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RADIONUCLIDES IN SEDIMENTS OF THE BALTIC SEA IN 1999 - 2006; FIRST DRAFT

Attached here to is the first draft of the sub-chapter "Radionuclides in sediments" of Chapter 3 ("Radioactivity in the Baltic Sea") for the thematic assessment of long-term changes in radioactivity in the Baltic Sea covering the period up to 2006, submitted by the chapter co-ordinators, Jukka Mattila and Erkki Ilus, STUK.

THE FIRST DRAFT

Radionuclides in sediments of the Baltic Sea in 1999 - 2006

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Introduction

Sediments are playing a significant role, when monitoring the state of the environment and radioactivity in the Baltic Sea. Many radionuclides tend to bind to the sediment particles creating a storage of radioactivity to the bottom. Many radionuclides could also be measured quite easily from sediment samples, because of the high K_d's (distribution coefficients). In a proper monitoring station, sediment layers have stored continuous deposition of sediment material and radionuclides for long time periods, that allow us to study the history and total amounts of certain radionuclides in the Baltic Sea.

In the recent years several articles and reports have been published, where the radioactivity in sediments have been used except for monitoring, also for scientific purposes. Both natural and artificial radionuclides have been used e.g. in estimation of sediment accumulation rates and dating of the sediments (Jensen et al. 2003, Mattila et al. 2006). Radioactivity has also been used as a tracer in intercomparison of sediment sampling devices (HELCOM 2000). The concentrations and amounts of transuranic elements as plutonium activities in the environment of the Baltic Sea have been summarized during the reporting period (Ikäheimonen 2003).

This report is a short summary of the radioactivity in the Baltic Sea sediments in the years 1999 - 2006. Radioactivity in sediments has been reported previously in HELCOM 1995 and 2003. Most recently the report "Long-lived radionuclides in the seabed of the Baltic Sea" has summarized the inventories of ^{137}Cs, gave new knowledge about the artificial radioactivity in sediments and took steps toward an estimation of the amounts of natural radioactivity as ^{40}K and ^{226}Ra activities in the surface sediments of the Baltic Sea (Ilus et al. 2007).

Material and methods

All the data is based on the HELCOM database, in which the Contracting Parties have yearly reported recent data. The amount of the sediment samples in the years 1999 - 2006 was (3461 + year 2006). The sampling techniques used by different countries are described previously (HELCOM 2003). Also the sediment types and the bottom morphologies have been described previously in many studies (Winterhalter 1972, Winterhalter et al. 1981, HELCOM 2003). The methods used in the radionuclide inventories are described in many context (Salo et al. 1986, HELCOM 2003, Ilus et al. 2007). In general, the inventories in sediments were based on mean total amounts of radionuclide activity concentrations in sediments and the surface areas of different sea areas taking into account the surface areas and the activities in soft and hard bottoms respectively.

The sources of artificial radioactivity

In 1999 - 2006 there were no major events increasing considerably the artificial radioactivity in the Baltic Sea sediments. In general, the main sources of artificial radioactivity have still been the fallout from the Chernobyl accident in year 1986 and the global fallout mostly in 1950's and 1960's (HELCOM 2003). Nuclear reprocessing plants (Sellafield in UK and La Hague in France) have had smaller influence on the radioactivity in the southern Baltic Sea. Also other nuclear facilities, such as nuclear power plants, hospitals and certain research institutes have had small inputs mainly locally in radionuclide concentrations in sediments. Furthermore, rivers are still bringing radionuclides from the drainage areas to the sea.

Results and discussion

The amounts of artificial radioactivity have not increased noticeably in the bottom sediments during the years 1999 - 2006. Most of the long lived artificial radioactivity is due the ^{137}Cs and mostly found in the bottom sediments in the Gulf of Bothnia (especially in the Bothnian Sea) and in the eastern part of the Gulf of Finland (figure 1). The amounts of the ^{137}Cs have been relatively even during the reporting period (figure 2). Artificial radioactivity from the Chernobyl and global fallouts is partly going to be buried to sediments in the accumulation bottoms. Nonetheless, a part of the activity bound in the sediments, is moving from the erosion/transportation bottoms toward the accumulation bottoms.

In spite of carefully planning of the monitoring programs, there are still a lot of factors affecting the results of the sediment studies and causing variations in time trends as could be seen in figure 2 (HELCOM 2000, HELCOM 2003, Mattila et al. 2006,). For example, variation inside the sedimentation basins and around the monitoring stations can be large due to the heterogeneity of soft sediment deposits. The differences in sampling techniques increase also variability in the results. The bottom dynamics affect accumulation, transportation/erosion, and hydraulic sorting of sediments. At the moment, we know relatively little about the variability in sediments around the monitoring stations.

Most of the radioactivity in the sediments in the Baltic Sea is originated from naturally occuring radionuclides. In the recent years, the activity concentrations of naturally occuring radionuclides with long half-lives as e.g. ^{40}K ($T_{½}$ 1,3x10^9 a), ^{226}Ra ($T_{½}$ 1600 a) and ^{232}Th ($T_{½}$ 1,4x10^{10} a) have been reported to the database. In the surface sediments, the concentrations of ^{40}K (in 0 - 10 cm layer) varied between 300 - 1100 Bq kg^{-1} d.w., those of ^{226}Ra (in 0 - 10 cm layer) between 10 - 90 Bq kg^{-1} d.w. and those of ^{232}Th (in 0 - 30 cm layer) between 13 - 42 Bq kg^{-1} d.w.. The activity levels of these nuclides depend on the type of sediments. In the sediment baseline study, we also tried to estimate for the first time the total amounts of ^{40}K and ^{226}Ra in the Baltic Sea sediments, but at this time the estimations had to be limited only the uppermost layers of the recently accumulated sediments. The total amounts in the 0 - 10 cm layer were estimated to be roughly about 8500 TBq for ^{40}K and about 420 TBq for ^{226}Ra.

In the Baltic Sea sediments, there are still considerable amounts of artificial radioactivity due to radionuclides with longer half-lives. Nonetheless, the artificial radioactivity is not expected to cause harmful effects to the Baltic Sea wildlife. After the Chernobyl fallout, there were also elevated concentrations of many short living radionuclides such as ^{60}Co ($T_{½}$ 5.3 a),^{103}Ru ($T_{½}$ 39.3 d), ^{106}Ru ($T_{½}$ 372,6 h), ^{110m}Ag ($T_{½}$ 249,8 d) and ^{125}Sb ($T_{½}$ 2,8 a), but because of their short half-lives, the activities of these radionuclides have decreased considerably or even mostly vanished (HELCOM 2003).

In the sediment baseline study (Ilus et al. 2007), it was also possible to get new knowledge about artificial radionuclides as ^{99}Tc ($T_{½}$ 2,1x10^5 a) and ^{237}Np ($T_{½}$ 2,1x10^6 a) by using the mass spectrometric methods. In the surface sediments, the concentrations of ^{99}Tc (in layer 0 - 10 cm) varied between 0.04 and 1.30 Bq kg^{-1} d.w., when the activities of ^{237}Np (in layer 0 - 20 cm) varied between 0.2 and 6.5 mBq kg^{-1} dry weight. The total amount of ^{237}Np in the sediments of the Baltic Sea was estimated to be about 0.02 TBq (ILUS ET AL. 2007).

In the years 1999 - 2006 the activities of ^{137}Cs ($T_{½}$ 30.2 a) have been relatively even in different monitoring stations of the Baltic Sea (figure 2). After the Chernobyl accident, activities of ^{137}Cs have been studied a lot, because there were lot of caesium activity in the fallout and because it has long half-live, high K_d's in brackish water environment and because it is relatively easy to measure. In the recent inventory, we estimated that the total amount of ^{137}Cs activity in the Baltic Sea sediments was about 2100 - 2400 TBq at the beginning of the 2000s. This amount was about 8 - 9 times higher in comparison to the amounts of pre-Chernobyl level in the beginning of the 1980s (table 1). In the recent years, caesium has continued deposit onto bottom sediments and, at same time, the physical half-live (30.2 a) reduces the activities slowly. Most of the ^{137}Cs activity is in the bottoms of the Bothnian Sea and in the eastern Gulf of Finland (figure 1). The new information achieved increased our knowledge about the distribution of ^{137}Cs activities between so called hard and soft bottoms in the open sea areas. The total amounts of ^{137}Cs activities on hard bottoms varied from 0.3 % to nearly 14 % of those on soft bottoms, when the average ratio was only about 4 % (Ilus et al. 2007).

The reported values of ^{239,240}Pu (^{239}Pu $T_{½}$ 2,4x10^4 a, ^{240}Pu $T_{½}$ 6563 a) activities have ranged between 0.01 and 14.10 Bq kg^{-1} d.w. and values of ^{238}Pu ($T_{½}$ 87,7 a) activities between 6 and 480 mBq kg^{-1}. New information about plutonium was also achieved recently. The total amount of the ^{239,240}Pu activities was estimated to be about 15.3 TBq (table 1). Most of the plutonium is originating from the global fallout, but there were small amounts of ^{238}Pu and ^{241}Pu in the fallout of Chernobyl accident that could be seen in the activity ratios of ^{238}Pu/^{239,240}Pu and ^{241}Pu/^{239,240}Pu and in the excess amounts of ^{241}Pu (Ikäheimonen 2003).

Table 1. Results of inventories of the total activities of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in the sediments of the Baltic Sea (Ilus et al. 2007). *=rough estimate, ** years 1987 - 1988

year / time period	Sr-90 (TBq)	Cs-137 (TBq)	Pu-239,240 (TBq)	Reference
begin of the 1980's	12	277	15	Salo et al. 1986
1990-1991	-	1200 - 1400	18**	HELCOM 1995
1998	-	1940 - 2210	-	HELCOM 2003
2000 - 2005	26*	2100 - 2400	15.3	HELCOM 2007

The activities of the artificial radionuclides ^{90}Sr ($T_{½}$ 28.5 a) and ^{241}Am ($T_{½}$ 432.7 a) are mostly originating from the global fallout. It has been estimated that the input of ^{90}Sr activity from the Chernobyl fallout to the Baltic Sea, has been only 13 % of the total amount of ^{90}Sr activity in the Baltic Sea (HELCOM 2003). Because of the small share in the Chernobyl fallout and the costly analytical methods, the interest on ^{90}Sr has been reduced and there are only relatively few data available. In the years 1999 - 2006 the reported ^{241}Am concentrations ranged from 0.08 to 4.80 Bq kg^{-1} d.w. and those of ^{90}Sr from 0.1 to 73.8 Bq kg^{-1} d.w.. Due the limited data, the present total amounts of these activities are difficult to estimate. A rough estimate for the total amount of ^{90}Sr in the sediments of the Baltic Sea was about 26 TBq (Ilus et al. 2007).

Future work and Recommendations

The monitoring work of radioactivities in the Baltic Sea is essential also in the future. The continuous monitoring work and time trends of the radioactive substances are the base for understanding the state of the Baltic Sea environment and radioactivity. However, there are still several gaps in our knowledge of radioactivity in the sediments of the Baltic Sea, as noted previously (HELCOM 2003, Ilus et al. 2007). In the near future, we should attempt to fill these gaps and increase our knowledge of the activities of e.g. Sr-90, Am-241 and natural radioactivities such as Pb-210 in the sediments of the Baltic Sea. Also the relevant works in the future include assessments of the impacts of ionising radiation on the environment of the Baltic Sea.

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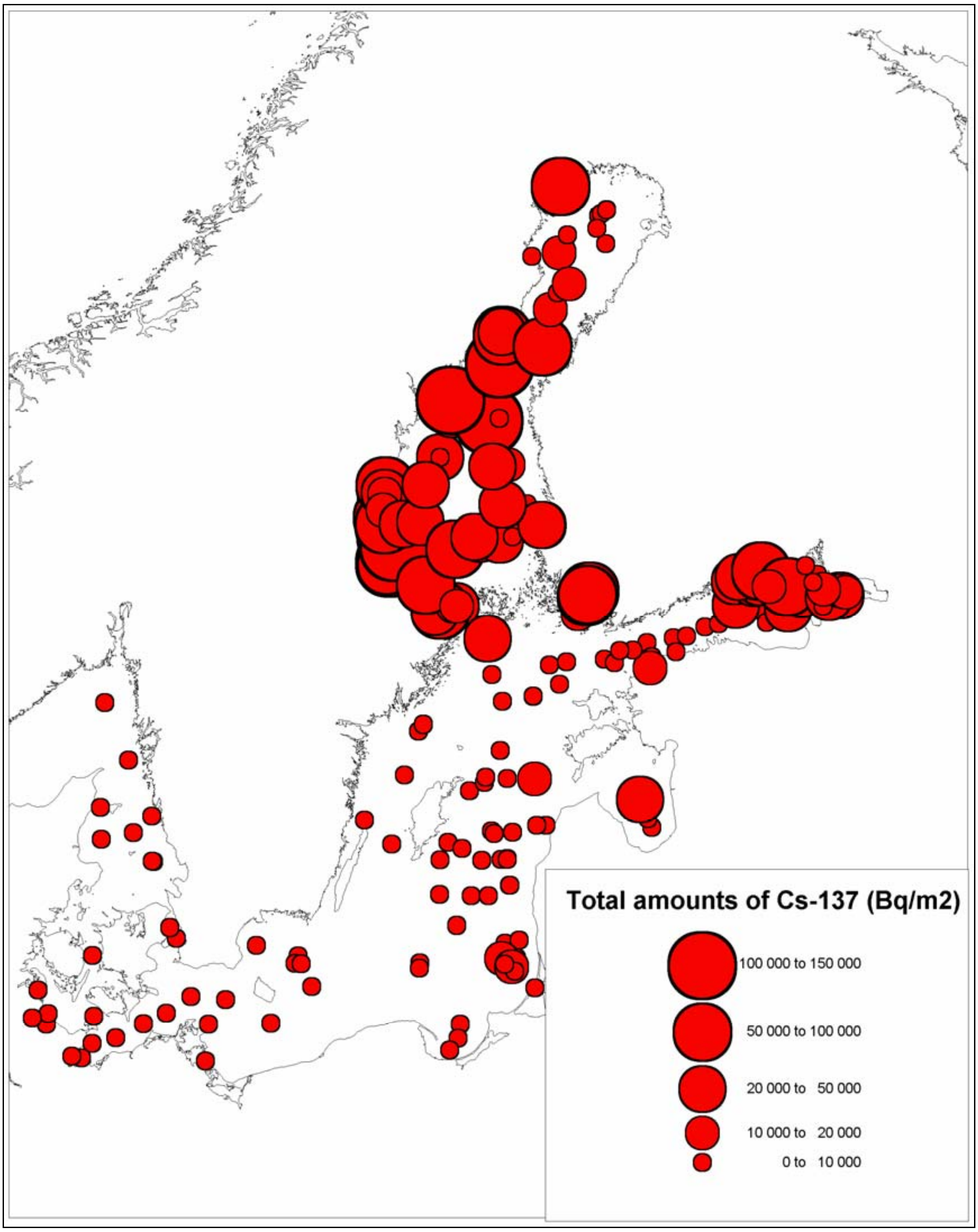

Figure 1. Total amounts of ^{137}Cs activities (Bq/m^2) in different sampling stations in the Baltic Sea in the late 1990s and the beginning of the 2000s (Ilus et al. 2007).

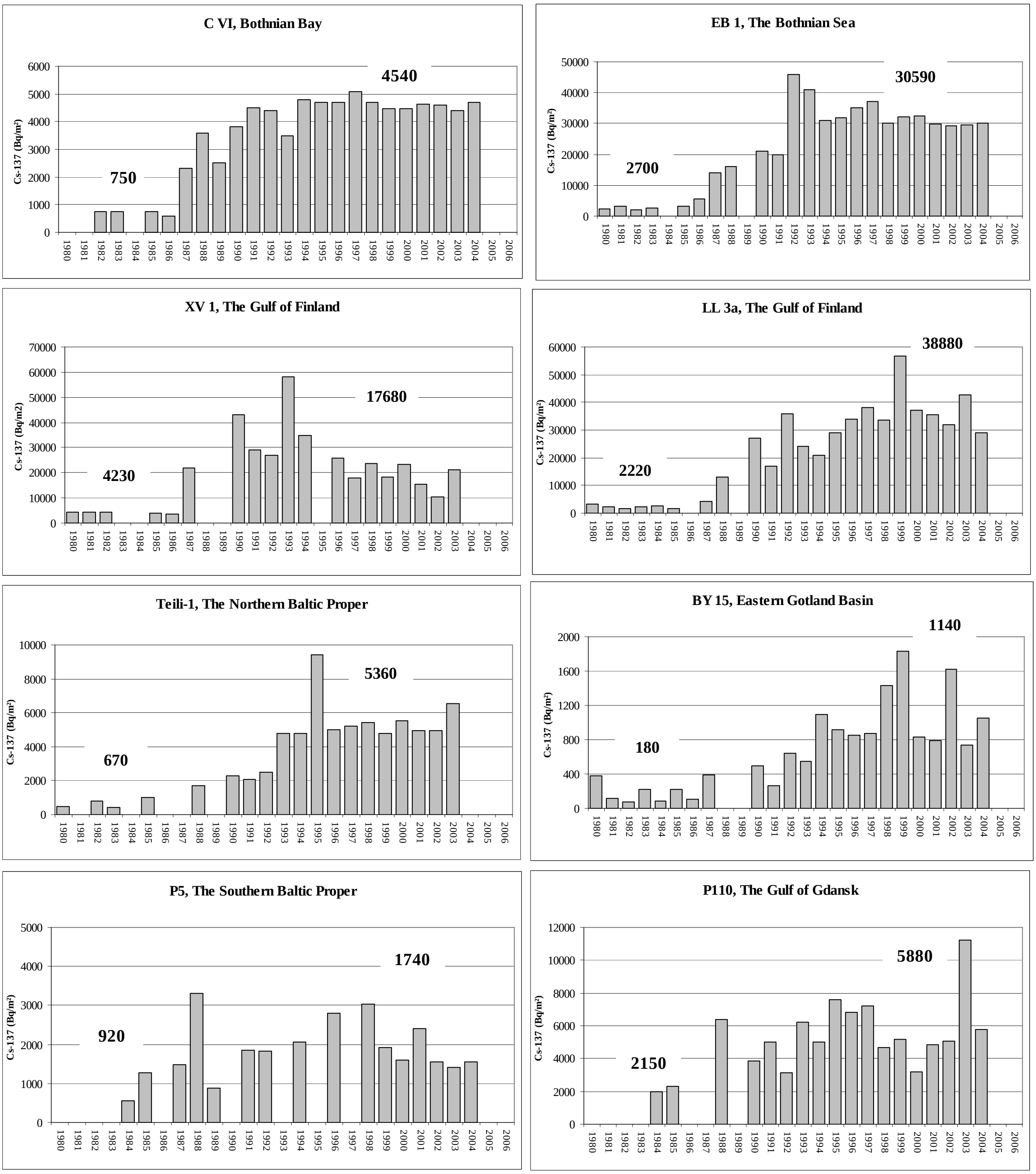

Figure 2. (Note the different scales in figures) Total amounts of ^{137}Cs activities (Bq/m^2) in some monitoring stations in the Baltic Sea. Pre-Chernobyl level (average in 1980 - 85) and the average level in 1999 - 2006 are indicated in numbers.

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