Journal of Environmental Radioactivity 127 (2014) 11–25

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Caesium-137 distribution, inventories and accumulation history in the Baltic Sea sediments

Agata Zaborska*, Aleksandra Winogradow, Janusz Pempkowiak

Institute of Oceanology, Polish Academy of Sciences, ul. Powstańców Warszawy 55, 81-712 Sopot, Poland

ARTICLE INFO

Article history:
Received 30 April 2013
Received in revised form
6 September 2013
Accepted 8 September 2013
Available online 8 October 2013

Keywords:
Baltic Sea
Cs-137
Radionuclides
Contamination
Nuclear power plant

ABSTRACT

The Baltic Sea is susceptible to pollution by hazardous substances due to limited water exchange, shallowness, and the large catchment area. Radionuclides, particularly ^{137}Cs, are one of the most hazardous anthropogenic substances present in the Baltic environment. This study was conducted to present ^{137}Cs present contamination that should further be a subject of reliable monitoring when the new Nuclear Power Plant is put into operation in the northern Poland. The sea-wide, up to date distribution of ^{137}Cs activities and inventories in the Baltic Sea bottom sediments are presented. The ^{137}Cs activity concentrations were measured in 30 cm long sediment cores collected at 22 sampling stations. Sediment accumulation rates were quantified by ^{210}Pb geochronology to follow the history of ^{137}Cs accumulation. The ^{137}Cs inventories and fluxes were calculated. Most of the Baltic Sea sediments accumulated ^{137}Cs in the range from 750 to 2675 Bq m^{-2}. The Bothnian Bay is severely contaminated by ^{137}Cs with inventories up to 95,191 Bq m^{-2}. This region is moreover characterized by extremely large patchiness of ^{137}Cs inventories. The ^{137}Cs annual fluxes are highest at the two stations located at the Bothnian Bay (342 Bq m^{-2} and 527 Bq m^{-2}) due to large Chernobyl ^{137}Cs contamination of that region and high sediment accumulation rates. When these stations are excluded, the recent, annual mean value of ^{137}Cs load to the Baltic Sea deposits is 38 ± 22 Bq m^{-2}. The distribution of radio-caesium inventories over the Baltic Sea nowadays reflects the pattern of Chernobyl contamination. The radio-caesium deposited in surface sediments is not permanently buried, but may be resuspended and redeposited by currents, bioturbation or anthropogenic activities.

1. Introduction

The Baltic Sea is susceptible to pollution by hazardous substances due to limited water exchange, shallowness, and the large catchment area. Radionuclides, particularly ^{137}Cs, are one of the most hazardous anthropogenic substances present in the Baltic environment (Nies and Wedekind, 1988; HELCOM, 1995; HELCOM, 2009a). Caesium-137 is a medium half-life (30 years) radionuclide, originating from anthropogenic sources. Starting from 1945 introduced to the environment as one of the nuclear fission products during nuclear bomb tests. The tests peaked in ′50 and ′60, when large amounts of this radionuclide were released into the atmosphere in the course of nuclear weapons race. Since then a slow deposition of the radionuclide from the atmosphere, the so-called fallout, has been observed. A small fraction of the total ^{137}Cs present in the atmosphere is deposited to the land and ocean surface yearly. Some amount is still present in the atmosphere, thus ^{137}Cs fallout is expected to be persistent for decades to come. Additionally, high ^{137}Cs loads were introduced to the environment, particularly to the Baltic Sea, as a result of the Chernobyl accident in 1986. The activity concentrations of both historical radiocaesium sources – global fallout and Chernobyl – have decreased significantly with time as the ^{137}Cs half-life is 30 years. Nowadays, however, newly produced caesium-137 is discharged to the marine environment with waste produced by nuclear reactors and nuclear fuel reprocessing plants (HELCOM, 2009a; Vartti, 2011). In the Baltic Sea, as much as 82% of these radionuclide inventories originate from the Chernobyl accident, while 14% originate from the global fallout following nuclear weapons testing. Discharges from nuclear power plants (mainly from Sweden and Finland) and nuclear fuel reprocessing facilities (Sellafield and Cap de la Hague) are responsible for 4% of the total ^{137}Cs present in the Baltic Sea (HELCOM, 2009a).

Nuclear power plants are low but continuous sources of ^{137}Cs radionuclide to the environment. The problem of nuclear power plants influence on the Baltic marine environment was raised by

* Corresponding author. Tel.: +48 587311736.
E-mail address: agata@iopan.gda.pl (A. Zaborska).

0265-931X/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jenvrad.2013.09.003

Skwarzec et al. (1994, 2006). They studied nickel (^{63}Ni), iron (^{55}Fe), and other radionuclides present in the effluents and cooling water from nuclear plants. These radionuclides were found to accumulate in marine organisms. Caesium-137 also accumulates readily in biota due to its chemical similarity to potassium (Dahlgaard et al., 1994). Moreover, recent studies show that this radionuclide biomagnifies through marine food chain (Carroll et al., 2002; Heldal et al., 2003).

Planned to be constructed in northern Poland, the nuclear power plant should be regarded as a new source of radiocaesium that is likely to affect the concentrations and distribution of the nuclide in the Baltic Sea. Although the effluent water from first Polish nuclear power plant to be built in Żarnowiec or Choczewo will contain dissolved radiocaesium at the low activity concentrations, it will be discharged into rivers draining directly to the Baltic Sea. Upon getting in contact with saline water, caesium will adsorb on suspended organic matter and clay minerals (Knapińka-Skiba et al., 1994). Both the riverine and the autochthonous suspended matter will eventually deposit on the sea bottom. However, since most of the south-west Polish coast is a highly energetic environment influenced by coastal currents, the fine sediments containing the radionuclide (and other contaminants) will be transported to the sediment accumulation areas eg. Gdańsk Basin (Bełdowski and Pempkowiak, 2007).

The aim of this paper is to present the sea-wide, up to date distribution of ^{137}Cs activities and inventories in the Baltic Sea bottom sediments. Since the last ^{137}Cs detailed studies covering the entire Baltic Sea were performed 10–20 years ago, this will bring the current results concerning ^{137}Cs distribution on the verge of constructing a new nuclear power plant. Moreover, as the study is devoted to the ^{137}Cs activity concentrations in sediments dated by means of the radiolead method, a detailed history of the Baltic Sea contamination with radioceasium will be revealed. This approach will help to understand the processes influencing the ^{137}Cs accumulation and distribution in sediments.

2. Radiocaesium in the Baltic Sea

The monitoring of radionuclides in the Baltic Sea seawater and sediments has been carried out by the HELCOM network for over 25 years. The monitoring of ^{137}Cs distribution in the Baltic seawater off the Polish coast has been conducted by the Institute of Meteorology and Water Management (Zalewska and Lipska, 2006; Zalewska and Saniewski, 2011) since 1985. Generally, ^{137}Cs activity concentration has been decreasing from some 100 Bq m^{-3} (in 1993–1998) to about 50 Bq m^{-3} (recently). Recent studies indicate that radiocaesium activity concentration in the seawater depends on the wind induced seawater transport from the strongly contaminated northern Baltic Sea (Zalewska and Lipska, 2006).

Upon entering marine waters, radiocaesium is readily absorbed by biota and/or is scavenged from the water column by sinking particles. The seawater/suspended particle ^{137}Cs activity equilibrium is reached within several hours (Knapińka-Skiba, 1994). As a consequence of the adsorption process, sediments reflect relative contamination of the sea by caesium radionuclide. Thus, marine sediments are the final sink of ^{137}Cs. The deposited radiocaesium can be buried in sediments and undergo the radioactive decay. It can be also re-introduced to the water column by sediment resuspension caused by waves, bottom currents, bioturbation or human activity (eg. trawling or underwater construction).

Several recent studies have concerned radiocaesium distribution in dated sediments. The ^{137}Cs inventories in seawater and sediments of the Baltic Sea were presented by Ikäheimonen et al. (2009) and Mattila et al. (2006). Unfortunately, little results from the southern Baltic off Poland were reported. Most of the authors concentrated on the Baltic Proper, the Gulf of Finland, and the heavily contaminated sediments of the Bothnian Bay. The ^{137}Cs inventory in bottom sediments there amounted to over 50,000 Bq m^{-2} (Ikäheimonen et al., 2009) and up to 120,000 Bq m^{-2} (Mattila et al., 2006). Radiocaesium activity concentrations in the central part of the Gulf of Finland ranged from 90 Bq kg^{-1} to 170 Bq kg^{-1}, while in the Kotka town neighborhood it reached as much as 2400 Bq kg^{-1} (Kankaapää et al., 1997). The ^{137}Cs activity concentrations ranging from 4 Bq kg^{-1} to 207 Bq kg^{-1} were reported along the Lithuanian coast sediments by Lujanienė et al. (2005) and Davuliene et al. (2006). The ^{137}Cs loads in sediments were assessed as ranging from 5120 Bq m^{-2} to 6180 Bq m^{-2} (Davuliene et al., 2006). The reviews prepared Szefer (2002) and Skwarzec (2011) present a literature on the radionuclides in the southern Baltic. At the Polish coast, a detailed study regarding the ^{137}Cs inventories in sediments was conducted by Suplińska (2002). Six sediment cores collected between 1998 and 2000 from the Gulf of Gdańsk and the Bornholm Basin were analyzed. The inventory of ^{137}Cs ranged from 1960 Bq m^{-2} in the Bornholm Basin to 4144 Bq m^{-2} in the Gulf of Gdańsk. Radiocaesium distribution in the southern Baltic was also studied by Suplińska and Pietrzak-Flis (2008) to validate ^{210}Pb dating. Additionally provided at six sampling sites was a ^{137}Cs inventory ranging from 1400 Bq m^{-2} in the Bornholm Basin to 4722 Bq m^{-2} in the Gdańsk Bay sediments. Most recently, Szczepańska et al. (2009, 2012) measured ^{137}Cs in several sediment cores collected in the central Baltic Sea. The radionuclide activity concentrations at the surface sediment layer ranged from 50 Bq kg^{-1} to 400 Bq kg^{-1}. Radiocaesium reached down to 10 cm below the sediment surface. ^{137}Cs distribution in the surface sediments of the Gulf of Gdańsk was recently presented by (Skwarzec 2011).

In the past, numerous and detailed studies of ^{137}Cs in the southern Baltic sediments were conducted by the Institute of Oceanology (eg. Pempkowiak, 1991; Knapińska-Skiba and Bojanowski, 1992; Bojanowski et al., 1994; Knapińska-Skiba et al., 1994; Bojanowski et al., 1995; Pempkowiak et al., 1996; Knapińska-Skiba et al., 2001). Unfortunately, ^{137}Cs activity concentrations presented there are no longer relevant as sediment accumulation and resuspension, and radiocaesium decay have changed the radionuclide distribution.

3. Materials and methods

3.1. Field work

Sediments for the ^{137}Cs study were collected at twenty-two sites in different Baltic Sea regions (Fig. 1). Sample collection was performed during r/v Oceania, r/v Aranda, and r/v Alkor cruises using a Niemistö type corer or a GEMAX twin corers. The collected sediment cores were sliced onboard into 10 mm thick layers (section 0–20 cm) and 20 mm layers (section 20–40 cm). The so obtained sediment samples were frozen onboard. After the cruise, samples were transported to the laboratory of the Institute of Oceanology, Sopot, Poland.

3.2. Laboratory

Sediment samples were freeze-dried and ground into fine particles. Sediment porosity and organic matter contents (ashing at 450 °C) were determined. Organic carbon concentration analysis was performed in an elemental analyzer combined with isotopic ratio mass spectrometer (Delta V Advantage, Thermo). The ^{137}Cs activity concentrations in the sediment samples were measured by gamma spectrometry. Known amounts (20–30 g) of sediments were packed in Cerbo counting vials. The weight and height of the