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The Chernobyl accident and the Baltic Sea

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The impact of the radioactive fallout caused by the accident at the Chernobyl NPP on the Baltic Sea is discussed in this paper. The fallout from Chernobyl was very unevenly distributed in the drainage area of the Baltic Sea; the Bothnian Sea and the eastern part of the Gulf of Finland received most of the deposition. This was reflected in the activity concentrations of the main fallout nuclides (especially ^{137}Cs) that have been found in the marine environment of the Baltic Sea since then. The maximum concentrations that were detected soon after the fallout decreased significantly in a short time, and the distribution pattern of the Chernobyl-derived ^{137}Cs has changed over the course of time as a consequence of river discharges, mixing of water masses, sea currents and sedimentation processes. Sea currents have transported caesium from the Gulf of Finland and the Gulf of Bothnia into the Baltic Proper and further out of the Baltic Sea into the North Sea. In addition, a considerable amount of ^{137}Cs has been bound in the seabed of the Baltic Sea. In general, the concentrations of man-made radionuclides in the sediments have been at or below the concentrations of naturally-occurring radionuclides, and are not expected to cause harmful effects on the wildlife in the Baltic Sea. The exposure of the population to radiation caused by the ingestion of Baltic Sea fish after the Chernobyl accident was considered to be low compared with the mean annual exposure of Finns to radiation or to the dose caused by natural radionuclides in the sea.

Introduction

Intensive radioecological research has been carried out in the Baltic Sea since the late 1950s. Atmospheric nuclear weapons tests carried out in the northern hemisphere in the 1950s and 1960s, and the proximity of the Baltic Sea to one of the nuclear test areas, Novaya Zemlya, prompted these studies. In Finland, the studies have been carried out through close co-operation between STUK (the Radiation and Nuclear Safety Authority) and the Finnish Institute of Marine Research (FIMR). Finland has always played a central role in the monitoring of radioactivity in the Baltic Sea. The location of the Baltic Sea and its special features (relatively small volume of water, slow exchange of water, climatic conditions, large proportion of river water, low salinity and, consequently, scarce biota and its sensitivity to changes in environmental circumstances) also aroused more extensive interest in the Baltic Sea as a radioecological object of study.

In 1981, the International Atomic Energy Agency (IAEA) initiated a coordinated research programme on radioactive materials in the Baltic Sea, in which all the Baltic Sea countries participated. The final meeting of the project was held in Helsinki in 1984 and the results of the

study were published as an IAEA-TECDOC (1986). It was decided that the beneficial international cooperation that was initiated by the IAEA project would be continued under the auspices of the Baltic Marine Environment Protection Commission (HELCOM). The first meeting of the HELCOM/MORS (Monitoring of Radioactive Substances) Expert Group was held in Helsinki on 8–11 April 1986, only two weeks prior to the accident at the Chernobyl nuclear power plant (NPP) in the Ukraine.

The accident at the Chernobyl NPP on 26 April 1986 further increased radioecological interest in the Baltic Sea because it was the marine area most affected by the Chernobyl fallout (Povinec *et al*. 1996). The first radioactive clouds from Chernobyl travelled north and caused high deposition in the Baltic Sea area. The impacts of the deposition were more clearly visible in the marine environment of the Baltic Sea than in the marine environments of the Black Sea or the Mediterranean Sea, for instance.

The studies of the Chernobyl fallout in the Baltic Sea had an impressive launch. When the accident occurred, 12 research vessels from different countries were by chance together in the southern Baltic Proper implementing a PEX study (patchiness experiment) coordinated by HELCOM. As news of the accident spread, the executive secretary of HELCOM asked the contracting parties to start intensified studies on the quantities and behaviour of the radioactive substances in the Baltic Sea. The sampling requests sent from the countries to the research ships prompted the crews to rush to the shops in Visby on Gotland and buy all the plastic cans of 10–30 litres that they could lay their hands on. The crew of the Finnish research vessel *Aranda* participated in this race, and on their return journey to Helsinki they took ten extra seawater samples to STUK for radionuclide analyses (Ilus *et al*. 1987b).

The aim of this paper was to make a general historical survey of the impacts of the Chernobyl fallout on the marine environment of the Baltic Sea based on the results of various Finnish studies. Some of the material presented here has not been published before, and some of it has been published in non-refereed papers, e.g. in HELCOM Proceedings.

Material and methods

The material for this summary report consists of the results from several Finnish studies carried out in the Baltic Sea after the Chernobyl accident, such as the Finnish contribution to the HELCOM/MORS monitoring programme, the monitoring programmes carried out in the sea areas off the Finnish nuclear power plants (NPP) and some other special studies. The samples for the HELCOM/MORS monitoring programme have been taken annually on board the Finnish research vessel *Aranda* by the staff of the Finnish Institute of Marine Research (FIMR), or in the coastal areas by the staff of STUK or other local people. The surveillance of radioactive substances in the vicinities of the Finnish NPPs has been carried out under extensive permanent monitoring programmes, in which the marine samples are taken by the staff of STUK. The sampling methods used on board *Aranda*, as well as the sampling, pre-treatment and analysis methods used at STUK, have been accredited by FINAS (accredited testing laboratories T40 (FIMR) and T167 (STUK)).

On board the *Aranda*, seawater samples were taken from various depths using the submerged pump-hose system of the ship or large water samplers. In the monitoring programmes of the NPPs, seawater samples were taken only from the surface by filling sample kegs with a pail through a large funnel. The volume of the seawater samples for gammanuclide analyses was 30 liters. The samples were acidified with concentrated nitric acid, and a carrier solution containing 100 mg of stable Cs was added to the samples after their arrival at the laboratory. The samples were concentrated by evaporation to 500 ml and analysed gammaspectrometrically using Marinelli geometry.

Fucus vesiculosus samples (1.5 kg fresh weight) were taken from a depth of 1–3 metres by scuba diving. The diver collected the algae one by one in a net bag. On the boat, the seaweeds were shaken onto porous paper and placed in plastic bags immediately after sampling. In the laboratory, the samples were preliminarily dried at room temperature and the final drying was performed overnight at 105 °C. After drying, the dry weight was recorded and the samples were

homogenized by milling. The homogenized samples were analysed gammaspectrometrically in Marinelli beakers.

The sediment samples have been taken with a Gemini Twin Corer since 1992 and with a Niemistö Corer before that. The corers are described in Ilus *et al*. (2000). The sediment cores were sliced into 1- or 5-cm sub-samples to a depth of 10–30 cm and put into plastic bowls. The samples were frozen immediately after sampling. In the laboratory the samples were freeze-dried and homogenized before the gammaspectrometric analysis in small cylindrical containers.

The fish samples were supplied to STUK by local research stations or fishermen along the Finnish coast, and the seal samples by the Finnish Game and Fisheries Institute from the south and southwest coast of Finland. The size of the fish samples was 5 kg of uncleaned fish. The heads and guts were removed from Baltic herring, whereas the pike were scaled and cut into fillets before freezing. In the laboratory, the samples were cut into pieces, dried at 105 °C and ashed (< 450 °C) before the gammaspetrometric analysis in small cylindrical containers. The seal flesh samples were cut into small pieces and analysed as fresh in cylindrical beakers.

The analysis of gamma-emitting radionuclides was performed using HPGe spectrometers in low-background shields (Rantavaara *et al*. 1994). The relative efficiencies of the detectors ranged from 20% to 78%. The spectra were analysed using the GAMMA-99 computer program, taking into account true coincidence summing, variable sample height and density (Sinkko 1981, Sinkko and Aaltonen 1985).

Results and discussion

The fallout from Chernobyl was very unevenly dispersed throughout the drainage area of the Baltic Sea, the Bothnian Sea and the eastern part of the Gulf of Finland receiving most of the deposition. The total injection of Chernobyl-derived ^{137}Cs was estimated at about 4700 TBq, while that of the global fallout caused by nuclear weapons tests was estimated at about 900 TBq. Until 1998, the proportion of Chernobyl fallout in the total input of ^{137}Cs into the Baltic Sea was 82%, while that of nuclear weapons tests was 14%. The cumulative amount of discharges from local nuclear power plants in the Baltic Sea region was only 0.03% (Nies *et al*. 1995, Ilus and Ilus 2000, Lüning and Ilus 2003).

Radionuclides in water

Soon after the accident, the activity concentrations of ^{137}Cs in Finnish coastal waters rose by a factor of about 100–500 as compared with those of the previous summer. During the first half of May, the highest ^{137}Cs concentration measured in surface seawater was 5200 Bq m^{-3}, but the caesium values also decreased very rapidly (i.e. in three months) to about a tenth of this. The activity concentrations of the fallout nuclides were generally higher in coastal waters than in the open sea due to the coastal rains, input of river waters and more effective mixing of surface waters in the open sea.

In the initial phase during the first weeks after the fallout situation, the marine samples also contained ^{131}I in sizeable quantities. The maximum value of ^{131}I observed in seawater samples was 24 000 Bq m^{-3} in a coastal sample from the Gulf of Finland (Ilus *et al*. 1987a). Because of its short half-life, iodine was not detected in seawater samples after the beginning of June (Ilus *et al*. 1987b).

The sinking rate of the fallout nuclides was relatively high owing to the coincidence of the end phase of the phytoplankton spring maximum, when the radionuclides were transported downwards by the dead plankton algae. In the first half of May, fresh fallout nuclides were already observed in water samples taken from a depth of 100 m in the southern Baltic Proper and in mid-June in samples of surface sediment at a depth of 170 m in the northern Baltic Proper (Ilus *et al*. 1989).

Since 1986, the distribution pattern of the Chernobyl-derived caesium has changed as a consequence of river discharges, mixing of water masses, sea currents and sedimentation processes. Saxén and Ilus (2001) estimated that the total amount of ^{137}Cs transported by rivers from Finland into the Baltic Sea in 1986–1996 was 65

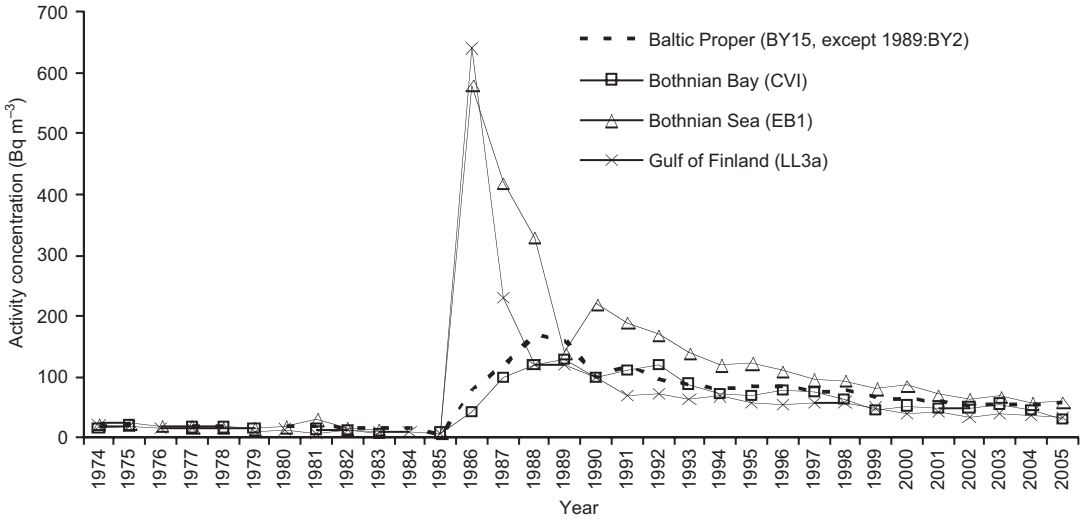

Fig. 1. Activity concentrations of ^{137}Cs (Bq m^{-3}) in surface water at four open sea stations of the Baltic Sea in 1974–2005.

TBq. In the sea, caesium was transported by sea currents from the Gulf of Finland and the Gulf of Bothnia into the Baltic Proper and further out from the Baltic Sea into the North Sea through the Danish Straits.

Due to better exchange of water, the Gulf of Finland has become cleansed of caesium faster than the Gulf of Bothnia (Fig. 1). At the same time as the ^{137}Cs concentrations decreased at stations LL3a and EB1 of the Gulf of Finland and the Bothnian Sea, they increased in the Baltic Proper (BY15) as a consequence of out-flowing waters from the gulfs containing higher quantities of caesium. In 2005, the activity concentrations of ^{137}Cs in surface water were 28–39 Bq m^{-3} in the Gulf of Finland and 45–60 Bq m^{-3} in the Bothnian Sea.

Radionuclides in algae

Bladder-wrack (*Fucus vesiculosus*) has widely been used as an indicator organism in radioecological studies, because it collects radioactive substances effectively from the surrounding seawater. During the initial phase after the Chernobyl accident, the vertical transport of fallout nuclides in the water column and their intake into algae was very rapid. The activity concentrations of gamma-emitting radionuclides in samples of bladder wrack taken from a regular sampling site (depth 2 m) in front of the Olkiluoto NPP (west coast of Finland) are given in Table 1. In August 1985, the sample contained only small amounts of discharge nuclides from the local power plant (^{60}Co, ^{54}Mn, ^{58}Co, ^{65}Zn and ^{110m}Ag), 7 Bq kg^{-1} of ^{137}Cs from weapons tests fallout and a usual amount of naturally-occurring ^{40}K. The sample taken at 18:00 on 28 April 1986 (only a few hours after the arrival of the fallout) contained a long list of fresh fallout nuclides which were exotic to this area and most of them short-lived. The activity concentration of ^{137}Cs was still only 25 Bq kg^{-1} and that of ^{131}I was 410 Bq kg^{-1}. It is worth noting that this sample contained only dry deposition because the first rain after the arrival of the fallout fell on the morning of 29 April. On 29 May, the concentrations of ^{131}I, the caesium and ruthenium isotopes and ^{140}Ba and ^{128m}Te had reached their maximum values (29 000 Bq kg^{-1} of ^{131}I and 1300 Bq kg^{-1} of ^{137}Cs), whereas many short-lived nuclides had already disappeared or started to diminish. In August 1986, the short-lived ^{131}I (half-live of eight days) had almost completely disappeared and the ^{137}Cs concentration was about a quarter of its highest value in May. In June 1987, the *Fucus vesiculosus* sample taken from the same sampling site contained only 220 Bq kg^{-1} of ^{137}Cs, 86 Bq kg^{-1} of ^{134}Cs and a slightly higher amount of ^{110m}Ag,

in addition to the background levels of the local discharge nuclides.

The highest ^{137}Cs concentration observed in *Fucus vesiculosus* samples from the Finnish coast was about 5000 Bq kg^{-1} dry weight at the end of May 1986. A joint Nordic study carried out in 1991 showed that the concentrations had clearly decreased and levelled out, but the highest values were still on the west coast of Finland and in the Åland archipelago (Fig. 2). The activity concentrations of ^{137}Cs varied on the Finnish coast between 47 and 170 Bq kg^{-1} dry weight and decreased toward the southern parts of the Baltic Sea and when passing out of the Baltic Sea into the North Sea (Carlson *et al*. 2002). The concentrations were only 1 Bq kg^{-1} or smaller in the Arctic sea areas. The Finnish contribution to this study was significant, consisting of *Fucus vesiculosus* samples taken from 26 sites along the Finnish coast.

Radionuclides in fish and seal

The highest activity concentrations of ^{137}Cs found in Baltic Sea fish after the Chernobyl accident were about 300 Bq kg^{-1} fresh weight in pike (*Esox lucius*) caught in 1990 from the sea area off Vaasa. In 2005, the caesium concentrations were 13–29 Bq kg^{-1} fresh weight in pike and 5–8 Bq kg^{-1} fresh weight in Baltic herring (*Clupea harengus membras*) caught along the Finnish coasts. Seals, which almost exclusively eat fish, represent the top consumers in the brackish water ecosystems of the northern Baltic Sea. Thus, the potential accumulation of radioactive substances in seals gave rise to public concern in Finland in the late 1980s after the Chernobyl accident. However, samples of seal flesh supplied by the Finnish Game and Fisheries Institute to STUK showed that the concentrations of ^{137}Cs were only 5–10 times higher than in Baltic herring at that

Table 1. Gamma-emitting radionuclides in *Fucus vesiculosus* (Bq kg^{-1} dry weight) at a sampling station in Olkiluoto (west coast of Finland) in 1985–1987.

	15 Aug. 1985	28 Apr. 1986 (18:00)	21 May 1986	14 Aug. 1986	9 July 1987
^{40}K	650	590	690	690	750
^{54}Mn	68	18	37	57	140
^{58}Co	25	0[a]	0	14	8
^{60}Co	100	70	110	94	84
^{65}Zn	21	3.3	23	26	12
^{95}Zr	0	690	110	4.2	0
^{95}Nb	0	920	220	7.6	0
^{95m}Nb	0	10	0	0	0
^{103}Ru	0	570	1900	52	0
^{106}Ru	0	110	590	61	0
^{110m}Ag	9.5	2.2	170	62	23
^{125}Sb	0	0	24	4.0	0
^{129m}Te	0	22	2800	0	0
^{131}I	0	410	29000	6.8	0
^{132}Te	0	500	300	0	0
^{134}Cs	0	8.3	710	140	86
^{136}Cs	0	3.4	150	0	0
^{137}Cs	7.0	25	1300	280	220
^{140}Ba	0	1000	4700	0	0
^{140}La	0	810	3700	17	0
^{141}Ce	0	690	94	1.7	0
^{144}Ce	0	360	130	7.1	0
^{144}Pr	0	320	65	0	0
^{147}Nd	0	230	0	0	0
^{237}U	0	38	0	0	0
^{239}Np	0	3500	0	0	0

[a] below the detection limit, [b] not analysed.

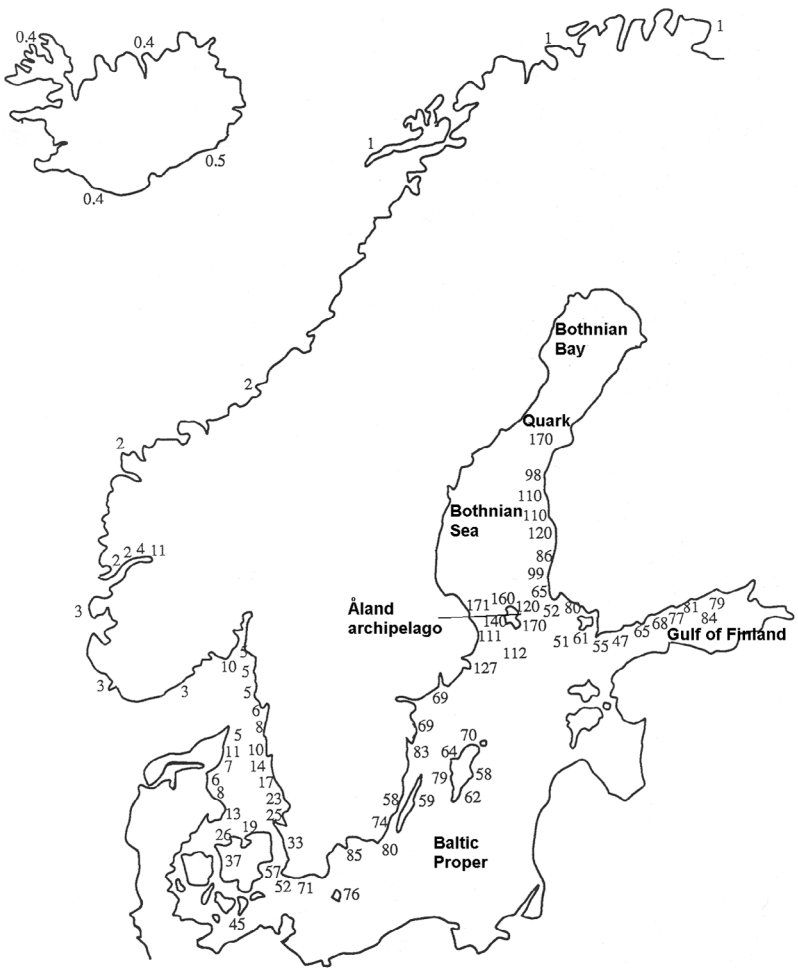

Fig. 2. Activity concentrations of ^{137}Cs (Bq kg^{-1} dry weight) in *Fucus vesiculosus* along the Nordic coasts in 1991 (Carlson *et al.* 1992).

time (Ilus *et al.* 2005). The average ^{137}Cs concentrations in grey seal and ringed seal and the concentration ratios between the seal flesh and seawater and Baltic herring are given in Table 2.

Radionuclides in sediments

Chernobyl-derived caesium is very unevenly distributed in the sediments of the Baltic Sea

Table 2. Activity concentrations of ^{137}Cs (Bq kg^{-1} fresh weight) in seal flesh on the Finnish coast after the Chernobyl accident and the concentration ratios between the seal flesh and seawater, and the seal flesh and the Baltic herring.

	Ringed seal (*Phoca hispida*) Gulf of Finland (May 1986–November 1988)	Grey seal (*Halichoerus grypus*) SW coast of Finland (1987–1988)
Activity concentration (average (range)) of ^{137}Cs (Bq kg^{-1} f.w.)	98 ± 5 (71–120)	176 ± 63 (117–280)
CR (Bq kg^{-1} f.w. in seal/ Bq kg^{-1} in seawater)	415 (310–520)	784 (420–1150)
CR (Bq kg^{-1} f.w. in seal flesh/ Bq kg^{-1} f.w. in Baltic herring)	4.8 ± 1.0	8.8 ± 3.2

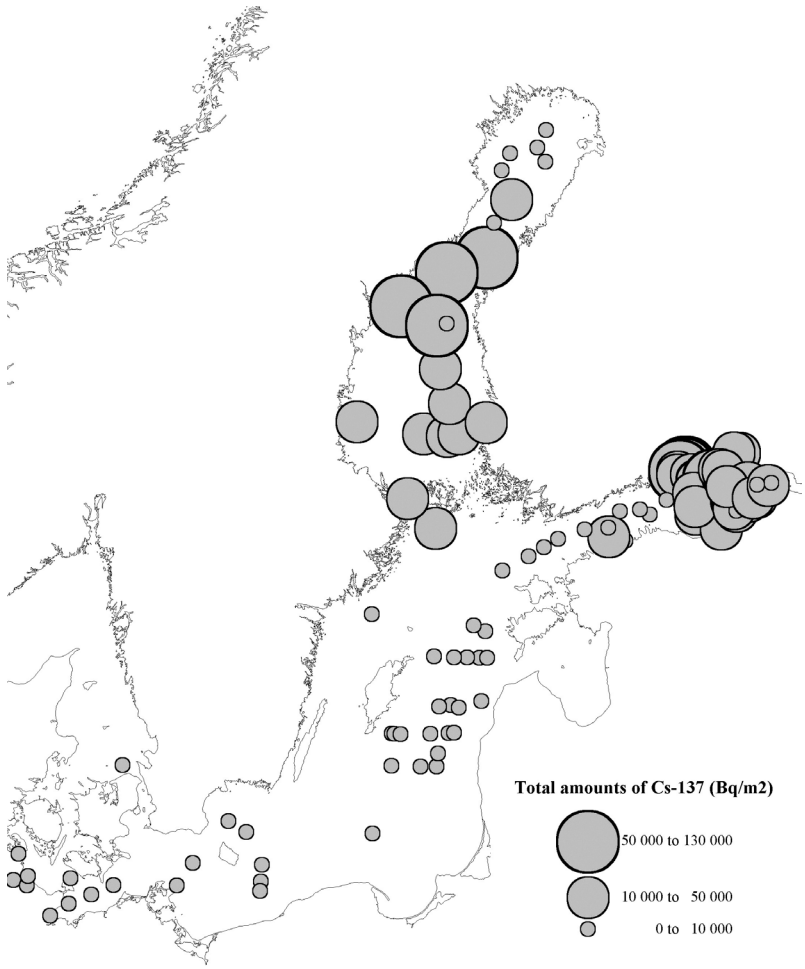

Fig. 3. Total amount of ^{137}Cs (Bq m^{-2}) in the bottom sediments of the Baltic Sea in 1998 (Ilus *et al.* 2003).

(Fig. 3). In addition to the scattered nature of the deposition, this has been affected by the character of the bottom, the sedimentation conditions and the sedimentation rate at the stations studied. Ilus *et al*. (2003) estimated that in total about 1940–2210 TBq of ^{137}Cs was bound in the seabed of the Baltic Sea in 1998. The highest amounts were measured in the bottom sediments in the northern parts of the Bothnian Sea, the southern parts of the Bothnian Bay and eastern parts of the Gulf of Finland (the maximum value in the northernmost part of the Bothnian Sea was 125 000 Bq m^{-2}). In the western Gulf of Finland and in the Baltic Proper the amount of ^{137}Cs in sediments was much lower (Fig. 3).

The Chernobyl fallout has created clear time markers in the sediments (Fig. 4), which can be used in the timing of sediments and determination of sedimentation and accumulation rates (e.g. Perttilä and Niemistö 1993, Kankaanpää 1997, Kankaanpää *et al*. 1997, Vallius 1999, Ilus 2001, Ilus *et al*. 2001, Perttilä *et al*. 2003, Mattila *et al*. 2006). In 1986, the sinking of Chernobyl caesium had just started on the surface of the sediment. In 1988 and 1990, the caesium peak was already buried to depths of 1–3 cm and 4–5 cm, respectively, when fresh particulate matter with lower contents of caesium had settled to the surface of the bottom. The slightly lower and broader peak in 1995 may give a hint of minor smearing of deeper sediment layers (below the peak in 7–8 cm) taking place during sampling (cf. Ilus *et al*. 2000). In 2002, the peak was at a depth of 13–14 cm, which means a sedimentation rate of about 0.9 cm year^{-1} at the site. As sediment compaction has not been taken into

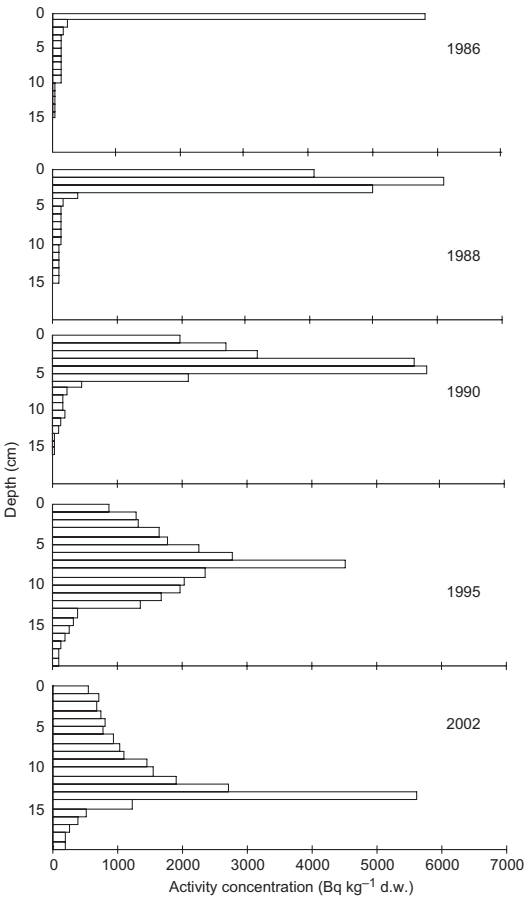

Fig. 4. Vertical distribution of ^{137}Cs (Bq kg^{-1} dry weight) in sediment profiles at a coastal station in the Gulf of Finland in 1986–2002.

account, this value may slightly overestimate the actual sedimentation rate over a longer period.

Radiation doses

The exposure of the population to radiation caused by ingesting Baltic Sea fish and other marine products and from living by the sea after the Chernobyl accident was evaluated in the EU sponsored Marina Balt Project in 1996–1998 (Nielsen 2000). The dose for the critical groups living on the coasts of the Bothnian Sea and the Gulf of Finland peaked in 1986 at a value of 0.2 mSv (a critical group is a theoretical group living by the sea and eating maximum amounts of fish and other marine products). This can be considered a very low value as on average a Finn receives an annual 3.7 mSv dose from other sources, and it should be borne in mind that since 1986 the effect of the Chernobyl fallout has significantly decreased in the northern Baltic Sea. At the same time, individuals in the critical groups had a dose rate of 0.7 mSv year^{-1} from ^{210}Po occurring naturally in the sea.

Conclusions

The most significant source of artificial radioactivity in the Baltic Sea has been the fallout from the Chernobyl accident in 1986. The distribution pattern of Chernobyl derived ^{137}Cs in the Baltic Sea area was very scattered, with the highest values occurring in the areas of the Bothnian Sea and the eastern Gulf of Finland. During the twenty years since the Chernobyl accident, the amounts of man-made radionuclides have decreased considerably in the marine environment of the Baltic Sea. Besides the reduction caused by the ecological half-life of ^{137}Cs, considerable amounts of it have been transported from the Baltic Sea within out-flowing water masses through the Danish Straits, or deposited into the seabed especially in the Gulf of Bothnia and the eastern Gulf of Finland. In the course of time, the amounts of caesium in the sediments will slowly decrease as a consequence of the radioactive decay of ^{137}Cs (physical half-life of 30 years), and pass out of biological availability when buried in deeper sediment layers. Accumulation of ^{137}Cs into biota should be considered, but it is relatively weak and partly reduced due to its effective removal from the organisms. In general, the concentrations of man-made radionuclides in the sediments have been at or below the concentrations of naturally occurring radionuclides, and are not expected to cause harmful effects on the wildlife of the Baltic Sea. The radiation dose received by people in the critical groups living on the coasts of the Bothnian Sea and the Gulf of Finland peaked in 1986, but was considered to be low compared with the mean annual exposure of Finns to radiation or the dose caused by natural radionuclides in the sea.

The Chernobyl accident provided scientists and decision-makers with knowledge of the behaviour and effects of radioactive fallout in the

marine environment of a semi-enclosed brackish-water area such as the Baltic Sea. In addition, it proved the importance of continuous monitoring of radioactive substances in the Baltic Sea by carrying out the HELCOM/MORS programme in normal circumstances as well.

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