Invited paper

Anthropogenic radionuclides ^{137}Cs and ^{90}Sr in the southern Baltic Sea ecosystem

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Abstract

The radioisotopes of caesium (^{137}Cs) and strontium (^{90}Sr) make the greatest contribution to the radioactivity level due to artificial radionuclides in the Baltic Sea, where the level of ^{137}Cs contamination is higher than in any other part of the world ocean. The main sources of man-made radionuclides are the Chernobyl accident in 1986 and the nuclear weapons tests carried out in the 1950s and 1960s. This study discusses the distribution patterns and trends in activity concentrations of ^{137}Cs and ^{90}Sr recorded in various compartments of the marine environment of the southern Baltic Sea. It is based on an investigation of radioactive substances as part of the Polish National Environmental Monitoring Programme. In 2010 the average concentration of ^{137}Cs in the southern Baltic was 35 Bq m^{-3}, while the level of ^{90}Sr in these waters has remained at much the same level in recent years (ca

The complete text of the paper is available at http://www.iopan.gda.pl/oceanologia/

8 Bq m^{-3}). The distribution of isotopes in the bottom sediments reflect historical events that can be identified in sediment profiles. The activity concentrations of the caesium isotope are the highest in sediments from the Gulf of Gdańsk, whereas the least polluted sediments are found in the Bornholm Basin, in the western part of the southern Baltic. The highest concentrations of ^{137}Cs in benthic plants were measured in the red alga *Polysiphonia fucoides*: 22.3 Bq kg^{-1} d.w. in June and 40.4 Bq kg^{-1} in September. These levels were much higher than those found in the bivalve *Mytilus trossulus* (7.3 Bq kg^{-1} d.w.). ^{137}Cs concentrations in fish have decreased in time, reflecting the trends recorded in seawater. In 2010 the respective ^{137}Cs activities in *Clupea harengus*, *Platichthys flesus* and *Gadus morhua* were 4.7, 4.9 and 6.6 Bq kg^{-1} w.w.

1. Introduction

When considering the level of radioactive pollution in the natural environment, including marine areas, attention is focused primarily on artificial radionuclides. The radioisotopes of caesium (^{137}Cs) and strontium (^{90}Sr) have made the greatest contribution to the radioactivity level due to artificial radionuclides in the Baltic Sea (HELCOM 1995, 2003, 2009, Nielsen et al. 1999, Zalewska & Lipska 2006, Ikaheimonen et al. 2009, Atwood 2010). This is the consequence of large quantities of these nuclides being introduced into the Baltic, and also because of the relatively long half-lives of these isotopes – 30.05 and 28 years respectively. The Baltic Sea is a unique marine ecosystem. It is an inland basin of temperate climate with such characteristic features as low salinity, a relatively large area and shallow average depth (Szefer 2002a). The annual freshwater input to the Baltic from riverine discharges and atmospheric precipitation amounts to 450–510 km^3. Water exchange with the world ocean – specifically with the North Sea and the NE Atlantic Ocean – is accomplished via the narrow and shallow Danish Straits (the Sund, and the Great and Little Belts); hence this exchange is much hampered. Nonetheless, this inflow activity is the only source of salt water transport from the North Sea to the Baltic Sea (Feistel et al. 2003, Piechura & Beszczyńska-Möller 2004).

At present, the Baltic Sea continues to show a higher level of ^{137}Cs contamination than any other part of the world ocean (HELCOM 1995, 2003, 2009, IAEA 2005, Zalewska & Lipska 2006, Ikaheimonen et al. 2009). In 2000 the average seawater concentration of ^{137}Cs (representative of the entire Baltic Sea area) was 60 Bq m^{-3}, while average ^{90}Sr levels were 10 Bq m^{-3} (IAEA 2005). In 2004, the average activity of ^{137}Cs in the southern Baltic was 45 Bq m^{-3} (Zalewska & Lipska 2006), a level three times higher than that prior to the Chernobyl accident of 1986.

Radioactivity levels and the distribution of radioisotopes in the Baltic Sea are influenced by their input sources and their behaviour in the marine environment, and also by the specific features of this sea. Anthropogenic radioisotopes, both those entering seawater with atmospheric fallout as well as those discharged in riverine outflow, spread into other compartments of the marine environment. Radionuclides can be bioaccumulated and/or absorbed by suspended particulate matter (SPM), which is composed mainly of plankton, biogenic detritus and mineral particles (Knapińska-Skiba et al. 1994, 1995, 2002, 2003, Szefer 2002b); sedimentation processes cause them to accumulate in bottom sediments (Knapińska-Skiba et al. 2001, Szefer 2002b). Radionuclides can also be bioaccumulated by higher flora and fauna. Knowledge of the radioisotope distribution in the biotic and abiotic compartments of the marine environment is fundamental to understanding the dynamics of their fate in that environment, and to determining the effects of environmental and physiological factors on radionuclide bioaccumulation and sedimentation. Radionuclide concentrations are key factors in any assessment of the radiological hazard to marine organisms and humans (Nielsen et al. 1999). They are indispensable in environmental status assessments, recently very frequently required on legal grounds, and also prompted by public interest. The best example of such regulatory requirements is the Marine Strategy Framework Directive (Anon 2008), which requires that concentrations of contaminants, including radionuclides, in the marine environment should be at levels not giving rise to pollution effects.

The main objective of this study is to assess the distribution patterns and trends in activity concentrations of the anthropogenic isotopes ^{137}Cs and ^{90}Sr, as recorded in various compartments of the southern Baltic Sea environment (Figure 1), i.e. seawater, sediments, submerged aquatic vegetation and benthic invertebrates, since the Chernobyl accident. The Chernobyl accident (1986) led to a significant increase in radioactivity levels in the Baltic Sea region, and the changes in radioactivity concentrations in various compartments of the southern Baltic Sea reflect the general trends noted in the entire Baltic basin. Measurement of radioactive pollution in the Baltic Sea basin was initiated in 1980. Between 1981 and 1984 the International Atomic Energy Agency coordinated these activities, but since 1985 the Helsinki Commission (HELCOM) has been responsible for monitoring radioactive substances. HELCOM established an Expert Group on the Monitoring of Radioactive Substances in the Baltic Sea. The data presented were obtained as part of the National Environmental Monitoring programme coordinated by the Chief Inspectorate for Environmental Protection in Poland.
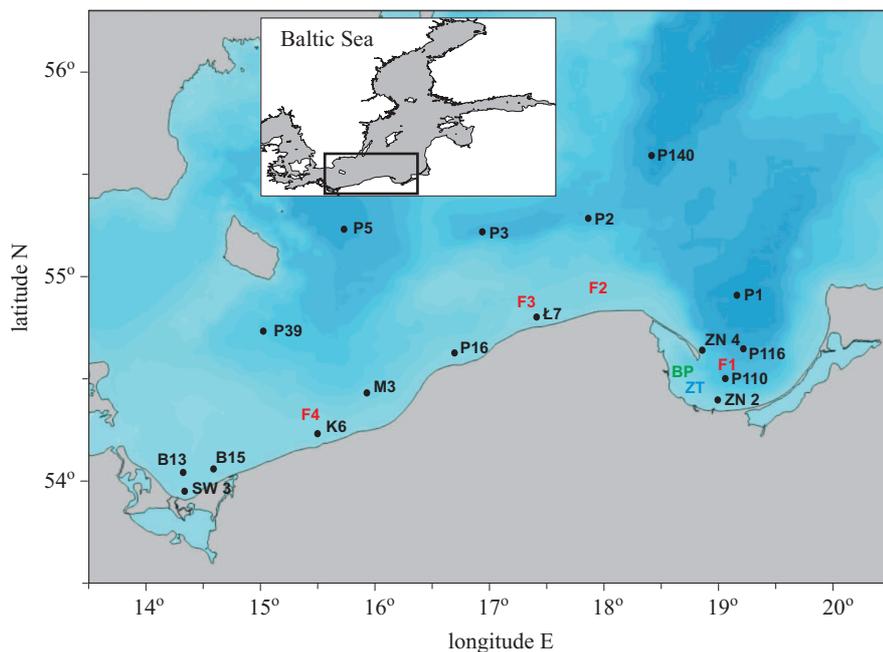
Figure 1. Sampling locations for seawater and bottom sediments (black), benthic plants (green), zoobenthic organisms (blue) and fish (red)

2. Sources of anthropogenic radionuclides

The main sources of anthropogenic radionuclides in the Baltic Sea can be categorized into four groups (HELCOM 2009): (i) discharges from nuclear facilities in the Baltic Sea drainage area (nuclear power plants (NPPs) and research reactors), (ii) discharges from nuclear reprocessing plants located beyond the Baltic Sea: Sellafield, on the north-west coast of England, and La Hague, on the north-west coast of France, (iii) the Chernobyl accident and (iv) atmospheric testing of nuclear weapons.

The atmospheric testing of nuclear weapons was done mainly in the northern hemisphere between 1945 and 1980, with particular intensity in 1951–1958 and later in 1961–1962. As a result of these tests, in addition to serious local pollution incidents, large quantities of radioactive isotopes were discharged into the troposphere and stratosphere, causing the global radioactive contamination of the atmosphere. It is assumed that the atmospheric testing of nuclear weapons delivered as much as 0.49–0.60 PBq of ^{90}Sr and 0.62–0.90 PBq of ^{137}Cs to the Baltic Sea.

Clearly, the Chernobyl nuclear plant accident in April 1986 was the largest source of radioactive isotopes supplied to the Baltic Sea environment, particularly in view of their contamination effects. The total activity of the

radioisotopes released amounted to ca 3000 PBq (UNSCEAR 1988). The majority of them were short-lived ones. Of the long-lived ones, ^{137}Cs was the most important as its total activity was approximately 38 PBq. Estimated amounts of ^{137}Cs entering the Baltic Sea as a consequence of this accident were of the order of 4.7 PBq (including 0.3 PBq via riverine discharges) (HELCOM 1995, 2003, 2009, Nielsen et al. 1999). This is equivalent to 82% of the total amount of ^{137}Cs accumulated in the Baltic (Figure 2).
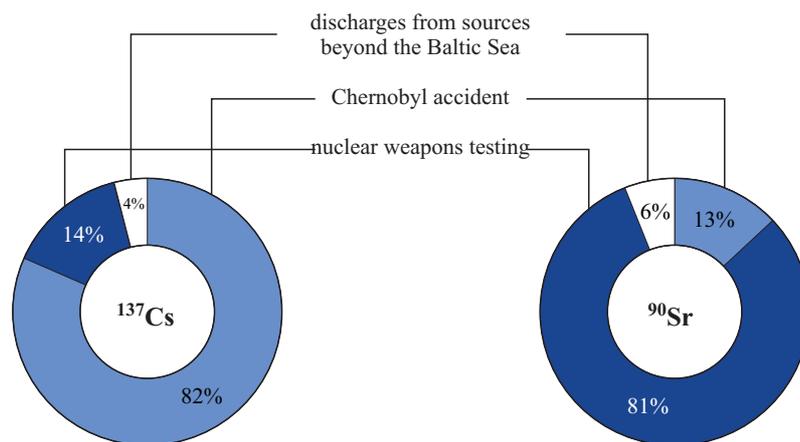
Figure 2. Main sources of ^{137}Cs and ^{90}Sr to the Baltic Sea (after HELCOM 2009)

The contribution of ^{90}Sr from the Chernobyl release relative to the total amount present in the Baltic Sea was much smaller – ca 13% – which is ca 80 PBq (HELCOM 2009, Nielsen et al. 1999). In the case of strontium the major supply source to the Baltic was the atmospheric testing of nuclear weapons (81%), in contrast to ^{137}Cs, the atmospheric contribution of which was only 14% (Figure 2).

As far as the magnitude of supply is concerned, the third-most efficient source of isotopes to the Baltic Sea is contamination by isotopes released from the reprocessing of nuclear fuel from the La Hague and Sellafield installations (HELCOM 2009, UNSCEAR 2000). The isotopes are transported from the North Sea to the Baltic via the Danish Straits with the inflows of saline water. Radioisotopes reach the Baltic Sea from the Irish Sea and the English Channel after 4–5 and 2 years respectively. The main components of the radioisotope mixture are ^{3}H, ^{99}Tc, ^{137}Cs, ^{90}Sr, ^{14}C and ^{129}I. It is estimated that to date these nuclear fuel reprocessing plants have supplied 250 TBq ^{137}Cs and 40 TBq ^{90}Sr to the Baltic Sea (HELCOM 2009), which is 4% and 6%, respectively, of the total amounts accumulated in this sea.

The least efficient and practically negligible source of isotopes is direct contamination from power plants constructed within the Baltic Sea region. Their effectiveness amounts to 0.03% in the case of ^{137}Cs (1.5 TBq) and 0.1% in the case of ^{90}Sr (0.73 TBq) (HELCOM 2009). The regional Baltic sources supply mainly ^{3}H, ^{60}Co, ^{137}Cs and ^{90}Sr.

Riverine discharges of radioactive isotopes and their atmospheric supply are relatively low at present when compared to the situation just after the Chernobyl accident. ^{137}Cs concentrations in Vistula (Wisła) river water are below the detection limits of the analytical methods used and, through dilution, have a considerable effect on activity in the areas of the Gulf of Gdańsk adjacent to the river mouth. A similar situation is observed in the Pomeranian Bay, which accumulates water from the second-largest Polish river, the Odra/Oder; in 1987 the ^{137}Cs concentration there reached 45.6 Bq m^{-3} (Tomczak 1988). In 1994, the respective mean activities of ^{137}Cs in the Vistula, Odra and Pomeranian rivers were 6.8, 9.6 and 4.4 Bq m^{-3} (Skwarzec 2011). In 2004, the weighted mean concentration of ^{137}Cs in Vistula water dropped to 1.34 Bq m^{-3} (Zalewska & Lipska 2006); taking into account the mean annual freshwater outflow from the Vistula of ca 34 km^3, this amounts to an annual fluvial flux of ^{137}Cs of 0.05 TBq. In 1994, the mean activity of ^{90}Sr in the Vistula was 5.4 Bq m^{-3} (Skwarzec 2011) and in 2005, at 3.8 Bq m^{-3}, it was still more than double that of radiocaesium in 2004; this produced an annual fluvial flux of radioactive strontium of 0.13 TBq. In 2010 the mean concentration of ^{90}Sr in the Vistula was even higher (ca 6.0 Bq m^{-3}), but this was probably the effect of the catastrophic flood that occurred in the Vistula catchment area in May of that year.

In 1987, the amount of ^{137}Cs deposited from the atmosphere onto 1 m^2 of the Baltic Sea was 26 Bq. Atmospheric depositions of other isotopes from the Chernobyl release were much lower: ^{134}Cs – 8.9 Bq m^{-2} and ^{90}Sr – 8.4 Bq m^{-2} (Tomczak 1988). Recent (2010) mean atmospheric depositions of these isotopes have decreased to 0.032 Bq m^{-2} of ^{137}Cs and 0.008 Bq m^{-2} of ^{90}Sr.

3. Material and methods

3.1. Seawater

Measurements of ^{137}Cs and ^{90}Sr radioactivity in the Polish economic zone of the Baltic Sea within the framework of the international HELCOM monitoring programme started in 1985. Seawater samples are collected once a year at 17 sampling stations located in the southern Baltic Sea (Figure 1). Seawater samples are taken from the surface and near-bottom

(2 m above the seabed) layers and along the water profile (at selected stations) with a rosette sampler, with simultaneous salinity and temperature profiling.

From 2000 to 2008, the ^{137}Cs activity concentration in seawater samples was determined radiochemically (Zalewska & Lipska 2006), followed by the measurement of β-radiation using a Low-Level Beta Counter FHT 7700T (ESM Eberline), with a background count rate of 0.01 counts s^{-1}, and the minimum detectable activity of 3 mBq per sample.

^{90}Sr was radiochemically determined in seawater samples (Zalewska & Lipska 2006), with ^{90}Y beta activity being measured with a Low-Level Beta Counter FHT 7700T (ESM Eberline).

Since 2009, ^{137}Cs activity in seawater has been determined by gamma spectrometry. Initially, 20 mg Cs$^+$ are added to each acidified seawater sample as a carrier. The caesium is absorbed in 10 g of ammonium phosphomolybdate (AMP) during the 20 minute stirring phase. Separated by decantation and filtration, the AMP is dried and the final activity is measured using a gamma spectrometry system: an HPGe detector with a relative efficiency of 18% and a resolution of 1.8 keV for the 1332 keV peak of 60°. The detector is coupled to an 8192-channel computer analyser and GENIE 2000 software.

The reliability of the methods was validated by participation in inter-comparison exercises organized by HELCOM (HELCOM/MORS Intercomparison Sea Water IC0008/04) and the Riso National Laboratory, Denmark (Intercomparison of Laboratory Analyses of Radionuclides in Environmental Samples – NKS-B Project LABINCO 2004–2005). The accuracy (0.4%) and precision (3%) of the method were assessed by replicate determinations of a standard solution (IAEA-50) supplied by IAEA-MEL (Monaco) with an assigned activity of ^{137}Cs of 50.12 0.12 Bq g^{-1}. Repeated analyses yielded a value of 49.9 ± 1.5 Bq g^{-1}.

3.2. Sediment

Sediment samples collected within the monitoring activities of radioactive substances in the Baltic Sea, related to HELCOM MORS PRO (HELCOM 1995, 2003, 2009), sponsored by the National Atomic Energy Agency in Poland were taken at 6 sampling stations in the southern Baltic Sea: in the Gulf of Gdańsk (station codes – P110, P116), the Gdańsk Deep (P1), the SE Gotland Basin (P140) and the Bornholm Basin (P5, P39) (Figure 1). The samples were taken with a Niemistö corer with an internal diameter of 5 cm from on board r/v 'Baltica' during routine monitoring cruises. For determining ^{137}Cs, 6 parallel sediment cores were taken at each station. The cores were divided into 1 cm thick slices down to 5 cm

depth, and 2 cm thick slices from greater depths. This yielded the following sediment layers: 0–1, 1–2, 2–3, 3–4, 4–5, 5–7, 7–9, 9–11, 11–13, 13–15, 15–17 and 17–19 cm. The corresponding slices/layers from the 6 parallel cores at each sampling station were pooled to produce a single analytical sample. These samples were initially deep-frozen on board ship and dried at a temperature of 105° and homogenized at the land-based laboratory prior to analysis. ^{137}Cs was determined using a gamma spectrometer with an HPGe detector with an energy resolution of 1.8 keV for 60° (1332 keV) and a relative efficiency of 30%, coupled to a Canberra MULTIPORT II MCA multichannel analyser with GENIE-2000 software at the Central Laboratory for Radiological Protection in Warsaw. The accuracy (0.2%) and precision (4.2%) of the method were validated by the determination of ^{137}Cs in reference material IAEA-300 (Baltic Sea sediments).

For the ^{90}Sr determination, one sediment core was taken at each station. The cores were divided into 5 cm thick slices down to 20 cm depth. The samples were then frozen, dried at 105°C and homogenized. ^{90}Sr was analysed radiochemically (Volchok et al. 1957). Extracted with nitric acid, strontium was separated as an oxalate precipitate and then incinerated at 830°C. The precipitate was dissolved in 1:1 water-dil. nitric acid. The solid residue containing barium and rare earth elements was then removed. The remaining solution was left for at least 18 days to establish equilibrium in the ^{90}Sr–^{90}Y radioactive system. Yttrium was then precipitated in oxalate form and the activity of ^{90}Y ($t_{1/2} = 64$ h, beta decay) was measured. In this method, a carrier of stable yttrium and the yield tracer of ^{85}Sr ($t_{1/2} = 64.85$ d) were applied to assess the efficiency of strontium and yttrium counting. The total measurement time was 12 h (4×3 h). The background level was 0.12–0.18 counts per minute, as determined in measurements with a 100 mm thick lead shielding.

3.3. Biota

Benthic plants were collected in the Gulf of Gdańsk in the southern Baltic in 2009 and 2010, at depths from 1 to 4 m (Figure 1 – BP). Within this depth range we were able to collect species representing all taxonomic groups, whose depth range is limited by assimilation pigments and the wavelengths of light penetrating to specific sea depths. The plant material was analysed taxonomically, i.e. the individual species occurring in the samples were identified, after which the biomass of each species was determined gravimetrically; taxonomic analyses and biomass determination were carried out according to the guidelines of the HELCOM COMBINE monitoring programme (HELCOM 1997).

For determining ^{137}Cs activity, the plant samples were dried, ashed at 450°C and homogenized. The ashed samples were placed in counting boxes of the appropriate shape and size.

The samples of benthic organisms (*Mytilus trossulus*, *Macoma balthica*, *Cerastoderma glaucum* and *Balanus improvisus*) were collected in the Gulf of Gdańsk using bottom trawls (Figure 1 – ZT (zoobenthos transect)). The soft tissue of *Mytilus trossulus* was separated, freeze-dried, ashed at 450° and homogenized. In the case of the other bivalves and the crustaceans, the whole body was used for radiocaesium determination owing to the relatively small mass of the soft tissue of these animals. The ashed samples were placed in counting boxes of the appropriate shape and size.

The biological material from fish was collected during the monitoring of radioactive substances in the Baltic Sea, related to HELCOM MORS PRO (HELCOM 1995, 2003, 2009), sponsored by the National Atomic Energy Agency in Poland. Herring (*Clupea harengus*), cod (*Gadus morhua*) and flounder (*Platichthys flesus*), caught in ICES squares F1, F2, F3 and F4, were obtained from commercial fishing catches (Figure 1) in February and March 1999–2010. From each catch 20–25 specimens of herring, 7–9 of flounder and 4–9 of cod were selected and subjected to ichthyological analysis. Muscles excised from these fish were pooled in one homogenized analytical sample. Each integrated sample was then ashed, digested in concentrated nitric acid and, in liquid form, poured into vessels of appropriate geometry for radioactivity measurement.

^{137}Cs activity in samples of biological material was measured using gamma spectrometry. Two gamma spectrometers were used: 1) an HPGe detector with a relative efficiency of 18% and a resolution of 1.8 keV for the 1332 keV peak of ^{60}Co, coupled to the multichannel analyser with GENIE 2000 software, installed at the Institute of Meteorology and Water Management – National Research Institute, Maritime Branch in Gdynia, and 2) an HPGe detector with an energy resolution of 1.8 keV for ^{60}Co (1332 keV), and a relative efficiency of 30%, coupled to a Canberra MULTIPORT II MCA multichannel analyser with GENIE 2000 at the Central Laboratory for Radiological Protection in Warsaw.

The reliability, accuracy and comparability of the measurements were validated by the participation of both laboratories in the HELCOM-MORS Proficiency Test: the Determination of Radionuclides in Fish Flesh Samples, organized by IAEA-MEL Monaco (IAEA-414, Irish and North Sea Fish). The IAEA-recommended ^{137}Cs concentration was 5.18 ± 0.10 Bq kg^{-1} d.w. Repeated analysis yielded a level of 5.06 ± 0.64 Bq kg^{-1} d.w. in the case of the IMWM laboratory (accuracy 2.3%, precision 12.8%) and

5.15 ± 0.20 Bq kg^{-1} d.w. in the case of the CLOR laboratory (accuracy 0.6%, precision 4.3%) (IAEA 2010).

4. ^{137}Cs and ^{90}Sr activity concentrations and trends in seawater

Radionuclide distribution in seawater is of great importance because water plays an intermediate role in the spreading of contaminants in the marine environment. Seawater acts as a sink for radionuclides discharged from different sources, predominantly from atmospheric fallout and water runoff. In the water column, radionuclide distribution is controlled by marine dynamic processes, such as horizontal advection and vertical mixing. Ultimately, the water is a source of radionuclides to biota and bottom sediments.

In consequence of the Chernobyl accident the Baltic Sea radioactivity levels were governed mainly by the distribution of ^{137}Cs, ^{90}Sr and ^{134}Cs in seawater (HELCOM 1995, 2003, Atwood 2010). The Chernobyl fallout was scattered very unevenly over the Baltic area. The Bothnian Sea, where according to experts' estimations nearly half of the released amount of ^{137}Cs was deposited, and the Gulf of Finland, where about 10% of the total ^{137}Cs entered surface waters (HELCOM 1995), were together regarded as the most contaminated regions of the Baltic. In 1986, the respective average concentrations of ^{137}Cs in these regions were 480 Bq m^{-3} and 500 Bq m^{-3}, whereas in the Baltic Proper the average activity concentration of this isotope was only 150 Bq m^{-3}. Much less contaminated were the Bothnian Bay, the Belt Sea and the southern Baltic Proper where, in 1986, the average concentration was 84 Bq m^{-3}. Hence in 1986, the mean concentration of ^{137}Cs (calculated as the arithmetic mean of all measurements in the year) increased in the Baltic Proper nearly sevenfold in comparison to the preceding year. The mean concentration of ^{134}Cs, which was below the detection limit in 1985, increased after the Chernobyl accident to 41.2 Bq m^{-3} (Figure 3). In the following year activity concentrations declined substantially – 30% in the case of ^{137}Cs and 50% in the case of ^{134}Cs – as a result of hydrological processes such as wave mixing, circulation and vertical water transport down to a depth below 30–40 m. A considerable amount of the radiocaesium load accumulated in planktonic organisms became buried in marine sediments.

^{137}Cs concentrations in seawater increased until 1991, when the average value in the southern Baltic Sea was of the order of 101 Bq m^{-3}. This increase was attributed to the transport of more polluted water from the northern Baltic as well as to considerable riverine discharges. Since 1991, ^{137}Cs activity has been falling continuously. This decrease is of an

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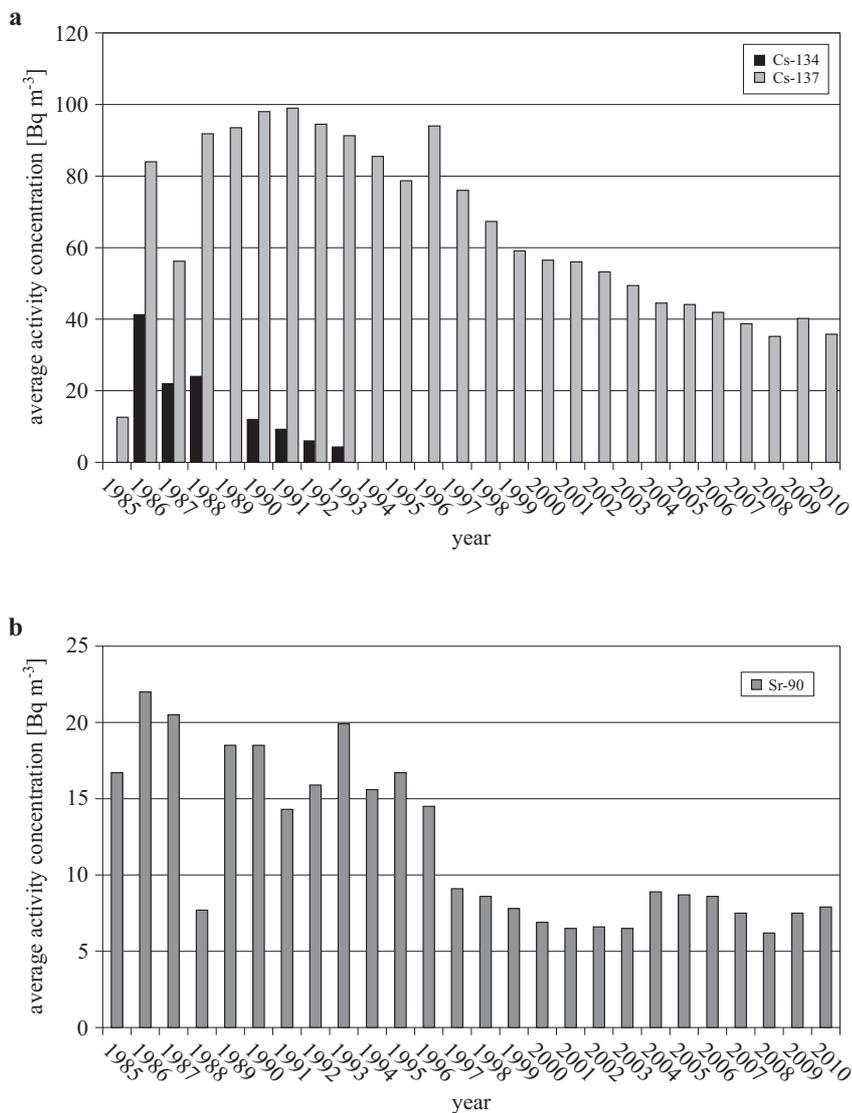
Figure 3. Changes in average activity concentrations of ^{134}Cs, ^{137}Cs (a) and ^{90}Sr (b) in seawater in the period 1985–2010 (data from 1985–1998 after Tomczak (1999), data from 1999–2010 after Zalewska & Saniewski (2012))

exponential character; assuming that the proportion between radiocaesium input and removal remains stable, pre-Chernobyl activity levels could be reached by around 2030.

In 2005 minimum activity concentrations of ^{137}Cs were measured in the Gulf of Finland, where the average concentration was 20 Bq m^{-3}, which is half the level in, for example, the southern Baltic Proper. The main driver

of these regional differences is the variable effective half-life observed in different sea regions (HELCOM 2009). The estimated effective half-life of an isotope is the time required for its concentration to be reduced to 50%, and depends on the radioactive decay half-life together with the specific physical, chemical and biological processes influencing the decrease in concentration (Atwood 2010). The effective half-lives of ^{137}Cs and ^{90}Sr, estimated in 2000 as representative of the entire Baltic Sea, were 14.1 ± 1.8 and 12.0 ± 1.7 years respectively (IAEA 2005).

At present, concentrations of ^{137}Cs are relatively uniform in all regions of the Baltic and remain at approximately similar levels mainly because of the transport and mixing of water masses. Besides these hydrological processes and radioactive decay, the decrease in activity concentrations of radioactive isotopes in seawater is affected by various processes, including sorption on suspended organic matter, bioaccumulation in flora and fauna, and sedimentation. Water exchange between the Baltic Sea and the North Sea via the Danish Straits should also be considered an important factor influencing the level of radioactivity in the Baltic. At the same time, the reason why the radioactivity level due to ^{137}Cs is still elevated after the Chernobyl accident – in 2010 the mean concentration of ^{137}Cs was 35.8 Bq m^{-3} and was still three times higher than in 1985 – is the fact that the Baltic is a semi-enclosed sea and that renewal of water as a result of exchange with the North Sea plays an important role in radionuclide distribution. Only major inflows, approximately 100 km^3 in volume, reach the Bornholm Basin. To renew the deep or intermediate water layers in the basins further east, i.e. the Gdańsk Basins and the Gotland Basin, even greater volumes of highly saline oceanic water are required (Szefer 2002a).

The increase in ^{90}Sr concentrations following the breakdown of the Chernobyl nuclear power plant was decidedly smaller. In 1986, the mean concentration of ^{90}Sr was 22 Bq m^{-3} (Figure 3), a level higher by ca 5 Bq m^{-3} than that in 1985. Since then, the mean content of strontium in seawater has varied within a relatively narrow range from 14.3 to 20.5 Bq m^{-3}, depending mainly on the equilibrium between the sources of ^{90}Sr, such as delayed transportation in river waters and atmospheric fallout, and its natural decay. It was only in 1997 that a substantial decrease in radioactive strontium concentration was noted, when its mean content fell to 9.1 Bq m^{-3}. Between 1998 and 2010, in contrast to ^{137}Cs, no unequivocal trends were observed in the ^{90}Sr activity concentration. The difference in the fate of both radionuclides in the aquatic environment is responsible for this variation.

Generally, pre-Chernobyl, the ^{137}Cs concentration increased with depth, reaching higher values in the bottom waters in accordance with the vertical stratification. This characteristic feature was due to the fact that waters flowing in from the North Sea were more contaminated with the caesium radioisotope than Baltic waters. Since the 1970s this has been due mainly to elevated contamination from the Sellafield nuclear reprocessing plant. After the Chernobyl accident, however, this situation reversed, that is, a higher level of ^{137}Cs activity was found in the upper layer of the Baltic Sea: at present, ^{137}Cs concentrations in the North Sea are lower than in the Baltic. At the same time, oceanic inflows affect mainly the water layers close to the sea floor; hence the lower concentrations of ^{137}Cs in near-bottom waters, especially in the western part of the southern Baltic (Figure 4). In the Bornholm Deep, radiocaesium concentrations are practically uniform from the surface down to 40 m; below that depth they decrease considerably, which is a common feature of recent years. In the Gdańsk Deep, where the halocline is much weaker, the vertical changes in ^{137}Cs concentration are much less pronounced (Figure 4).

A marked decline in ^{137}Cs activities along the depth profiles in both locations was observed in 2003 as a direct consequence of the inflow of North Sea water that took place in January 2003 (Feistel 2003, Piechura & Beszczyńska-Möller 2004). About 200 km^3 of highly saline and cold waters, enriched with oxygen, entered the Baltic Sea from the Kattegat. The inflow of 2003 caused a decline in ^{90}Sr concentrations along vertical profiles in both offshore areas (Figure 5). Needless to say, the effects were more pronounced in the Bornholm Deep, especially in the water layers beneath the halocline. The diluting effect of the saline water from the North Sea resulted in a relationship between salinity and ^{137}Cs concentrations (Figure 6). The calculated (based on data from 2010) correlation coefficient was -0.8668 at the confidence level $p = 0.0000$.

The diluting effect of water flowing in from the North Sea is well marked in the near-bottom layer of the offshore region of the southern Baltic Sea, where concentrations of ^{137}Cs are distinctly higher than in the surface water (Figure 7). At the same time, ^{137}Cs increases in the bottom water from west to east, whereas its activity levels in the surface layer remain much the same. The situation in the case of ^{90}Sr is quite similar: concentrations are higher in the surface water layers than in the near-bottom water, although the differences are much smaller. In coastal areas, activities in

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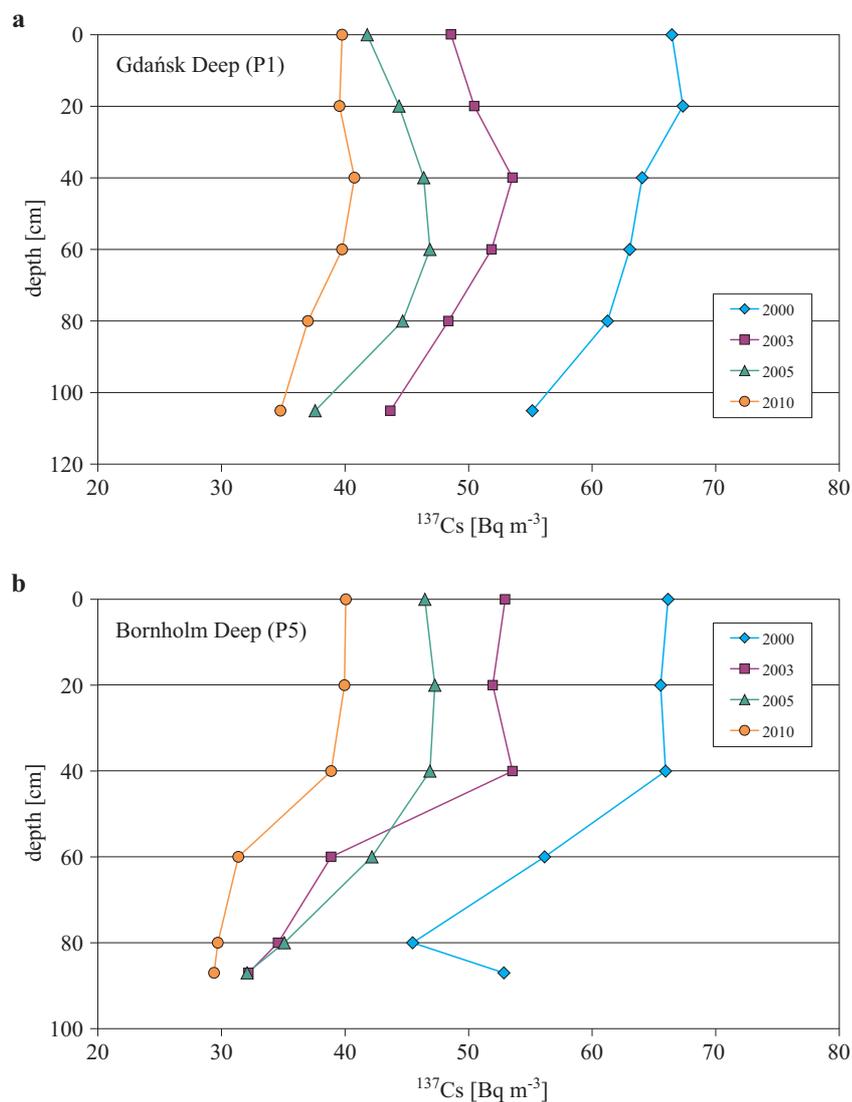
Figure 4. Vertical distribution of ^{137}Cs activity concentrations in the Bornholm and Gdańsk Deeps (measurement uncertainty 1.8–2.5%)

surface water are only slightly higher than those measured near the bottom, most probably as a result of unobstructed mixing between the layers.

In areas influenced by strong riverine discharges, activity concentrations of both radionuclides decline considerably, this being particularly marked in surface concentrations of ^{137}Cs. At present, riverine water contains much lower concentrations of radioisotopes and exerts a diluting effect on seawater, as already mentioned in the section on contemporary sources of

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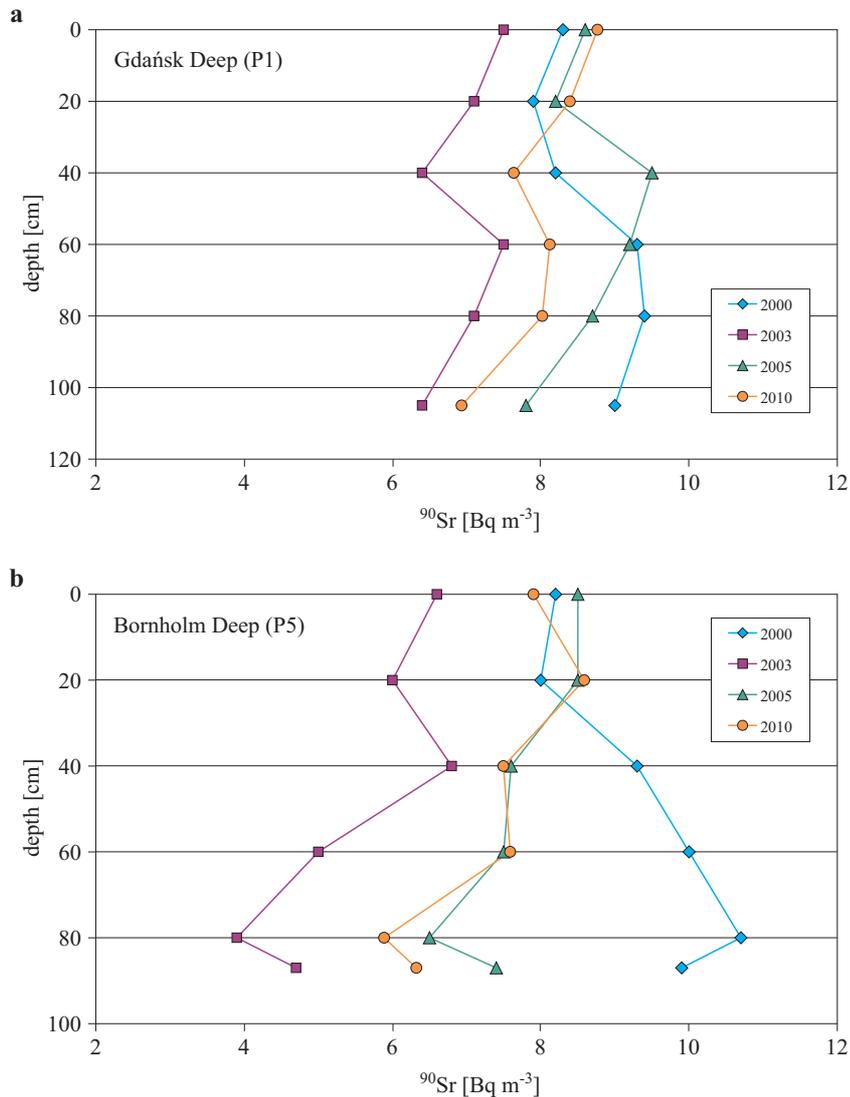
Figure 5. Vertical distribution of ^{90}Sr activity concentrations in the Bornholm and Gdańsk Deeps (measurement uncertainty 4–7%)

radionuclides. This effect was very well documented during the flood that occurred in the Vistula catchment area in May 2010. The huge volume of floodwater caused a substantial decrease in the activity concentration of ^{137}Cs close to the river mouth in the Gulf of Gdańsk, and the plume of Vistula water was perceptible far into the Gulf. In 2010 diminished concentrations of ^{137}Cs were also measured in the Pomeranian Bay, close to mouth of the other large Polish river, the Odra.
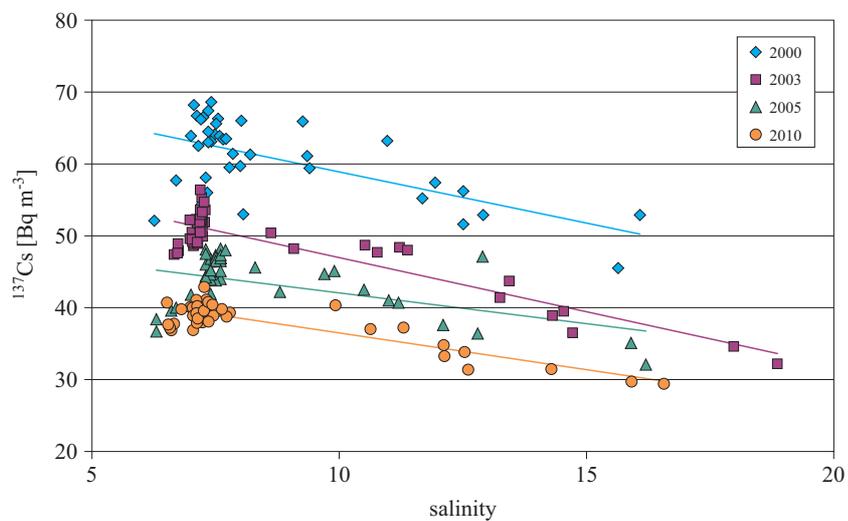
Figure 6. Relationships between activity concentrations of ^{137}Cs and salinity of Baltic Sea water
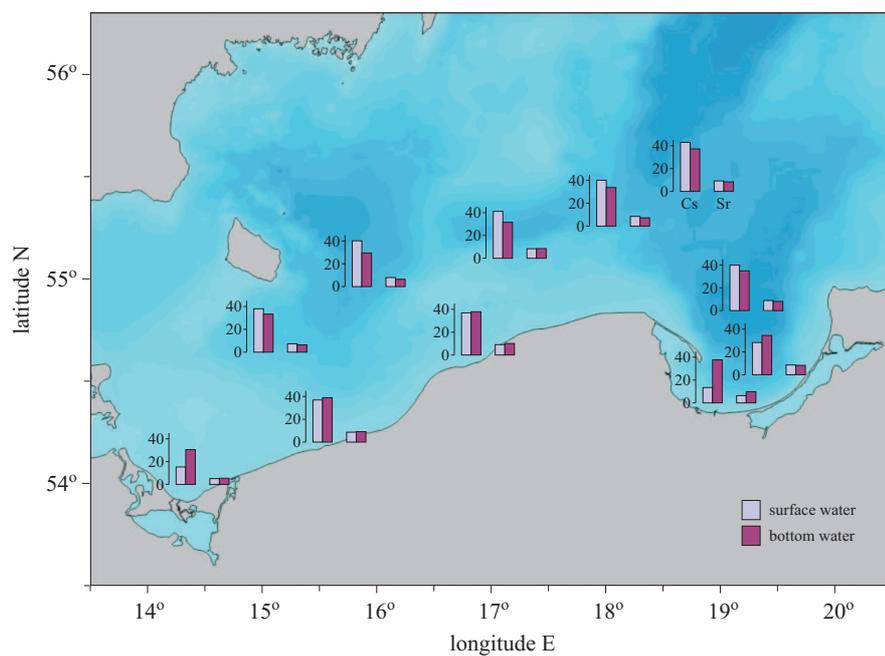
Figure 7. Activity concentrations of ^{137}Cs and ^{90}Sr in the surface and bottom waters of the southern Baltic Sea in 2010 (Zalewska & Saniewski 2012)

5. ^{137}Cs and ^{90}Sr levels in sediments

Pollution of bottom sediments with anthropogenic radioactive isotopes in the southern Baltic Sea is uneven. The variability of isotope distribution with geographical location resulted mainly from the uneven pollution of Baltic seawater directly after the Chernobyl accident, as well as the distribution along vertical profiles in the sediment, where the changes were the consequence of historical events and radioactive decay (Suplińska 2002, Suplińska & Pietrzak-Flis 2008).

Within the southern Baltic Sea, ^{137}Cs activity concentrations are the highest in sediments from the Gulf of Gdańsk (Figure 8). Values are slightly lower in the offshore regions of the Gdańsk Deep and SE Gotland Basin, while the least polluted sediments are found in the western part of the southern Baltic, i.e. in the Bornholm Basin (Suplińska 2002, Suplińska & Pietrzak-Flis 2008).

In the early 1990s, maximum activities of ^{137}Cs were measured in the eastern part of the southern Baltic Sea in the uppermost 1 cm sediment layer (Figure 8a). The concentration was highest (385 Bq ^{137}Cs kg^{-1}) in the Gulf of Gdańsk and only slightly lower (303 Bq kg^{-1}) in the Gdańsk Deep; sediment from the SE Gotland Basin, located further east, was much less contaminated (233 Bq kg^{-1}). Activity concentrations of ^{137}Cs at that time, measured in sediments from western areas of the southern Baltic Sea, were decidedly lower, e.g. 77 Bq kg^{-1} in the Bornholm Deep (P5) and 114 Bq kg^{-1} in the Bornholm Basin (P39).

Generally, activity concentrations of ^{137}Cs fell quickly along the sediment profiles (Figure 8a), indicating the immediate effect of the Chernobyl accident input, and below 18 cm of depth were beyond the method's limit of detection.
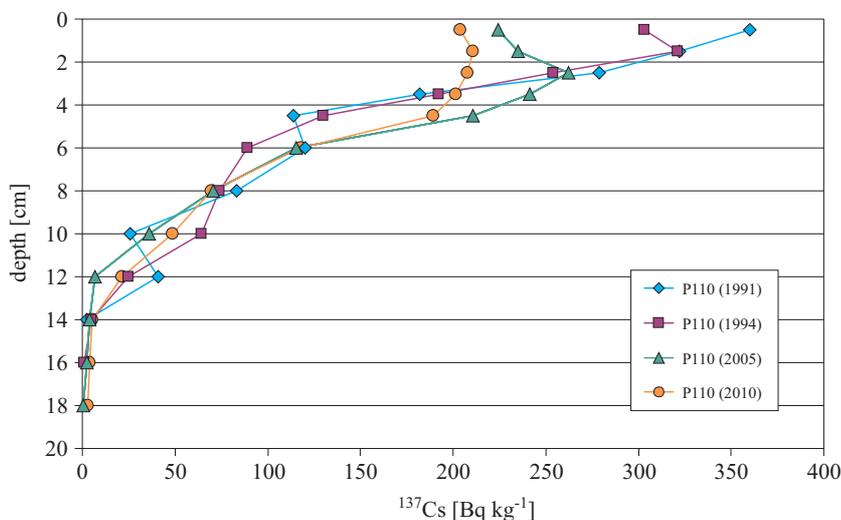
In 2010 the vertical distribution of ^{137}Cs in marine sediments in western parts of the southern Baltic appeared to be much less random (Figure 8b). The highest concentrations were recorded in surface sediment layers but were only 53 and 79 Bq kg^{-1} in the Bornholm Deep and Bornholm Basin respectively. In the Gulf of Gdańsk, the maximum radiocaesium concentration (260 Bq kg^{-1}) was found in the sediment layer just 3 cm below the sediment surface (Figure 8b). In 2010 all the sediments exhibited a decrease in radioactivity compared to the early 1990s, this being attributed mainly to radioactive decay.

In the Gulf of Gdańsk, the maximum of the measured activity concentrations of ^{137}Cs shifted from the surface layer in 1991 to a 2–3 cm deep sediment layer in 2005 (Figure 9). In 2010 there was no longer a well-defined peak, and there was a marked decrease in isotope activity concentrations compared to 2005. Below 5 cm sediment depth, activity
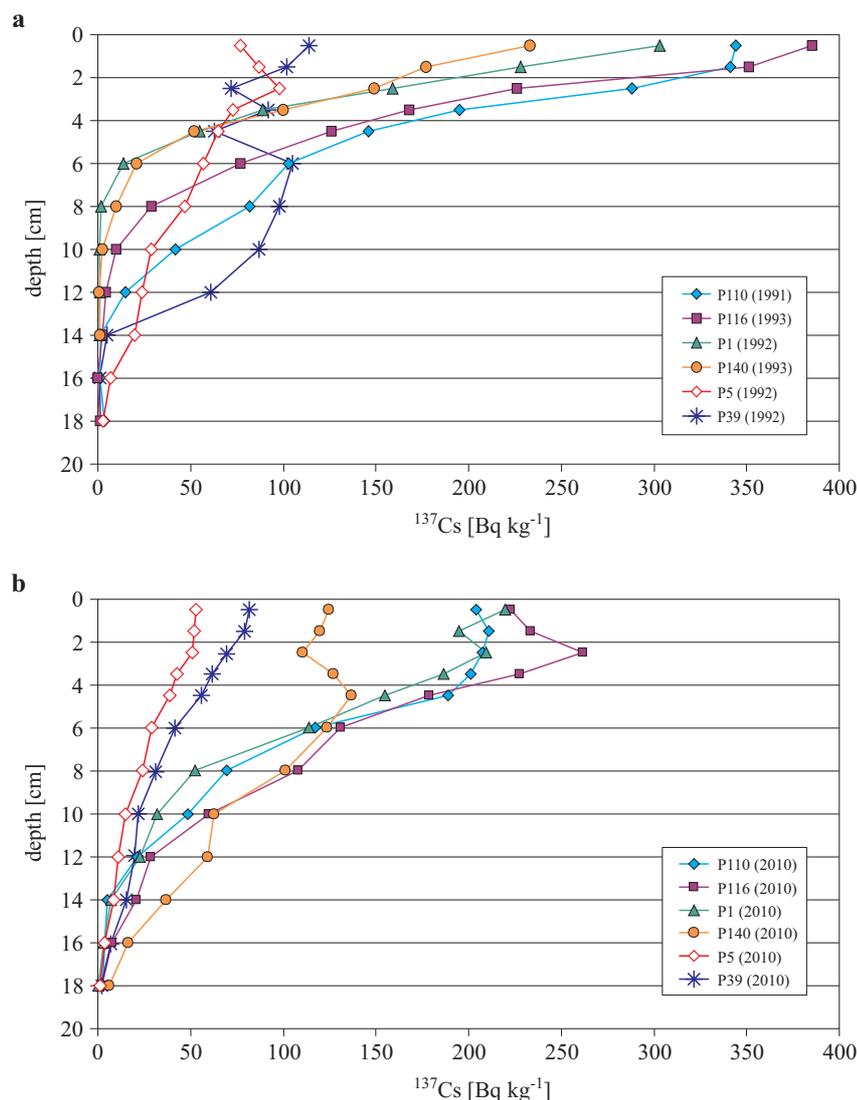
Figure 8. Vertical distribution of ^{137}Cs in marine sediments at six locations in the southern Baltic Sea in the early 1990s (a) and in 2010 (b) (measurement uncertainty 3–7%)

concentrations of ^{137}Cs were very much alike from year to year. The somewhat higher concentrations recorded in deeper sediment layers may be attributed to ^{137}Cs transport to these layers resulting from diffusion, bioturbation and sorption of radiocaesium present in interstitial water, the release of gaseous products, as well as the action of bottom currents. The decline in ^{137}Cs activity concentrations in the surface sediment layer reflects
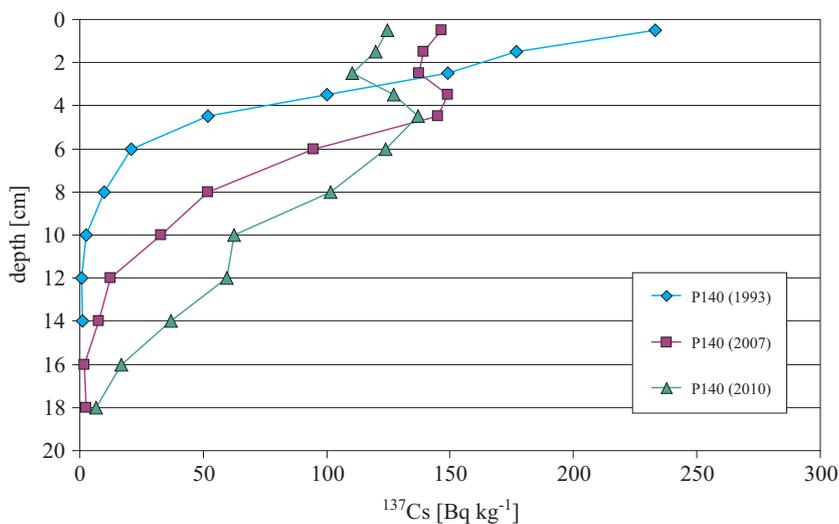
Figure 9. Vertical distribution of ^{137}Cs in sediments from the Gulf of Gdańsk in 1991, 1994, 2005 and 2010 (measurement uncertainty 3–7%)

changes occurring in the near-bottom water from one year to the next and the continuing, gradual decrease in ^{137}Cs.

A shift of maximum ^{137}Cs concentrations, as recorded in 1993, from the surface sediment layer to the layer at 5 cm depth in 2010, was also noted in the SE Gotland Basin. The activity of ^{137}Cs dropped in this region from the initial 233 Bq kg^{-1} (1993) to 137 Bq kg^{-1} (2010) (Figure 10). Simultaneously, ^{137}Cs concentrations in the upper sediment layers, down to 7 cm depth, became comparable. The significant difference in comparison to the changes observed in sediment profiles from the Gulf of Gdańsk is the fact that the decline in activity concentrations of ^{137}Cs in the deeper sediment layers of the SE Gotland Basin was less dynamic in 2007 and 2010 than in 1993.

In 2005 and 2008, sediments from the Bornholm Basin showed rather even activity concentrations of ^{137}Cs along the entire profile (Figure 11). Down to 7 cm depth, values in the upper sediment layer were nearly uniform, varying from 64 to 92 Bq kg^{-1} in 2005 and from 67 to 83 Bq kg^{-1} in 2008.

In the Bornholm Basin area in 1997, the maximum ^{137}C concentrations, approximately 123 Bq kg^{-1}, were recorded in the upper 2 cm of the sediment; below that level the activity was less. The lack of clearly marked concentration peaks along the sediment profiles in this Basin results mainly from the mixing of the surface sediments as a result of strong currents and bioturbation.

Figure 10. Vertical distribution of ^{137}Cs in sediments from the SE Gotland Basin in 1993, 2007 and 2010 (measurement uncertainty 3–7%)
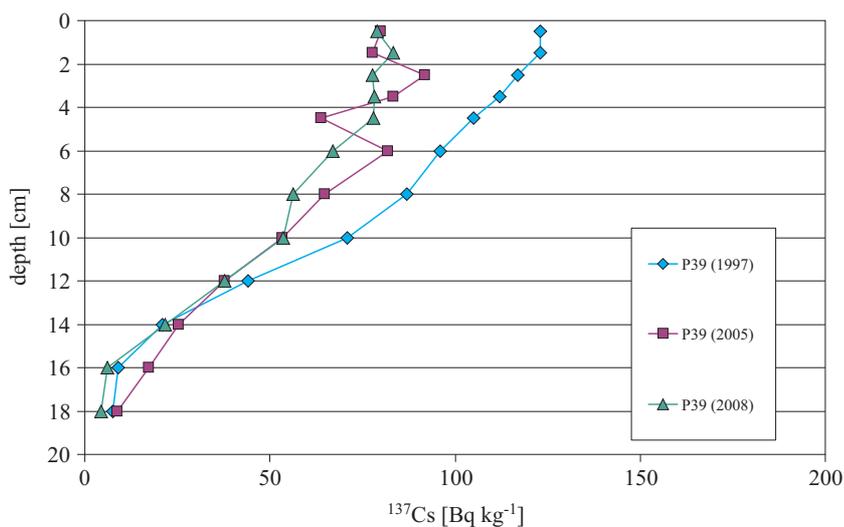
Figure 11. Vertical distribution of ^{137}Cs in sediments from the Bornholm Basin in 1997, 2005 and 2008 (measurement uncertainty 3–7%)

The magnitude of radionuclide deposition was calculated on the basis of ^{137}Cs activities in sediment cores (Table 1). The calculated depositions provide evidence for the highly differentiated pollution of sediments with anthropogenic ^{137}Cs in the southern Baltic Sea, indicating the Gulf of Gdańsk as the most polluted region. As anticipated, the Bornholm

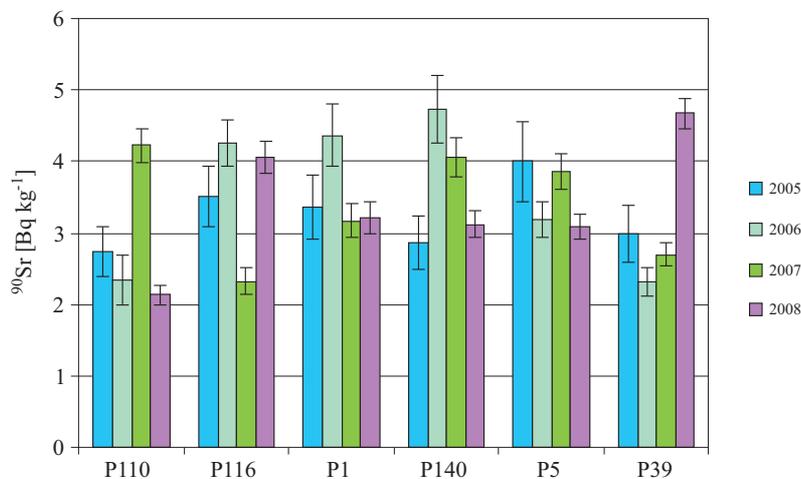**Table 1.** Mean deposition of ^{137}Cs in selected locations of the southern Baltic Sea

Area	Deposition [Bq m^{-2}]	Range [Bq m^{-2}]
Gulf of Gdańsk (P110)	$5451 \pm 265^{b)}$ $(15)^{a)}$	4204–7597
Gulf of Gdańsk (P116)	2763 ± 253 (14)	1819–5288
Gdańsk Deep (P1)	2608 ± 386 (14)	1214–5020
SE Gotland Basin (P140)	3318 ± 160 (14)	2558–4404
Bornholm Deep (P5)	1897 ± 144 (14)	1273–3022
Bornholm Basin (P39)	2465 ± 213 (14)	1093–3441

$^{a)}$number of years; $^{b)}$mean $\pm$ SD

Basin turned out to be the least polluted area, with deposition in the Bornholm Deep (station P5) lower by 30% than that at station P39.

It has to be emphasized that the initial atmospheric deposition was much lower in the southern Baltic Sea than in northern areas, owing to the rather uneven transport of the isotope load following the Chernobyl accident. In 1992, atmospheric deposition of ^{137}Cs in the Gulf of Gdańsk was five to ten times less than in the Gulf of Finland (28.25 kBq m^{-2}) or in the Bothnian Bay (27.57 kBq m^{-2}) (Suplińska & Grzybowska 2000).

The spatial distribution of ^{90}Sr in sediments was definitely more uniform than in the case of ^{137}Cs, as illustrated by the mean activity of ^{90}Sr in the sediment profiles at six locations in the southern Baltic (Figure 12). ^{90}Sr depositions confirmed the evenness of the spatial distribution of this isotope (Table 2). Between 2005 and 2008, mean ^{90}Sr concentrations lay within the 2–4 Bq kg^{-1} range; maximum concentrations (4.7 Bq kg^{-1}) were measured in the SE Gotland Basin in 2006 and in the Bornholm Basin (4.6 Bq kg^{-1}) in 2008. The minimum concentration (2.1 Bq kg^{-1}) was recorded in the Gulf of Gdańsk in 2008. The vertical distribution of ^{90}Sr activity in 5 cm sediment layers was also relatively uniform, as evidenced by the small standard deviations (Figure 12). The results did not indicate any significant temporal changes in activity concentrations of ^{90}Sr between 2005 and 2008.

Figure 12. Average activity concentrations of ^{90}Sr in marine sediments at six locations in the southern Baltic Sea in 2005–2008 (the bars are related to the standard deviation of the mean)

Table 2. Mean deposition of ^{90}Sr in selected locations of the southern Baltic Sea

Area	Deposition [Bq m^{-2}]	Range [Bq m^{-2}]
Gulf of Gdańsk (P110)	$204 \pm 73^{b)}$ $(4)^{a)}$	140–310
Gulf of Gdańsk (P116)	163 ± 50 (4)	109–230
Gdańsk Deep (P1)	153 ± 30 (4)	115–180
SE Gotland Basin (P140)	213 ± 64 (4)	151–291
Bornholm Deep (P5)	150 ± 9 (4)	138–161
Bornholm Basin (P39)	169 ± 51 (4)	119–230

$^{a)}$number of years; $^{b)}$mean $\pm$ SD

6. ^{137}Cs and ^{90}Sr levels in marine biota: benthic plants, macroinvertebrates and fish

Man-made radioisotopes, as well as other chemical elements discharged into seawater from different sources, become distributed within other com-

partments of the marine environment. Marine floral and faunal organisms are among the most important components of the ecosystem because of their functions and environmental status, and also because of their potential importance for humans. Caesium ions can be bioaccumulated by higher flora (Boisson et al. 1997, Sawidis et al. 2003, Burger et al. 2006, Pinder et al. 2006, Zalewska & Saniewski 2011a,b, Zalewska 2012a,b) and fauna (Kryshev & Ryabov 2000, Smith et al. 2002, Malek et al. 2004, Zalewska & Suplińska 2012, 2013). Bioaccumulation and absorption participate in diminishing ^{137}Cs levels in the marine environment. However, the importance of biota in ^{137}Cs removal from seawater in the Baltic is rather low in comparison to other processes such as radioactive decay, sedimentation and water exchange (Ikaheimonen et al. 2009).

The data presented here give an account of ^{137}Cs activity concentrations in marine biota: certain benthic plants, representing green, red and brown algae as well as vascular plants, and macroinvertebrates, such as bivalves, crustaceans and fish.

6.1. Benthic plants

All the species of benthic plants are specific to the southern Baltic Sea (Figure 13). The highest ^{137}Cs activity concentration was measured
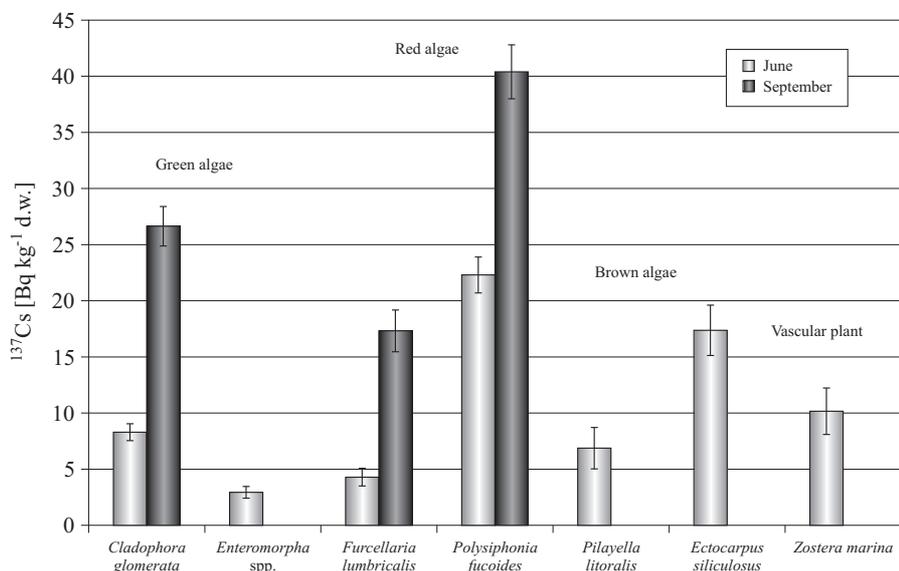
Figure 13. ^{137}Cs activity concentrations in benthic plants from the southern Baltic Sea in June and September 2009 and 2010

in *Polysiphonia fucoides*, a red alga, in both summer and autumn: in June it was 22.3 ± 1.6 Bq kg^{-1} d.w., whereas it was nearly twice that amount (40.4 ± 2.4 Bq kg^{-1} d.w.) in September. An increase in ^{137}Cs content in plants was also observed in other species. ^{137}Cs activity in *Cladophora glomerata* increased more than threefold from 8.3 ± 0.8 Bq kg^{-1} d.w. in June, to 26.6 ± 1.8 Bq kg^{-1} d.w. in September. In the case of *Furcellaria lumbricalis* the increment was more than fourfold, from 4.3 ± 0.8 to 17.3 ± 1.9 Bq kg^{-1} d.w. These observations indicate a close relationship between external environmental factors, such as temperature and light conditions, and the life cycles of these plants (Zalewska 2012a,b). This, however, is detrimental to radiocaesium bioaccumulation. In summer, under optimal growth conditions, intensive plant growth and fruiting result in great increases in biomass. These growth processes are directly related to photosynthesis and respiration. Simultaneously, as the intensification of these metabolic processes results in enhanced metal uptake, the augmentation of biomass causes a diluting effect. Therefore, final ^{137}Cs concentrations in plants in summer should be regarded as the result of these two processes. In autumn, as in summer, there are two opposite tendencies that affect the radiocaesium content in plant tissues. Metabolic processes are still quite intensive and enhance bioaccumulation, but the decline in thermal and insulating conditions retards their activity. This slowing down reduces the rate of bioaccumulation and is also reflected in a biomass decline, although the latter effect can be considered a concentrating process (as opposed to the diluting process associated with biomass increase). The ultimate effect of these opposing activities is the continued increase of ^{137}Cs concentration in plant tissue in autumn. The results of the studies clearly indicate that radiocaesium concentrations in particular plant species are mostly dependent on the plant biomass (Zalewska 2012a,b).

Comparison of ^{137}Cs activities in particular plant species shows without any doubt that *P. fucoides* has the best bioaccumulative properties for Cs$^+$ ions. *Ectocarpus siliculosus*, a brown alga, and *C. glomerata*, representing the green algae, follows close behind. The main feature linking these three benthic plants is the large external surface exchange area of their thalli, which is the one of the most crucial morphological parameters affecting the bioaccumulative process. This is especially important in the light of the fact that foliar uptake is the main means by which elements in algae are exchanged with the environment. Vascular plants exchange elements with their environment mainly through root systems embedded in sand, which is a limited source of ^{137}Cs,; this is reflected in the lower concentration (10.2 ± 2.0 Bq kg^{-1} d.w.) found in *Zostera marina*, representing this group.

Additionally, *P. fucoides*, *E. siliculosus* and *C. glomerata* possess very large surface exchange areas, as these algae are filamentous, one of the groups within the functional-form model proposed by Littler (Littler & Littler 1980, Lobban & Harrison 1997).

6.2. Benthic macroinvertebrates

The blue mussel – *Mytilus trossulus* – is commonly regarded as a model bioindicator of marine environmental pollution. This is because of its efficient bioaccumulation of pollutants through the filtration of large volumes of seawater, its feeding mechanism, and its widespread occurrence. The changes in mean concentrations of ^{137}Cs, monitored since 1989, provide evidence of the applicability of *M. trossulus* as an indicator of radioactive pollution as well (Figure 14).
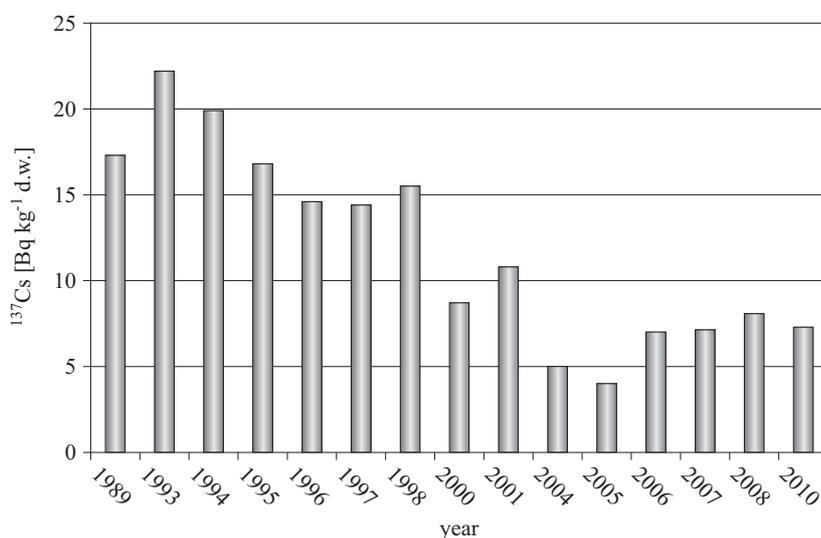
Figure 14. Changes in ^{137}Cs activity concentrations in *Mytilus trossulus* in the southern Baltic Sea in 1989–2010

In 1989, the mean activity concentration of ^{137}Cs was 17.3 Bq kg^{-1} d.w., while in 1993 it reached a maximum of 22.2 Bq kg^{-1} d.w., which is a delay of two years after the maximum activity concentrations in southern Baltic seawater occurred. In subsequent years, activity concentrations of ^{137}Cs in the soft tissue of the blue mussel closely followed the trend observed in seawater concentrations. The minimum (4 Bq kg^{-1} d.w.) occurred in 2005, followed by a slight increase to 7 Bq kg^{-1} d.w. in 2006; thereafter the level remained relatively uniform. In 2010 the mean activity concentration of ^{137}Cs in blue mussel was 7.3 Bq kg^{-1} d.w.

The levels of ^{137}Cs activity found in other bivalves – *Cardium glaucum* and *Macoma balthica* (Table 3) – were lower than those observed in the blue mussels. It has to be emphasized, however, that in contrast to the blue mussel, the analyses were done on whole organisms (soft tissue + shell). The presence of shells could have affected the final result in that the whole sample mass would have been larger, even though they do not accumulate caesium ions. Bioconcentration factors (BCFs), calculated as the ratio between the ^{137}Cs concentration in an organism and in seawater, based on data from 2004 (Table 3), reflect the differences in concentrations. BCFs were also calculated for macrophytobenthic organisms (Zalewska & Saniewski 2011a). Using these values, the bioaccumulative abilities of the organisms towards ^{137}Cs can ordered as follows: $\text{BCF}_{P.\ fucoides} > \text{BCF}_{E.\ siliculosus} > \text{BCF}_{C.\ glomerata} > \text{BCF}_{F.\ lumbricalis} > \text{BCF}_{Z.\ marina} > \text{BCF}_{P.\ littoralis} > \text{BCF}_{M.\ trossulus} > \text{BCF}_{Enteromorpha\ spp.} > \text{BCF}_{M.\ balthica} > \text{BCF}_{B.\ improvisus} > \text{BCF}_{C.\ glaucum}$.

Table 3. ^{137}Cs and ^{90}Sr activity concentrations and bioconcentration factors in macrozoobenthic organisms in 2004

| Class	Species	^{137}Cs* [Bq kg^{-1} d.w.]	BCF* [dm^{-3} kg^{-1}]	^{90}Sr [Bq kg^{-1} d.w.]	BCF [dm^{-3} kg^{-1}]
Bivalves	**Blue mussel** (*Mytilus trossulus*)	4.96 ± 0.49	110	1.19 ± 0.22	129
	Baltic cockle (*Cerastoderma glaucum*)	1.05 ± 0.17	23	0.93 ± 0.14	101
	Baltic macoma (*Macoma balthica*)	3.08 ± 0.24	68	0.57 ± 0.12	61
Crustacean	**Barnacles** (*Balanus improvisus*)	1.53 ± 0.16	34	–	–

* after Zalewska & Saniewski 2011a

Comparison of the BCFs calculated for macroalgae and zoobenthic organisms leads to the conclusion that phytobenthic plants may be regarded as much better bioindicators when their bioaccumulative abilities are taken into account. This is especially important given that, in the case of macrophytobenthic organisms, uptake processes are practically limited to

one period of the year, when intensive life processes are taking place, whereas mussels accumulate pollutants over a longer time. The relatively rapid response time with regard to pollution is also very important.

The activities of ^{90}Sr in the bivalves were lower than the ^{137}Cs activity concentrations (Table 3), although concentration coefficients were comparable between the blue mussel and the Baltic macoma, but much higher in the case of the Baltic cockle.

6.3. Fish

The breakdown of the Chernobyl nuclear power plant in 1986 is the main source of ^{137}Cs accumulated in the tissues of fish from the southern Baltic Sea. An increase in activity concentrations of this radionuclide was observed in such commercial fish species as herring (*Clupea harengus*), flounder (*Platichthys flesus*) and cod (*Gadus morhua*). In 1989, the mean activity concentrations reached their maxima: 15.0 Bq kg^{-1} w.w. in *C. harengus*, denoting a sixfold increment compared to the pre-Chernobyl level (HELCOM 2009), 13.6 Bq kg^{-1} w.w. in *P. flesus* and 17.5 Bq kg^{-1} w.w. in *G. morhua*. Since 1989 the changes in mean ^{137}Cs concentrations in specific fish species have shown explicit declining trends (Grzybowska 1989), characterized by high correlation coefficients (Figures 15–17). In 2010 the mean concentration of radioactive caesium in herring muscle (4.7 Bq kg^{-1} w.w.) was $< 30\%$ of the level determined in 1989 (Zalewska & Suplińska 2013). In the case of flounder and cod, the respective mean activities of this
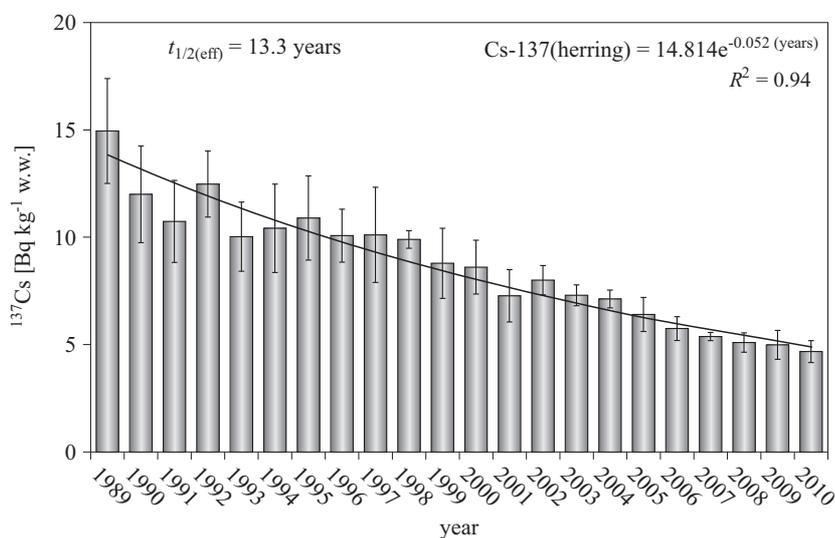
Figure 15. ^{137}Cs activity concentrations in *Clupea harengus* from the southern Baltic Sea in 1989–2010
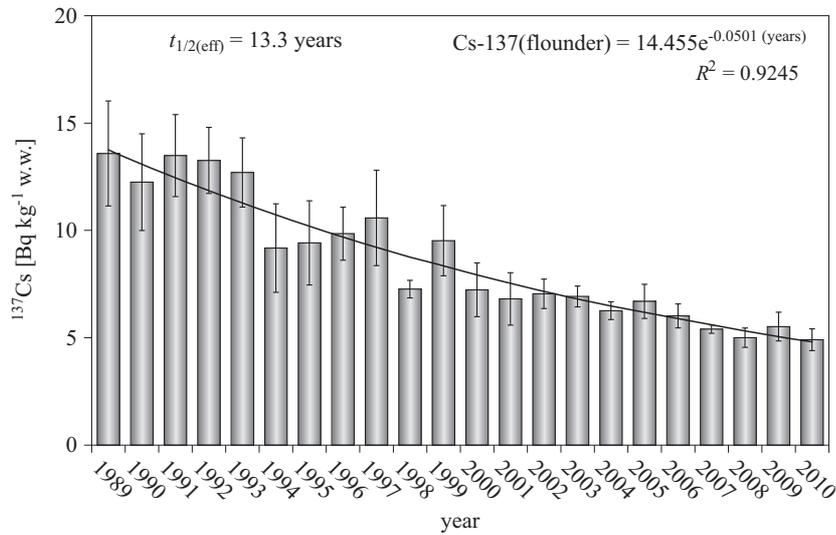
Figure 16. ^{137}Cs activity concentrations in *Platichthys flesus* from the southern Baltic Sea in 1989–2010
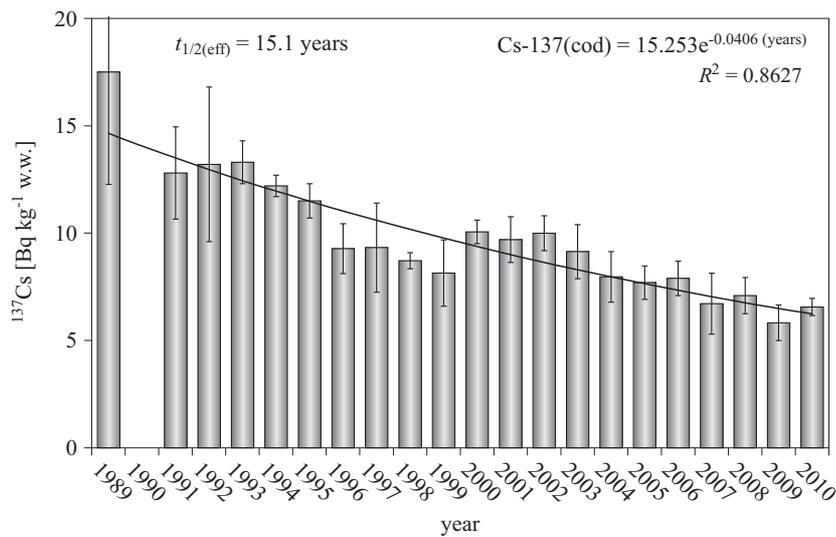
Figure 17. ^{137}Cs activity concentrations in *Gadus morhua* from the southern Baltic Sea in 1989–2010

radionuclide were 4.9 and 6.6 Bq kg^{-1} w.w. and had declined by ca 60% compared to the 1989 levels. Generally, ^{137}Cs concentrations in *G. morhua*, a top predator, were higher than in *P. flesus* and *C. harengus* during the entire study period. The effective half-lives of ^{137}Cs in these fish species were

determined on basis of these trends: 13.3 years in the case of *C. harengus* and *P. flesus*, and slightly longer (15.1 years) for *G. morhua.*

The bioaccumulation of various substances in fish is much more complex than in, for example, macrophytes (Zalewska 2012a,b), in which the exchange of elements takes place directly between the organism and the surrounding water. The main source of radiocaesium in fish is food, and the build-up of its content in fish tissue is due to bioaccumulation and biomagnification, the latter process being related to an increase in ^{137}Cs content in successive elements of the trophic chain. Despite the fact that the importance of seawater as a direct source of ^{137}Cs contamination in fish is more limited, ^{137}Cs concentrations in fish tissues accurately reflected the decreasing trend of its activity, as observed in seawater between 1991–2010. Therefore, the content of radioactive caesium in fish can be considered a reliable indicator in the radioactivity status assessment of the marine environment, especially – taking into account the biomagnification factor – when the concentrations in seawater fall below the detection limit.

7. Conclusions

The radioactivity level in the southern Baltic Sea is due mainly to the distribution of anthropogenic radioactive isotopes – ^{137}Cs and ^{90}Sr – in seawater, marine sediments and biota. The main input sources of these isotopes to marine sediments were the nuclear weapons tests carried out in the 1950s and 1960s and, especially in the case of radiocaesium, the Chernobyl accident. The activity concentration of ^{137}Cs in seawater has been decreasing since peaking in 1991; the major processes involved are radioactive decay and bioaccumulation, together with sedimentation and water exchange with the North Sea. The decline in radioactivity is exponential in character. In 2010 the mean concentration of ^{137}Cs in southern Baltic seawater was 35.8 Bq m^{-3}, that of ^{90}Sr was 7.9 Bq m^{-3}. Strontium concentrations did not show a marked temporal trend, but a significant decline in its activity in seawater was observed in 1997; since then the concentrations of this isotope have varied within a narrow range from 6.2 to 8.7 Bq m^{-3}.

Activity concentrations of ^{137}Cs in the southern Baltic are the highest in sediments from the Gulf of Gdańsk (385 Bq kg^{-1} in the surface layer). Values are slightly lower in the offshore regions of the Gdańsk Deep (303 Bq kg^{-1}) and the SE Gotland Basin (233 Bq kg^{-1}), while the least polluted sediments are found in the western part of the southern Baltic, i.e. the Bornholm Deep (77 Bq kg^{-1}). In general, ^{137}Cs activity concentrations decreased along the sediment profiles, indicating the immediate effect of the Chernobyl accident input. In contrast to ^{137}Cs, the distribution

of ^{90}Sr is not spatially differentiated, either horizontally or in the vertical sediment profiles. The mean activities of ^{90}Sr in marine sediments varied between 2.1 Bq kg^{-1} in the Gulf of Gdańsk to 4.7 Bq kg^{-1} in the Bornholm Basin.

^{137}Cs is bioaccumulated in macroalgae with considerable efficiency, mainly due to foliar uptake, an effective means of element exchange with the environment. *Polysiphonia fucoides*, a red alga, contained the highest activity concentrations of ^{137}Cs throughout the growing season: 22.3 Bq kg^{-1} d.w. in late spring (June) and 40.2 Bq kg^{-1} in early autumn (September). This algae is recommended as a bioindicator of radioactive isotope pollution in the marine environment.

The concentration of ^{137}Cs in the soft tissues of the blue mussel (*Mytilus trossulus*), a benthic macroinvertebrate, was 7.3 Bq kg^{-1} in 2010, a value conspicuously lower than in *Polysiphonia fucoides*.

The Chernobyl accident caused an increase in ^{137}Cs activity concentrations in commercial fish such as herring (*Clupea harengus*), flounder (*Platichthys flesus*) and cod (*Gadus morhua*). In all three species the decline in ^{137}Cs activity between 2000 and 2010 closely reflects the situation in seawater. In 2010 the mean concentration of radioactive caesium in herring muscle (4.7 Bq kg^{-1} w.w.) was $< 30\%$ of the level determined in 1989. In the case of flounder and cod, the mean activities of the radionuclide were 4.9 and 6.6 Bq kg^{-1} w.w. respectively and had declined by ca 60% compared to the 1989 values.

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