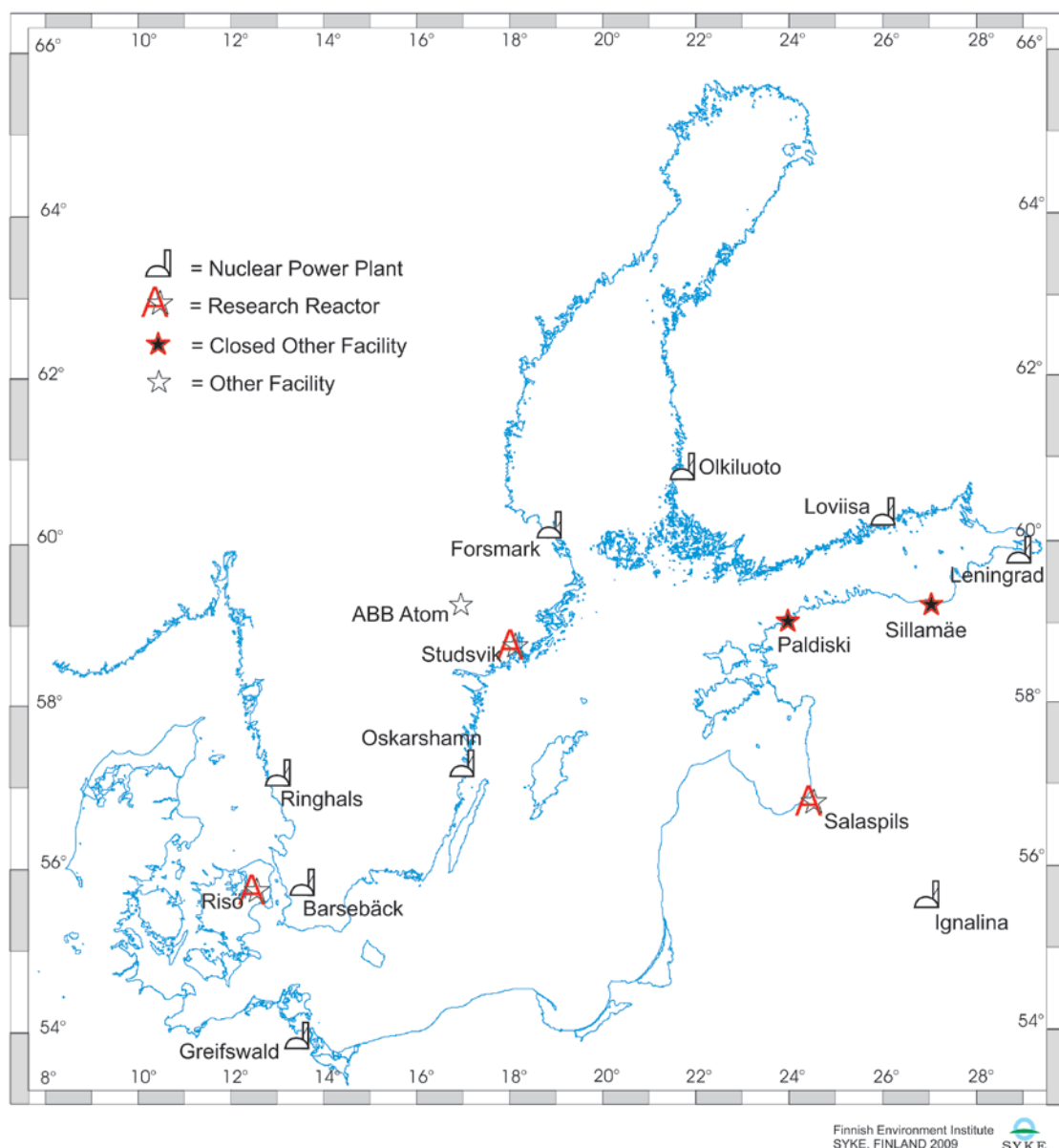
Kattegat and the Belt Sea, most of this activity returns to the Skagerrak and only about 1% enters the Baltic Sea (Nielsen et al. 1995). See **Figures 11 and 12**.

2.3.2 Chernobyl accident

Since April 1986 the accident at the Chernobyl NPP has been the main source of man-made radioactivity in the Baltic Sea (**Table 4**).

The total input of ¹³⁷Cs from the Chernobyl accident into the Baltic Sea area was estimated at 4,500 TBq by the CEC (1991). This estimate was later adjusted to 4,700 TBq (Nielsen et al. 1999). The HELCOM/MORS Expert Group estimated in its Joint Report of 1995 that 4,100-5,100 TBq of ¹³⁷Cs and 80 TBq of ⁹⁰Sr (decay-corrected to 1991) entered the Baltic Sea as a consequence of the Chernobyl accident (Nies et al. 1995). The deposition of fallout from Chernobyl was very une-

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



venly 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}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. Thorough calculations showed that a total of 65 TBq of ^{137}Cs was discharged by Finnish rivers into the Baltic Sea during the period 1986-1996 (Saxen & Ilus 2000). Corresponding calculations performed in Russia showed that 14 TBq were discharged by five large rivers

from the territory of the former Soviet Union during the years 1986-1988 (Gavrilov et al. 1990). A Polish estimate made for the River Vistula showed that a total of 18 TBq of ^{137}Cs were discharged during the period 1986-1996 (Ilus & Ilus 2000). The total river input of ^{137}Cs was estimated at 300 TBq during the decade 1986-1996 (Ilus & Ilus 2000).

2.3.3 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 was 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 inputs of weapons-test ^{90}Sr and ^{137}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 from the Baltic Sea resulted in quite similar values: 490 TBq for ^{90}Sr , and 620 TBq for ^{137}Cs (calculated to 1981) (Salo et al. 1986).

2.3.4 Dumping of radioactive waste

Five officially confirmed dumpings of radioactive waste at three different dump sites have been reported in the Baltic Sea region. All of these small-scale dumpings were performed in the late 1950s or early 1960s. A radiological assessment of these dumpings 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

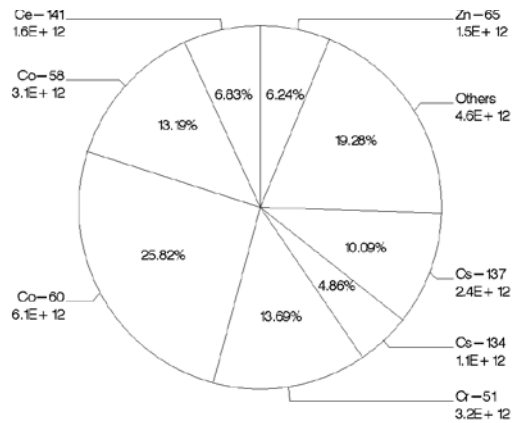


Figure 2: Total aquatic discharges from local nuclear facilities into the Baltic Sea until the end of 2006, excluding H-3.

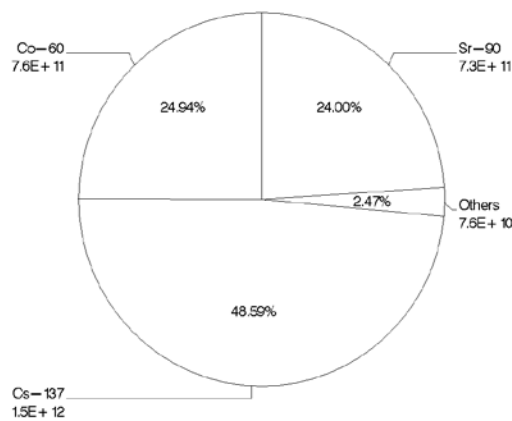


Figure 3: Cumulative aquatic discharges into the Baltic Sea, decay-corrected to the end of 2006, excluding H-3.

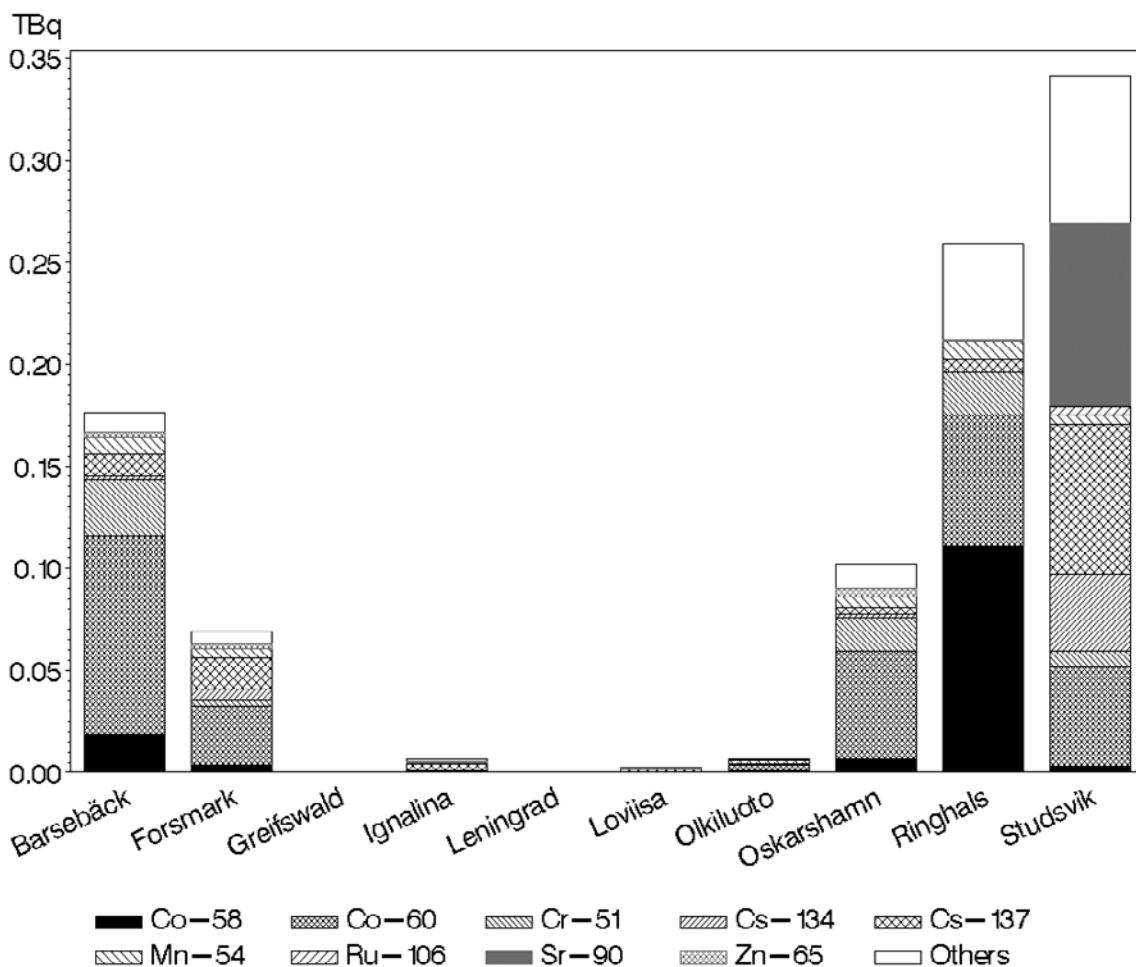


Figure 4: Total aquatic discharges from local nuclear power plants into the Baltic Sea 1999-2006, excluding H-3.

Figure 5:
Annual Co-60 discharges from local nuclear facilities into the Baltic Sea 1999-2006.

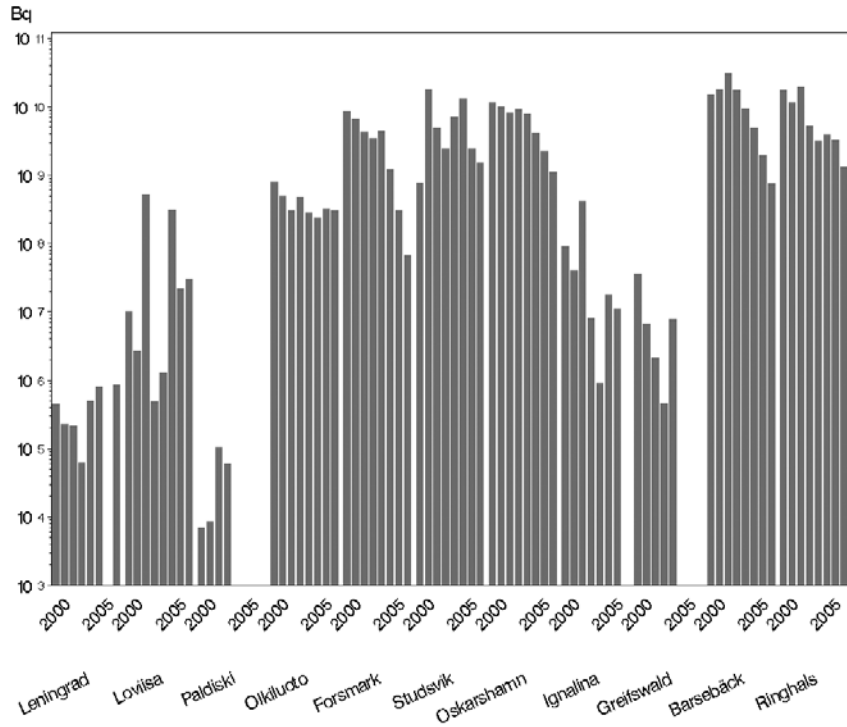
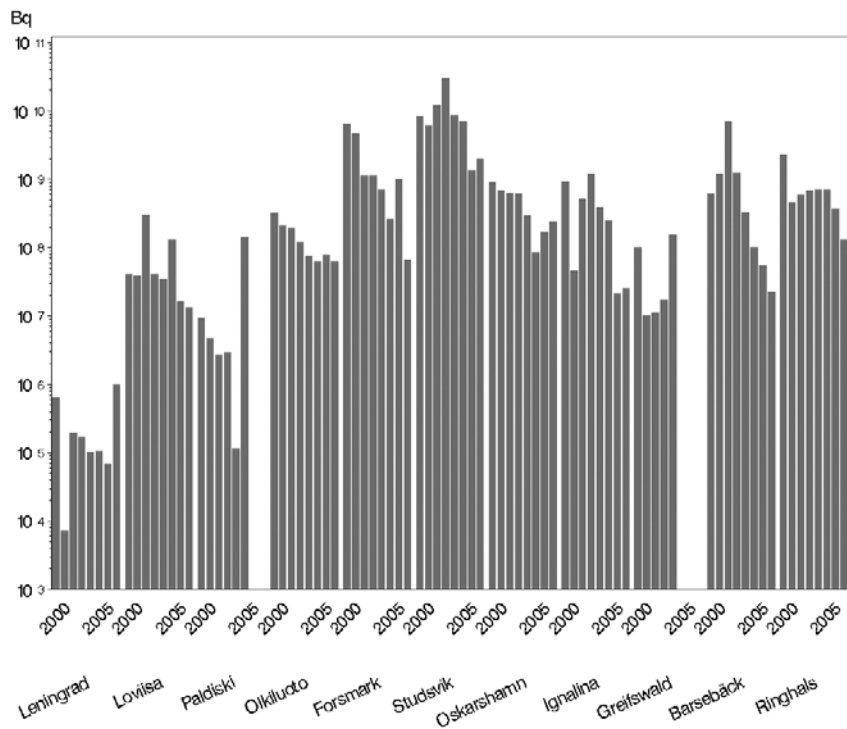


Figure 6:
Annual Cs-137 discharges from local nuclear facilities into the Baltic Sea 1999-2006.



deposition were ¹³⁷Cs and ¹³⁴Cs. The total input of ¹³⁷Cs from Chernobyl to the Baltic Sea has been estimated at 4,700 TBq. The post-Chernobyl river discharges of ¹³⁷Cs were estimated in the Marina Balt Study at 300 TBq, thus comprising 6-7% of the total input.

The second most important source is global fallout from the atmospheric nuclear weapons

tests carried out during the late 1950s and early 1960s. The predominant radionuclides in the consequent global fallout were ¹³⁷Cs and ⁹⁰Sr, in an activity ratio of about 1.6. During the late 1990s the decay-corrected amounts of weapons-test ¹³⁷Cs and ⁹⁰Sr in the Baltic Sea have been evaluated at 800 and 500 TBq, respectively.

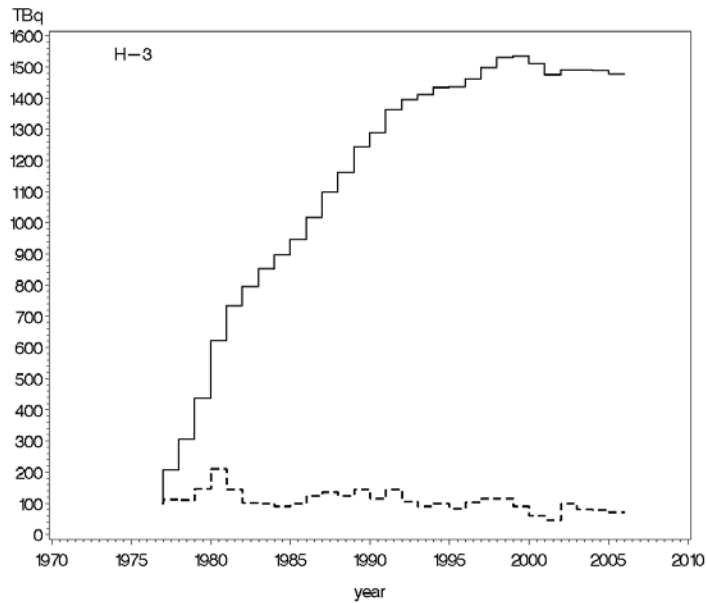


Figure 7: Annual (dotted line) and cumulative aquatic H-3 discharges from local nuclear facilities into the Baltic Sea 1977-2006.

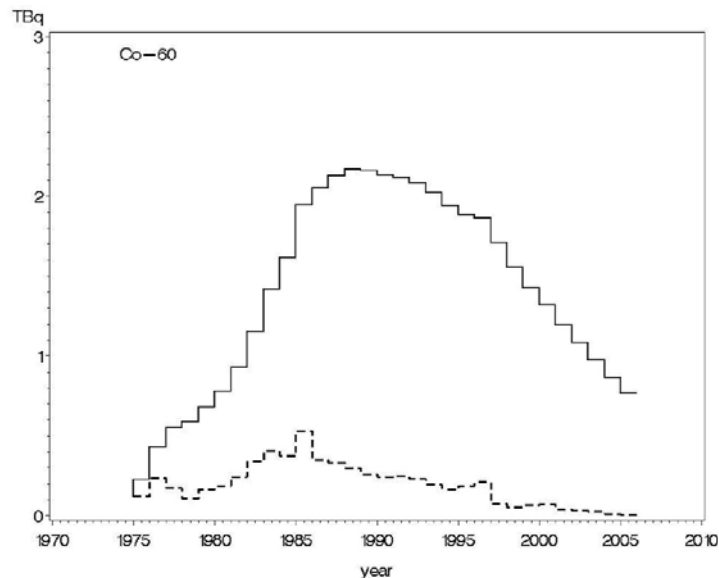


Figure 8: Annual (dotted line) and cumulative aquatic Co-60 discharges from local nuclear facilities into the Baltic Sea in 1975-2006.

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

The predominant radionuclide in the discharges from nuclear power plants and research reactors in the Baltic Sea region is ^3H . The total discharges of ^3H from these local sources have amounted to 3,200 TBq, while discharges of other beta-gamma nuclides have amounted to about 24 TBq, by the end of 2006. The total discharges of alpha nuclides have been 0.005 TBq.

For ^{137}Cs , the main source is fallout from Chernobyl (82%), followed by nuclear weapons test fallout (14%). For ^{90}Sr , the main source of contamination is fallout from nuclear weapons tests (81%), while the proportion originating from Chernobyl fallout was smaller (13%).

Figure 9:
Annual (dotted line) and cumulative aquatic Cs-137 discharges from local nuclear facilities into the Baltic Sea 1975-2006.

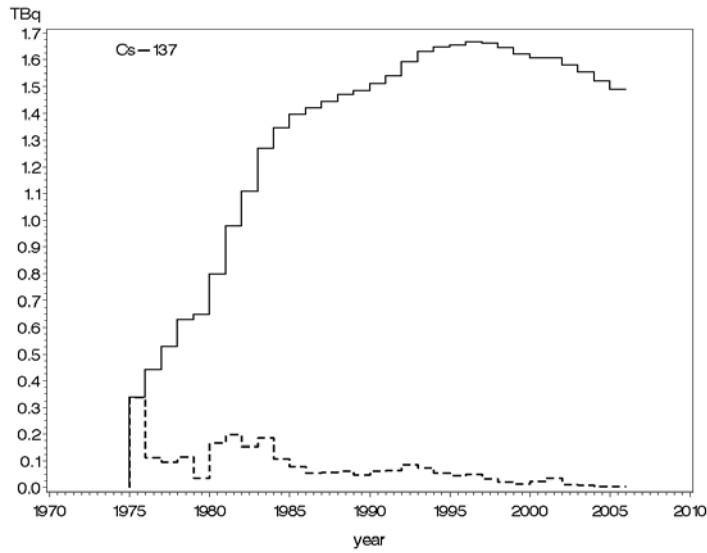


Figure 10:
Annual (dotted line) and cumulative aquatic Sr-90 discharges from local nuclear facilities into the Baltic Sea 1981-2006.

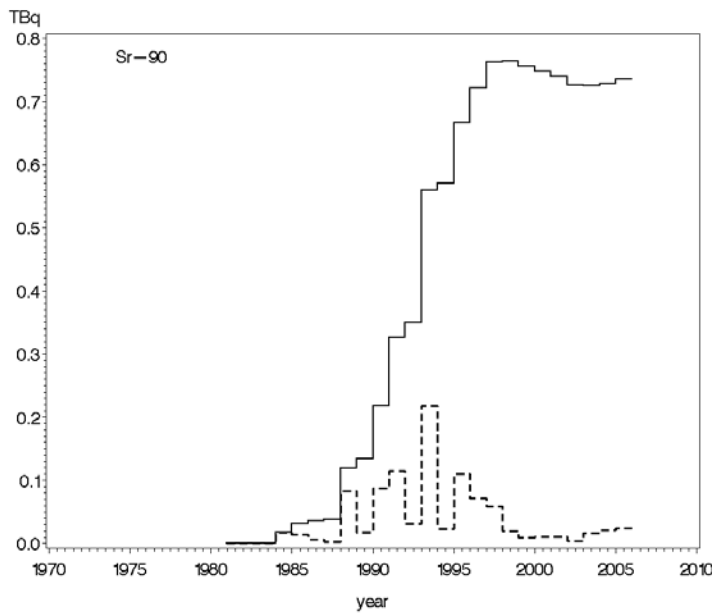
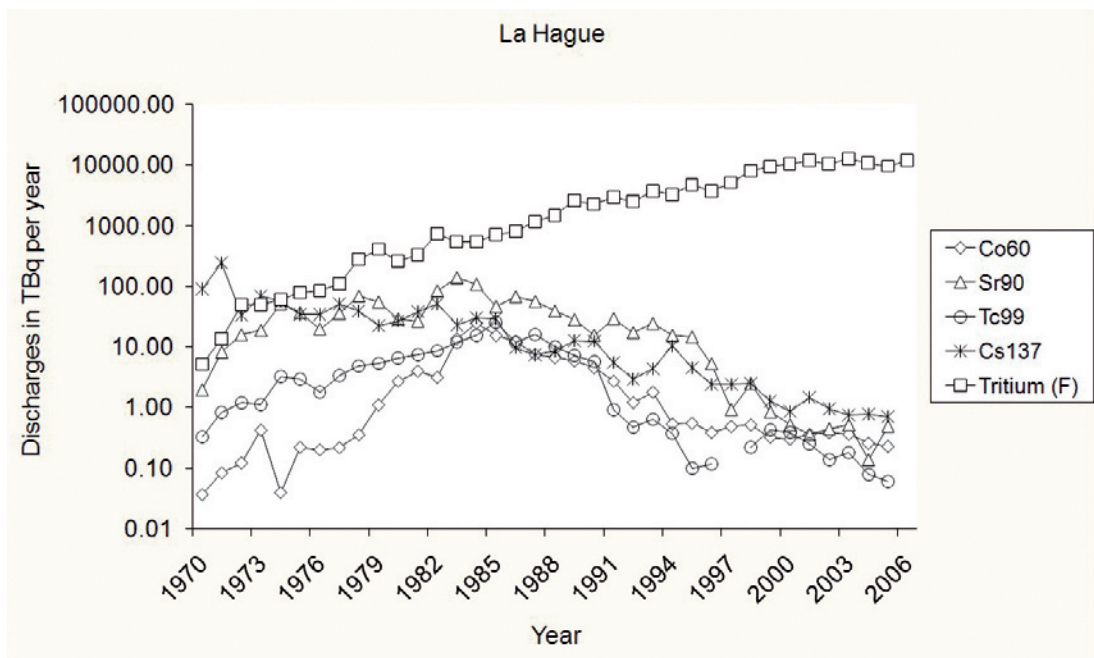


Figure 11:
Discharges of selected radionuclides from La Hague into the English Channel.



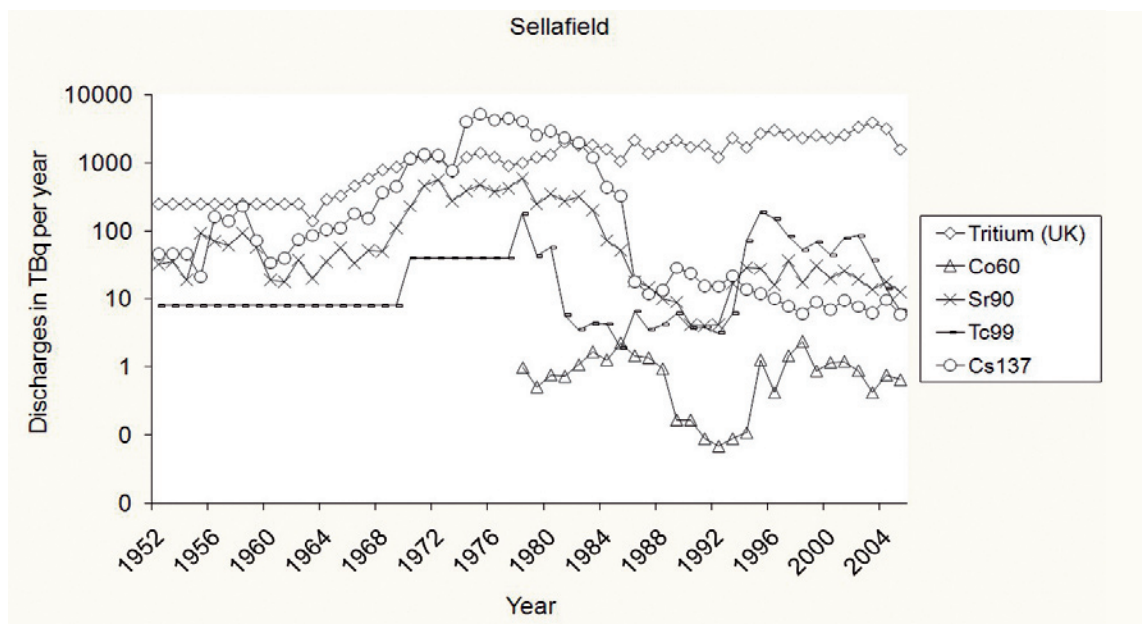


Figure 12:
Discharges of selected radionuclides from Sellafield into the Irish Sea.

References

CEC, Commission of the European Communities (1991): Proceedings of Seminar on Comparative Assessment of the Environmental Impact of Radionuclides Released during Three Major Nuclear Accidents: Kyshtym, Windscale, Chernobyl, Vol. I and II, EUR 1374.

EC, European Commission (2003): Effluent and dose control from European Union NORM Industries: Assessment of current situation and proposal for a harmonised Community approach, Radiation Protection 135, Vol 1-2.

Gavrilov, V.M., Z.G. Gritchenko, L.M. Ivanova, T.E. Orlova, V.P. Tishkov & N.A. Tishkova (1990): Strontium-90, caesium-134 and caesium-137 in water reservoirs of the Soviet Union's Baltic region (1986-1988), Radiochemistry, No. 3:171-179. (in Russian).

Ilus, E. & T. Ilus (2000): Sources of Radioactivity. In: Nielsen, S.P. (ed.): The radiological exposure of the population of the European Community to radioactivity in the Baltic Sea, Marina-Balt project, Radiation Protection 110:9-76, EUR 19200, European Commission, Luxembourg.

Nielsen, S.P., O. Karlberg & M. Øhlenschläger (1995): Modelling the transfer of radionuclides in the Baltic Sea. In: HELCOM (1995): Radioactivity in the Baltic Sea 1984-1991. Balt. Sea Environ. Proc. No. 61:121-148.

Nielsen, S.P., P. Bengtson, R. Bojanowski, P. Hagel, J. Herrmann, E. Ilus, E. Jakobson, S. Motiejunas, Y. Panteleev, A. Skujina & M. Suplinska (1999): The radiological exposure of man from radioactivity in the Baltic Sea, The Science of the Total Environment 237/238:133-141.

Nies, H., R. Bojanowski, O. Karlberg & S.P. Nielsen (1995): Sources of radioactivity in the Baltic Sea. In: HELCOM (1995): Radioactivity in the Baltic Sea 1984-1991. Balt. Sea Environ. Proc. No. 61:6-18.

Salo, A., K. Tuomainen & A. Voipio (1986): Inventories of some long-lived radionuclides in the Baltic Sea, The Science of the Total Environment 54:247-260, Elsevier Science publishers, Amsterdam.

Saxén, R. & E. Ilus (2000): Discharge of ¹³⁷Cs by Finnish rivers to the Baltic Sea in 1986-1996. In: Nielsen, S.P. (ed.): The radiological exposure of the population of the European Community to radioactivity in the Baltic Sea, Marina-Balt project, Radiation Protection 110:333-347, EUR 19200, European Commission, Luxembourg.

UNSCEAR (2000): Sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, Vol. 1. United Nations, New York.

3A Radionuclides in Seawater

Jürgen Herrmann¹, Iisa Outola², Tarja K. Ikäheimonen²

¹ BSH, Federal Maritime and Hydrographic Agency, Germany

² STUK, Radiation and Nuclear Safety Authority, Finland

3A.1 Introduction

This chapter describes the distribution of artificial radionuclides in seawater in the Baltic Sea over the years 1999-2006. During this period nine countries contributed their results for a total of almost 2,000 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 some gaps in the Archipelago Sea (see **Figure 1**). As presented in earlier reports (HELCOM 1995a, 2003) the predominant radionuclide in the Baltic Sea is ¹³⁷Cs, as this radionuclide was released in great amounts by the Chernobyl accident in 1986. The other main contaminant released in the Chernobyl event, ¹³⁴Cs, has practically vanished to concentrations below the 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 ⁹⁰Sr, ²³⁹Pu and ⁹⁹Tc. The sources of these radionuclides are described in Chapter 2. The concentrations mentioned in this chapter are generally understood as activity concentrations. A detailed description of methods was given in the earlier joint report (HELCOM 1995b) and updated in the Appendix of this report. The collecting of monitoring data was accompanied by a thorough programme of quality assurance, covering both ¹³⁷Cs and ⁹⁰Sr in seawater in annual exercises, also shown in the Appendix.

3A.2 Distribution and temporal evolution of ¹³⁷Cs

The fate of any pollutant introduced into the sea is determined by both its own chemical properties and hydrographical conditions of the sea itself. As a relatively small, semi-enclosed, brackish sea, which is 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 any form

of pollution. The Chernobyl accident made this situation most clear, as its sorry legacy is still abundant 20 years after the event.

The Chernobyl accident resulted in the very uneven ¹³⁷Cs 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 ¹³⁷Cs has changed as a consequence of river discharges, the mixing of water masses, sea currents, and sedimentation processes (Ilus 2007). In the early phase after Chernobyl, ¹³⁷Cs concentrations decreased rapidly in the Gulf of Finland and in the Bothnian Sea, while at the same time increasing in the Baltic Proper (**Figures 1 and 2**).

During the period 1999-2006 concentrations of ¹³⁷Cs have continued to decrease in all regions of the Baltic Sea. In the beginning of this period, the highest ¹³⁷Cs concentrations were reported in the Bothnian Sea, where concentrations decreased from 82 to 49 Bq/m³ during the monitoring period. In the Baltic Proper, concentrations decreased from 69 to 47 Bq/m³, and by 2006 concentrations were at the same level in both the Bothnian Sea and the Baltic Proper. In both the Gulf of Finland and in the Bothnian Bay, concentrations were lower, at around 37 Bq/m³ in 2006. Concentrations in the Western Baltic have been lower overall, decreasing from 53 to 31 Bq/m³ during the monitoring period. Variations in ¹³⁷Cs concentrations in surface water between different seawater regions are becoming less pronounced. In 2006 ¹³⁷Cs concentrations varied by a factor of 1.5 between the Baltic's most contaminated area, the Bothnian Sea, and the least contaminated waters in the Western Baltic.

The vertical distribution of radionuclides in the water column is influenced by physical and biological processes as described above. The distribution of ¹³⁷Cs between surface and near-bottom water in different basins is shown in **Figures 3 and 4**. The average ratio of ¹³⁷Cs

Figure 1: ^{137}Cs concentrations (Bq/m³) in surface water (sampling depth $\leq 10\text{m}$) 1984-2006, as annual mean values by basin. Target values have been calculated as averages of pre-Chernobyl (1984-1985) concentrations. (Note: logarithmic scales used in the graphs).

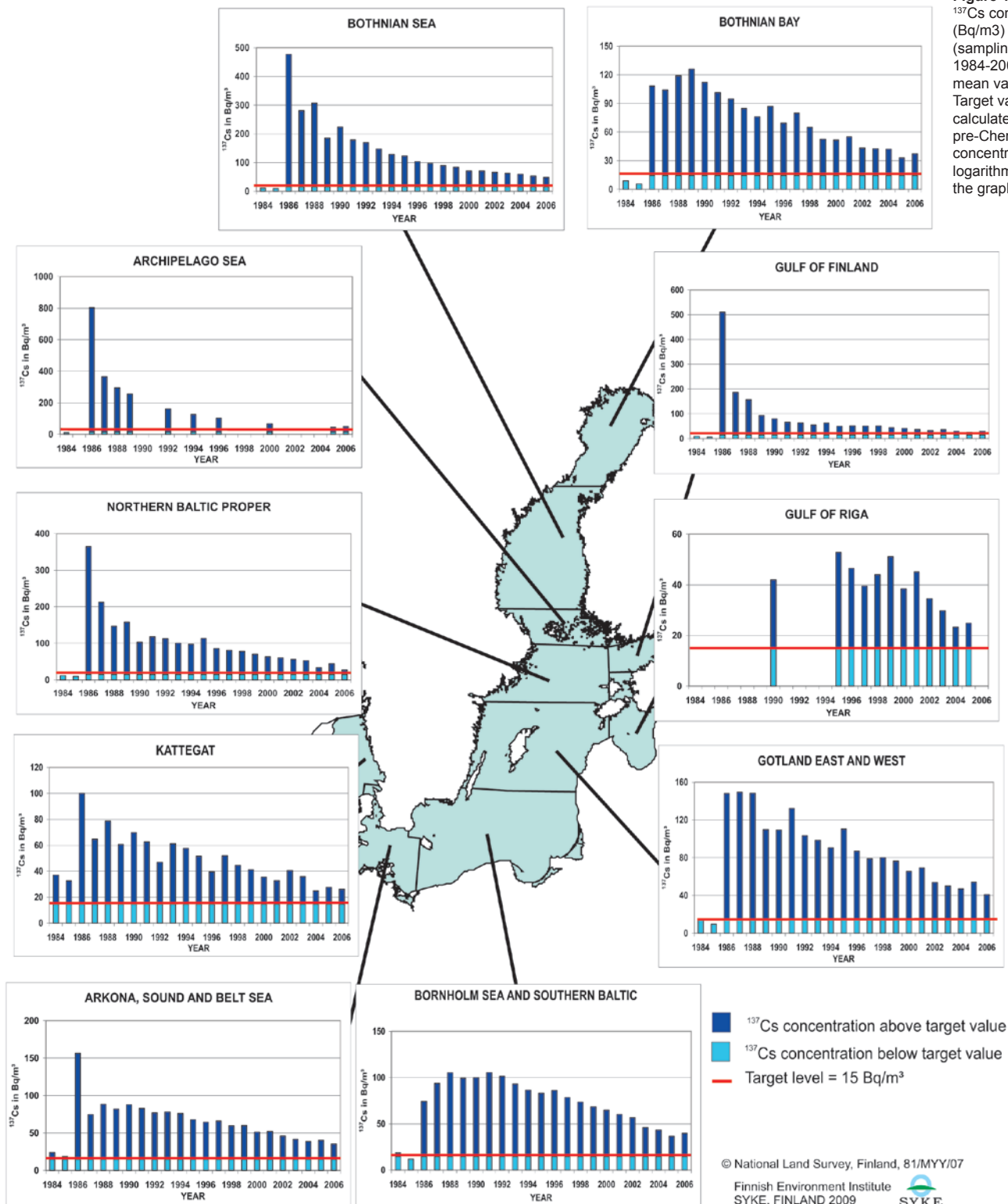


Figure 2:
Temporal variations in ^{137}Cs concentrations (Bq/m^3 in surface water) at five stations in various parts of the Baltic Sea.

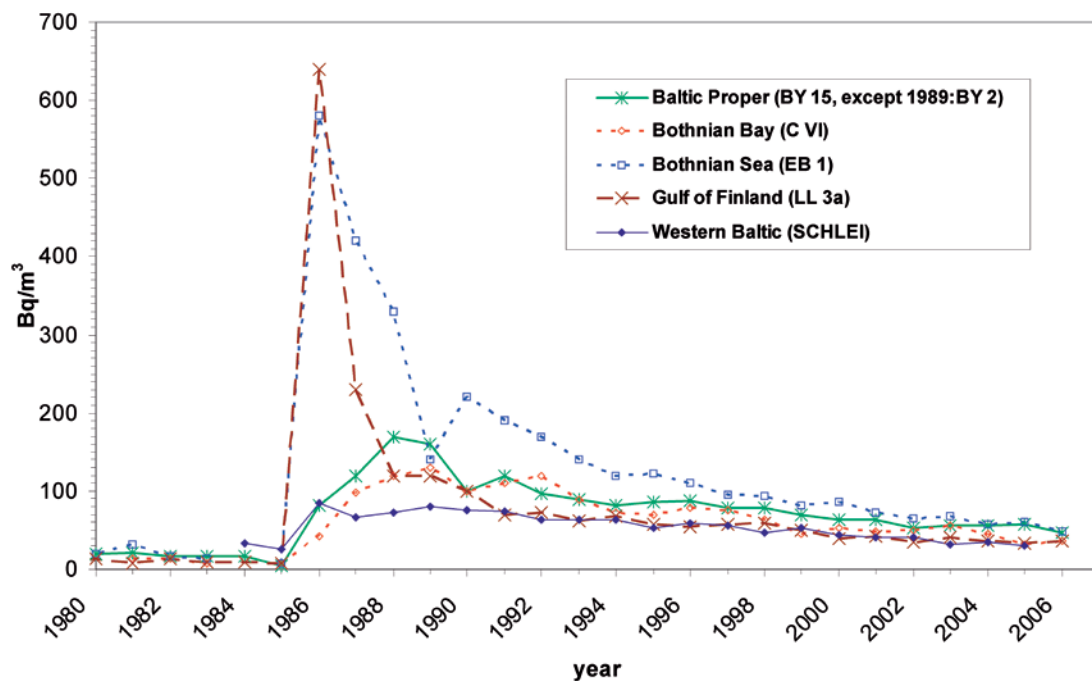
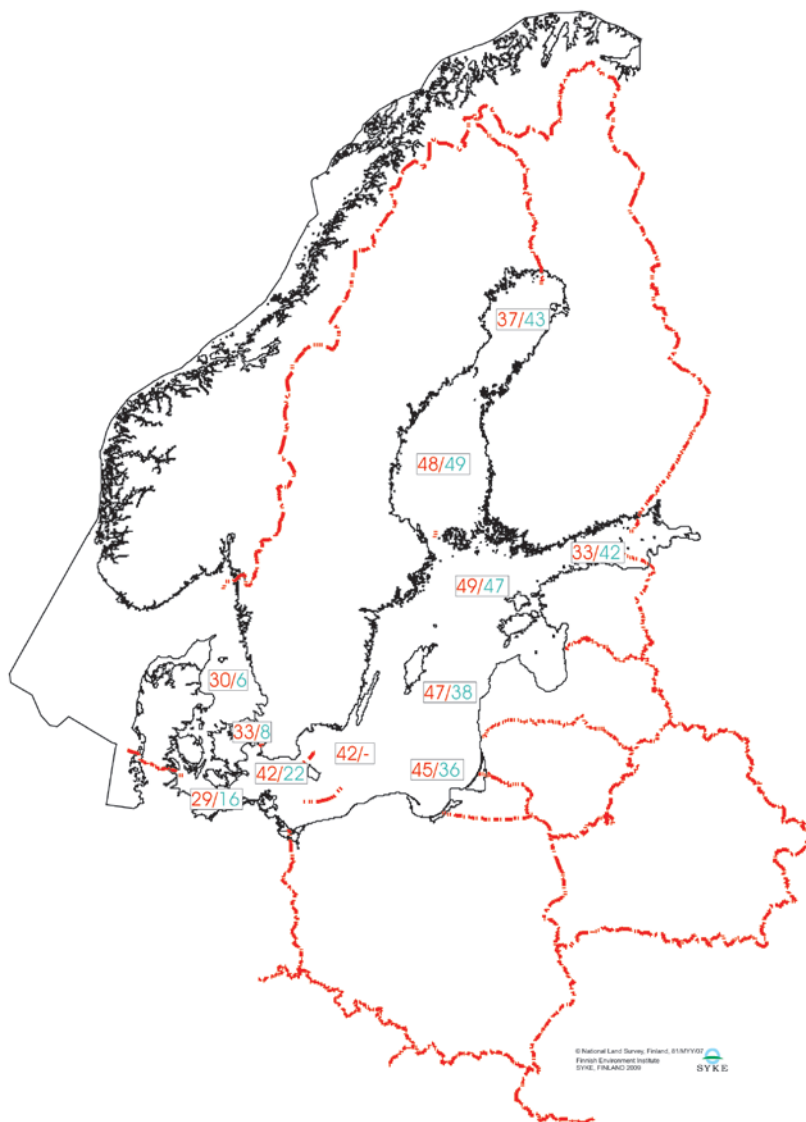


Figure 3:
 ^{137}Cs activity (Bq/m^3) in surface/bottom water as mean values by basin in 2006.



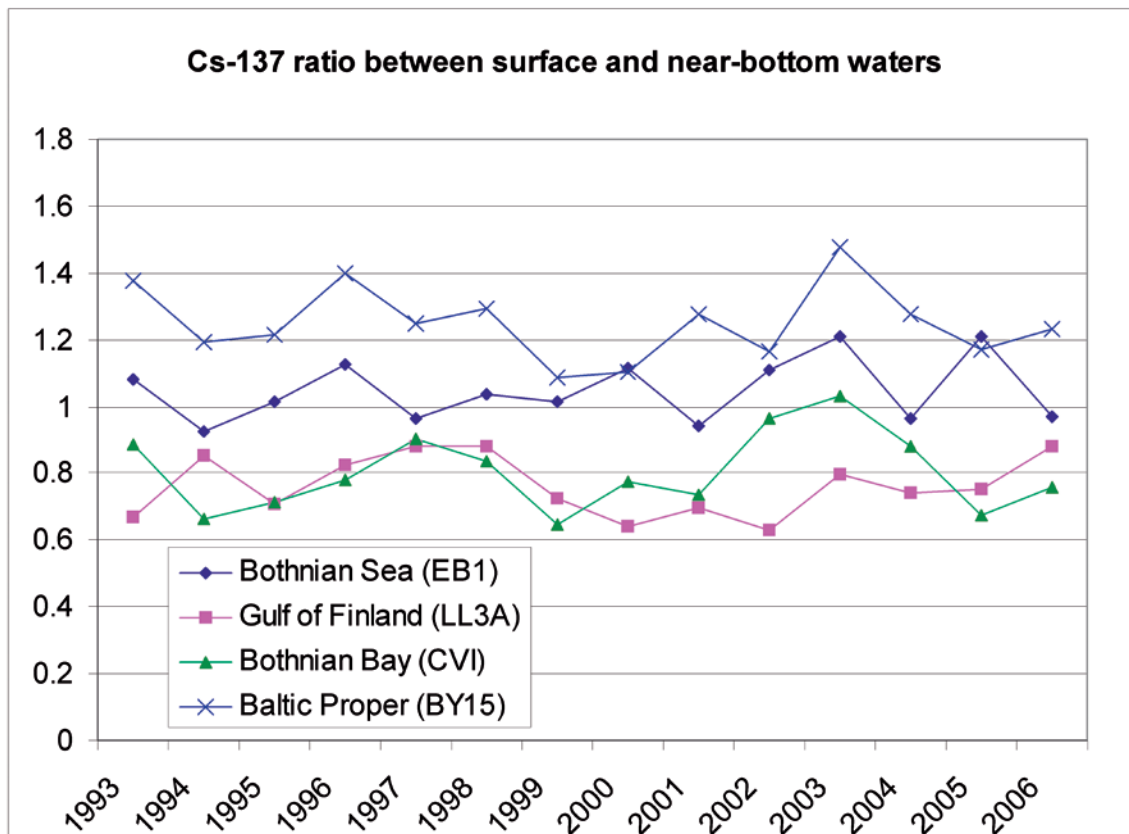


Figure 4: Temporal variations in ratios of ^{137}Cs concentrations in surface water to those in near-bottom waters at four stations in various parts of the Baltic Sea.

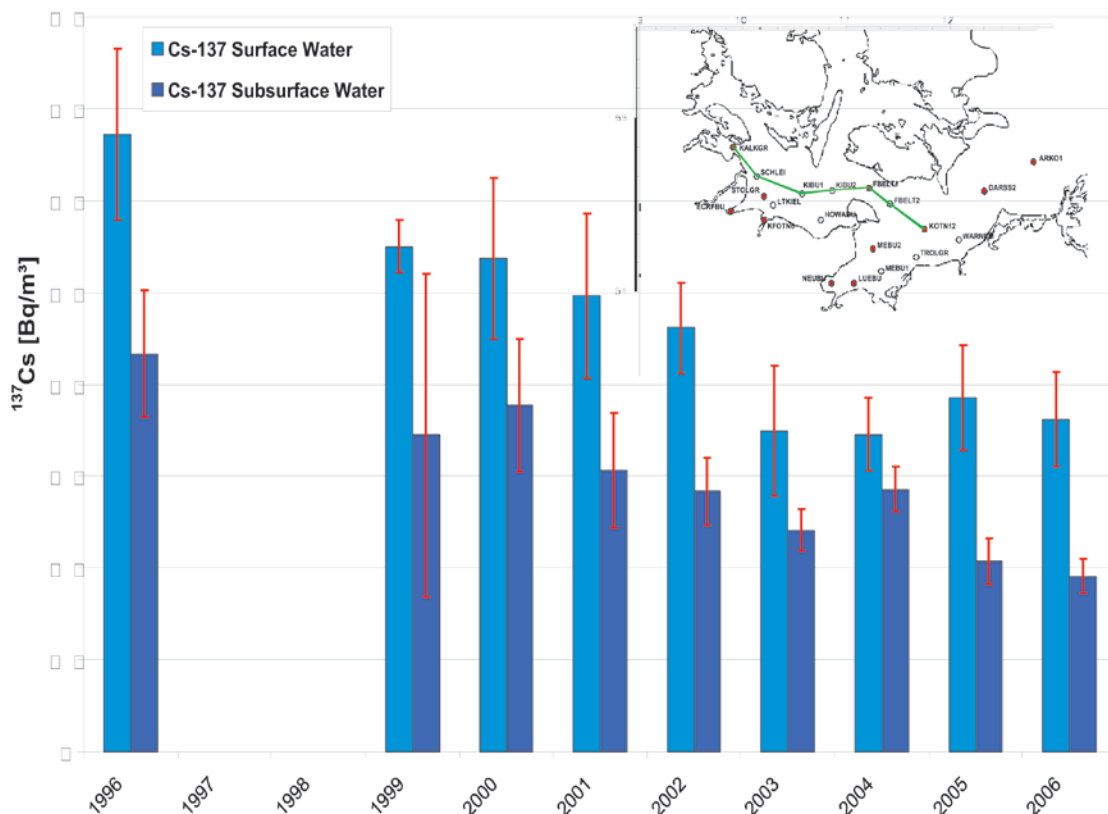
concentrations in surface water to those in near-bottom water was 1.3 in the Baltic Proper during the period 1993–2006, due to the inhibiting effect of the pronounced halocline on mixing between different water layers. In the Bothnian Sea the average ratio was 1.0, while in both the Gulf of Finland and the Bothnian Bay, the ratio was 0.8. Vertical exchange is much more efficient in these water basins, because of the lack of stratification.

Both the Gulf of Finland and the Bothnian Bay are shallow basins, with an average water depth of around 40 metres. The Bothnian Sea is generally deeper, with an average depth of around 70 m. Large freshwater inflows may contribute to lower ^{137}Cs concentrations in surface water in the Gulf of Bothnia and the Gulf of Finland. No evidence of any remobilization of ^{137}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 ^{137}Cs contamination by transferring contaminated near-bottom water from the Bothnian Sea to the Baltic Proper. As illustrated in **Figure 3**, ^{137}Cs concentrations in near-bottom water are highest in the Bothnian Sea, and decrease towards the Sound and the Kattegat.

The Western Baltic, as a transitional area between the North Sea and the Baltic Sea, has special hydrographic conditions different from the rest of the Baltic Sea. First of all it is shallow, with an average depth of 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 which results in a current system of high intra-annual and inter-annual variability. As an indicator of the inflow of bottom water, mean ^{137}Cs concentrations from seven selected stations are shown in **Figure 5**. In addition to the general decreasing trend in ^{137}Cs concentrations, this graph shows information about the variability of surface and bottom water concentrations, which has been much less in recent years in comparison with the year 1999. The year with the smallest difference between concentrations in surface and bottom waters was 2004, when bottom water flowing in from the Kattegat with low ^{137}Cs concentrations evidently did not reach the indicator stations.

The bottom water from the North Sea supplies the Baltic Sea with contaminants originating from the La Hague and Sellafield

Figure 5:
Time series of ^{137}Cs mean concentrations from stations characterising the inflow of bottom water into the Western Baltic.



reprocessing plants (see Chapter 2) such as ^{239}Pu , ^{99}Tc and ^{129}I , whereas the outflowing surface waters from the Baltic Sea represent a significant source of ^{137}Cs for the North Sea. The consequent impacts are detectable along the entire south coast of Norway at least until 60°N . Today the Baltic Sea can be regarded as the most significant source of ^{137}Cs for the North Atlantic, only comparable with the sediments of the Irish Sea. The quantification of this source is still unclear, because of the high variability of outflow rates, but it is estimated at tens of TBqs per year.

radionuclide and each environment where they may occur. Effective half-lives have been calculated for ^{137}Cs in various parts of the Baltic Sea, as shown in **Table 1**. Currently, the effective half-lives of ^{137}Cs in surface water vary from 9 years in the Bothnian Bay to 15 years in the Baltic Proper. The longer residence time of ^{137}Cs in the Baltic Proper is most likely due to inflows of more contaminated water from the northern part of the Baltic Sea. In the time period following Chernobyl, 1986-1988, the effective half-lives of ^{137}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}Cs in Gulf of Finland as compared to the Bothnian Sea during 1986-1988 was probably due to different water exchange and sedimentation processes in these two regions (Ilus et al. 1993). Over time the effective half-lives have increased in both regions, and currently the residence times of ^{137}Cs are 13 and 9 years in the Gulf of Finland and in the Bothnian Sea respectively. The target level for ^{137}Cs concentrations in Baltic seawater is defined as 15 Bq/m^3 , equivalent to average pre-Chernobyl concentrations. Based on the calculated effective half-lives, this level will be reached by the year 2020 in both the Gulf of Finland and the Gulf of Bothnia. But it will take

Table 1:
Effective half-lives of ^{137}Cs in surface water in different basins of the Baltic Sea (1993-2006)

Baltic Sea Region	Time period	
	1986-1988 (years)	1993-2006 (years)
Bay of Bothnia	-	10
Bothnian Sea	2.5	9
Gulf of Finland	0.8	13
Baltic Proper	-	15

3A.3 Effective half-life and target levels of ^{137}Cs

The effective half-life of a radioactive contaminant is the time required for its concentrations to decrease by 50% as a result of physical, chemical and biological processes. Half-lives are specific to each

10 more years, until 2030, to reach the target level in the Baltic Proper. These estimates are only valid if the effective half-lives remain constant. If they increase, as often happens with time, the target levels will only be reached at a later date.

3A.4 Inventories of ¹³⁷Cs in seawater

Inventories of ¹³⁷Cs in the Baltic seawater are given in **Table 2**. These estimates show that the inventory of ¹³⁷Cs in the Baltic water mass was approximately 1,540 TBq in 1999, and had decreased to 870 TBq in 2006. The inventories were estimated by calculating ¹³⁷Cs inventories for various seawater regions of Baltic Sea (Bothnian Bay, Bothnian Sea, Gulf of Finland, Gulf of Riga, Baltic Proper, Kattegat, Belt Sea) and then combining these estimates. Inventories for different basins were calculated using their volumes (HELCOM, 1996), and their average ¹³⁷Cs concentrations, which were calculated from the observation data. The temporal evolution of ¹³⁷Cs inventory in the Baltic seawater is presented in **Figure 6**. Previous estimates for ¹³⁷Cs inventories (HELCOM 1995, 1998; Dahlgard 1989) are shown together with the new estimates. The HELCOM 1995 and 1998 estimates were calculated with the assumption that the estimated mean concentration of ¹³⁷Cs was the same for the entire Baltic Sea. The new estimates appear to be more consistent as

Year	¹³⁷ Cs (TBq)
1999	1540
2000	1380
2001	1320
2002	1230
2003	1090
2004	1030
2005	950
2006	870

Table 2:
Estimated ¹³⁷Cs inventories in Baltic seawater.

compared to the earlier ones (HELCOM 1995, 1998). Based on the inventory estimates, the effective half-life of ¹³⁷Cs in Baltic seawater during the period 1993-2006 has been 9.6 years. With this decay rate, the ¹³⁷Cs inventory in the Baltic Sea would reach pre-Chernobyl levels (250 TBq) by the year 2020, presuming that the effective half-life will stay constant, and no substantial remobilization of ¹³⁷Cs from sediments will occur.

3A.5 Other radionuclides

⁹⁰Sr

⁹⁰Sr concentrations in Baltic seawater varied in general from 5 to 15 Bq/m³ in surface water during the period 1999-2006. Similar levels were detected in near-bottom water. The lowest ⁹⁰Sr concentrations were detected in the Kattegat, where only 2 Bq/m³ of ⁹⁰Sr was detected in 2006 in near-bottom water. The ⁹⁰Sr inventory in Baltic Sea amounted to about

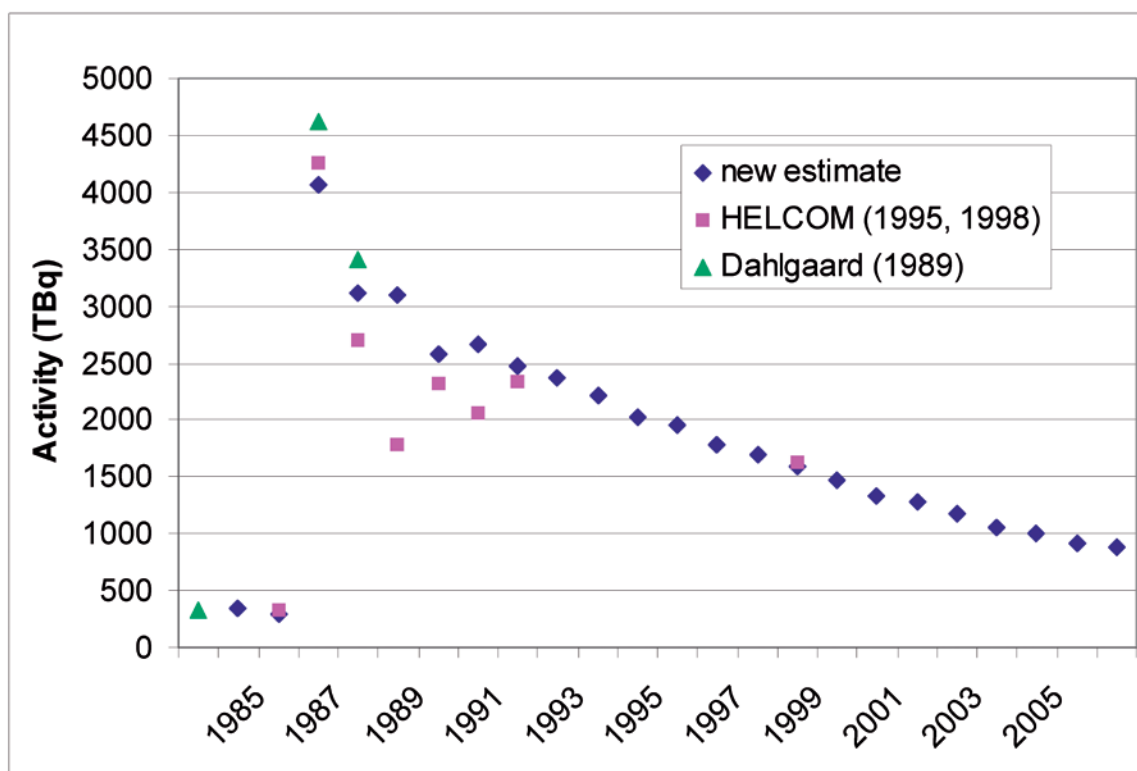


Figure 6:
¹³⁷Cs inventories in the Baltic seawater during 1984-2006.

200 TBq in 2006, corresponding to half of the ^{90}Sr inventory in 1985, the year before the Chernobyl accident. Concentrations of ^{90}Sr decrease slowly with time, and its behaviour in seawater is different from ^{137}Cs . ^{90}Sr is more soluble in water, and its effective half-life in seawater is longer than that of ^{137}Cs , at around 20 years during 1987-2006.

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

Concentrations of $^{239,240}\text{Pu}$ were very low, generally varying from 1 to 10 mBq/m³.

^{99}Tc

^{99}Tc was only analyzed for the Bornholm Sea, the Arkona Sea, the Kattegat and the Sound. Concentrations varied from 0.04 to 3 Bq/m³, with the highest levels detected in the Kattegat. The main source of ^{99}Tc is inflows of contaminated waters from the North Sea originating from the Sellafield nuclear reprocessing plant. ^{99}Tc concentrations have decreased during the study period, and levels in the Kattegat in 2006 were 6 times lower than in 1999.

^3H

The tritium concentration in surface water was only reported in 1999 and 2000, when it varied from 1,000 to 2,000 Bq/m³.

3A.6 Conclusions

^{137}Cs concentrations are the main indicator of the radioactive status of the waters of the Baltic Sea. The highest concentrations during 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/m³ will be reached between 2020 and 2030. First estimates for effective half-lives for different parts of the Baltic Sea have range between 9 and 15 years. An updated calculation of the total inventory of ^{137}Cs in the seawater of the Baltic Sea resulted in a figure of 870 TBq for 2006.

With regard to ^{137}Cs , the Baltic Sea is still one of the most contaminated areas of the World Ocean, even 20 years after the Chernobyl accident.

References

- Dahlgaard H. (1989):
Personal communication.
- HELCOM (1995a): Panteleev Y., H. Nies, T.K. Ikäheimonen & V. Tishkov. Radionuclides in seawater. In: Radioactivity in the Baltic Sea 1984-1991. Balt. Sea Environ. Proc. No. 61:59-68.
- HELCOM (1995b): Ballestra S., J. Herrmann & T.K. Ikäheimonen. Quality of data. In: Radioactivity in the Baltic Sea 1984-1991. Balt. Sea Environ. Proc. No. 61:30-58.
- HELCOM (1996): Third periodic assessment of the state of the marine environment of the Baltic Sea, 1989-1993. Balt. Sea Environ. Proc. No. 64B:28-128.
- HELCOM (2003): Mulsow, S., P. Kotilainen & T.K. Ikäheimonen. Radionuclides in seawater. In: Radioactivity in the Baltic Sea 1992-1998. Balt. Sea Environ. Proc. No. 85:49-54.
- Ilus E., K.L. Sjöblom, T.K. Ikäheimonen, R. Saxén & S. Klemola (1993): Monitoring of radionuclides in the Baltic Sea in 1989-1990. STUK-A103, Helsinki, 35 pp.
- Ilus E. (2007): The Chernobyl accident and the Baltic Sea. Boreal Environ Research 12:1-10.

3B Radionuclides In Sediments

Jukka Mattila and Erkki Ilus

STUK, Radiation and Nuclear Safety Authority, Finland

3B.1 Introduction

Sediments play a significant role, when monitoring the state of the environment and radioactivity in the Baltic Sea. Many radionuclides tend to bind to sediment particles, and are accumulated in bottom sediments. They can therefore be analysed quite easily from sediment samples. At a proper monitoring station and in favourable conditions, sediment layers have stored continuous deposition of suspended matter and radionuclides for long time periods, which allows us to study the history and total amounts of certain radionuclides in sediments.

In recent years several published articles have used radioactivity in sediments for monitoring and scientific purposes. Both natural and artificial radionuclides have been used, e.g. in estimations of sediment accumulation rates and in the dating of sediments (e.g. Jensen et al. 2003, Mattila et al. 2006). Radionuclides have also been used as tracers in intercomparisons of sediment sampling devices (Ilus et al. 2000). The concentrations and amounts of transuranic elements (e.g. plutonium activities) in the environment of the Baltic Sea have been summarized during the reporting period (Ikäheimonen 2003).

This report is a summary of the studies carried out as part of MORS-PRO on radioactivity in Baltic Sea sediments during the years 1999-2006, and is a sequel to the previous reports of Ilus et al. (1995 and 2003). Most recently the report "Long-lived radionuclides in the seabed of the Baltic Sea" summarized the inventories of ¹³⁷Cs, gave new knowledge about artificial radioactivity in sediments, and took steps towards estimations of the amounts of natural radioactivity (e.g. ⁴⁰K and ²²⁶Ra activities) in the surface sediments of the Baltic Sea (Ilus et al. 2007).

3B.2 Material and methods

Data has been based on the HELCOM database, into which the Contracting Parties

have submitted their annual results. Additional data from STUK, outside of the HELCOM/MORS monitoring programme, has also been used in the inventory calculations. The total number of sediment samples reported over the period 1999-2006 was 3,934. The sampling techniques used by different countries have been described previously (Ilus et al. 2003). The sediment types and the bottom morphologies have been described previously in many articles (Winterhalter 1972, Winterhalter et al. 1981, Ilus et al. 2003). The methods used in the radionuclide inventories are described in Salo et al. (1986), Ilus et al. (2003) and Ilus et al. (2007). In general, the sediment inventories were based on mean total amounts of radionuclides in sediments (Bq m⁻²) and the surface areas of different basins, taking into account the surface areas and the activities in soft and hard bottoms, respectively.

3B.3 Sources of artificial radioactivity

Over the period 1999-2006, no major events occurred that would have significantly increased artificial radioactivity in sediments in the Baltic Sea. In general, the main sources of artificial radionuclides were still the fallout from the Chernobyl accident in 1986 and the global fallout from atmospheric nuclear weapons testing in the 1950s and 1960s (Lüning and Ilus 2003). Europe's two nuclear reprocessing plants (Sellafield in the UK and La Hague in France) have had a minor influence on radioactivity in the southern Baltic Sea. Nuclear power plants and research reactors have only resulted in small local inputs to radionuclide concentrations in sediments in the vicinity of these facilities. Rivers are meanwhile still bringing radionuclides from their drainage areas into the sea.

3B.4 Results and discussion

The amounts of artificial radionuclides in bottom sediments did not increase noticeably during the years 1999-2006. Most of the artificial radioactivity was due to ¹³⁷Cs, which

Table 1:
Details of some naturally occurring and artificial radionuclides observed in the sediments of the Baltic Sea and discussed in the text. The half-lives ($T_{1/2}$) of these radionuclides generally vary from 370 hours to 14,000,000,000 years. The main sources of artificial radionuclides have been the fallout from the Chernobyl accident in 1986 and the global fallout from atmospheric nuclear weapons testing in the 1950s and 1960s. Rivers are still transporting fallout nuclides from their drainage areas to the sea. Other sources have only had minor impacts on radioactivity levels, either in the southern Baltic Sea (West European Nuclear Reprocessing Plants) or only very locally (nearby NPPs).

Nuclide	Origin	$T_{1/2}$
^{40}K	natural	300,000,000 yrs
^{210}Pb	natural	22.2 yrs
^{226}Ra	natural	1,600 yrs
^{232}Th	natural	14,000,000,000 yrs
^{60}Co	artificial	5.3 yrs
^{103}Ru	artificial	39.3 days
^{106}Ru	artificial	372.6 hrs
$^{110\text{m}}\text{Ag}$	artificial	249.8 days
^{125}Sb	artificial	2.8 yrs
^{90}Sr	artificial	28.5 yrs
^{90}Tc	artificial	210,000 yrs
^{137}Cs	artificial	30.2 yrs
^{237}Np	artificial	2,100,000 yrs
^{238}Pu	artificial	87.7 yrs
^{239}Pu	artificial	24,000 yrs
^{240}Pu	artificial	6,563 yrs
^{241}Pu	artificial	14.4 yrs
^{241}Am	artificial	432.7 yrs

was mostly found in the bottom sediments of the Gulf of Bothnia (especially in the Bothnian Sea) and in the eastern part of the Gulf of Finland (**Figure 1**). The amounts of ^{137}Cs have remained relatively unchanged during the reporting period (**Figure 2**). Artificial radionuclides from the Chernobyl and global fallouts are partly going to be buried deeper into the sediments on the accumulation bottoms. Nonetheless, part of the activity bound up in the sediments will still be transported from the erosion/transportation bottoms towards the accumulation bottoms. In spite of careful planning of the monitoring programmes, there are still a lot of factors

Table 2:
Total inventories (TBq) of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the sediments of the Baltic Sea have been estimated several times. The total amount of ^{137}Cs in the bottom sediments increased considerably after the Chernobyl fallout, and today is about 8-9 times higher than pre-Chernobyl levels in the beginning of the 1980s. The ^{90}Sr and $^{239,240}\text{Pu}$ inventories are only rough estimates because of the limited amount of data.

Year	^{90}Sr	^{137}Cs	$^{239,240}\text{Pu}$	Reference
Early 1980s	12	277	15	Salo et al. 1986
1990-1991	-	1200-1400	18**	Ilus et al. 1995
1998	-	1940-2210	-	Ilus et al. 2003
2000-2005	26*	2100-2400	15	Ilus et al. 2007

* = rough estimate
** = 1987-1988

affecting the results of the sediment studies and causing variation in the results and time trends (Ilus et al. 2000 and 2003, Mattila et al. 2006). For example, variations inside the sedimentation basins and around the monitoring stations can be large, due to the heterogeneity of soft sediment deposits. The differences in sampling techniques, as well as in analysis methods, may also increase the variability in the results. The bottom dynamics have an influence on the accumulation, transportation/erosion and hydraulic sorting of the sediments. As yet we evidently know relatively little about the variability in the sediments around our monitoring stations.

Most of the radioactivity in the sediments of the Baltic Sea originates from naturally

occurring radionuclides. In recent years, the activity concentrations of naturally occurring radionuclides with long half-lives, such as ^{40}K , ^{226}Ra and ^{232}Th (**Table 1**), have been reported into the database. In the surface sediments, most of the ^{40}K concentrations (in the 0-10 cm layer) varied between 200 and 1,400 Bq kg^{-1} d.w.; concentrations of ^{226}Ra (in the 0-10 cm layer) amounted to 10-100 Bq kg^{-1} d.w.; and ^{232}Th concentrations (in the 0-30 cm layer) varied between 10 and 50 Bq kg^{-1} d.w.. The activity levels of these nuclides depend on the type of the sediment. In the sediment baseline study, we tried to estimate the total amounts of ^{40}K and ^{226}Ra in the seabed of the Baltic Sea, but at this time the estimations had to be limited only to the uppermost layers of recently accumulated sediments. The total amounts in the 0-10 cm layer were estimated to be roughly 8,500 TBq for ^{40}K , and 420 TBq for ^{226}Ra . In the 0-20 cm layer, these amounts were over twice as high as in the uppermost 10 cm.

Although there are considerable amounts of artificial radioactivity due to the presence of long-lived radionuclides in the Baltic Sea sediments, they are not expected to cause harmful effects to marine life in the Baltic Sea. After the Chernobyl fallout, elevated concentrations were also detected of many other radionuclides, such as ^{60}Co , ^{103}Ru , ^{106}Ru , $^{110\text{m}}\text{Ag}$ and ^{125}Sb (**Table 1**), but because of their short half-lives, the activities of these radionuclides have decreased considerably, or they have vanished (Ilus et al. 2003).

In the sediment baseline study (Ilus et al. 2007), it was also possible to get new knowledge of ^{99}Tc and ^{237}Np (**Table 1**) in Baltic Sea sediments by using mass spectrometric analysis methods. In surface sediments, the concentrations of ^{99}Tc (in layer 0-10 cm) varied between 0.04 and 1.30 Bq kg^{-1} d.w., whereas the activities of ^{237}Np (in layer 0-20 cm) varied between 0.2 and 6.5 mBq kg^{-1} d.w.. The total amount of ^{237}Np in the sediments of the Baltic Sea was estimated to be about 0.02 TBq (Ilus et al. 2007).

Over the period 1999-2006, the activities of ^{137}Cs remained relatively unchanged at different monitoring stations in the Baltic Sea, although there were considerable differences between the stations (**Figure 2**). After the Chernobyl accident, the activity of ^{137}Cs has been intensively studied, since there was a lot of caesium activity in the fallout, it has a long half-life and high K_d values (Bq kg^{-1} in

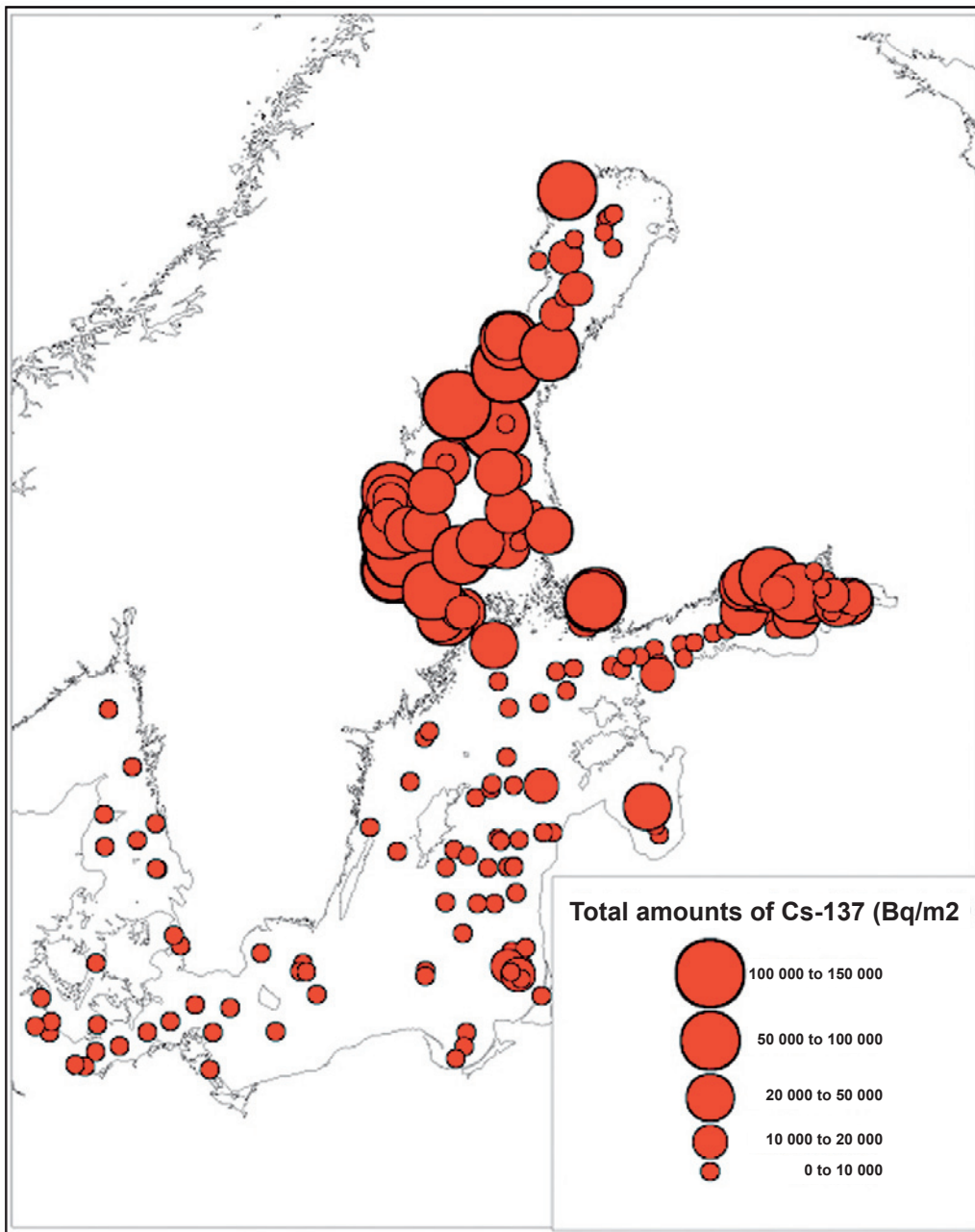


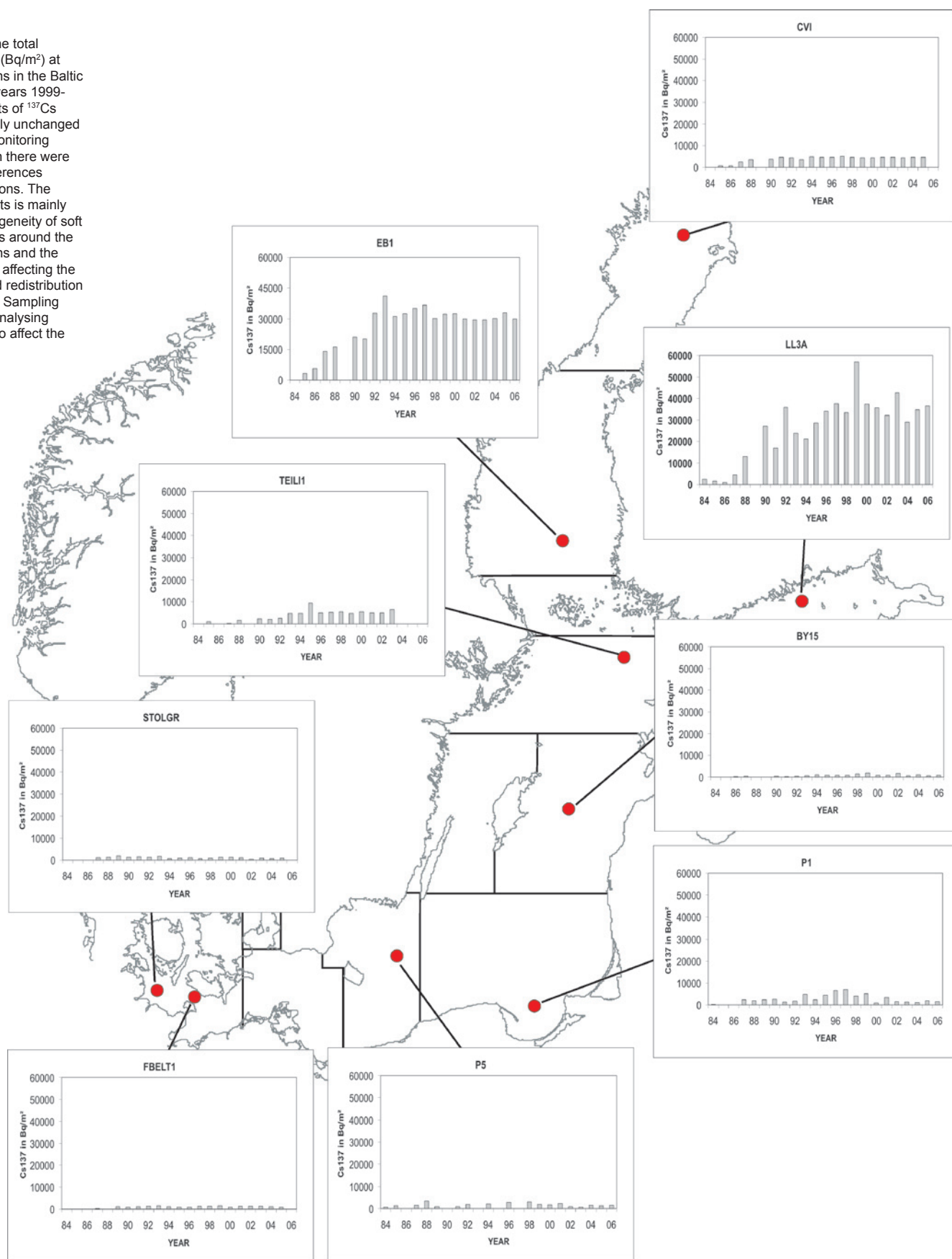
Figure 1: Total amounts of ^{137}Cs (Bq/m^2) at different sampling stations in the Baltic Sea in the late 1990s and the beginning of the 2000s (Illus et al. 2007). Most of ^{137}Cs is in the sediments of the Bothnian Sea and in the eastern Gulf of Finland, due the high deposition of the Chernobyl fallout 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 (Illus 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% (Illus et al. 2007).

Additional data on transuranic elements was also compiled during the study period. The reported values of $^{239,240}\text{Pu}$ 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 $^{239,240}\text{Pu}$ was estimated to be about 15.3 TBq (**Table 2**). Most of the plutonium originated from the

Figure 2:
 Time trends for the total amounts of ^{137}Cs (Bq/m^2) at monitoring stations in the Baltic Sea. During the years 1999-2006, the amounts of ^{137}Cs remained relatively unchanged at the different monitoring stations, although there were considerable differences between the stations. The variability in results is mainly due to the heterogeneity of soft sediment deposits around the monitoring stations and the bottom dynamics affecting the accumulation and redistribution of the sediments. Sampling techniques and analysing methods may also affect the results.



global fallout, but small amounts of ^{238}Pu and ^{241}Pu in the fallout of the Chernobyl accident could be noticed 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}Pu (Ikäheimonen 2003). In the years 1999-2006, the reported ^{241}Am concentrations ranged from 0.08 to 4.80 Bq kg^{-1} d.w..

The activities of ^{90}Sr mostly originate from global fallout. It has been estimated that the input to the Baltic Sea of ^{90}Sr activity from the Chernobyl fallout only accounts for 13% of the total ^{90}Sr activity in the Baltic Sea (Lüning and Ilus 2003). Because of the small share in the Chernobyl fallout and the costly analytical methods, the focus on ^{90}Sr has been reduced, and relatively little sediment data is available. Over the period 1999-2006, reported ^{90}Sr concentrations ranged from 0.1 to 73.8 Bq kg^{-1} d.w.. A rough estimate for the total amount of ^{90}Sr in the sediments of the Baltic Sea was about 26 TBq (Ilus et al. 2007).

3B.5 Recommendations and future work

It will continue to be essential to monitor radioactive substances in sediments in the Baltic Sea in the future. The continuous monitoring work and the compiling of time trends for radioactive substances provide a basis for understanding the state of the Baltic Sea environment and the radioactivity it contains. However, there are still several significant gaps in our knowledge (Ilus et al. 2003, Ilus et al. 2007). In the near future, we should attempt to fill these gaps and increase our knowledge of the activities of e.g. ^{90}Sr , ^{241}Am and natural radionuclides such as ^{210}Pb , in sediments in the Baltic Sea. Plans for the future should also include assessments of the impacts of ionising radiation on the Baltic marine environment.

References

Ikäheimonen, T.K. (2003): Determination of transuranic elements, their behaviour and sources in the aquatic environment (thesis). Report STUK-A194, Helsinki, 82 p. + Annexes.

Ilus E., L. Niemistö & R. Bojanowski (1995): Radionuclides in sediment and suspended particulate matter. In: HELCOM (1995): Radioactivity in the Baltic Sea 1984-1991. Balt. Sea Environ. Proc. No. 61:69-91.

Ilus E., T. Ilus, T.K. Ikäheimonen, L. Niemistö, J. Herrmann, M. Suplinska, Y. Panteleev, L. Ivanova, Z.G. Gritchenko & G. Neumann (2000): In: HELCOM (2000): Intercomparison of sediment sampling devices using artificial radionuclides in Baltic Sea sediments.- The MOSSIE Report. Balt. Sea Environ. Proc. No. 80. 76 pp.

Ilus E., M. Suplinska & J. Mattila (2003): Radionuclides in sediments. In: HELCOM (2003): Radioactivity in the Baltic Sea 1992-2006. Balt. Sea Environ. Proc. No. 85:55-75.

Ilus E., J. Mattila, S.P. Nielsen, E. Jakobson, J. Herrmann, V. Graveris, B. Vilimaite-Silobriene, M. Suplinska, A. Stepanov & M. Lüning (2007): In: HELCOM (2007): Long-lived radionuclides in the seabed of the Baltic Sea. Balt. Sea Environ. Proc. No. 110:1-41.

Jensen A., B. Larsen, P. Jonsson & M. Perttilä (2003): In: Perttilä, M. (ed.): Contaminants in the Baltic Sea sediments, Results of the 1993 ICES/HELCOM Sediment Baseline Study. Report Series of the Finnish Institute of Marine Research, MERI 50:58-64.

Lüning M. & E. Ilus (2003): Sources of radioactivity in the Baltic Sea. In: HELCOM (2003): Radioactivity in the Baltic Sea 1992-2006. Balt. Sea Environ. Proc. No. 85:5-15,.

Mattila, J., H. Kankaanpää & E. Ilus (2006): Estimation of recent sediment accumulation rates in the Baltic Sea using artificial radionuclides ^{137}Cs and $^{239,240}\text{Pu}$ as time markers. Boreal Env. Res. 11:95-107.

Salo, A., K. Tuomainen & A. Voipio (1986): Inventories of some long-lived radionuclides in the Baltic Sea. The Science of the Total Environment, 54:247-260, Amsterdam.

Winterhalter, B. (1972): On the geology of the Bothnian Sea, an epeiric sea that has undergone Pleistocene glaciation. Geol. Survey Finland, Bull. 258:1-66, Helsinki.

Winterhalter, B., T. Floden, H. Ignatius, S. Axberg & L. Niemistö (1981): Geology of the Baltic Sea. In Voipio, A. (ed.): The Baltic Sea. Elsevier Oceanography Series, 30:69-100, Amsterdam.

3C Radionuclides in Biota

Günter Kanisch

vTI, Johann Heinrich von Thünen-Institute, Germany

3C.1 Introduction

The biota of the Baltic Sea received the most significant contribution to their radionuclide levels following the Chernobyl accident in 1986, predominantly in the form of ^{137}Cs and ^{134}Cs . The ratio $^{134}\text{Cs}/^{137}\text{Cs}$ in Baltic biota agreed very well with that of the Chernobyl fallout. High trophic level species, including predators such as cod and pike, showed the highest ^{137}Cs levels, but there was some delay in reaching their maximum values after 1986, when compared to trends in seawater. In the long-term, ^{137}Cs time trends in biota closely follow the trends in seawater.

Levels of radionuclides in marine biota are linked to the corresponding levels in seawater and sediments, via accumulation through food chains. The complexity of food chains increases with the trophic level of the species considered. Fish, the biota type in the Baltic Sea most important for human consumption, accumulate most of their radionuclides from food, not from water. Within the existing box model (see Chapter 4), which is also used for describing activity concentrations in biota, however, the simpler concentration factor concept is applied. This relates the biota activity concentrations directly to those of seawater, which is the simplest approximation to a food chain sub-model. Concentration factors specific to the different model boxes are derived from measured data. Results of model predictions based on estimated concentration factors are included in the following summary where appropriate.

In this report, the evaluation of biota data is restricted to fish and the bladder wrack *Fucus vesiculosus*, which is a well established biological indicator for radionuclides.

3C.2 Material

The number of biota samples collected annually by the MORS-PRO group decreased slightly through the sampling period 1999-2006 from about 120 initially to about 90

ultimately. Over the whole period the numbers of samples collected were 615 for fish, 126 for *Fucus vesiculosus*, and 102 for benthic animals.

Fish flesh samples were measured from the marine species: herring (138), cod (166) and whiting (3), of the round fish; and flounder (68) and plaice (24), of the flat fish.

Fish flesh sample numbers were measured from the freshwater species: Pike (48) and perch (6).

Additionally 45 herring samples were analysed examining their “edible parts” (fish without head and entrails).

The main artificial radionuclides measured in biota during the assessment period were: ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am in fish, and ^{99}Tc in *Fucus vesiculosus*.

3C.3 Use of a box model for comparison with measured data

Trends for the activity values of ^{137}Cs and ^{90}Sr may be also simulated by using the box model described briefly in Chapter 4. Because ^{137}Cs showed a sharp peak in seawater concentrations in 1986 (HELCOM 1995), a small sub-model for fish is included in the model, which allows the fish to take up and excrete the activity with an appropriate biological half-life (250 days), thereby transforming the sharp seawater peak into a smooth increase in fish and a slightly delayed decrease after the peak.

Concentration factors updated from the MORS-PRO database are required for this purpose. The results of the consequent estimations are included in the next section.

3C.4 Results and discussion

3C.4.1 Concentration factors

Because of area-dependent significant

Type of Biota	71 Kattegat	75 Belt Sea	81 Baltic Sea West	83 Baltic Sea East	85 Bothnian Sea	87 Bothnian Bay	89 Gulf of Finland	91 Gulf of Riga
¹³⁷Cs:								
<i>Marine fish:</i>								
round fish, flesh	35 ± 8	125 ± 36	164 ± 18	164 ± 20	188 ± 26	261 ± 28	194 ± 67	
round fish, edible parts				153 ± 32	154 ± 18	230 ± 29	206 ± 28	211 ± 8
flat fish, flesh		63 ± 33	215 ± 6	129 ± 15				160 ± 48
<i>Freshwater fish:</i>								
Pike, flesh <i>Esox lucius</i>				310 ± 50	564 ± 220	555 ± 111	602 ± 142	
Perch, flesh <i>Perca fluviatilis</i>								285 ± 76
<i>Aquatic Plants:</i>								
Brown algae, <i>Fucus vesiculosus</i>	188 ± 39	215 ± 26	453 ± 145	522 ± 73	622 ± 100		1247 ± 424	390 ± 156
⁹⁰Sr:								
<i>Marine fish:</i>								
round fish, flesh		0.23 ± 0.06	0.39 ± 0.13	0.34 ± 0.15				
round fish, edible parts				3.0 ± 0.5	3.1 ± 0.7	6.4 ± 1.6	2.7 ± 0.5	4.2 (1 value)
flat fish, flesh		1.1 ± 1.2		1.2 ± 1.1				3.4 ± 2.4
<i>Freshwater fish:</i>								
Pike, flesh <i>Esox lucius</i>				1.7 ± 0.5	1.5 ± 0.4	4.0 ± 1.5		
Perch, flesh <i>Perca fluviatilis</i>								3.1 ± 1.4
<i>Aquatic Plants:</i>								
Brown algae, <i>Fucus vesiculosus</i>		206 ± 165		162 ± 94	821 ± 174		858 ± 96	664 ± 163

Table 1: ¹³⁷Cs and ⁹⁰Sr concentration factors and standard uncertainties of fish (in l kg⁻¹ wet) and *Fucus vesiculosus* (in l kg⁻¹ dry) estimated in different model-related Baltic Sea areas for the years 1988/1990 - 2006. Marine fish have been grouped as follows: **Round fish:** herring, cod and whiting; **Flat fish:** flounder, plaice and dab.

freshwater contributions to the Baltic seawater, the marine biota concentration factors (CF) as recommended by the International Atomic Energy Agency IAEA (1985 and 2004) cannot be used. As the earlier determinations of concentration factors (HELCOM 1995) suffered partly from non-equilibrium states of the Chernobyl-derived Cs in biota, new estimates of the CFs were obtained from biota and seawater data from the years up to 2006, starting from 1990 (¹³⁷Cs) and 1988 (⁹⁰Sr). For each year, nuclide and water box, a CF value was calculated using the ratio between the annual means of the biota specific activity and surface seawater activity concentrations. The various time series of CF values were treated statistically including robust methods ensuring reduced influence from outliers. **Table 1** shows the results of estimated concentration factors of fish and *Fucus vesiculosus*.

It is clear that for marine fish species the ¹³⁷Cs CF values increase from western Baltic Sea areas to eastern/northern areas, which is explained by the corresponding increase of freshwater contributions to the seawater. The values from **Table 1** have been used for comparing the modelled and measured ¹³⁷Cs and ⁹⁰Sr activities of fish and *Fucus vesiculosus*, respectively. See Chapter 4 (modelling/dose) for these results.

3C.4.2 Activity concentrations

The following tables show extreme values of activity concentrations for the representative beta/gamma-emitting radionuclides, ¹³⁷Cs and ⁹⁰Sr, and the alpha emitters, ^{239,240}Pu and ²⁴¹Am, as observed during the period 1999-2006.

The values obtained for the dominant fish species are shown in **Table 2**. One outstanding ¹³⁷Cs value of 41 Bq kg⁻¹ wet weight was observed for cod; it belonged to a single large specimen (collected in 2000, Southern Baltic Proper); the next highest value was 23 Bq kg⁻¹ wet weight. The latter value then represents the maximum value for the marine fish species. Among the freshwater fish, represented by pike and perch, pike showed even larger ¹³⁷Cs values due to their higher concentration factors. While perch were collected in the Gulf of Riga, pike were collected in waters adjacent to the Finnish coast. The larger ¹³⁷Cs values for pike were observed near the coast of the Bothnian Sea.

For ⁹⁰Sr, a radionuclide mainly derived from nuclear weapons testing fallout and related runoff from rivers, values for specific activity are more than two orders of magnitude lower than those of ¹³⁷Cs. The ⁹⁰Sr activities in the flesh of flat fish species (flounder, plaice) exhibit slightly larger values than in round fish

Table 2:
Extreme activity values of artificial radionuclides in fish, in Bq kg⁻¹ wet weight (1999-2006)

Species	¹³⁷ Cs		⁹⁰ Sr		^{239,240} Pu		²⁴¹ Am	
	min.	max.	min.	max.	min.	max.	min.	max.
Herring	0.5	21	0.0028	0.0049	0.000019	0.0000912	0.0000137	0.000143
Cod	0.61	41	0.0015	0.0067	0.000021	0.000021	0.0000186	0.0000186
Whiting	6.5	11	0.0013	0.0020				
Mackerel	4.5	4.5						
Flounder	1.9	11	0.01	0.047	0.000044	0.000045	0.000157	0.000157
Plaice	0.15	14	0.016	0.016	0.0000201	0.000026	0.0000218	0.000196
Pike	9.2	73	0.0098	0.33				
Perch	0.35	12	0.016	0.05				
Herring, (edible parts)	4.6	19	0.011	0.103				

Table 3:
Extreme activity values of artificial radionuclides in *Fucus vesiculosus*, in Bq kg⁻¹ dry weight (1999-2006)

¹³⁷ Cs		⁹⁰ Sr		^{239,240} Pu		²⁴¹ Am		⁹⁹ Tc	
min.	max.	min.	max.	min.	max.	min.	max.	min.	max.
1.3	66	4	10	0.023	0.12	0.0020	0.0069	8.2	168

Table 4:
Activity values of the naturally occurring alpha emitting radionuclide ²¹⁰Po in Baltic Sea fish, in Bq kg⁻¹ wet weight (1990-2006)

Species (flesh)	²¹⁰ Po		
	min.	max.	Mean
Herring	0.19	8.5	1.3
Cod	0.043	1.5	0.41
Flounder	0.26	3.4	1.1

species (herring, cod, whiting and mackerel), because the analysed samples of the former may contain more small parts of bones which have accumulated bone-seeking ⁹⁰Sr much more than fish muscle. The ⁹⁰Sr values of herring analysed as “edible parts” (fish without head and entrails, i.e. containing larger fractions of bones) are significantly larger than those of herring flesh alone. Samples of the flesh of flat fish also suffer from increased bone fractions leading to slightly larger ⁹⁰Sr activities.

The radionuclides ^{239,240}Pu and ²⁴¹Am exhibit maximum activity values which are even one to two orders of magnitude lower than those of ⁹⁰Sr; so, they do not have any significance with respect to the doses caused by fish consumption.

Table 3 shows the corresponding ranges of radionuclide activity concentrations in bladder wrack of the species *Fucus vesiculosus*. Practically all of the radionuclides shown are significantly accumulated from seawater. Besides ¹³⁷Cs and ⁹⁰Sr, the alpha-emitting radionuclides Pu and Am can also be safely detected. The dominant sources of Pu and Am isotopes are global fallout as well as discharges from nuclear reprocessing facilities; additionally, ²⁴¹Am is slowly in-growing from the radioactive decay of the beta-emitter ²⁴¹Pu, which has originated from the same sources. The very long-lived beta-emitter ⁹⁹Tc, which originates from nuclear reprocessing discharges and has been

measured in samples from Danish waters, is also accumulated significantly.

The radionuclide which is the most important with respect to doses caused by fish consumption is the alpha-emitting ²¹⁰Po, which belongs to the naturally-occurring radionuclides. The activity values of 104 samples of fish flesh collected in the Kattegat and the Bornholm Sea since 1990 have been summarised in **Table 4**. Maximum values, between 1.5 and 8.5 Bq kg⁻¹ wet weight, were slightly different between the three species considered in these measurements. The average of the three mean values is about 0.94 Bq kg⁻¹ wet. In a former evaluation of ²¹⁰Po in fish from Danish waters including the North Sea (Dahlgard 1996) average values of 0.35, 0.65 and 0.96 Bq kg⁻¹ wet weight were observed for cod, herring and plaice fillets, respectively.

3C.4.3 Activity ratios

Some of the biota species serve as good biological indicators of activity concentration ratios of alpha emitting radionuclides, i.e. the ratios ²³⁸Pu/^{239,240}Pu and ²⁴¹Am/^{239,240}Pu. Measured results of these ratios may be compared with values known to be characteristic for their sources, such as global fallout or discharges from nuclear reprocessing. Activity ratios have been measured within the MORS-PRO monitoring project in samples of bladder wrack (*Fucus vesiculosus*), blue mussel (*Mytilus edulis*) and small sprat (*Clupea harengus*). These organisms were analysed as whole body samples, so the blue mussel samples (small organisms) mainly consisted of shells. Some of the measured ²³⁸Pu activity values were not significant, i.e. their uncertainties were too large and thereby reported as “less-than” values, which leads to “less-than” values

Species	Model box	Time range	Mean \pm 1s SD	n	ns	min value	max value
ratio $^{238}\text{Pu} / ^{239,240}\text{Pu}$							
Sprat	75	1991 - 2006	0.041 \pm 0.018	9	6	0.030	< 0.14
	(81, 83)	1991 - 2006	0.038 \pm 0.008	10	4	0.031	< 0.13
Blue mussel	75	1991 - 1997	0.065 \pm 0.024	10	7	0.056	< 0.17
	83	1989 - 2005	0.044 \pm 0.016	12	2	0.027	0.078
Bladder wrack	75	1988 - 1998	0.043 \pm 0.008	9	0	0.034	0.057
	83	1995 - 2006	0.034 \pm 0.007	16	0	0.019	0.044
ratio $^{241}\text{Am} / ^{239,240}\text{Pu}$							
Sprat	75	1995 - 2005	0.47 \pm 0.11	6	0	0.36	0.63
	(81, 83)	1995 - 2005	0.41 \pm 0.07	9	0	0.34	0.53
Blue mussel	75	1995 - 1997	0.25 \pm 0.09	8	0	0.15	0.35
	83	1995 - 2005	0.28 \pm 0.07	10	0	0.15	0.37
Bladder wrack	75	1995 - 1998	0.24 \pm 0.14	6	0	0.12	0.50
	83	1995 - 2006	0.12 \pm 0.09	14	0	0.04	0.33

Table 5: Results from a statistical evaluation of radionuclide activity ratios measured in samples of sprat, blue mussel and *Fucus vesiculosus*; n and ns are the numbers of significant and non-significant values. All samples were measured as whole body samples.

for the activity ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$. Data sets containing “less-thans” are called “censored”. Their statistical evaluation requires special statistical procedures (Helsel and Cohn, 1988; Lee and Helsel 2005). The former method by Helsel and Cohn was used for this purpose, programmed in FORTRAN 90. **Table 5** shows the results of this evaluation.

Average values found for the ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$ are in the range of 0.034 to 0.065. Earlier (1988-1990), in *Fucus vesiculosus* from the Belt Sea a few values around 0.054 had been found (HELCOM 1995). Global nuclear weapons fallout can be assumed to be the major contribution to this ratio, for which values around 0.034 (as of 1971, i.e. 0.026 decay-corrected to 2006) are typical for this hemisphere (c.f. Pentreath 1988). Discharges from the fuel reprocessing plants of Sellafield and La Hague lead to activity ratios between 0.1 and 0.5 in North Sea water (BMU 2006). Small contributions from Sellafield and La Hague to western Baltic Sea biota are to be expected; this may be indicated by the measured data showing slightly larger averages in Box 75 (the Belt Sea), closer to the North Sea, compared to those from the Box 83 (Baltic Sea east).

Regarding these two major sources for plutonium and americium, a similar argumentation for the ratio $^{241}\text{Am}/^{239,240}\text{Pu}$ is not as easy, because the two involved radionuclides, ^{241}Am and $^{239,240}\text{Pu}$, are expected to have different CF values for accumulation from seawater. While for fish the same CF of 100 for both, Pu and Am, is recommended by the IAEA (2004), the corresponding recommended CF values for macroalgae are 4×10^3 and 8×10^3 for Pu and Am, respectively. Additionally, the geochemical behaviour of Pu and Am in the sea is different.

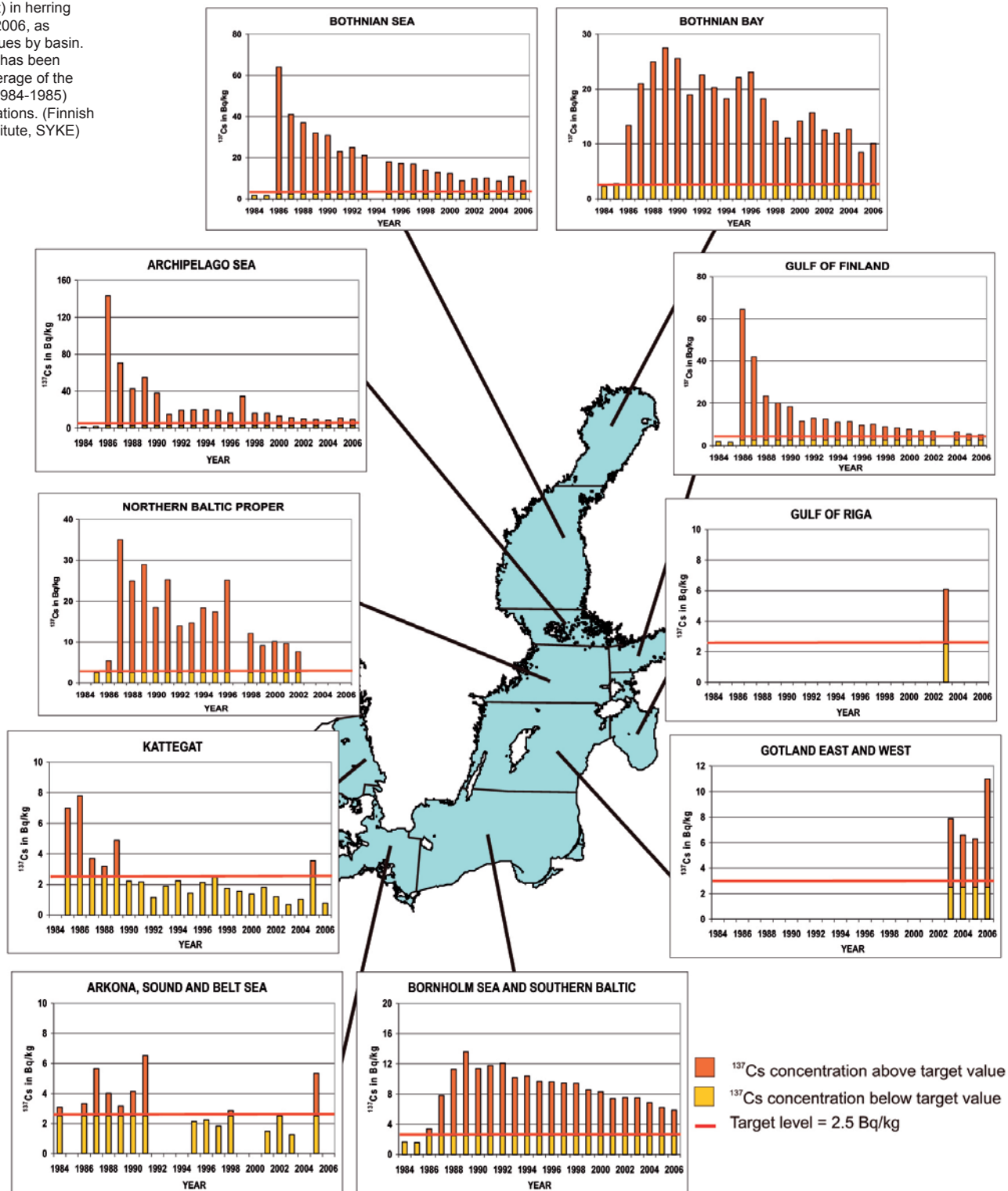
3C.4.4 Trends in activity concentrations

^{137}Cs , the most important radionuclide in fish since Chernobyl, is continuing to show generally slowly decreasing trends, as demonstrated by **Figure 1**, which shows such trends for **herring muscle** (*Clupea harengus*). In the western parts of the Baltic Sea, i.e. the Kattegat, the Sound, the Belt Sea and the Arkona Sea, the values already show levels slightly below the target value of 2.5 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.

The diagrams show comparisons between calculated (red lines and squares) and observed (black circles) concentrations of ^{137}Cs or ^{90}Sr in Baltic Sea sub-regions, i.e. the model boxes. Figure 1 in Chapter 4 shows the geographical locations of these boxes. Each Figure consists of a collection of model box related graphs. The observed concentrations are annual average values of data collected by MORS-PRO. The vertical bars represent the variability of the observed concentrations within a single year. The modeled curves are included here in order to demonstrate agreement or disagreement of the model with measured data. The box- and species-specific concentration factors (CF, in l kg⁻¹) are shown in the title lines of the individual graphs, which also specify the values used for the biological half-lives in the model (T-bio, in days).

Figure 2 shows measured and modeled ^{137}Cs activity concentrations in muscle tissue from (**marine**) **round fish** (cod, herring and whiting). A slight increase in the values is observed when moving from the Kattegat (box 71) over the Belt Sea (box 75) to the southeast boxes 81 and 83 (Baltic Sea west and east). Furthermore, in the Bothnian Sea and Bothnian Bay, boxes 85 and 87

Figure 1:
 ^{137}Cs activity concentrations (in Bq/kg wet weight) in herring muscle in 1984-2006, as annual mean values by basin. The target value has been calculated as average of the pre-Chernobyl (1984-1985) activity concentrations. (Finnish Environment Institute, SYKE)



respectively, slightly higher values are observed. At the end of this assessment period, means of measured values between 1 and 10 Bq kg⁻¹ wet weight are observed. For samples of the edible parts of herring (head without entrails), which were analysed mainly from basins with box numbers from 83 upwards, slightly lower ¹³⁷Cs activity concentrations were found compared to the values for combined round fish muscle shown in Figure 2.

Figure 3 shows the ¹³⁷Cs time series for the **flat fish** group, consisting of flounder (*Platichthys flesus*), plaice (*Pleuronectes platessa*) and dab (*Limanda limanda*), in the western and southern Baltic Sea areas. Samples of fillets/flesh were used for these measurements. At the end of the assessment period, the values were below about 8 Bq kg⁻¹ wet weight.

In the coastal areas of Baltic Sea basins around Finland (boxes 83, 85, 87 and 89), **freshwater fish**, i.e. pike (*Esox lucius*), have also been used for monitoring measurements since 1985. Due to larger freshwater contents in this seawater, their concentration factors are larger, resulting in slightly larger ¹³⁷Cs values. **Figure 4** shows the corresponding comparison of measured and modeled ¹³⁷Cs in fillets of pike; the trends are clearly decreasing. By the end of the assessment period the annual means approached values between about 10 and 25 Bq kg⁻¹ wet weight.

The corresponding measurement/model comparison for ⁹⁰Sr in pike muscle tissue is illustrated in **Figure 5**. The levels of activity concentrations in pike, which in the assessment period are about 200-fold smaller than those of ¹³⁷Cs, are slowly decreasing. By the end of the assessment period, levels of about 0.02-0.03 Bq kg⁻¹ wet weight were measured. As ⁹⁰Sr is a strongly bone-seeking radionuclide, its concentrations in fillets are rather small. The presence of even small amounts of bone in the fillet samples will result in increased observed concentration values. This explains why fluctuations are observed in some cases. As suggested by the trends of the model curves in Figure 5, the impact of Chernobyl fallout was much less than for ¹³⁷Cs.

Bladder wrack, *Fucus vesiculosus*, accumulates radionuclides (IAEA, 2004) to such an extent that it is a good measurable indicator, especially for gamma-emitting

radionuclides. *Fucus* can be collected over the whole Baltic Sea area. ¹³⁷Cs in *Fucus* has been monitored since 1984 in practically all considered Baltic basins, from the Kattegat (71) to the Gulf of Finland (89); with the exception of the Bothnian Bay, where no samples were taken.

The corresponding measurement/model comparison for ¹³⁷Cs in *Fucus vesiculosus* is shown in **Figure 6** for seven Baltic Sea basins. The trends within the period of this assessment in the various basins show a slow decrease. By the end of this period, activity concentrations in *Fucus* approached values (in Bq kg⁻¹ dry weight) of 5 (Kattegat), 7 (Belt Sea) and 20 in the eastern and northern basins (Baltic Sea east and west, Bothnian Bay, Gulfs of Riga and Finland).

The modeled curve for the Gulf of Finland (box 89) shows in 1986-2006 some deviation from measured values during this time, initially (until 1990) underestimating the measured values, while after about 1998 producing overestimates. This discrepancy will have to be considered for future modeling work.

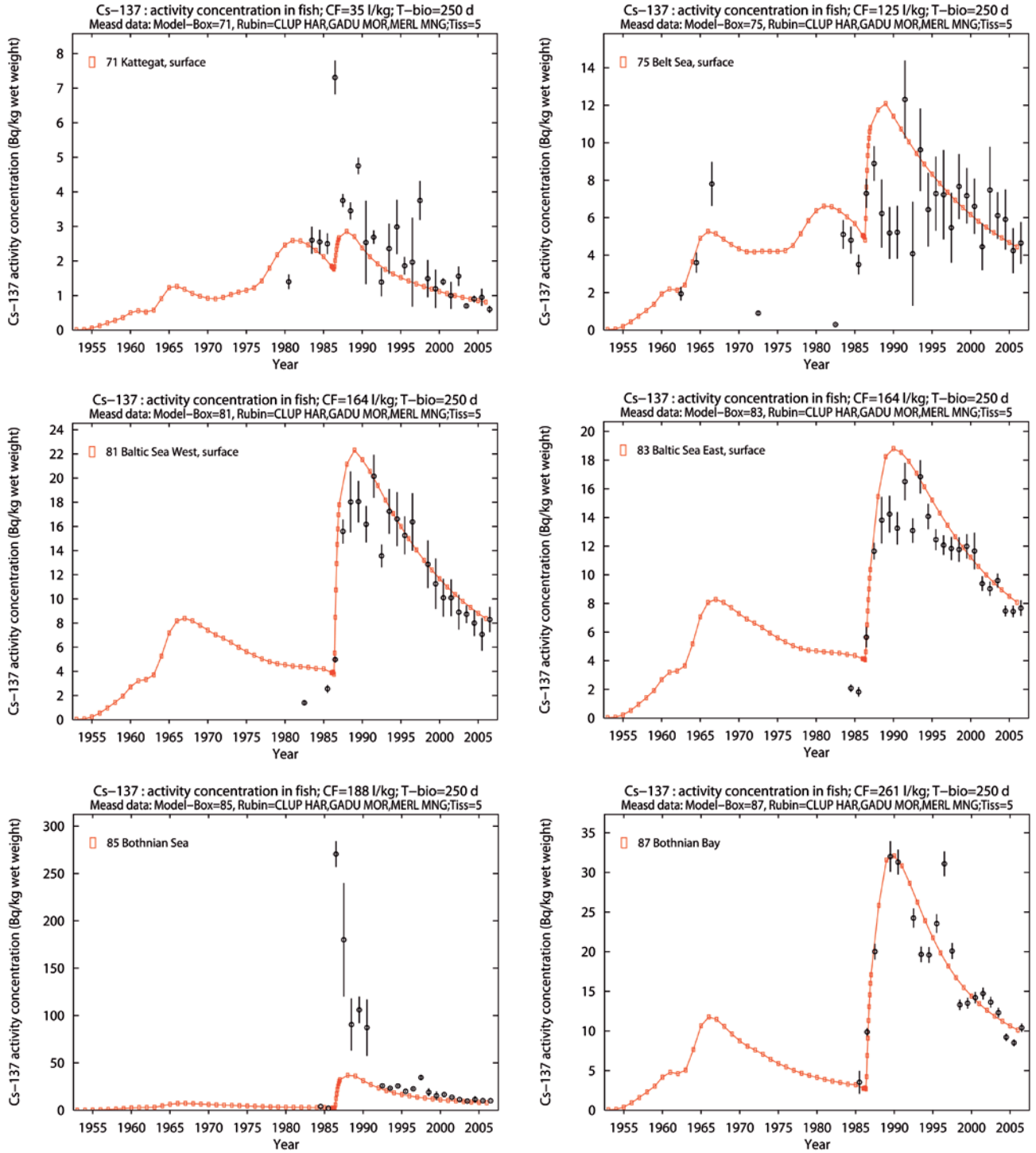
3C.4.5 Conclusions

Within the current assessment period, Chernobyl-derived ¹³⁷Cs continued to be the most dominant man-made radionuclide in Baltic Sea fish regarding activity concentrations. By the end of the period, mean values of 1-10 Bq kg⁻¹ wet weight were found in (marine) round fish (cod, herring, whiting) in various Baltic Sea basins, while freshwater pike sampled in Finnish coastal waters, due to its higher concentration factors, showed values of 10-25 Bq kg⁻¹ wet weight. In (marine) flat fish (plaice, flounder, dab), slightly lower mean values were found than in (marine) round fish.

Baltic Sea basin specific concentration factors of ¹³⁷Cs and ⁹⁰Sr have been derived for the three above-mentioned groups of fish and for bladder wrack, *Fucus vesiculosus*, with the latter covering practically the whole of the Baltic Sea. The findings were based on seawater and biota data compiled by the MORS-PRO Group over the years 1988/1990-2006.

Activity ratios ²³⁸Pu/^{239,240}Pu and ²⁴¹Am/^{239,240}Pu were evaluated in samples of sprat, blue mussel and *Fucus vesiculosus*, collected in the Belt Sea and the Southern Baltic Proper.

Figure 2: Comparison between calculated and observed activity concentrations of ¹³⁷Cs in muscle tissue of (marine) round fish (cod, herring and whiting) for sub-regions 71, 75, 81, 83, 85 and 87.



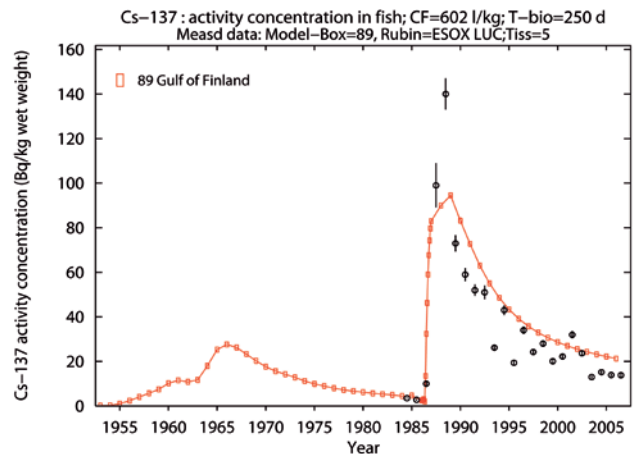
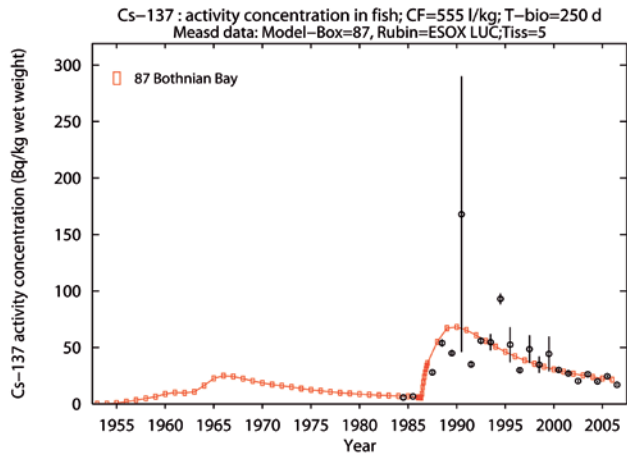
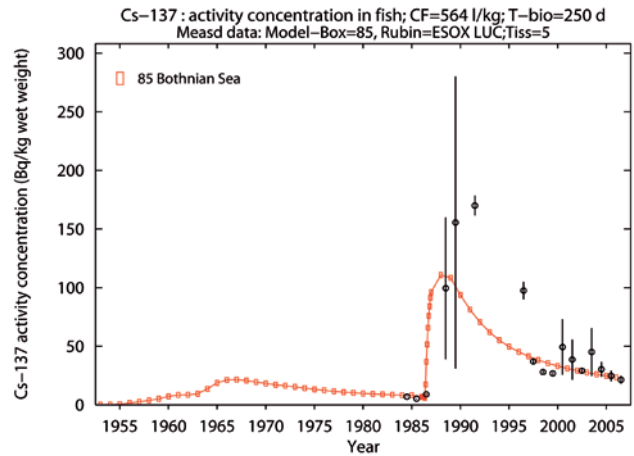
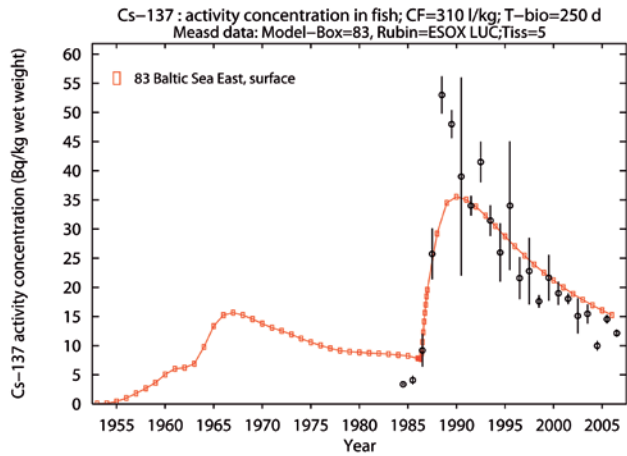
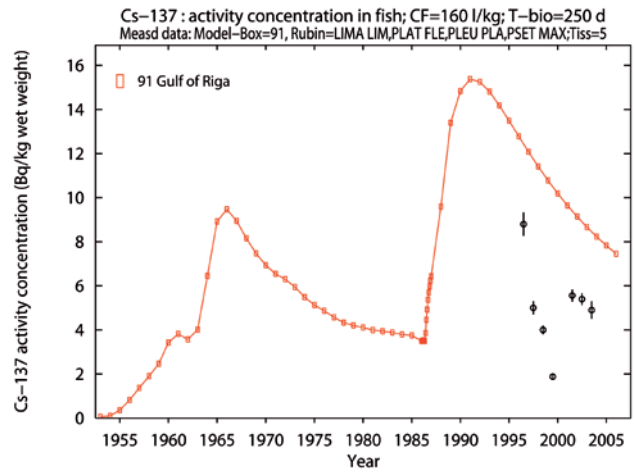
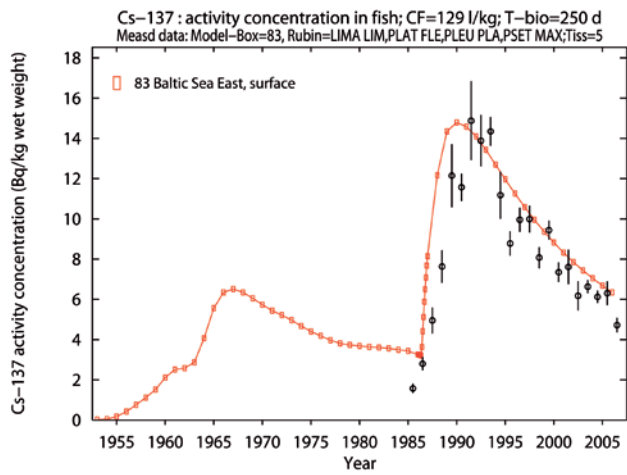
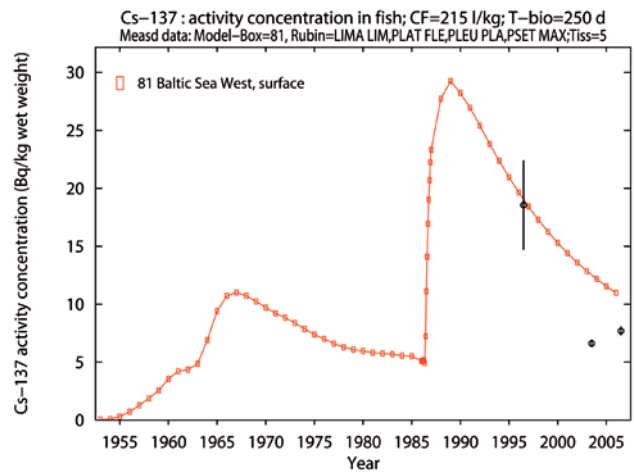
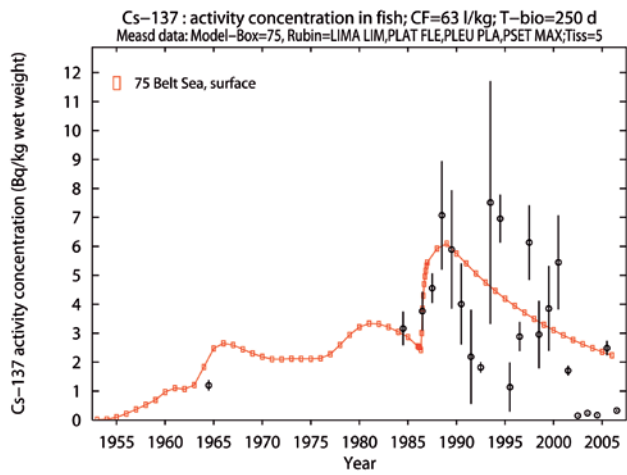
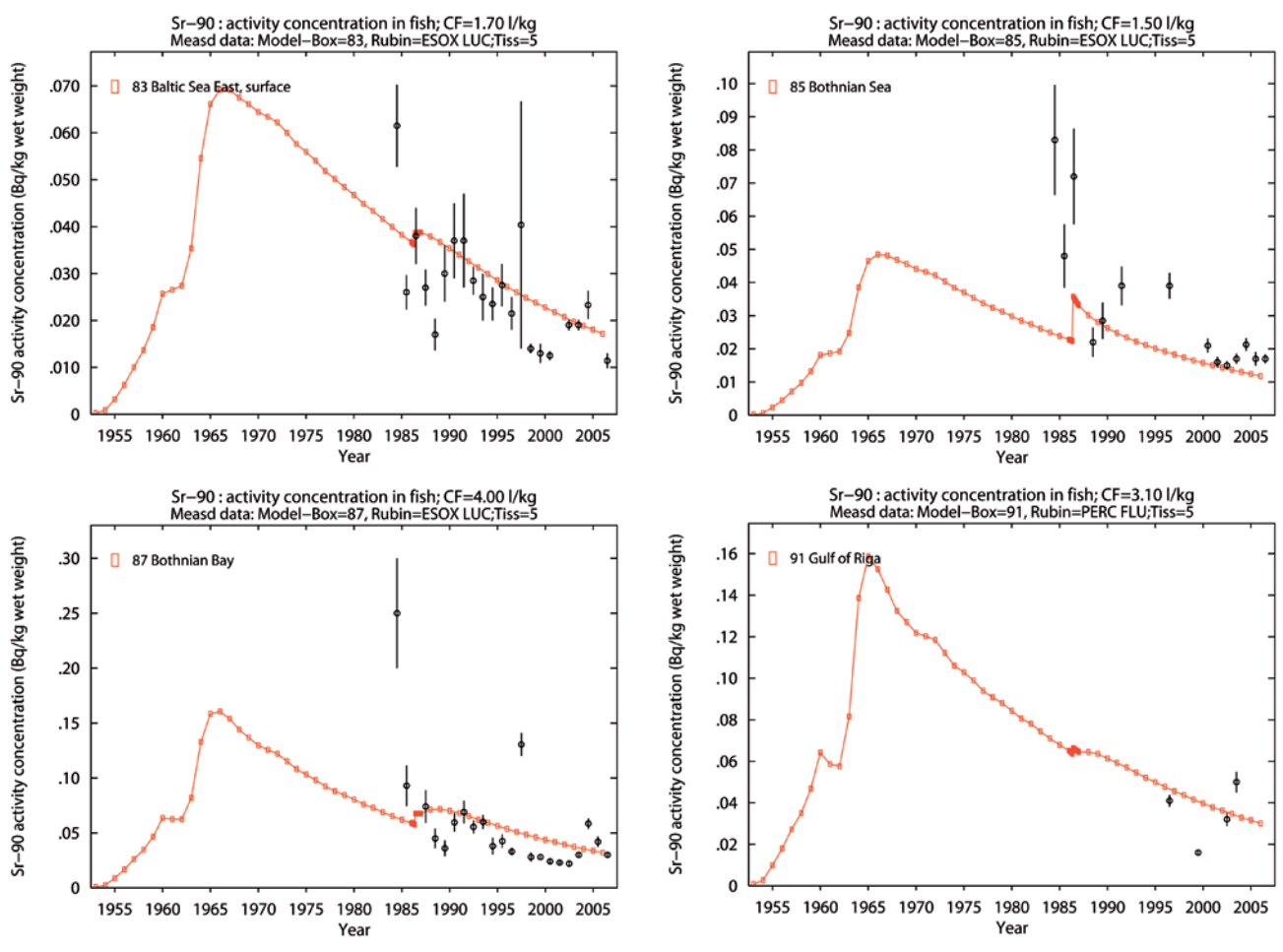


Figure 3: Comparison between calculated and observed activity concentrations of ¹³⁷Cs in flat fish flesh (flounder, plaice, dab) for sub-regions 75, 81, 83 and 91.

Figure 4: Comparison between calculated and observed activity concentrations of ¹³⁷Cs in freshwater pike flesh for sub-regions 83, 85, 87 and 89.

Figure 5: Comparison between calculated and observed activity concentrations of ⁹⁰Sr in pike flesh (freshwater fish) for sub-regions 83, 85, 87 and 89; and in perch for sub-region 91.



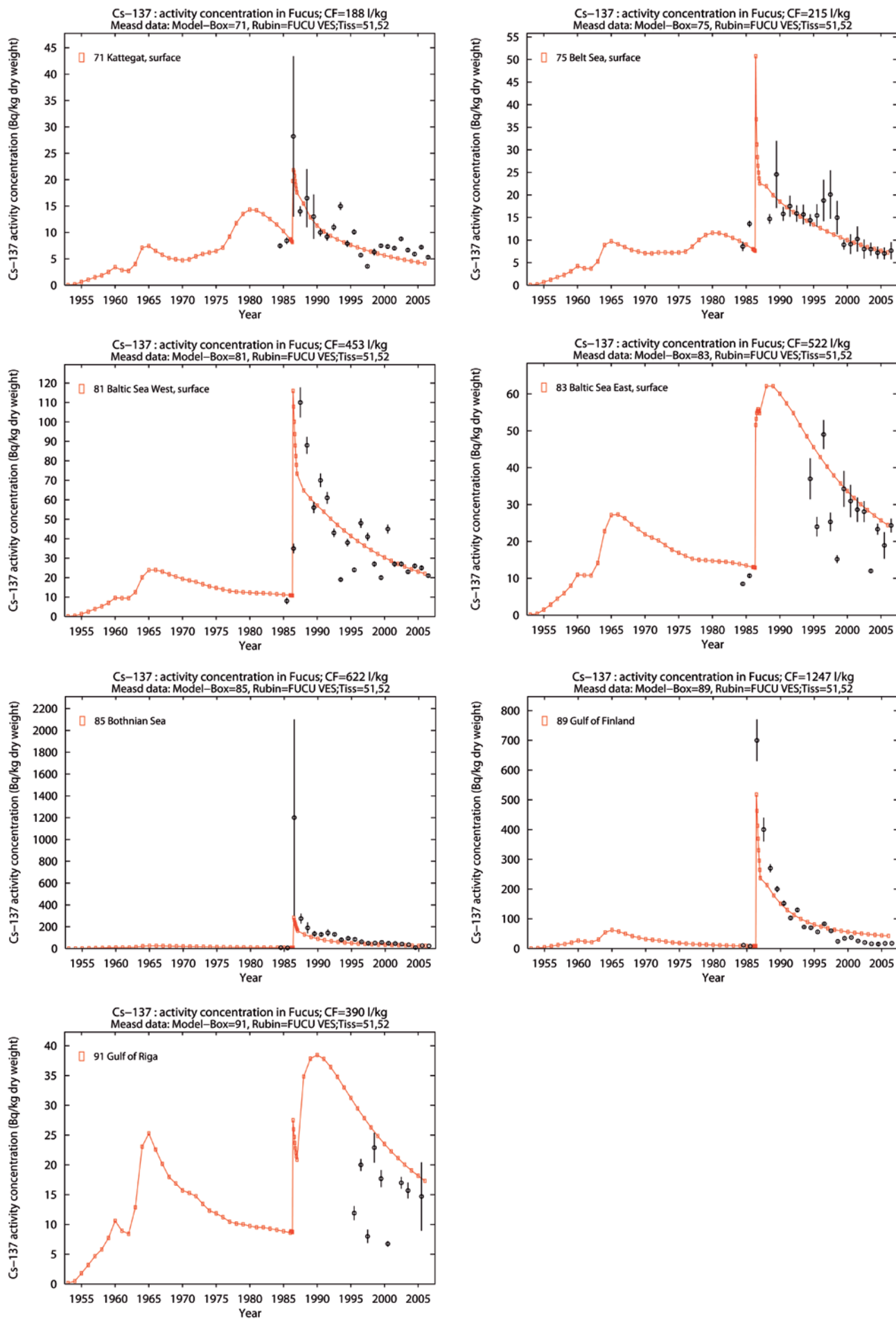


Figure 6: Comparison between calculated and observed activity concentrations of ¹³⁷Cs in *Fucus vesiculosus* for sub-regions 71, 75, 81, 83, 85, 87, 89 and 91.

The resulting mean values of $^{238}\text{Pu}/^{239,240}\text{Pu}$, with a range 0.034-0.065, lead to the conclusion that global weapons fallout is still expected to be the main source for plutonium isotopes in the Baltic Sea. Mean values of the ratio $^{241}\text{Am}/^{239,240}\text{Pu}$ were found between 0.12 (*Fucus vesiculosus*) and 0.47 (whole-body sprat).

References

- BMU (2006): Meerwasser, Schwebstoff, Sediment. In: Umweltpolitik: Umweltradioaktivität und Strahlenbelastung, Jahresbericht 2005. Ed.: Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU), Bonn. (In German).
- Dahlgaard, H. (1996): Polonium-210 in Mussels and Fish from the Baltic-North Sea Estuary. *J. Environmental Radioactivity* 32 (1-2):91-96.
- HELCOM (1995): Radioactivity in the Baltic Sea 1984-1991. *Balt. Sea Environ. Proc.* No. 61. 182 pp.
- Helsel, D.R. & T.A. Cohn (1988): Estimation of descriptive statistics for multiply-censored water quality data. *Water Resources Research* 24 (12), 1997-2004.
- IAEA (1985): Sediment K_d 's and Concentration Factors for Radionuclides in the Marine Environment. IAEA Technical Report Series, 247, Vienna.
- IAEA (2004): Sediment Distribution Coefficients and Concentration Factors for Biota in the Marine Environment. IAEA Technical Report Series, 422, Vienna.
- Lee, L. & D.R. Helsel (2005): Statistical analysis of water-quality data containing multiple detection limits: S-language software for regression on order statistics. *Computers & Geosciences* 31:1241-1248.
- Pentreath, R.J. (1988): Sources of artificial radionuclides in the marine environment. In: J.C. Guary, P. Guegueniat and R.J. Pentreath (Eds.): *Radionuclides: A tool for oceanography*. London: Elsevier Applied Science.



4 Modelling and Dose Calculations

Günter Kanisch¹ and Sven Nielsen²

¹) *vTI, Johann Heinrich von Thünen-Institute, Germany*

²) *Risø DTU, National Laboratory for Sustainable Energy, Denmark*

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

to simulate levels of ⁹⁰Sr and ¹³⁷Cs in Baltic seawater based on known inputs from the dominating sources, i.e. atmospheric fallout from nuclear weapons testing, atmospheric fallout from the Chernobyl accident, and discharges into the sea from the European reprocessing facilities at Sellafield and La Hague. The model also includes the runoff

Environmental compartment	¹³⁷ Cs			⁹⁰ Sr		
	P/O ratio	geometric SD of P/O	No. of values	P/O ratio	geometric SD of P/O	No. of values
Seawater	1.05	1.16	396	0.99	1.10	367
Fish	1.03	1.29	362	0.97	1.30	178
<i>Fucus vesiculosus</i>	1.01	1.23	150	0.98	1.27	39

Table 1:

Summary of comparisons between model predictions (P) and observations (O) in environmental compartments of the Baltic Sea. Geometric means and geometric standard deviations were calculated for the P/O ratios. The concentration factors considered for fish, differentiated by marine and freshwater fish, round fish and flat fish, fillets and edible parts, as well as for the bladder wrack *Fucus vesiculosus*, were taken from Chapter 3.

2003) that was implemented in software which does not run on the latest computers. Work is presently in progress to implement 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.

The model has been developed to assess the radiological consequences of releases of radioactive material into 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. The association of radionuclides with suspended sediment material is taken into consideration, in addition to any subsequent transfer into sediments through particle scavenging. Starting with specified inputs of radioactivity into the marine environment, the model calculates time-dependent concentrations in seawater and sediments. This data may be used to calculate doses to man from a range of exposure pathways. The locations of the water boxes defined for these studies are shown in **Figure 1**.

The new implementation of the model (Kanisch et al. 2000) has made it possible

of radioactivity from land to sea via rivers. **Figures 2a and 2b** show comparisons between calculated (red lines and squares) and observed (black circles) concentrations of ¹³⁷Cs in seawater in various sub-regions. The observed concentrations are the

Range of years	Effective half-life (years)	
	¹³⁷ Cs	⁹⁰ Sr
1988 - 1996	9.0	15.2
1999 - 2006	12.8	14.5
1993 - 2006	11.5	14.6

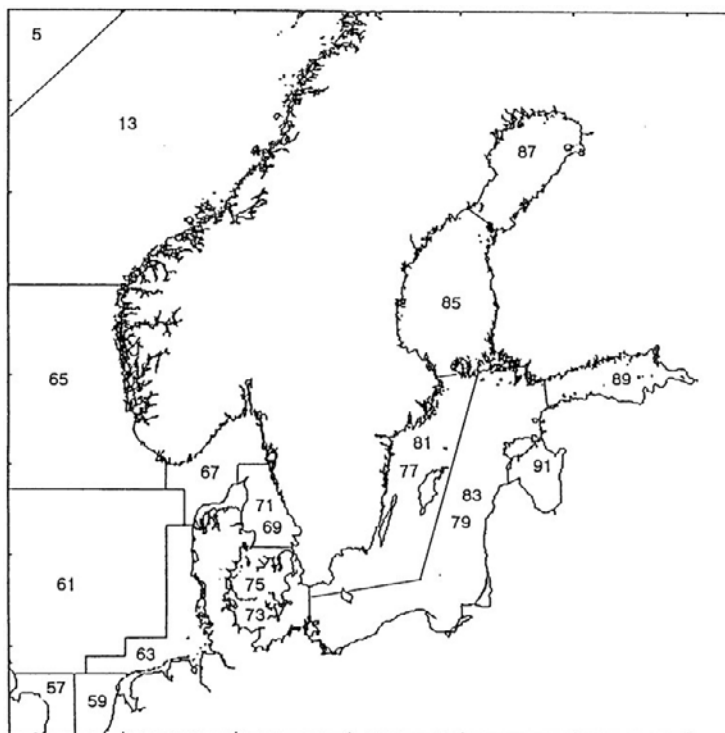
Table 2:

Rates of reduction of ⁹⁰Sr and ¹³⁷Cs in the Baltic Sea, in terms of effective half-lives derived from calculated total inventories during different time periods.

annual average values of data collected by MORS-PRO. The vertical bars represent the variability of the observed concentrations within a single year.

The reliability of the model calculations is illustrated in **Figures 3a and 3b** showing scatter plots of observed and calculated seawater concentrations of ⁹⁰Sr and ¹³⁷Cs across all regions of the Baltic Sea for the years 1965-2006. The data points are distributed both above and below the line of the 1:1 relationship thus indicating the model's overall unbiased quality. **Table 1** shows the corresponding results of the statistical evaluation of the ratios predicted/observed; with the results for fish and *Fucus vesiculosus* included. **Table 2** shows the rates of reduction

Figure 1: Map of the Baltic Sea showing water sub-regions applied in the model. Sub-regions no. 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.



of ^{90}Sr and ^{137}Cs in Baltic seawater from inventories calculated by the model expressed in terms of effective half-lives. The values for ^{137}Cs compare well with corresponding values derived from observed inventories, showing values in the range of 9 to 15 years, as described in Chapter 3A on seawater.

Important aspects to incorporate in further modelling work include river run-off in agreement with available observed data, and the transfer of radioactivity from contaminated seabed sediments to the water column. Both of these processes contribute to the fact that the concentrations of ^{137}Cs in Baltic seawater remain relatively high more than twenty years after the Chernobyl accident in 1986.

4.2 Dose calculations

Estimates of radiation doses accumulated until the year 2000 by human individuals and populations from radioactivity in the Baltic Sea were made by the MORS Group in HELCOM (2003). These 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}Cs and the ingestion of fish.

The concentrations of the dominating man-made radionuclides in the Baltic Sea, ^{90}Sr and ^{137}Cs , have been declining since 2000. The only man-made radionuclides showing increasing trends in Baltic seawater are the small quantities of radionuclides originating from discharges from Sellafield and La Hague (^{99}Tc and ^{129}I), but in terms of radiation doses to humans these are insignificant.

For the reporting period covered by the present report, we may estimate an upper limit for individual doses from man-made radionuclides in the Baltic Sea. During the period 1999-2006 the concentrations of ^{137}Cs in fish from the Baltic Sea have been below 20 Bq/kg. For an individual having a high-rate consumption of 90 kg fish per year this concentration corresponds to an annual radiation dose of 20 μSv . This dose is well below the safety limit for the annual radiation dose to a member of the public of 1000 μSv (EC 1996). The corresponding annual dose from naturally occurring radionuclides in fish is about 100 μSv , of which the dominating contribution is from polonium-210. The annual dose contribution from tritium (^3H) is insignificant by comparison, less than 0.01 μSv from natural and man-made sources combined.

The annual radiation doses calculated for the period 1955-2005 for individuals consuming 9 kg fish flesh per year are shown in **Figure 4**. The calculations show that the annual

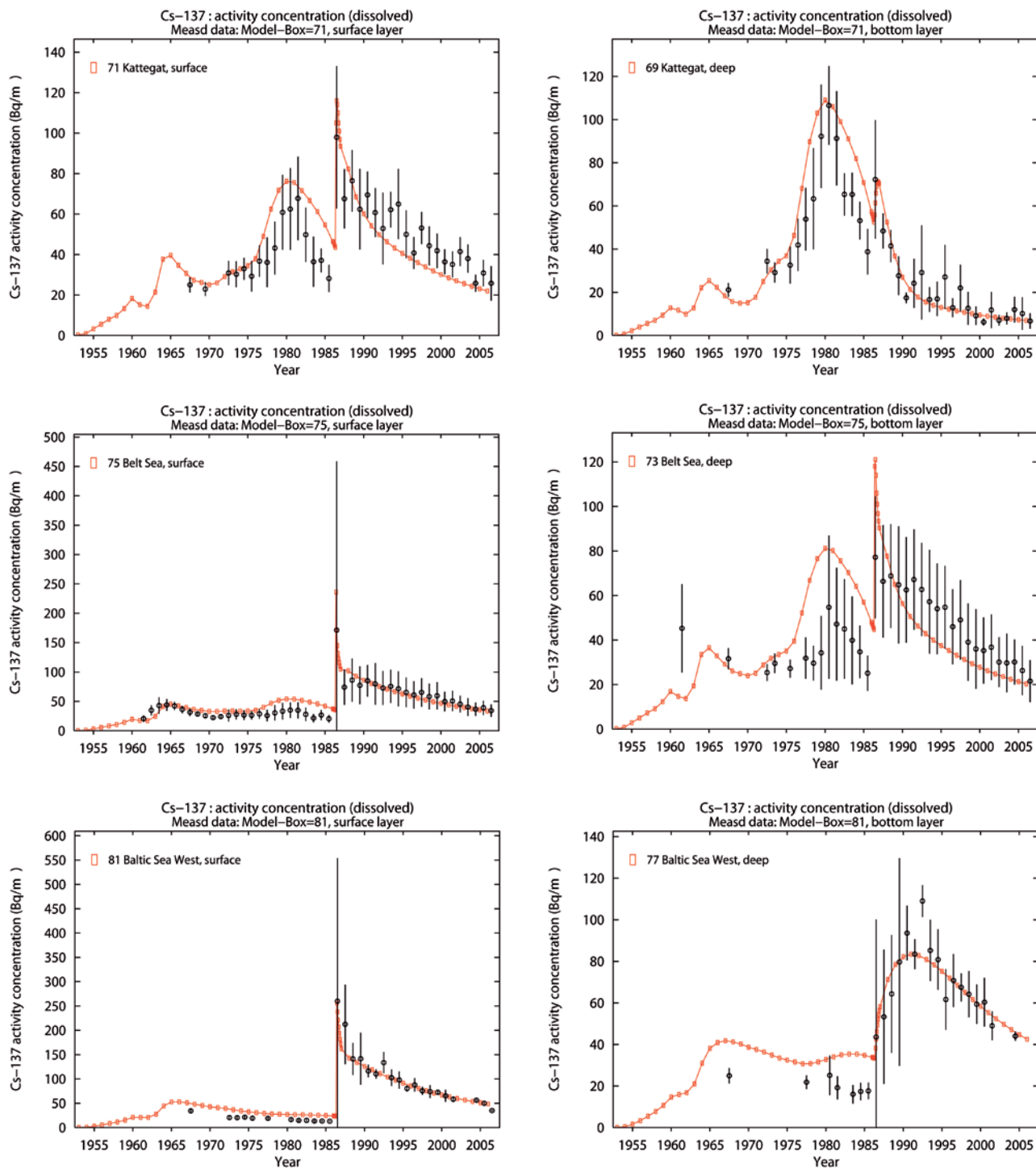


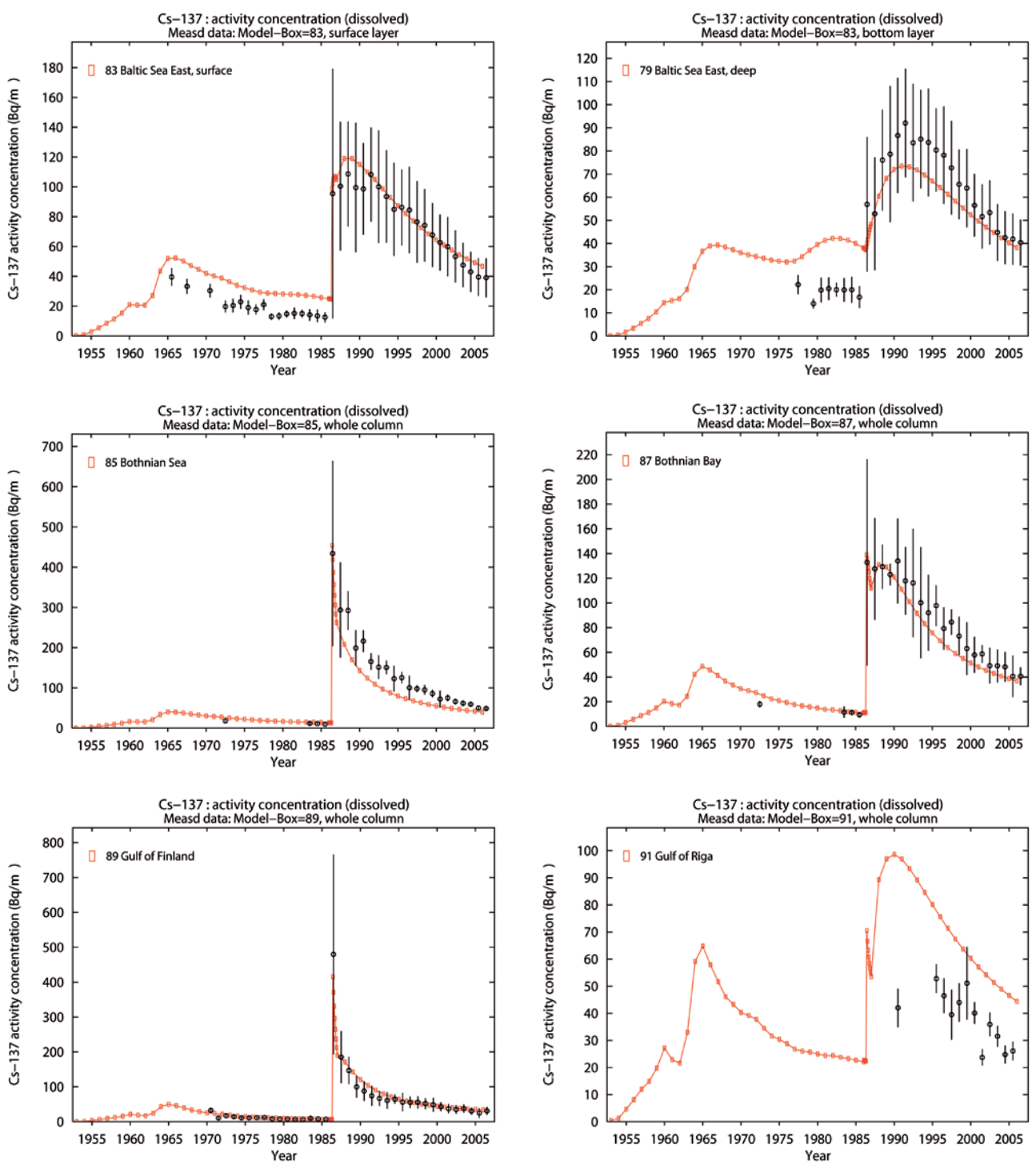
Figure 2a: Comparison between calculated and observed concentrations of ¹³⁷Cs in Baltic seawater for sub-regions 69, 71, 73, 75, 77 and 81.

individual doses from man-made radionuclides peak in 1986 at 14 μ Sv. The corresponding dose from naturally occurring radioactivity in fish flesh is about 10 μ Sv.

may be based on work carried out in the ERICA Project (2004-2007) using a software tool developed for that purpose.

Future work will include assessments of the radiological risk to the environment from radioactive substances in the Baltic Sea. Assessments of the radiological risks to marine biota including mammals, fish, molluscs, crustaceans, algae and plankton

Figure 2b:
 Comparison between calculated and observed concentrations of ^{137}Cs in Baltic seawater for sub-regions 79, 83, 85, 87, 89 and 91.



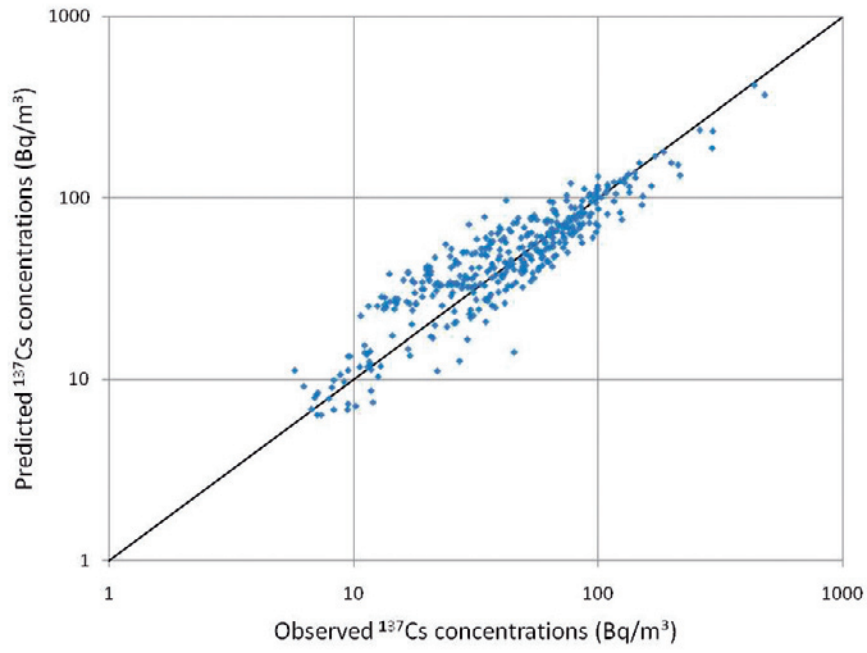


Figure 3a: Scatter plots of observed and calculated concentrations of ^{137}Cs in Baltic seawater covering all regions of the Baltic Sea, 1965-2006.

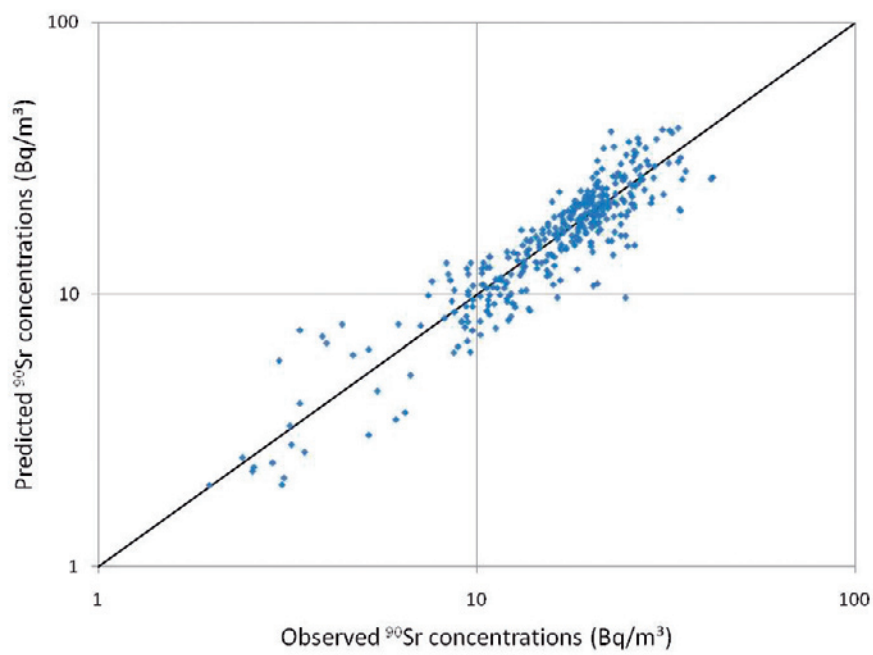
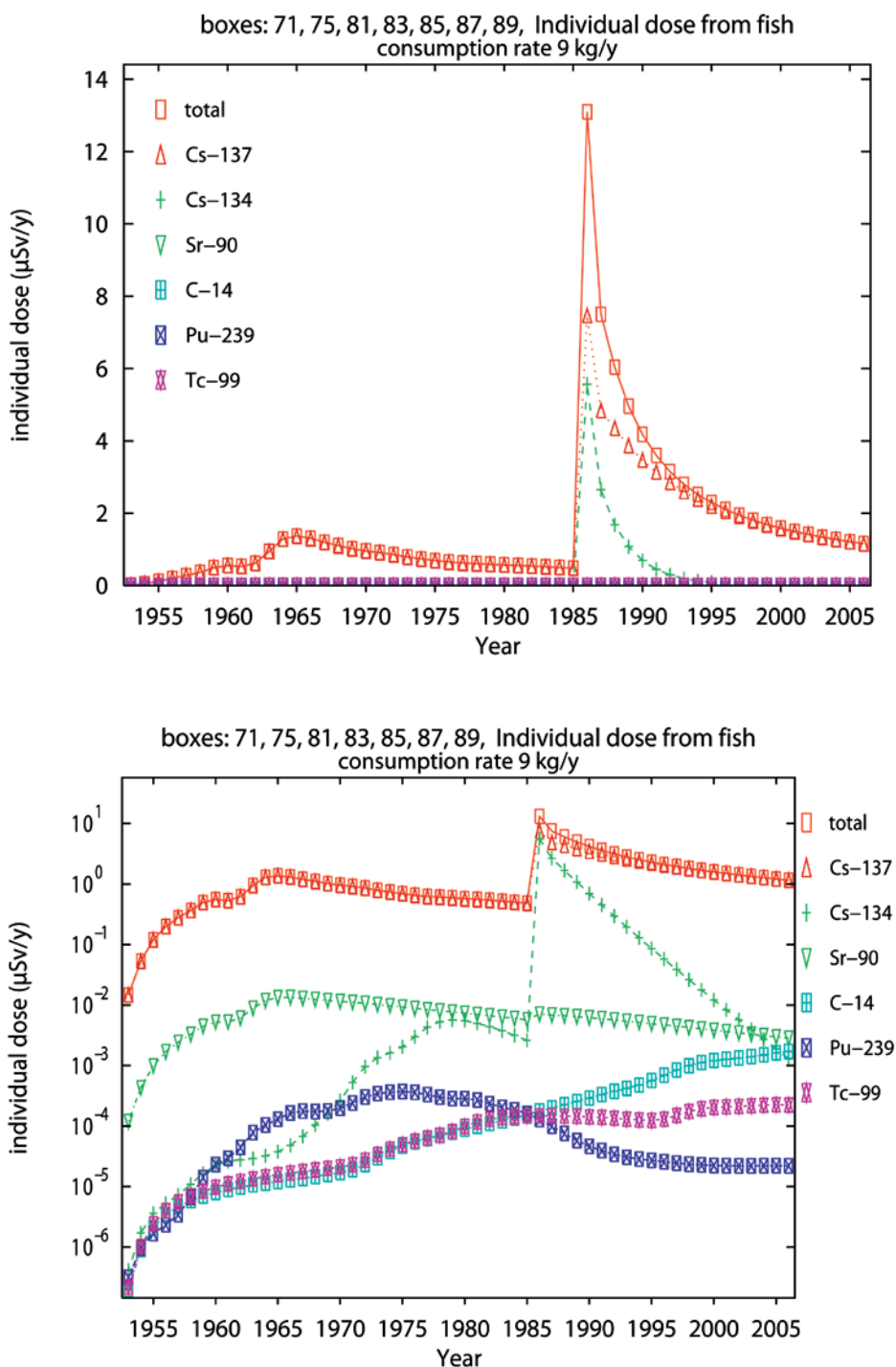


Figure 3b: Scatter plots of observed and calculated concentrations of ^{90}Sr in Baltic seawater covering all regions of the Baltic Sea, 1965-2006.

Figure 4: Calculated annual radiation doses for individuals in the Baltic Sea region consuming 9 kg/y of fish flesh. The graphs show the same data, but the graph on the top uses a logarithmic scale on its vertical axis. The six radionuclides shown are those with the largest contributions to the dose; the sum of the radionuclide contributions is shown as "total".



References

EC (1996): Basic Safety Standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. Council Directive 96/29/EURATOM, European Council, Brussels.

ERICA (2004-2007): Environmental Risk from Ionising Contaminants: Assessment and Management. Project supported by European Commission within the Sixth Framework Programme.

HELCOM (2003): Radioactivity in the Baltic Sea 1992-1998. Balt. Sea Environ. Proc. No. 85. 102 pp.

Kanisch, G., G. Nagel, A. Krüger & H.-J. Kellermann (2000): Radiological implications from the temporal development of radioactivity in marine food from the North Sea. Kerntechnik, 65:183-189.

5 Radioactivity in the Baltic Sea compared to other sea regions

Iolanda Osvath

*International Atomic Energy Agency (IAEA)
Marine Environment Laboratories*

A recently published study on worldwide marine radioactivity (IAEA, 2005) enables a comparison of levels of anthropogenic radionuclides in Baltic seawater against those in other areas of the world ocean. The Baltic Sea ranked highest for average Cs-137 levels in surface water in the year 2000 (**Figure 1**), closely followed by the Irish Sea. In terms of Sr-90 levels, the Baltic Sea ranks third, after the Irish Sea and the Black Sea. Average Pu-239,240 levels in surface water in the Baltic Sea are similar to those measured in other areas of the northern ocean at large, but two orders of magnitude below those estimated for the Irish Sea.

These levels clearly reflect the dominant regional sources for the respective radionuclides, as described in detail for the Baltic Sea in Chapter 2, as well as the processes affecting the fates of these radionuclides, quite different in terms of physico-chemical properties, after their release to the environment. A comparison with other semi-enclosed European Seas, such as the Black Sea and the Mediterranean Sea, helps set this in perspective. The following comparison will focus on Cs-137, which has been considered as the most significant man-made radionuclide in the marine environment in terms of inventory, transfer and global-scale dosimetry (IAEA, 1995). Input from the Chernobyl accident is significantly higher for the Baltic Sea than the contribution of global fallout from nuclear weapons testing, while for the Black Sea these two sources contributed more equally to the inventory of Cs-137 (**Table 1**). Although it drains a smaller land area than the Black Sea, the Baltic Sea also received larger long-term post-Chernobyl inputs of Cs-137 through rivers than the Black Sea. This

reflects the pattern of contamination by fallout from Chernobyl of the respective drainage basins (OECD, 1996; EC, 1998) and the latitudinal distribution of global fallout (UNSCEAR, 2000). The contribution of global fallout is considerably higher for the Mediterranean Sea, given the large surface area of this sea and of its drainage basin. Basin-specific freshwater inflow patterns, seawater circulation and mixing as well as geochemical processes further control the transport and transfer of radionuclides through the marine environment and explain the observed distributions and differences from region to region.

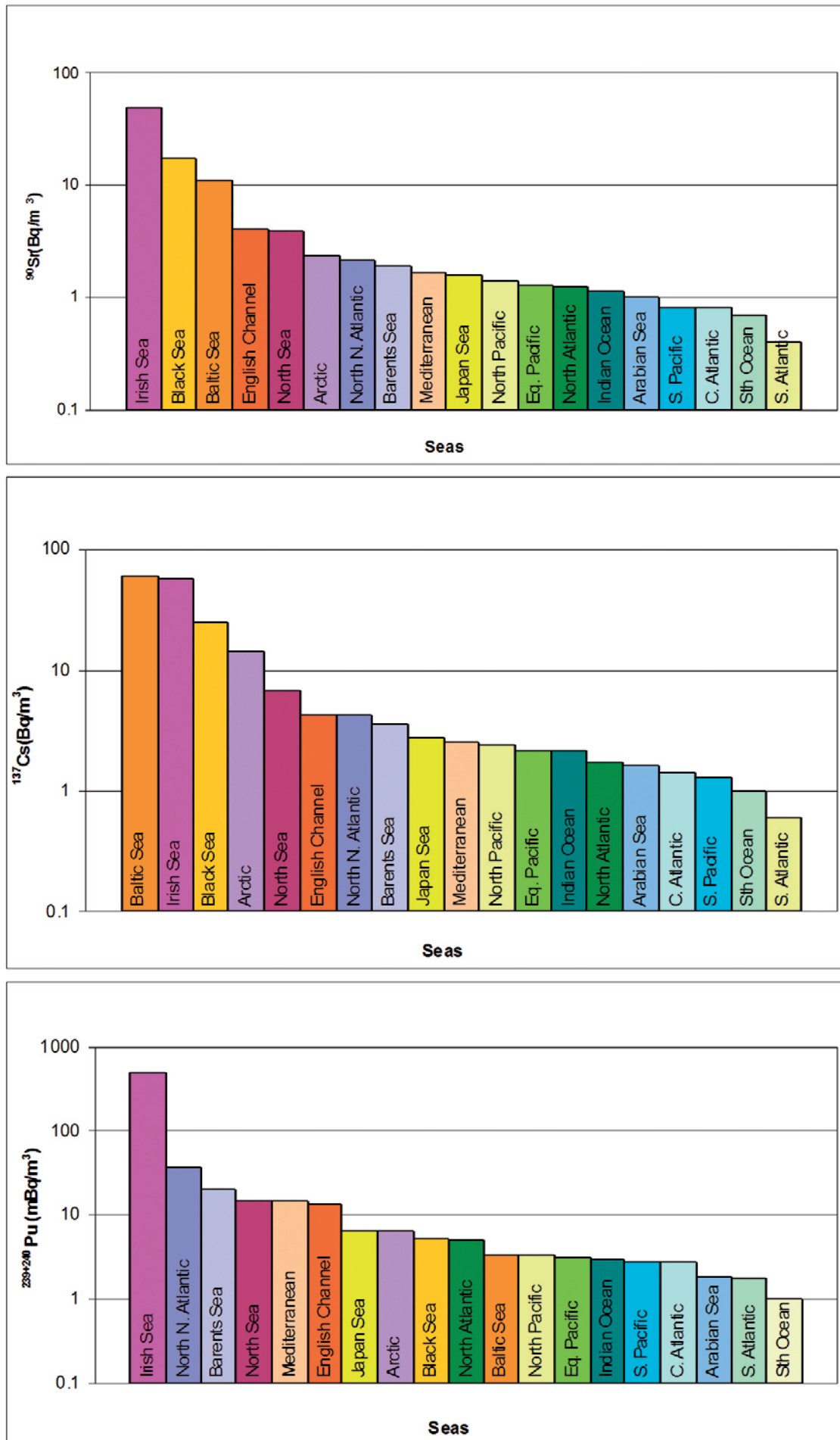
During the period 2000-2005, Cs-137 levels of up to 125 kBq·m⁻² were found in Baltic Sea bottom sediments, with median levels of 36 kBq·m⁻² in the Bothnian Sea and 10 kBq·m⁻² in the Gulf of Finland (HELCOM, 2007). For comparison, Cs-137 inventories in Black Sea bottom sediments reported by Egorov et al. (2006) for the years 1990-1994 range between 0.2 kBq·m⁻² in the abyssal basin and continental slope areas, and 30 kBq·m⁻² at the mouths of the River Danube. Sediment inventories up to 18 kBq·m⁻² Cs-137 were reported for the latter area for 2003 (Laptev and Voitsekhovitch, 2006). Western Mediterranean inventories of Cs-137 in deep-sea sediment of 0.2 – 0.3 kBq·m⁻² were reported for the early 1990s (Delfanti et al., 1995).

A comprehensive study of radioactivity in the marine environment and doses from marine exposure pathways published in 1995 (IAEA, 1995) shows that average levels of Cs-137 in fish in the Baltic Sea in 1990 were similar to

Source	Baltic Sea	Black Sea	Mediterranean Sea
Chernobyl atmospheric deposition on the sea surface, PBq	4.1-5.1	1.7-2.4	3-5
Chernobyl river inflow (1), inflow from the Black Sea through the Bosphorus Strait (2), PBq	0.3 (1)	0.03 (1)	0.3 (2)
Global fallout/atmospheric nuclear weapons tests, PBq	0.8	1.4	12
Nuclear waste reprocessing plants, PBq	0.25	-	0.03
Sea surface area, 10 ³ km ² (average depth, m)	415 (55)	436.4 (1253)	2500 (1500)
Drainage basin surface area, 10 ³ km ²	1700	2400	3300

Table 1: Contributions to cumulative Cs-137 inventory: comparative table for semi-enclosed European seas (This report; Egorov et al, 2007; IAEA 2005)

Figure 1:
Average surface levels of ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in the world's oceans and seas (estimates for 01.01.2000)



those in the Irish Sea, approximately 4 times higher than in the Black Sea, and some 30 times higher than in the Mediterranean.

References

- Delfanti, R., C. Papucci, M. Alboni, R. Lorenzelli & S. Salvi (1995): ¹³⁷Cs inventories in the water column and in sediments of the Western Mediterranean Sea. *Rapp. Comm. Int. Mer Medit.* 34, 226.
- EC (1998): Atlas of caesium deposition on Europe after the Chernobyl accident. EUR 16733, Luxembourg.
- Egorov, V.N., G.G. Polikarpov, S.B. Gulin, I. Osvath, N.A. Stokozov & G.E. Lazorenko (2006): XX years of radioecological response studies of the Black Sea to the Chernobyl NPP accident. Presented at the First Biannual Scientific Conference "Black Sea Ecosystem 2005 and Beyond", Istanbul, 8-10 May 2006.
- HELCOM (2007): Long-lived radionuclides in the seabed of the Baltic Sea. Report of the Sediment Baseline Study of HELCOM MORS-PRO in 2000-2005. *Balt. Sea Environ. Proc.* No. 110. 41 pp.
- IAEA (1995): Sources of radioactivity in the marine environment and their relative contributions to overall dose assessment from marine radioactivity (MARDOS). IAEA-TECDOC-838.
- IAEA (2005): Worldwide marine radioactivity studies (WOMARS). Radionuclide levels in oceans and seas. IAEA-TECDOC-1429.
- Laptev G. & O. Voitsekhovitch (2006): Contribution to RER/2/003 report. IAEA RER/2/003 working document.
- Laptev, G. & O. Voitsekhovitch (2006): Contribution to RER/2/003 report. IAEA RER/2/003 working document.
- OECD (1996): Chernobyl - Ten years on radiological and health impact. NEA-OECD.
- UNSCEAR (2000): Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation – UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes, Vol. I: Sources. United Nations, New York.



6 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}Cs and ^{134}Cs . The total input of ^{137}Cs from Chernobyl to the Baltic Sea has been estimated at 4,700 TBq, and the post-Chernobyl river discharges of ^{137}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. The predominant radionuclides in the global fallout were ^{137}Cs and ^{90}Sr , in an activity ratio of about 1.6. During the late 1990s the decay-corrected amounts of weapons-test ^{137}Cs and ^{90}Sr in the Baltic Sea have been evaluated at 800 and 500 TBq, respectively.

The corresponding decay-corrected total inputs to the Baltic Sea of ^{137}Cs and ^{90}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 ^3H . Total discharges of ^3H from these local sources have amounted to 3,200 TBq, and those of other beta-gamma emitting radionuclides amounted to about 24 TBq by the end of 2006. The total discharges of alpha emitting radionuclides have been 0.005 TBq.

For ^{137}Cs in the Baltic Sea, the main source is fallout from Chernobyl (82%), followed by nuclear weapons test fallout (14%). For ^{90}Sr , the main source of contamination is fallout from nuclear weapons tests (81%), while the proportion from Chernobyl fallout was smaller (13%).

Today, ^{137}Cs is the main indicator of man-made radioactivity in Baltic seawater. The highest concentrations observed in seawater during the period 1999-2006 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/m³, corresponding to pre-Chernobyl levels, will be reached between 2020 and 2030. First estimates of effective half-lives for different parts of the Baltic Sea have been calculated. The inventory of ^{137}Cs in the Baltic seawater in 2006 is estimated at 870 TBq.

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. However, the concentrations of man-made radionuclides are still higher than the target specified in HELCOM's ecological objective of "radioactivity at 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}Cs in the Baltic Sea sediments was estimated at 2,100-2,400 TBq at the beginning of the 2000s, while the inventories of the naturally occurring radionuclides ^{40}K and ^{226}Ra were estimated to be about 8,500 and 420 TBq, respectively, in the uppermost 0-10 cm sediment layer. Hence, the concentrations of man-made radionuclides in sediments were generally at or below the concentrations of naturally occurring radionuclides.

It remains essential to monitor radioactive substances in the Baltic Sea to provide a basis for evaluating time trends and for understanding the state of the Baltic marine environment and various aspects of radioactivity. However, there are still gaps in our knowledge of radioactivity in the sediments 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}Cs continued to be the most dominant man-made radionuclide in Baltic Sea fish. By the end of the reporting period, mean values of 1-10 Bq kg^{-1} wet weight were found in marine round fish (cod, herring, whiting) in various Baltic Sea basins, while the concentrations in pike were 10-25 Bq kg^{-1} wet weight on the Finnish coast. In marine flat fish (plaice, flounder, dab) slightly lower mean values were found than in marine round fish.

The Baltic Sea has the highest concentrations of ^{137}Cs of any regional sea around the world, due to radioactive fallout from the Chernobyl accident. The Baltic Sea ranks third in the world with respect to ^{90}Sr in seawater, with only the Irish Sea and the Black Sea showing higher levels. Average concentrations of ^{137}Cs in fish from the Baltic Sea in 1990 were similar to those in the Irish Sea, about 4 times higher than in the Black Sea, and about 30 times higher than in the Mediterranean Sea.

Radiation doses to humans from man-made radionuclides in the Baltic Sea are due mainly to ingestion of ^{137}Cs in fish. Doses from ^3H are lower by several orders of magnitude. During 1999-2006 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).

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.

References

- EC (1996): Basic Safety Standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation. Council Directive 96/29/EURATOM, European Council, Brussels.
- IAEA (1996): International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation sources, Safety Series No. 115. International Atomic Energy Agency, Vienna.

7 Recommendations

The objective of HELCOM MORS-PRO 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-PRO's responsibilities include the following activities:

- 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;
- to compile annually data on discharges of radioactivity from civil nuclear facilities to the Baltic Sea reported by the Contracting Parties;
- to compile annually data on discharges and environmental levels of radioactivity in the Baltic Sea submitted to the HELCOM databases, in accordance with HELCOM Recommendation 26/3;
- to validate annually all data in the HELCOM MORS databases and to make it available on CD-ROMs to MORS-PRO Members;
- to update annually HELCOM Indicator Fact Sheets on radioactive substances in the Baltic Sea;
- to keep the Guidelines on the Monitoring of Radioactive Substances in the Baltic Sea updated;
- to coordinate and organise inter-comparison exercises on seawater, sediments and biota to assure the high quality of monitoring data;
- to 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. technetium-99 and iodine-129) from Sellafield to the Baltic Sea and the outflow of Chernobyl radioactivity from the Baltic Sea to the Skagerrak;
- to produce thematic reports, e.g. on naturally occurring radionuclides in the Baltic Sea, releases of man-made radionuclides from non-nuclear activities (e.g. hospitals), and simple procedures for assessing doses to man from radioactivity in the Baltic Sea;
- to finalise in 2011 the periodic assessment on radioactivity in the Baltic Sea covering data up to 2009. This assessment will include levels, inventories and trends for radioactivity in the Baltic Sea and its radiological impact on man and environment.

8 Acknowledgements

The authors wish to express their sincere thanks to HELCOM MONAS for providing the organisational framework and financial support for the work of the MORS Project. In particular, the authors wish to thank the HELCOM Secretariat for their invaluable support during and in between the annual meetings of the group, and for their considerable patience and understanding.

The authors also wish to express their gratitude to Prof. Pavel P. Povinec, University of Bratislava, Slovakia for providing scientific review of the Assessment and his valuable comments both on the scope and contents of the current assessment, as well as sound recommendations for the future work of the MORS Project.

Tarja K. Ikäheimonen, Iisa Outola
STUK, Radiation and Nuclear Safety Authority, Finland

1 Introduction

More than ten laboratories from the nine countries bordering Baltic Sea have contributed to the monitoring programmes of Baltic Sea by analyzing radionuclides from marine samples. The various analytical methods used in the different laboratories are summarized in the table below. Quality assurance is a fundamental part of radioanalytical work, needed to confirm the precision and the long-term repeatability of analyses. Eight intercomparisons were organised during the project period 1999-2006 for seawater and sediment samples, and their results are also presented below.

2 Analytical procedures used by MORS laboratories

The analytical methods in use (**Table 1**) have not changed notably since the last report (Ikäheimonen and Mulsow, 2003). Pretreatment of water samples is performed either by evaporation, adsorption on AMP, or using ferro-cyanide precipitation. Biota and sediment samples are generally dried and ashed. ^{137}Cs is measured by a high-purity Ge-detector, except in one country that used beta counting. ^{90}Sr is chemically separated using extraction, Sr-resin or various precipitations is monitored through proportional counting or LS counting. Traditional fuming nitric acid precipitation has generally been replaced by other chemical separation methods. U, Pu and Am are separated using anion exchange, extraction chromatography resins or liquid-liquid extraction followed by electroplating and alpha counting. A short description of the radiochemical methods used for natural radionuclides (Po, U, Th), ^3H and ^{99}Tc is also given in Table 1 for the first time.

3 Quality assurance, internal and external checking

The quality systems in use in different MORS laboratories are summarised in **Table 2**. Six laboratories have quality management systems in place, and four of these also

employ accredited methods in their analytical work. All of the laboratories have participated in numerous intercomparisons during the study period, assuring good data quality.

4 Intercalibration exercises (1999-2006) organized by the MORS group

4.1 Sediment

In June 2000 the MORS group arranged an intercomparison exercise together with IAEA using a sediment sample taken from the Bothnian Sea. A detailed description of the intercomparison is given by Ikäheimonen and Vartti (2007). Eight laboratories from Denmark, Estonian, Finland, Germany, Latvia, Poland and Sweden participated in the intercomparison, and reported results for gamma nuclides, transuranic elements and natural radionuclides (**Table 3**). Results for ^{137}Cs and $^{239,240}\text{Pu}$ are shown in **Figures 1 and 2**. The results indicated that the measurements by these eight laboratories are in quite good agreement. The difference from the mean value was statistically significant only in two cases among 41 results evaluated based on the z-scores.

4.2 Seawater

The capabilities of the MORS group in terms of analyzing seawater were tested with two different seawater samples. An IAEA-337 sample was collected from the Gotland Deep (sampling site BY15) in July 1996 and then analyzed during the period 1999-2002 for ^{137}Cs and ^{90}Sr . During the years 2004-2006 the laboratories were asked to analyze ^{137}Cs and ^{90}Sr once a year from a seawater sample collected from the Bothnian Sea (sampling site EB1) in June 2004. The results of these intercomparisons are illustrated in **Figures 3-6**. The results indicate that the measurements are in very good agreement. The results were unsatisfactory only in 3 cases among 68 results evaluated based on the z-scores. In addition to the intercomparisons organised by the MORS group, the laboratories

have actively participated in many other intercomparisons, in many cases coordinated by the IAEA and NKS (Nordic Nuclear Safety Research).

5 Conclusions

The radiochemical procedures and counting techniques used by laboratories are well-tested, up-to-date, and similar to those used by laboratories worldwide. The eight intercomparisons organized by the MORS group during the period 1999-2006 confirm that the data produced by the MORS group is of very good quality and can be considered comparable. Less than five percent of the results were considered outliers. At the same time the reported uncertainties vary considerably between laboratories. Each laboratory calculates uncertainties in its own particular way, and the harmonization of uncertainty calculations would improve the comparability of the data.

References

- Ikäheimonen, T.K. & V. Vartti (2007):
An intercomparison of Radionuclide Analyses in a Baltic Sea Sediment Sample. In: HELCOM (2007): Long-lived radionuclides in the seabed of the Baltic Sea. Balt. Sea Environ. Proc. No. 110:25-41.
- Ikäheimonen, T.K. & S. Mulsow (2003): Data quality. In: HELCOM (2003): Radioactivity in the Baltic Sea 1992-1998. Balt. Sea Environ. Proc. No. 85:49-54.

Table 1: Analytical methods used by MORS laboratories

Lab/ Country	Method used for Cs-137	Method used for Sr-90	Method used for Pu (Am)	Methods for natural and other radionuclides
Risoe/ Denmark	sediment, biota: HPGe gamma spectrometry water: absorption on AMP + HPGe gamma spectrometry	Ca(OH) ₂ + Y-90 counting with low-level GM counter	water: Fe(OH) ₂ /Fe(OH) ₃ + TIOA solvent extraction + anion exchange + electroplating + α spectrometry (or ICPMS)	Tc-99 (water): anion exchange + Fe(OH) ₃ + TIOA/xylene extraction + low-level GM counting (biota); acid digestion + GM counting
Estonian Radiation Protection Center / Estonia	evaporation (water), drying (sediment), drying + ashing (biota) + HPGe gamma spectrometry	sediment: drying + ashing + HDEHP extraction + cerenkov counting in liquid scintillation counter		
STUK /Finland	seawater: evaporation + HPGe gamma spectrometry solid: drying + HPGe gamma spectrometry (gamma-emitting radionuclides in energy range of 30-2700 keV)	evaporation/drying/ashing/microwave leaching + Sr-resin purification + beta counting by betacounter/liquid scintillation counting	precipitation/microwave leaching + anion exchange purification (Pu) + TRU-TEVA purification (Am) + α -spectrometry	evaporation/drying/ashing/microwave leaching; spontaneous deposition (Po), anion exchange (U) + α spectrometry; Sr resin purification (Pb, if not gamma spectrometry) + liquid scintillation counting
BFFG/ VTIG Germany	biota: drying + ashing; Ge + HPGe gamma spectrometry	biota: drying + ashing + wet digestion + HDEHP extraction + purification with Adogen 464 + hydroxide precipitation + Y-oxalate precipitation + beta counting	biota: drying + ashing + wet digestion + TOPO/cyclohexane extraction; LaF ₃ precipitation + anion exchange (Pu) cation/anion exchange + DDCP extraction + anion exchange (Am) electrodeposition + α -spectrometry	
BSH/ Germany	seawater: adsorption on K ₂ Co(Fe(CN) ₆) + HPGe gamma spectrometry	seawater: HDEHP extraction + cleaning with Adogen 464 + oxalate precipitation + thermal decomposition + beta counting	NH ₂ OH precipitation (water), freeze-drying, acid leach (sediment) + HDEHP extraction + anion exchange + electrodepositing + α - spectrometry	
Environmental Protection Agency/ Lithuania	sediment: drying + HPGe gamma spectrometry biota: drying + ashing + HPGe gamma spectrometry water: double sorbent cartridge method Cu ₂ Fe(CN) ₆ + drying + HPGe gamma spectrometry or concentrating of Cs-137 (together with Sr-90) with HCl, SrCl ₂ , CsCl, FeCl ₃ , CaCl ₂ , K ₂ Fe(CN) ₆ , Na ₂ CO ₃ + drying + HPGe gamma spectrometry	concentration with K ₂ Fe(CN) ₆ , Na ₂ CO ₃ (water)/ drying + ashing + dissolution (sediment, biota) + HDEHP extraction + hydroxide precipitation + oxalate precipitation + Y counting by proportional counter		
CLOR /Poland	sediment: drying + HPGe gamma spectrometry biota: drying + ashing + HPGe gamma spectrometry benthic animals (small size): dissolution + adsorption on AMP + gas-flow beta counting	sediment: acid leach + oxalate precipitation + thermal decomposition + Fe(OH) ₃ + Y oxalate precipitation + gas-flow beta counting	sediment: wet ashing + ion exchange + electrodepositon + α - spectrometry	Radium: acid leach (HNO ₃ +HF) + BaSO ₄ precipitation + de-emanation + Lucas cell counting Tritium: distillation + electrolytic enrichment + distillation + liquid scintillation counting
IMGW/ Poland	water: absorption on AMP + ion exchange + Cs ₂ PfCl ₆ precipitation + beta counting	water: oxalate precipitation + thermal decomposition + nitric acid precipitation + hydroxide precipitation + chromate precipitation + carbonate precipitation + Y hydroxide precipitation + oxalate precipitation + beta counting		
Khlopin Radium Institute/ Russia	water: Ferro-cyanide and carbonate precipitation + Ge(Li) gamma spectrometry solid: drying + Ge(Li) gamma spectrometry	Ferro-cyanide precipitation (water) drying+ashing+acid leach+ sulphate precipitation (solid) + carbonate precipitation + hydroxide precipitation + Y hydroxide + Y oxalate + beta counting	water: Fe(OH) ₃ precipitation + anion exchange + electrodepositon + α - spectrometry solid: drying + ashing + acid digestion. + anion exchange + electrodepositon + α - spectrometry	Ra-226, Th-232: gamma-spectrometry; Tritium: liquid scintillation counting
SSI/ Sweden	water: filtration through impregnated Cu ₂ (Fe(CN) ₆) filter + HPGe gamma spectrometry sediment: drying + HPGe spectrometry	dissolution in acid + HDEHP extraction + cerenkov counting of Y-90 with liquid scintillation counter		water: HPGe gamma spectrometry Tritium: distillation or shake with cation exchanger + LS counting

Laboratory/Country	Quality management system in laboratory/institute	Accredited methods	Total number of participations in intercomparisons 1999-2006:
Risoe/ Denmark	no	no	25 gamma 19 strontium 7 Tc 15 transuranic 8 natural radionuclides
Estonian Radiation Protection Center/ Estonia	yes/ no	gamma-emitting radionuclides in energy range of 88-1836 keV	46 gamma 44 strontium 6 transuranic 6 natural radionuclides
STUK /Finland	yes/yes	<ul style="list-style-type: none"> • gamma-emitting radionuclides in energy range of 30-2700 keV • Strontium • Pu/Am/Cm • U/Po/Pb/ • tritium 	19 gamma 17 strontium 6 transuranic 11 nat. rad. nuclides 10 tritium
BFFG/ VTIG Germany	no/no	no	14 gamma (including natural radionuclides) 6 strontium 9 transuranic
BSH/ Germany	yes/yes	<ul style="list-style-type: none"> • gamma spectrometry on water and sediment • Sr in seawater • Pu/Am/Cm in water and sediment 	16 gamma 10 strontium 12 transuranic 2 tritium
Environmental Protection Agency/ Lithuania	yes/yes	The department is accredited for chemical analyses in water and waste water and sampling. The Laboratory is not accredited.	18 gamma 16 strontium
CLOR /Poland	no/no	no	7 gamma 6 strontium 5 transuranic 5 nat. radionuclides
IMGW/ Poland	no/no	no	6 gamma 12 Cs by beta measurements 10 strontium
Khlopin Radium Institute/ Russia	yes/no	<ul style="list-style-type: none"> • Gamma spectrometry • Sr-90 • Pu • Tritium 	20 gamma 10 strontium 10 transuranic 2 tritium 2 C-14
SSI/ Sweden	yes	-	7 gamma 6 strontium 2 transuranic 2 nat. radionuclides

Table 2:
Quality assurance in MORS laboratories

Laboratory	Country	Nuclides reported
RISO	Denmark	^{40}K , ^{137}Cs , ^{134}Cs , ^{210}Pb , ^{226}Ra , ^{228}Th , ^{232}Th , $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am
ERPC	Estonian	^{40}K , ^{137}Cs , ^{134}Cs
STUK	Finland	^{40}K , ^{137}Cs , ^{134}Cs , ^{210}Pb , ^{226}Ra , ^{232}Th , ^{238}U , $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am
DHIG	Germany	^{40}K , ^{137}Cs , ^{134}Cs , ^{210}Pb , ^{226}Ra , ^{238}U , $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am
BFFG	Germany	^{40}K , ^{137}Cs , ^{134}Cs , ^{210}Pb , ^{226}Ra , ^{228}Th , ^{238}U , $^{239,240}\text{Pu}$, ^{238}Pu , ^{241}Am
LVEA	Latvia	^{40}K , ^{137}Cs , ^{134}Cs
CLOR	Poland	^{40}K , ^{137}Cs , $^{239,240}\text{Pu}$
SSI	Sweden	^{40}K , ^{137}Cs , ^{134}Cs , $^{239,240}\text{Pu}$

Table 3:
Laboratories participating in the sediment intercomparison in 2000

Figure 1:
Cs-137 results with 2 sigma
uncertainties in sea sediment
intercomparison

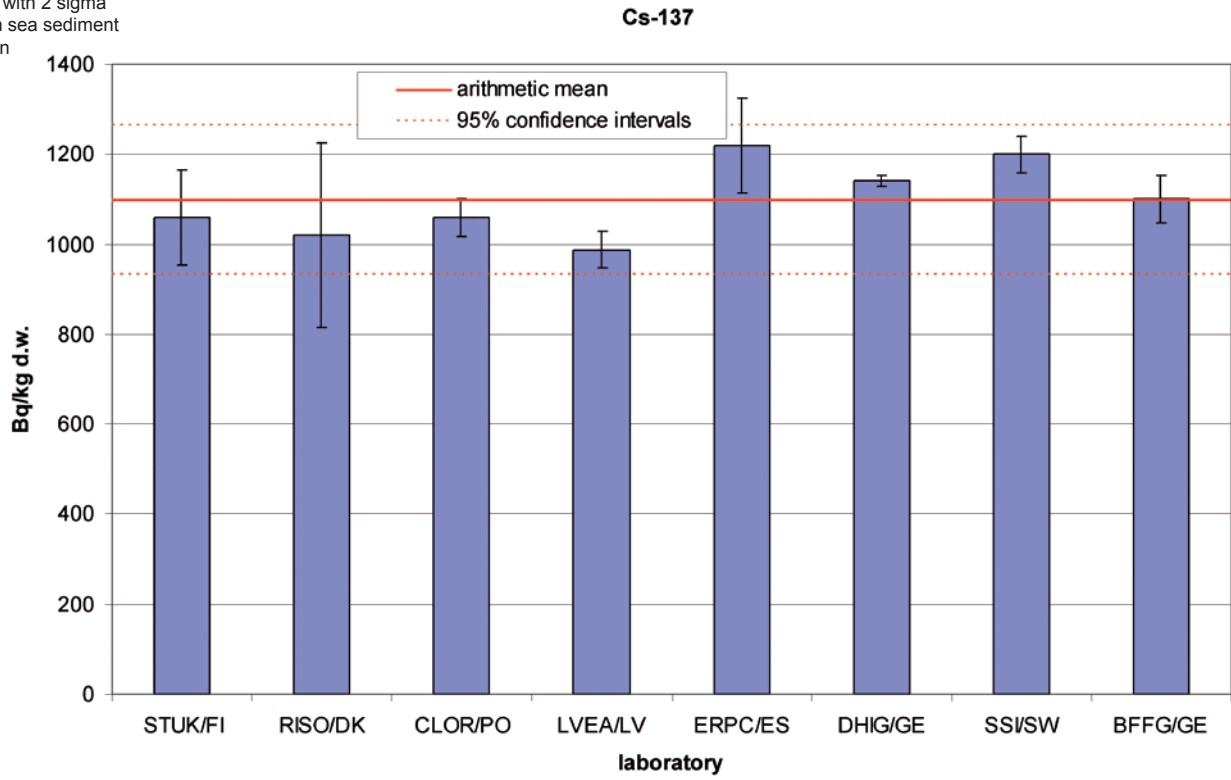
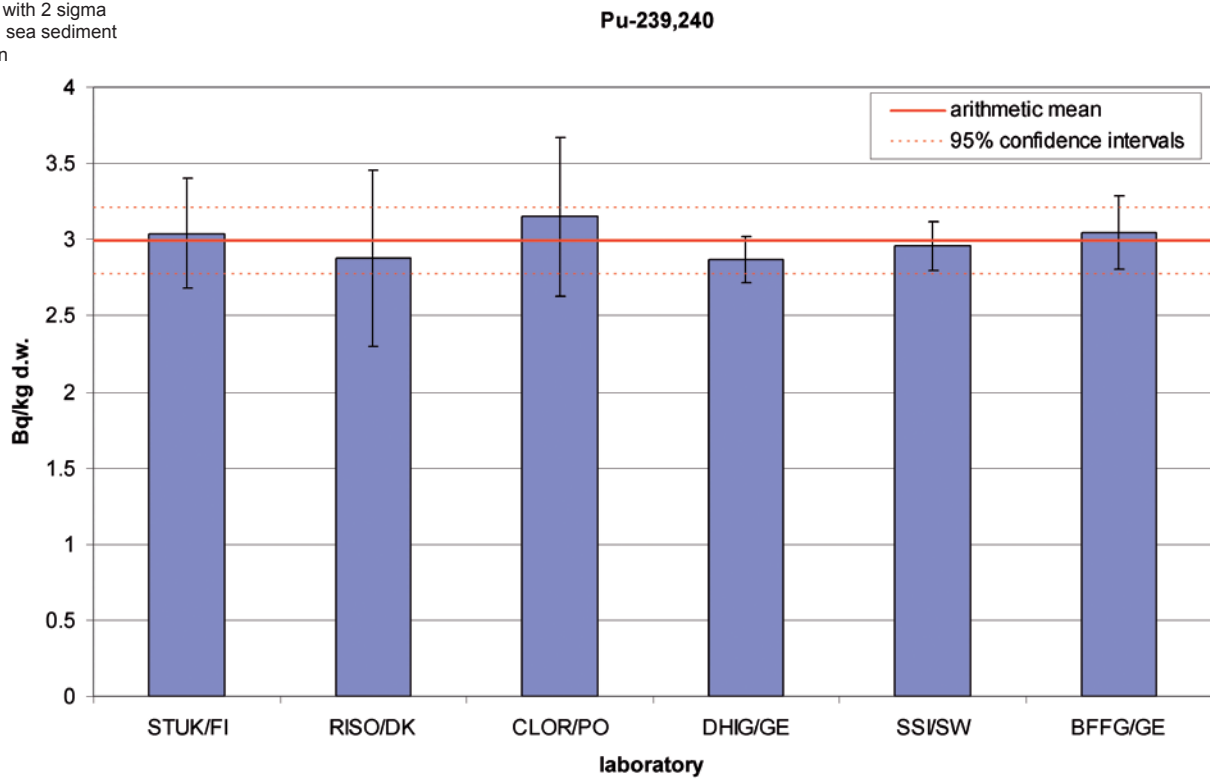


Figure 2:
^{239,240}Pu results with 2 sigma
uncertainties in sea sediment
intercomparison



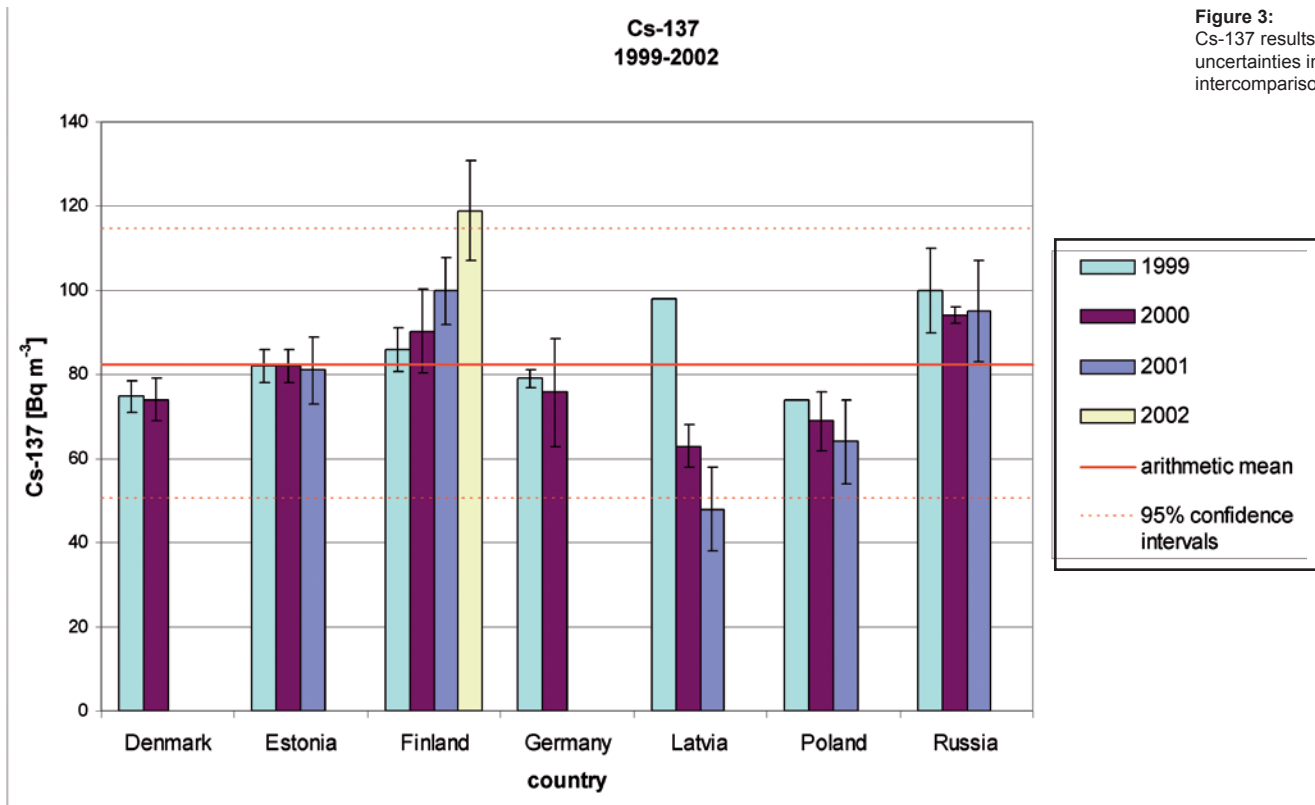


Figure 3:
Cs-137 results with 2 sigma uncertainties in seawater intercomparisons, 1999-2002

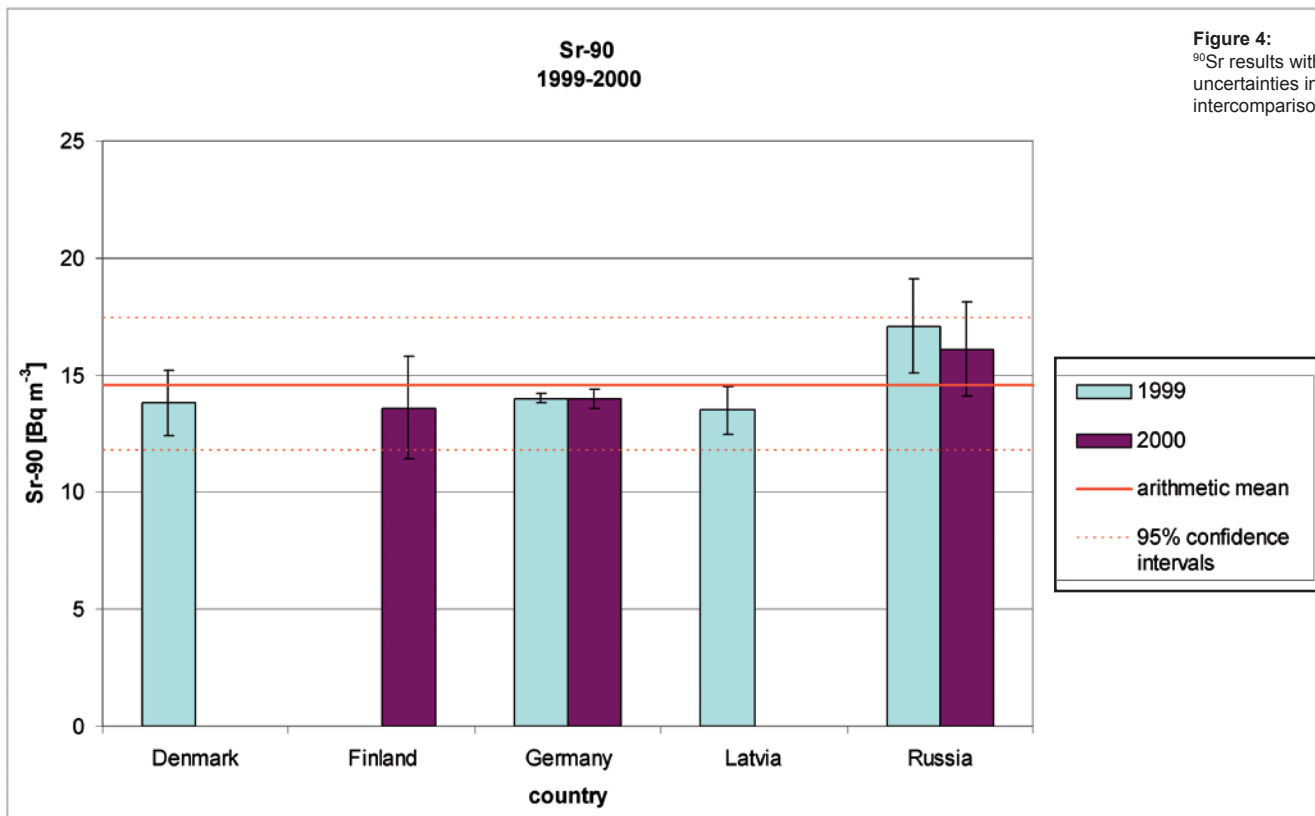


Figure 4:
⁹⁰Sr results with 2 sigma uncertainties in seawater intercomparisons, 1999-2000

Figure 5:
Cs-137 results with 2 sigma
uncertainties in seawater
intercomparisons, 2004-2006

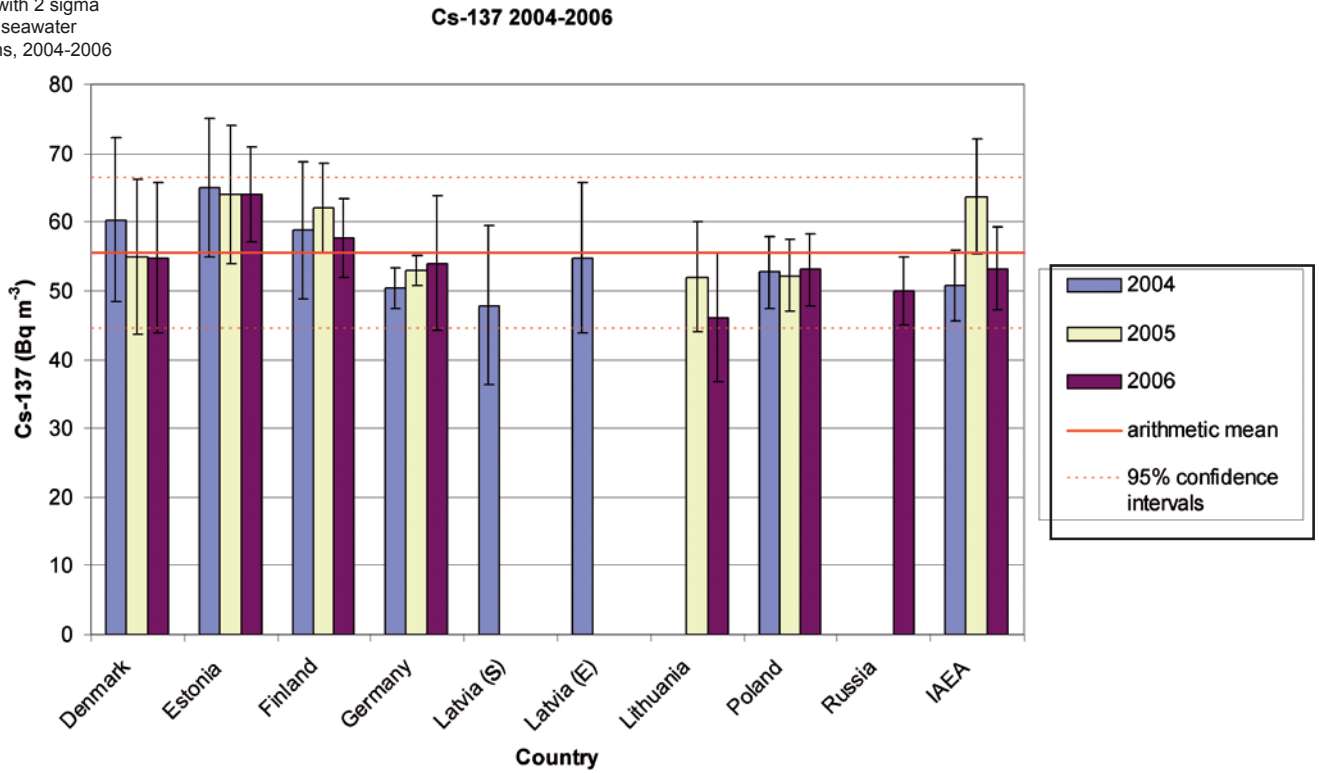
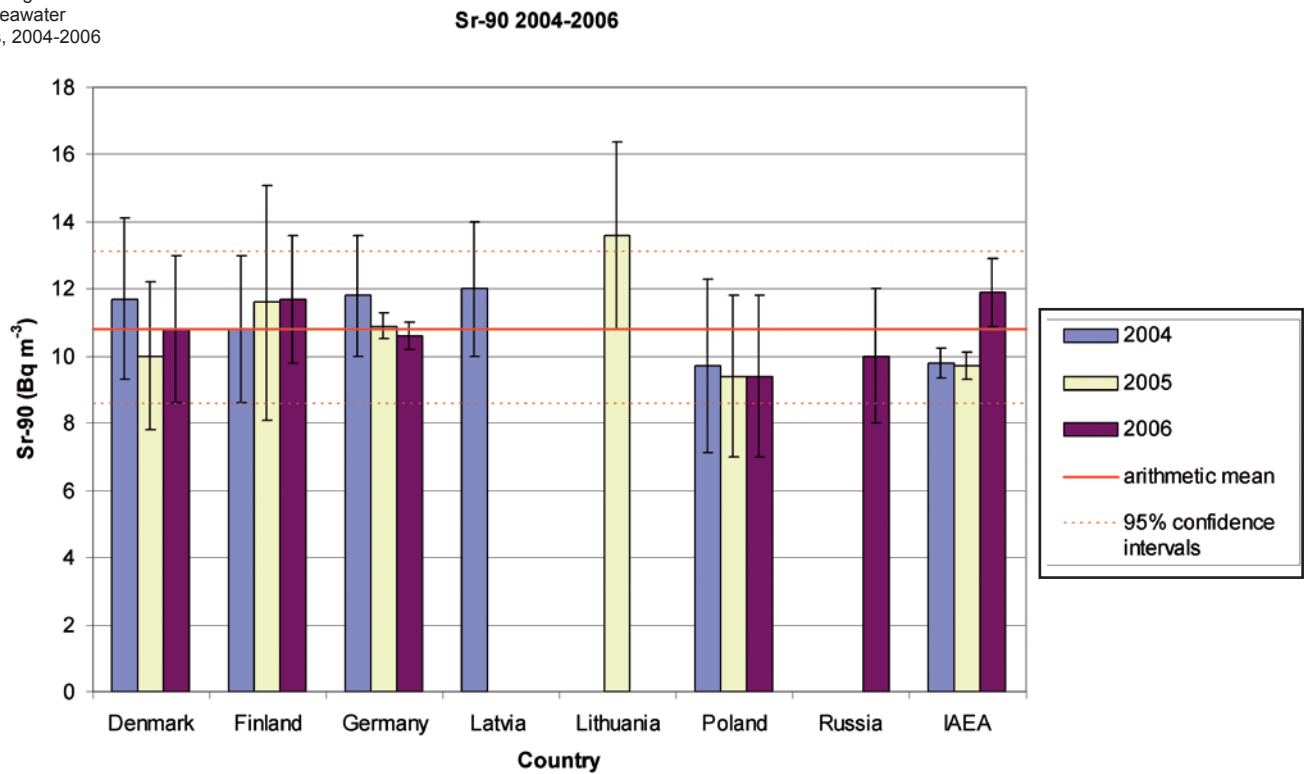
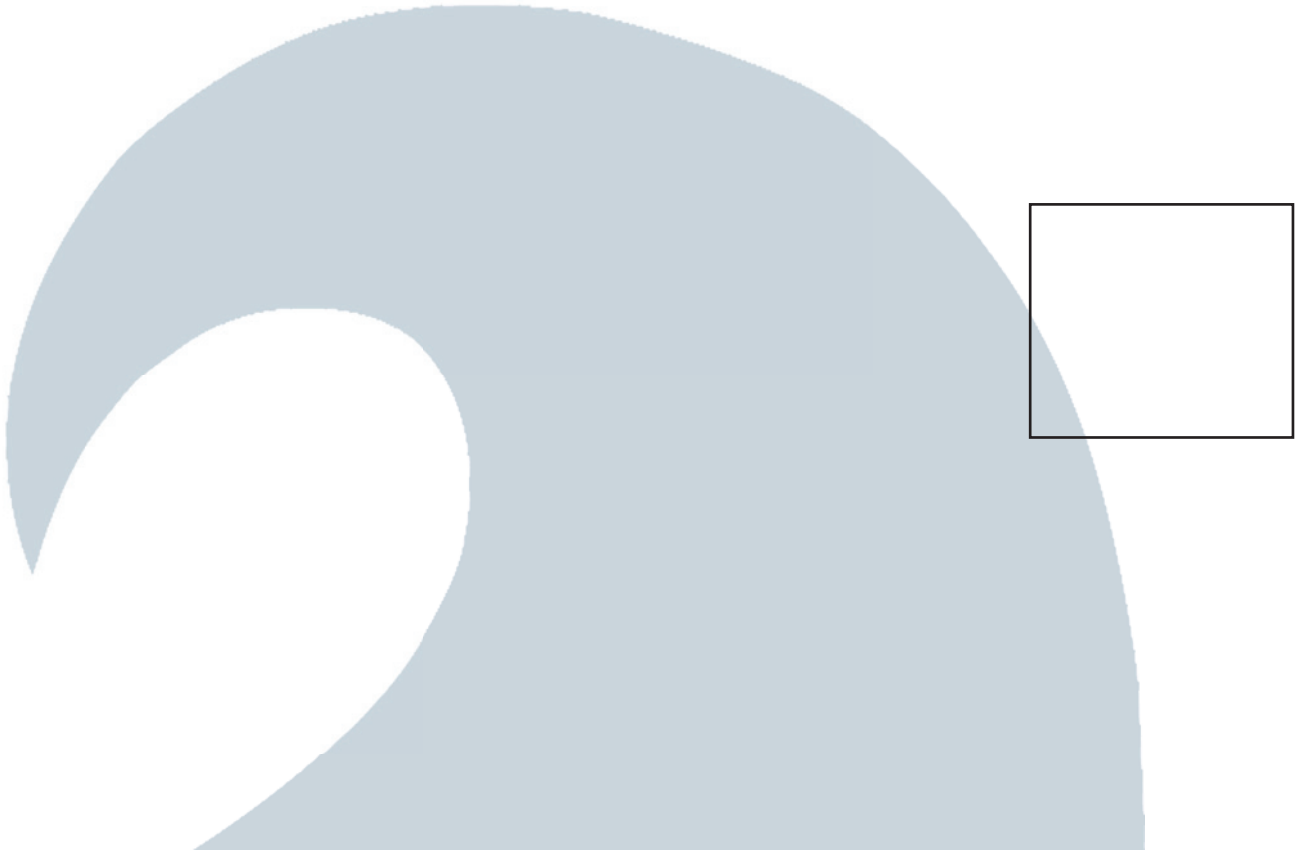


Figure 6:
⁹⁰Sr results with 2 sigma
uncertainties in seawater
intercomparisons, 2004-2006















HELSINKI COMMISSION
Baltic Marine Environment Protection Commission

Katajanokanlaituri 6 B
FI-00160 Helsinki
Finland

ISSN 0357-2994