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Erkki Ilus[1], Jukka Mattila[1], Sven P. Nielsen[2], Eia Jakobson[3], Jürgen Herrmann[4], Visvaldis Graveris[5], Beata Vilimaite-Silobritiene[6], Maria Suplinska[7], Andrey Stepanov[8], Maria Lüning[9]

[1] STUK - Radiation and Nuclear Safety Authority, Finland, [2] Risø National Laboratory, Denmark, [3] Estonian Radiation Protection Centre, [4] Federal Maritime and Hydrographic Agency, Germany, [5] Latvian Environment, Geology and Meteorology Agency, [6] Lithuanian Environment Protection Agency, [7] Central Laboratory for Radiological Protection, Poland, [8] V.G. Khlopin Radium Institute, Russia, [9] Swedish Radiation Protection Authority

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Language revision: Janet Pawlak, Charlottenlund, Denmark
Design and layout: Leena Närhi, Bitdesign, Vantaa, Finland
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Photos: Erkki Ilus

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1 Introduction

Bottom sediments play an important role in radioecological studies of the marine environment because a large proportion of radioactive substances entering the sea is adsorbed over time onto suspended particulate matter and deposited in sediments. Generally, this is the final sink for most of the organic material produced in the water phase, as well as for other particles transported by water currents from other sea areas and from adjacent terrestrial areas. During their slow settling, the particles tend to bind radionuclides from the water phase and carry them to the bottom.

Under favourable conditions, the deposited particles form undisturbed laminae in a stratigraphic sequence on the seabed, and the bottom sediments create an archive from which the history of the area can be read. Various particle-bound substances can be identified as markers of specific historical events and, with the aid of marker horizons, the laminae can be dated. Modern dating methods provide reliable time scales in which time-dependent changes in the concentrations of radionuclides can be recorded. As an example, the radioactive fallout from atmospheric nuclear weapons tests in the 1950s and 1960s and the accident at the Chernobyl Nuclear Power Plant in April 1986 have created useful markers in the sediments of many sea areas, especially in the Baltic Sea.

The Baltic Sea offers exceptionally good opportunities to conduct sedimentological studies because the average rate of sedimentation there is much higher than in the oceans and in most other coastal seas. The anoxic conditions in the near-bottom water of the Baltic Proper, and hence the lack of benthic animals over large bottom areas, result in the formation of an undisturbed sedimentary medium, which enables the sampling of sediment laminae in an undisturbed stratigraphic sequence. However, sediment sampling is extremely sensitive to errors, which can cause substantial differences in results. This fact should always be taken into account when considering sediment results.

This report presents the results of a Sediment Baseline Study carried out by the HELCOM Project Group for Monitoring of Radioactive Substances in the Baltic Sea (HELCOM MORS-PRO) in 2000–2005. The goal of the study was to complement the knowledge and inventories of long-lived radionuclides in the seabed of the Baltic Sea by providing additional data from so-called “white areas” lacking data, and from radionuclides that were not measured in earlier surveys.

2 Sources of long-lived radionuclides in the Baltic Sea sediments

The most remarkable event affecting the quantities of artificial radionuclides in the Baltic Sea sediments was the accident at the Chernobyl Nuclear Power Plant in April 1986. The global fallout from atmospheric nuclear weapons tests, as well as discharges from the reprocessing plants in Western Europe and from nuclear facilities in the Baltic Sea region, have had markedly smaller consequences for radionuclide concentrations in sediments, especially in the northern parts of the Baltic Sea.

The Chernobyl accident was not the first event resulting in the dispersion of artificial radionuclides into the atmosphere. Atmospheric nuclear weapons tests giving rise to radioactive pollution of the air, and subsequently of the earth's surface and the marine environment, were conducted beginning in 1945. Most of the tests were carried out during 1955–1958 and 1961–1962. One of the test sites was Novaya Zemlya in the former USSR. By late 1962, the tests were reduced significantly owing to the signing of the Nuclear Weapons Test Ban Treaty of 1963, and since 1980 no atmospheric testing has been conducted in the northern hemisphere. The total depositions of weapons test Cs-137 and Sr-90 into the Baltic Sea have been estimated at 900 TBq and 600 TBq, respectively (decay corrected to 1991).

Chernobyl Accident

On 26 April 1986, a severe accident occurred at the nuclear power plant of Chernobyl, in the former USSR. The accident resulted in widespread contamination, mainly by I-131, Cs-134, and Cs-137. Owing to its long half-life, only Cs-137 is currently of interest.

Global fallout

Beginning with the use of nuclear weapons at the end of the Second World War, artificial radionuclides were transported towards the upper atmosphere. This continued on a far greater scale during the atmospheric testing of nuclear weapons until the 1960s. The radioactive material released was distributed worldwide via the atmosphere and ultimately precipitated down to land or sea.

Reprocessing plants

Reprocessing plants are a part of the so-called nuclear fuel cycle. The Western European plants are located in the UK (Sellafield) and in France (La Hague). Used fuel rods from nuclear power plants are processed physically and chemically in order to recover unused fuel. This has resulted in regulated discharges of effluents containing numerous radionuclides into the marine environment that peaked in the 1970s and 1980s. Currently, such discharges have decreased considerably.

Nuclear facilities

Nuclear facilities comprise mainly nuclear power plants, fuel reprocessing plants, fuel fabrication plants, radioactive waste handling factories, and certain research institutes and military installations. These facilities handle radionuclides, some of which may be released into the environment.

In addition to the above, no other sources of radioactivity noticeably contributed to atmospheric fallout prior to the Chernobyl accident, except, e.g., the Windscale reactor fire in 1957, which mainly had a local impact. However, aquatic discharges from the Western European nuclear reprocessing plants (Sellafield, UK, and La Hague, France) have influenced the concentrations of Cs-137 and some other radionuclides in the marine environment of the southern Baltic Sea via the inflow of saline waters through the Danish Straits.

The Baltic Sea was the marine area most affected by the Chernobyl accident because the first radioactive clouds from Chernobyl travelled north and caused strong deposition in the Baltic Sea region (Povinec et al., 1996). The deposition was greater there than, e.g., in the Black Sea, the Mediterranean Sea or any other marine or brackish water area.

The International Atomic Energy Agency, IAEA, (1986) estimated that the total amount of radioactive substances released to the atmosphere as a result of the Chernobyl accident was 1–2 x 10^{18} Bq. Most of the radionuclides released were short-lived, and their impact on the environment was negligible. Among the longer-lived radionuclides, Cs-137 was the most important owing to its relatively long half-life (30 years) and its relevance with respect to radiation doses to man. The total amount of Cs-137 released to the environment has been estimated to vary between 38 000 TBq ± 50% and 167 000 TBq, with the most frequently used value 70 000 TBq. The total input of Cs-137 from the Chernobyl accident into the Baltic Sea area has been estimated at 4 100–5 100 TBq (decay corrected to 1991).

During the acute fallout situation, Chernobyl-derived caesium was dispersed directly onto the surface of the sea and, with a delay, throughout the entire drainage area. In the course of time, the Baltic Sea has received caesium from the surrounding terrestrial and adjacent coastal areas as a result of runoff, river discharges, and coastal currents. A large part of this caesium has been deposited in the sediments, but a significant amount of Chernobyl caesium has also been transported by sea currents from the Gulf of Bothnia and the Gulf of Finland into the Baltic Proper and further out from the Baltic Sea through the Danish Straits. Currently, the proportion of Chernobyl-derived caesium is clearly dominant in the Baltic Sea environment, while that originating from other sources is decreasing, and it is often difficult to distinguish the ”old” caesium from that of Chernobyl.

The distribution pattern of Chernobyl-derived Cs-137 in the catchment area of the Baltic Sea was very patchy, with the highest deposition values occurring in the areas surrounding the Gulf of Bothnia and the Gulf of Finland (Figure 1). The highest Cs-137 concentrations and total amounts (per square metre) in bottom sediments also occur in these gulfs, but the patchy distribution has been further emphasized as a consequence of river discharges, sea currents, and varying sedimentation rates on hard (erosion) and soft (sedimentation) bottoms.

Radioactivity
Natural phenomenon which is a property of many elements. It consists of a spontaneous emission of charged particles or radiation. After this event the nucleus has changed, which results in a different element or a different state of the same element. Elements with this property are called radionuclides. The “half-life” of a radionuclide describes the time after which half of the material has undergone a radioactive transformation. Radioactivity is measured in becquerels (Bq), which is defined as the activity of a quantity of radioactive material in which one nucleus decays per second. Units used in this report include terabecquerel (TBq), or 10^{12} Bq, and millibecquerel (mBq), or 10^{-3} Bq.

Natural and artificial radionuclides
Some radionuclides have been present from the beginning of the universe, while others are produced by natural processes; both are called natural radionuclides. Other radionuclides are produced by human activities such as in the operation of nuclear power plants or explosion of nuclear weapons. These are called artificial radionuclides.

Radionuclides of interest
As for other types of contaminants in the sea, relevant properties for the definition of a “nuclide of interest” are abundance, longevity and affinity of accumulation. Owing to the slow transport processes in the marine environment, only certain radionuclides are of interest.

Because Cs-137 and Cs-134 were the most abundant constituents in the Chernobyl fallout, the main emphasis in the monitoring has focused on these nuclides. In addition, the amounts of Ru-103, Ru-106, Ag-110, and Sb-125 clearly increased in Baltic Sea sediments as a consequence of the Chernobyl accident. The occurrence of these nuclides followed the distribution pattern of the caesium isotopes rather well, generally being most abundant in 1987; but due to their relatively short half-lives, they already started to decrease by 1988/1989.

Observations of Co-60 (half-life 5.3 years) in sediments also increased in various sub-regions of the Baltic Sea in the late 1980s.

The studies have focused on the occurrence and behaviour in the sediments of the fallout nuclides originating from the above-mentioned events. In this sense, the sedimentation processes and sedimentation rates of the radionuclides, as well as the accumulation of the fallout nuclides into sediments, have been monitored.

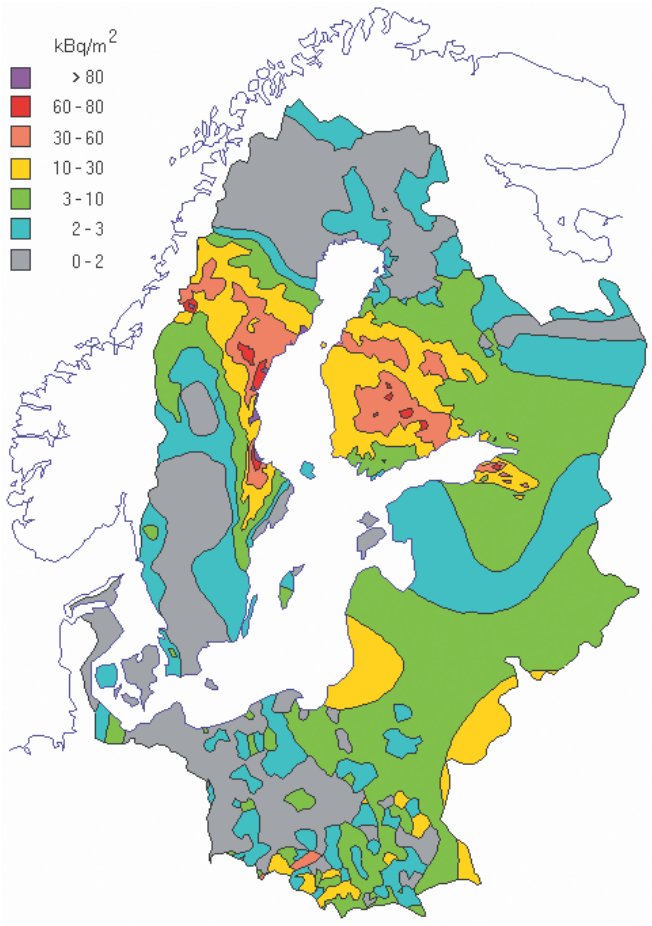


Figure 1.
Terrestrial deposition of Cs-137 in the Baltic Sea drainage area (kBq m^{-2}); compiled by STUK.

3 Previous inventories of long-lived radionuclides in the Baltic Sea sediments

In recent years, several investigations have been carried out to evaluate total inventories of certain long-lived radionuclides in the Baltic Sea sediments. Salo et al. (1986) estimated the total amounts of Sr-90, Cs-137, and Pu-239,240 bound to bottom sediments in the Baltic Sea at the beginning of the 1980s. They used two different methods in their calculations. In the first method, they estimated the average contents of radionuclides (Bq m^{-2}) in soft and hard sediments in various sea areas and multiplied them by the areas (Table 1) of soft and hard bottoms in each. The second method was based on an estimation of approximate mean concentrations of radionuclides in sinking matter and assumed sedimentation rates in different areas of the Baltic Sea. They concluded that at the beginning of the 1980s the total amounts of Cs-137, Sr-90, and Pu-239,240 bound in Baltic Sea sediments were 277 TBq, 12 TBq, and 15 TBq, respectively.

Inventory
An inventory is the estimate of the total amount of a radionuclide in a certain compartment, for example, sediment or water.

The uneven distribution of Chernobyl fallout in the drainage area of the Baltic Sea has made the calculations more difficult. On the other hand, although the global fallout of the nuclear weapons tests was more or less evenly deposited in the northern hemisphere, the deposition of radionuclides into sediments was not evenly distributed at that time either owing to differences in the sedimentation rate in different areas.

The first preliminary calculations of the Cs-137 inventory in the Baltic Sea sediments, made by the MORS Group soon after the Chernobyl accident, resulted in values of 520 TBq in 1986 and 856 TBq in August 1987 (HELCOM, 1989). These figures were based on measurements of Cs-134 and an initial activity ratio of 0.52 between Cs-134 and Cs-137. Direct calculations from measured Cs-137 concentrations in sediments gave an inventory of 1466 TBq in August 1987, as the sum of Cs-137 from all sources. However, these calculations were based only on soft-bottom values and the fact that lower values are associated with hard bottoms was not taken into account.

Sediment
Sediment covers the seafloor. It can consist of sand, gravel or mud, depending on the different conditions in the environment. In general, hard bottoms consist of sand and gravel, and the soft bottoms are muddy.

Sedimentation
As a result of a combination of biological and physical processes, solid material is transported from the water column towards the seafloor. This material accumulates in layers which can provide information about the history of the water body.

Using the same method as Salo et al. (1986) based on the average contents of radionuclides (Bq m^{-2}) in soft and hard sediments in various sea areas, Ilus et al. (1995) estimated that the total amount of Cs-137 in the Baltic Sea sediments was 1 400 TBq in 1990–1991 and that of Pu-239,240 was 18 TBq in 1987–1988. In comparison with the values given by Salo et al. (1986), this meant that the Chernobyl fallout had increased the total amount of Cs-137 by a factor of 5, whereas the increase of Pu-239,240 was negligible. Salo et al. (1986) assumed that Sr-90 and Cs-137 were distributed between hard and soft bottoms in

Sub-region	Total (km^2)	Soft bottom (km^2)	Hard bottom (km^2)
Bothnian Bay	37 000	16 000	21 000
Bothnian Sea	79 000	40 000	39 000
Gulf of Finland	30 000	16 000	14 000
Baltic Proper	209 000	99 000	110 000
Gulf of Riga	19 000	7 000	12 000
Total	374 000	178 000	196 000

Table 1. Areas of soft and hard bottoms (km^2) in different regions of the Baltic Sea (Salo et al., 1986). The values were measured planimetrically from maps of Quaternary deposits in the Baltic Sea (Winterhalter et al., 1981).

a ratio of 1:5, while the ratio was about 1:10 for Pu-239,240. Later results from the Finnish coastal areas have shown that the ratio for Cs-137 may be much smaller (1:20), which would lower the total inventory. By using this value, the estimate for Cs-137 in 1990–1991 was 1 200 TBq.

Because very few data are available on radionuclide concentrations in sinking matter and the use of concentrations observed in the surface sediment layer is doubtful for this purpose, the method based on concentrations in sinking matter and the sedimentation rate was not used. Furthermore, it was not possible to estimate the total amount of Sr-90 in the seabed because very few data on strontium in sediments have been reported since 1986. However, it is well known that the proportion of Sr-90 was quite small in the Chernobyl fallout.

The number of observations used in the above evaluation was relatively small. In the second evaluation of Ilus et al. (1999), the study material was more comprehensive, consisting of 129 sampling stations and 180 sediment cores. These were taken by STUK and the Finnish Institute of Marine Research in 1993–1997 from different sub-regions of the Baltic Sea. In this evaluation, the activity concentrations were time-corrected to 26 April 1996 (tenth anniversary of the Chernobyl accident) and the ratio of 1:20 was used to calculate Cs-137 values for hard bottoms. According to this investigation, the total inventory of Cs-137 in Baltic Sea sediments was 2 140 TBq in 1996. The significant difference between this value compared with that given before was supposed to result from the additional data on Cs-137 in sediments and the fact that Chernobyl-derived caesium had continued to be deposited onto the seabed.

In the third evaluation of Ilus et al. (2003), the total inventory of Cs-137 in the seabed of the Baltic Sea was estimated at 1 940–2 210 TBq in 1998. This was about eight times higher than the inventory made at the beginning of the 1980s (277 TBq) and about one and one half times higher than our estimate made in 1990–1991. The study was based on the Cs-137 data reported by all the Contracting Parties to the HELCOM/MORS database, enhanced with additional data from STUK and the Finnish Institute of Marine Research. Before the calculations were made, the quality of data was checked and the obviously questionable values were eliminated. The questionable values were identified, e.g., by comparing the results given by different laboratories for the same sampling stations. Then the latest observations reported by the laboratories for each station were chosen for manual checking of the results. After checking, the accepted values were used for calculating averages for each station. The sampling stations were grouped according to the sub-regions of the Baltic Sea and the median value of each sub-region was chosen to represent the area in question. The median was used because the averages were dominated by a few, very high “hot spot” values.

In this study (Ilus et al., 2003), two alternative ratios (1:5 and 1:20) were used to calculate Cs-137 values for hard bottoms. The values for hard bottoms were calculated from the above-mentioned median values of each sub-region. The content of Cs-137 (Bq m^{-2}) on soft and hard bottoms in different sub-basins was multiplied by the area of soft and hard bottoms in each according to the values given by Salo et al. (1986). The Belt Sea, the Kattegat, and the Sound were not included in the inventory owing to a lack of quantitative data on the area of soft and hard bottoms. Bojanowski et al. (1995a, 1995b) estimated that the total inventory of Cs-137 in the sediments of the Polish Economic Zone increased from 10 TBq to 45 TBq as a consequence of the Chernobyl accident. This area forms about 8% of the total area of the Baltic Sea. This estimation was in good agreement with the total inventory, taking into account that the Chernobyl fallout was clearly lower in the area surrounding the southern Baltic Proper than, e.g., in the areas surrounding the Bothnian Sea and the Gulf of Finland.

Sediment samples are usually taken from soft bottoms, i.e., from real sedimentation bottoms of sedimentation basins. Soft bottoms very often act as “sinks” for radionuclides, whereas hard bottoms are regarded as transport bottoms with very little accumulation of sinking matter. However, erosion bottoms are very seldom truly uncontaminated because bioturbation caused by benthic fauna may transfer contaminants and organic material into deeper sediment layers. Studies carried out on the Polish coast have shown that Cs-137 penetrates effectively into nearshore sandy sediments, and that rapidly accumulating sediments affected by river discharges have much higher contents of exchangeable radio-caesium than slowly accumulating marine sediments (Knapinska-Skiba et al., 1994, 1995, 1997).

It should be kept in mind that the calculations presented above are very rough because the uneven distribution of the Chernobyl fallout has created an additional difficulty in the calculations.

4 Project plan of the Sediment Baseline Study

In 2001 at its sixth meeting, the Project Group MORS identified the need for a Sediment Baseline Study as a future activity of the Group to provide a more detailed description of the spatial distribution of radionuclides in the Baltic Sea sediments. The Group noted that especially the western part of the Baltic Proper had not been covered in an adequate way in the preceding studies. Other areas for additional sampling were also considered.

The Chernobyl fallout is very unevenly distributed in the Baltic Sea area. During the 20 years since the accident, the MORS Group has made several efforts to evaluate the total inventories of the most important fallout nuclides (especially Cs-137) bound in the Baltic Sea sediments. With reference to the existing data, there were huge “white areas” lacking data especially on the Swedish side of the Baltic Proper, in the southeastern parts of the Baltic Proper, and in the Gulf of Riga.

One of the main objectives of the study was to obtain more information about the amounts of radionuclides in the sediments of the “white areas”. In addition to Cs-137, more information about Sr-90, Tc-99, I-129, Np-237, and other transuranic elements was needed. The study aimed to be arranged cooperatively with the planned sediment monitoring programme under HELCOM COMBINE and several national sediment monitoring programmes being planned, e.g., in Sweden.

The study was planned to cover all the sampling stations listed in the regular monitoring programme of MORS, with additional stations as appropriate. The western and southeastern parts of the Baltic Proper would be the main target areas in planning for additional samples. Furthermore, special attention was to be directed to coastal areas, to areas with hard bottoms, and to anoxic sediments. The resources among the Contracting Parties to increase sampling at additional stations were recognized to be limited. However, many Contracting Parties announced that they were prepared to consider their possibilities to provide additional data.

A more detailed Project Plan for the study was made in 2002 at the seventh meeting of the MORS-PRO Group. At that time, the project period was planned to span three years (2002–2004) and the general objectives of the project were defined as follows:

– to provide additional data on radionuclides in Baltic Sea sediments;
– the sampling and analysis activities should mainly be based on the permanent HELCOM/MORS Monitoring Programme. Additional samples would be taken with the aim of providing new data from so-called “white areas”, coastal areas, areas with hard bottoms, and anoxic sediments;
– special attention should be directed to Sr-90, Tc-99, I-129, and Np-237 analyses;
– the sampling and analysis activities would be coordinated by Finland so that the whole Baltic Sea area would be covered (as fully as possible) by new data on Cs-137, Sr-90, Tc-99, I-129, Np-237, and Pu and Am isotopes.

A preliminary plan of national contributions to the project was also defined in the MORS-PRO 7 meeting.

Due to the delay in sampling and other problems that arose in many countries, the eighth meeting of the Group decided to prolong the duration of the Study by one year. At the same meeting, a detailed list of national contributions was updated. The Risø National Laboratory promised to contribute to the project by analysing about 100 sediment samples taken by the other participants for Np-237, Tc-99, and transuranics. The ninth meeting prolonged the timetable of the project further until 2006.

5 Results and discussion

In this section, primary attention has been given to Cs-137 because it was the main long-lived radionuclide in the Chernobyl fallout. The role of Cs-137 is particularly important in sedimentological studies because the affinity of caesium to clay particles is well known. Certain amounts of Cs-137 already occurred in the sediments of the Baltic Sea before the Chernobyl accident, resulting from the nuclear weapons tests in the 1950s and 1960s. Nevertheless, the proportion of the "old" caesium is beginning to be insignificant, particularly since the caesium peak of the global fallout is already buried into deeper sediment layers.

The sampling and analysis activities were coordinated by STUK (Finland) so that the Baltic Sea area was covered as fully as possible. The Risø National Laboratory (Denmark) contributed to the project by analysing samples taken by the other participants for Sr-90, Tc-99, Np-237 and Pu-239, Pu-240 and Pu-239 + 240.

5.1 Cs-137

A large number of additional caesium-137 results were reported in the Sediment Baseline Study and used in the present update of inventory calculations. New data were supplied especially for the Swedish side of the Bothnian Sea and Bothnian Bay, and for the southeastern part of the Baltic Proper. Unfortunately, however, the western Baltic Proper (Gotland west and south) still remained relatively poorly investigated. The new data also provided additional information from coastal areas and from hard bottoms.

The present evaluation of the Cs-137 inventory is based on the data reported by all the Contracting Parties to the HELCOM/MORS database, enhanced with additional data from STUK for 1995–2005 and with Swedish data from the Gulf of Bothnia. Data were reported from 190 stations in the Baltic Sea, excluding 20 stations in the Belt Sea and the Danish Straits, which were not included in the inventory calculations due to the lack of data on the distribution of soft and hard bottoms in these sub-regions. Since many of the stations were sampled annually in 2000–2005, and the total amounts per square metre were based on numerous superimposed sediment slides, the total number of results was considerable. In total, the inventory was based on the results of 309 sediment cores taken from different sub-regions of the Baltic Sea.

Prior to starting the calculations, the quality of the data was checked and the obviously questionable values were eliminated. Questionable values were identified, for example, by comparing the results reported by different laboratories for the same sampling station. The most recent observations reported by the laboratories for each station were selected for manual checking of the results. After checking, the accepted values were used in calculating averages for each station. The sampling stations were grouped according to the respective sub-regions of the Baltic Sea, and the median value for each sub-region was chosen to represent the area in question. The median was used because the averages were dominated by a few, very high "hot spot" values, which were shown to misrepresent the results.

Sediment samples are usually taken from soft bottoms and, thus, a majority of the results in the database represent concentrations of radionuclides in soft sediments. However, hard bottoms contain much less particle-bound radionuclides than soft bottoms. Therefore, two alternative ratios (1:5 or 1:20) were used to calculate Cs-137 values for hard bottoms analogously with the earlier calculations. The values for hard bottoms were calculated from the above-mentioned median values for each sub-region. The content of Cs-137 (Bq m^{-2}) on soft and hard bottoms in the different sub-basins was multiplied by the area of soft and hard bottoms in each, according to the values given by Salo et al. (1986) (see Table 1). These values were measured planimetrically from maps of Quaternary deposits in the Baltic Sea (Winterhalter et al., 1981).

According to recent Finnish results from the southeastern Baltic Proper, the ratio 1:5 (20%) seems to overestimate the amounts of Cs-137 on hard bottoms compared to those on soft bottoms. At the eight stations taken in 2004 by the STUK from the southeastern Baltic Proper, the total amounts of Cs-137 on hard bottoms were 1–14%

(average 3%) of those on soft bottoms. Thus, the lower value given in our inventory estimates may be more realistic, but since the inventory is still based on a limited number of observations, and taking into account the considerable variability of the data, it is more relevant to give a range rather than a single value as a result.

The spatial distribution of the total amounts of Cs-137 at different sampling stations in the Baltic Sea is shown in Figure 2. The highest total amounts of Cs-137 per m^2 were observed in the Bothnian Sea. The maximum value, 125 000 Bq m^{-2}, was recorded in 1998 from the northernmost part of the Bothnian Sea (Ilus et al., 2003), and values exceeding 110 000 Bq m^{-2} were also recorded in the sea area off Gävle. The median total amount of Cs-137 was 36 400 Bq m^{-2} on the soft bottoms of the Bothnian Sea. The corresponding value for the soft sediments in the Bothnian Bay was 9 700 Bq m^{-2}.

In the Gulf of Finland, the maximum amount of Cs-137 was 42 700 Bq m^{-2} in the middle of the eastern part in 2003, and values exceeding 38 000 Bq m^{-2} were also recorded in the easternmost parts of the Gulf. The median total amount in the soft sediments of the Gulf of Finland was 10 400 Bq m^{-2}. In the Gulf of Riga, the maximum total amount of Cs-137 was 21 800 Bq m^{-2} at a

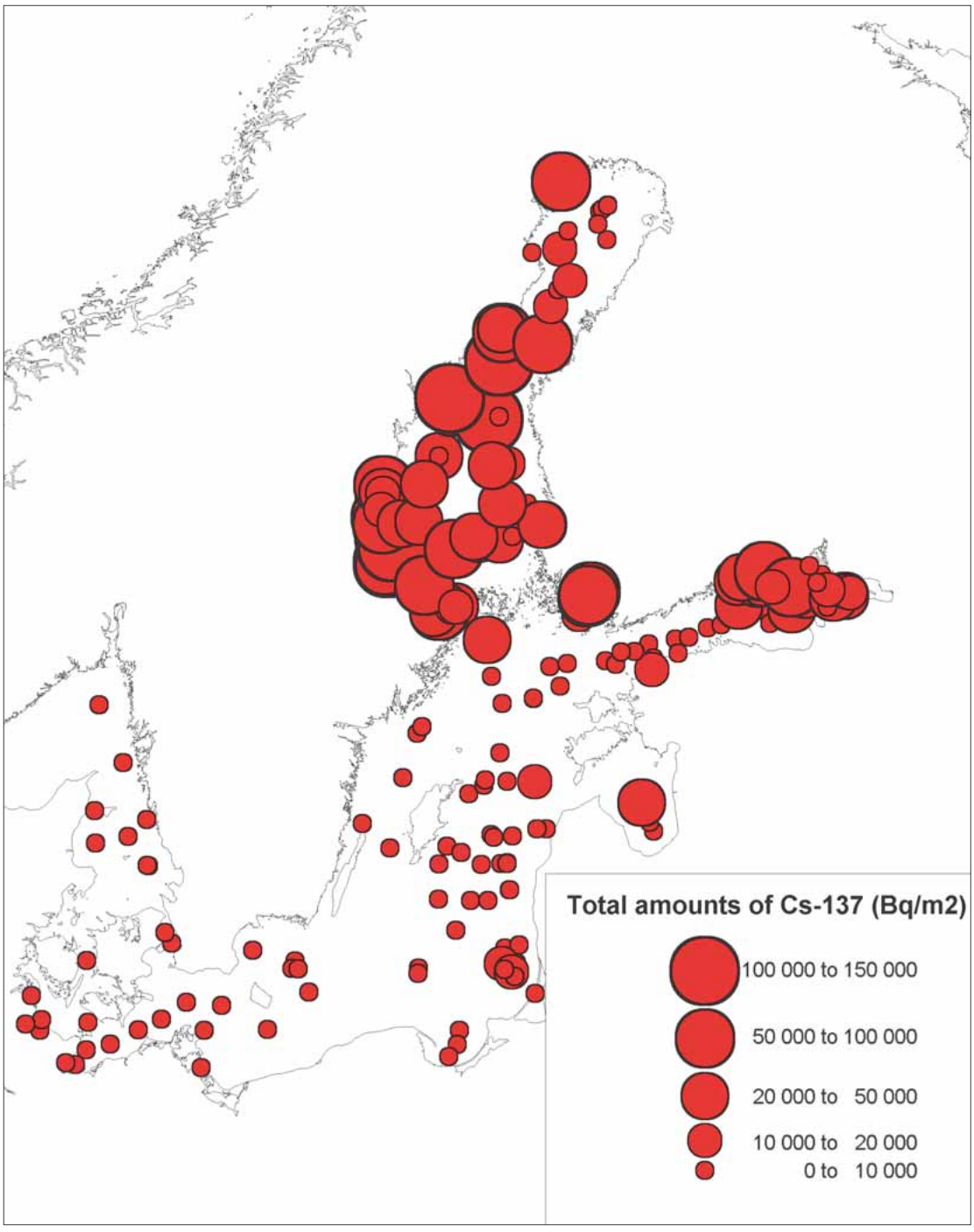


Figure 2.
Total amounts of Cs-137 (Bq m^{-2}) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

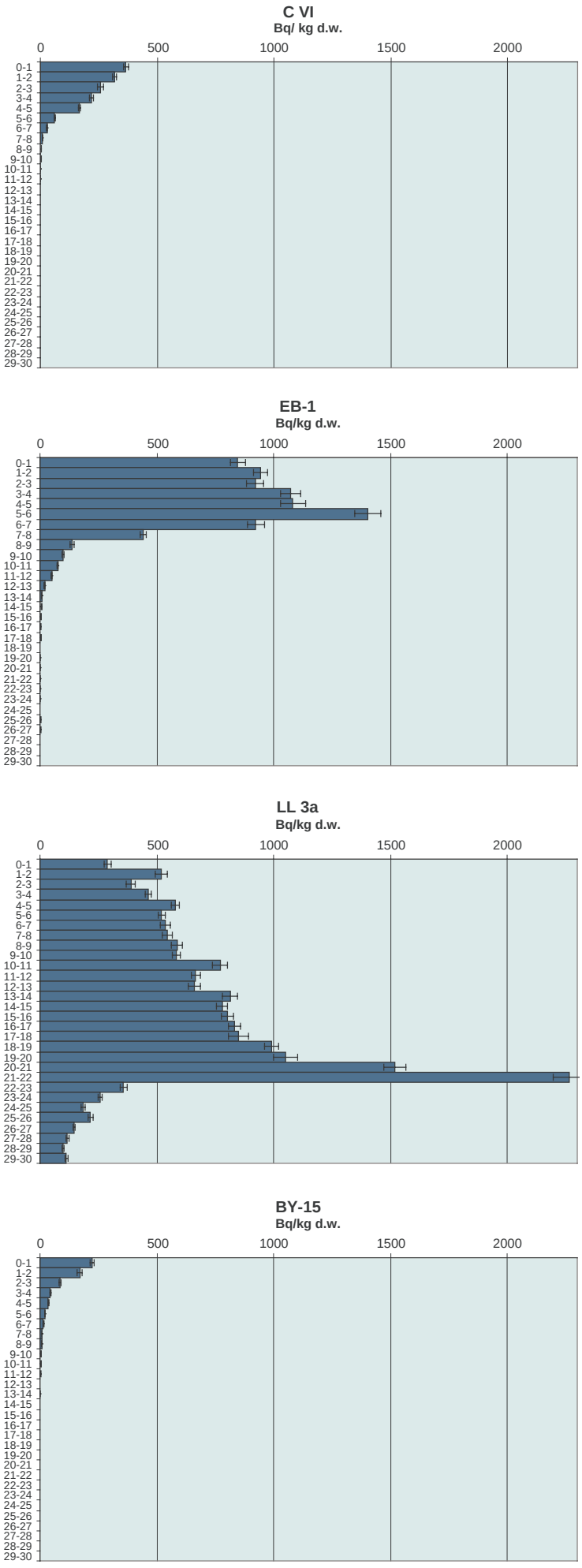


Figure 3.
Vertical distribution of Cs-137 (Bq kg^{-1} d.w.) at some stations in the Bothnian Bay (CVI), Bothnian Sea (EB1), Gulf of Finland (LL3a), and Baltic Proper (BY15) in 2003.

soft bottom station in the middle of the Gulf. In the Baltic Proper, the amounts of Cs-137 in sediments varied considerably, from 40 Bq m^{-2} to 14 700 Bq m^{-2} with a median value of 1 970 Bq m^{-2}. In the Belt Sea, the Kattegat, and the Sound, the median value was 1 370 Bq m^{-2}.

The vertical distribution of Cs-137 in sediments (Figure 3) reflects large differences in the sedimentation conditions at different stations, which depend on the bottom topography, sedimentation rate, bioturbation (which may partly explain the peak concentrations in the uppermost sediment layer), etc.

Depending on the method used (either 1:20 or 1:5 for the total amounts on hard bottoms), the total inventory of Cs-137 in the seabed of the Baltic Sea was estimated at 2 100–2 400 TBq. This is about 8–9% more than in the previous evaluation in 1998 (Ilus et al., 2003). The difference is explained by the additional new data, which have given greater precision to the calculations, and by the fact that Chernobyl-derived caesium has continued to be deposited onto the seabed. The inclusion of new data from the Gulf of Riga has also increased the total inventory.

The inventories for the different sub-regions are given in Table 2. The dominant role of the Bothnian Sea as an accumulation basin for Chernobyl caesium has continued to strengthen, amounting to 73% of the total inventory (1 530–1 740 TBq). The proportion occurring in the Gulf of Finland has clearly decreased from the previous estimate, while proportions in the Baltic Proper and the Bothnian Bay have increased slightly.

Sub-region	Number of stations	Sum (hard 1:20) (TBq)	Sum (hard 1:5) (TBq)	Percent of total (%)
Bothnian Bay	17	165	195	8
Bothnian Sea	58	1530	1740	73
Gulf of Finland	46	175	195	8
Baltic Proper + Gulf of Riga	69	230	270	11
Total	190	2100	2400	100

Table 2. Inventories of Cs-137 in different sub-regions of the Baltic Sea based on the areas of soft and hard bottoms in each.

The results were not decay-corrected in this evaluation because very often the total amounts of Cs-137 seemed to vary at the same station in consecutive years without any trend, or remained unchanged during the whole study period. The variability seemed to be caused by other factors than the half-life of Cs-137, for example, differences in the exact sampling position, success of sampling, etc. Figure 4 shows the variability of results at the stations LL3a (Gulf of Finland) and EB1 (Bothnian Sea) in 2000–2005. In both cases, the total amounts of Cs-137 were relatively stable during the whole project period. However, if the results were to be decay-corrected to the middle of the study period (2002), the total inventory would be reduced by about 3%, and if they were to be decay-corrected to the end of 2005, the total inventory would be reduced by about 9%. The physical half-life of Cs-137 is 30.1 years.

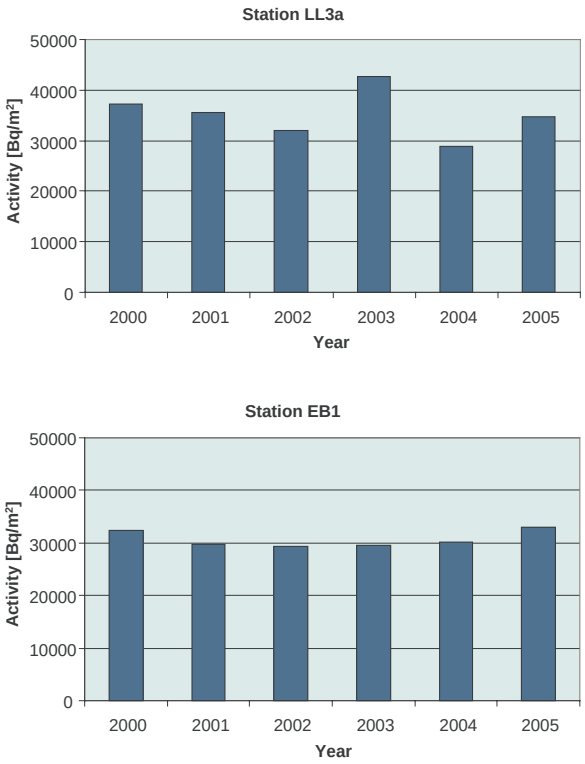


Figure 4. Total amounts of Cs-137 (Bq m^{-2}) at the stations LL3a (Gulf of Finland) and EB1 (Bothnian Sea) in 2000–2005.

5.2 K-40

Potassium-40 is a naturally occurring primordial radionuclide and forms a major source of radioactivity in the environment. Average activity concentrations of K-40 (Bq kg^{-1} dry weight) in the surface sediments (0–10 cm) of the Baltic Sea are shown in Figure 5. The spatial differences are due to the type of sediment at different sampling stations. Higher concentrations tend to occur in finer sediments. Typical activity concentrations of K-40 in soft sediments of the Baltic Sea are 600–1 100 Bq kg^{-1} dry weight.

In general, K-40 is quite evenly distributed in the seabed. Consequently, since samples are usually taken only from the surface layers of the sediments and the lengths of the sediment cores vary considerably, it is not relevant to evaluate the total inventory of K-40 in the Baltic Sea sediments based on the results from the surficial sediments only. In spite of that, supposing that the total amounts of K-40 would be equal on soft and hard bottoms (a not-proven starting hypothesis), a rough estimate results in a total inventory of about 8 500 TBq of K-40 in the 0–10 cm surface layer of the Baltic Sea sediments. This is about four times higher than the total inventory of Cs-137 given above.

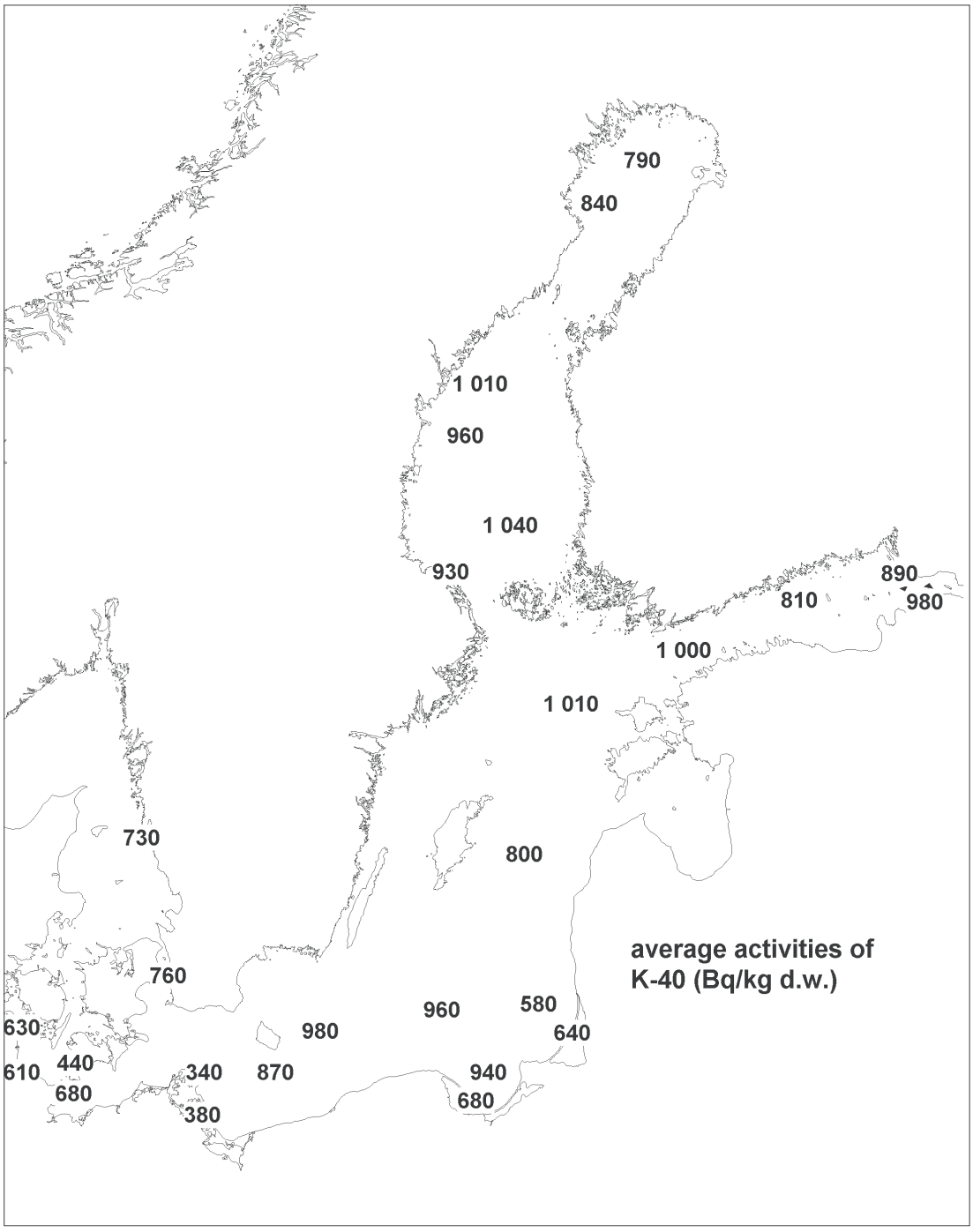


Figure 5.
Average activity concentrations of K-40 (Bq kg^{-1} d.w.) in surface sediments (0–10 cm) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

5.3 Sr-90

Traditionally, strontium-90 has been, together with Cs-137, the other intensively monitored artificial radionuclide in radioecological studies because it is one of the most important fission products and its half-life is almost as long (28.5 years) as that of Cs-137. In the atmospheric weapons tests during the 1950s and 1960s, the dominant fallout nuclides were Cs-137 and Sr-90 in an activity ratio of about 1.6. Consequently, Sr-90 was intensively monitored in the environment after that. In recent years, the frequency of analysis of strontium has been significantly reduced. One reason is the laborious and time-consuming analytical method for Sr-90; however, in addition, there is less interest in strontium because its share in the Chernobyl fallout was much smaller than that of Cs-137. The total input of Sr-90 from the Chernobyl fallout into the Baltic Sea area was estimated at 80 TBq (decay corrected to 1991), while it was 4 100–5 100 TBq for Cs-137 (Nies et al., 1995).

Although the acquisition of recent data on Sr-90 in the Baltic Sea sediments was set as a target for the Sediment Baseline Study, relatively few results were reported to the database during the project. Furthermore, due to the wide diversity of the data reported (e.g., the length of the sediment props analysed varied from 3 cm to 40 cm), it was difficult to estimate the total current inventory of Sr-90 in the seabed of the Baltic Sea. Typical total amounts of Sr-90 in sediments were from about 50 Bq m^{-2} to 200 Bq m^{-2}. A rough estimate of the total inventory was calculated as 26 TBq, which is about two times higher than that (12 TBq) given by Salo et al. (1986) for the beginning of the 1980s. This seems to be in agreement with the deposition values and the concentrations in sediments.

5.4 Tc-99

The concentrations of technetium-99 were low in the sediments of the Baltic Sea. In four samples analysed, the activity concentrations of Tc-99 in surface layers of the sediments (0–10 cm) varied between 0.040 Bq kg^{-1} and 1.3 Bq kg^{-1} d.w. The highest value was in the Belt Sea and the lowest in the Gulf of Finland.

5.5 Ra-226

Radium-226 is a naturally occurring radionuclide in soils and sediments. Average activity concentrations of Ra-226 (Bq kg^{-1} dry weight) in surface sediments (0–10 cm) of the Baltic Sea are shown in Figure 6. In general, Ra-226 is quite evenly distributed in the seabed; typical activity concentrations in soft sediments of the Baltic Sea are 30–70 Bq kg^{-1} dry weight. Since samples are usually taken only from the surface layers of the sediments and the lengths of the sediment cores may vary considerably, it is not relevant to evaluate the total inventory of Ra-226 in the Baltic Sea sediments based on the results from the surficial sediments only. In spite of that, supposing that the total amounts of Ra-226 would be equal on soft and hard bottoms (a not-proven starting hypothesis), a rough estimate results in a total inventory of about 420 TBq of Ra-226 in the 0–10 cm surface layer of the Baltic Sea sediments.

5.6 Th-232

Thorium-232 is a long-lived naturally occurring radioactive metal in soil, rocks, water, and sediments. Results on Th-232 were reported only from the southern Baltic Proper, the Belt Sea, the Kattegat, and the Sound. The activity concentrations varied between 13 Bq kg^{-1} and 42 Bq kg^{-1} dry weight and the total amounts in the surface

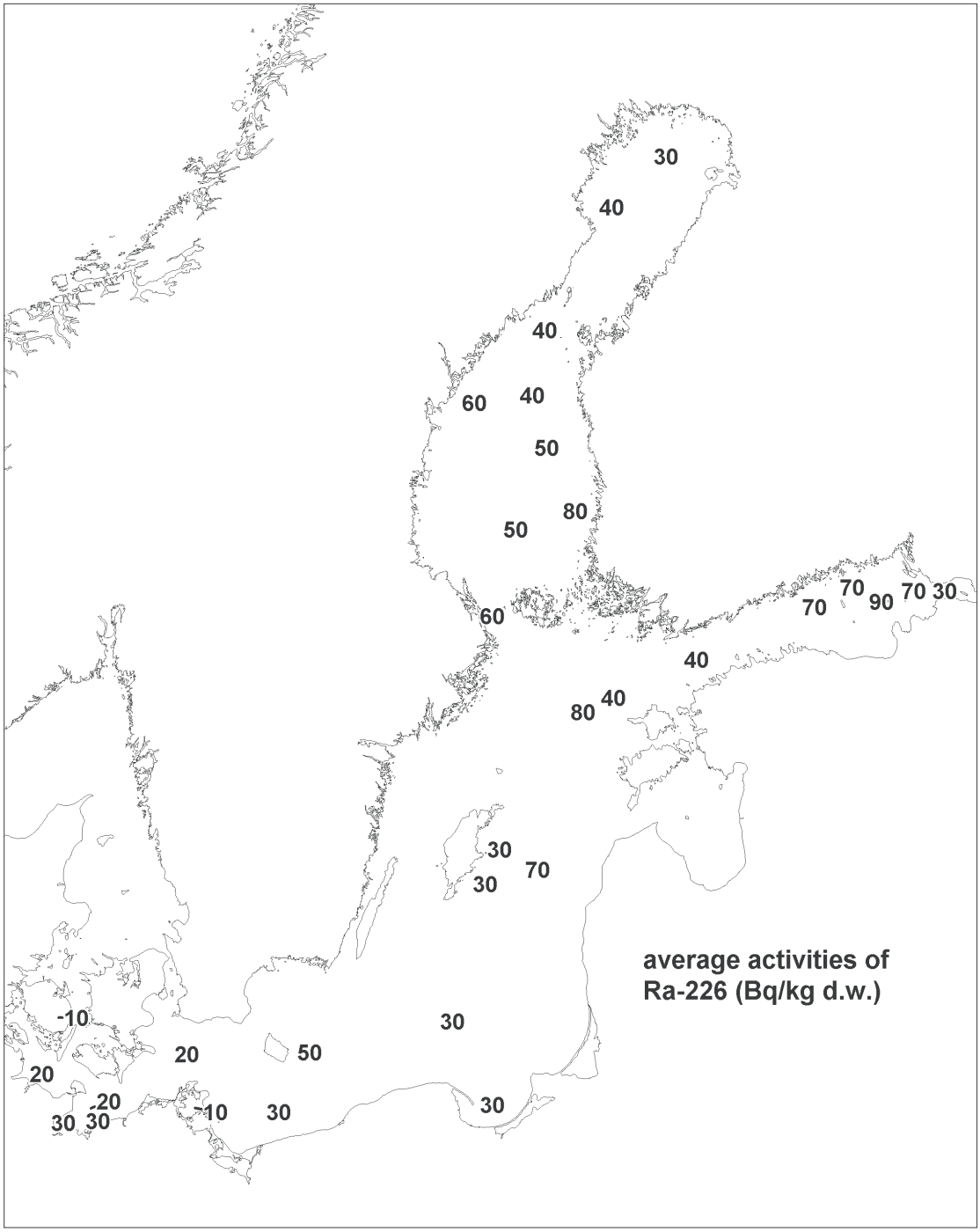


Figure 6. Average activity concentrations of Ra-226 (Bq kg^{-1} d.w.) in surface sediments (0–10 cm) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

sediment layers (0–30 cm) ranged between 1 900 Bq m^{-2} and 3 900 Bq m^{-2}.

5.7 Np-237

Neptunium-237 is an artificial radionuclide produced in nuclear reactors as a result of the irradiation of uranium-235 and uranium-238. Large amounts are found in spent nuclear fuel. The activity concentrations of Np-237 in surface sediments (0–20 cm) of the Baltic Sea were very low: 0.2–6.5 millibequerels per kg dry weight. The total amounts of Np-237 (mBq m^{-2}) at 26 sampling stations are shown in Figure 7. The total inventory of Np-237 in the seabed of the Baltic Sea was estimated at 0.02 TBq.

5.8 Pu-238

Results on plutonium-238 were reported from 11 stations in the southern Baltic Proper and four stations in the northern areas of the Baltic Sea. In the southern Baltic Proper, the activity concentrations of Pu-238 ranged from 6 mBq kg^{-1} to 250 mBq kg^{-1} dry weight in the surface sediments (0–15 cm) and the total amounts were generally from 0.5 Bq m^{-2} to 4.1 Bq m^{-2}. Only at one station in the southern Baltic Proper was the total amount of Pu-238 regularly higher (4.8–9.1 Bq m^{-2}). In the

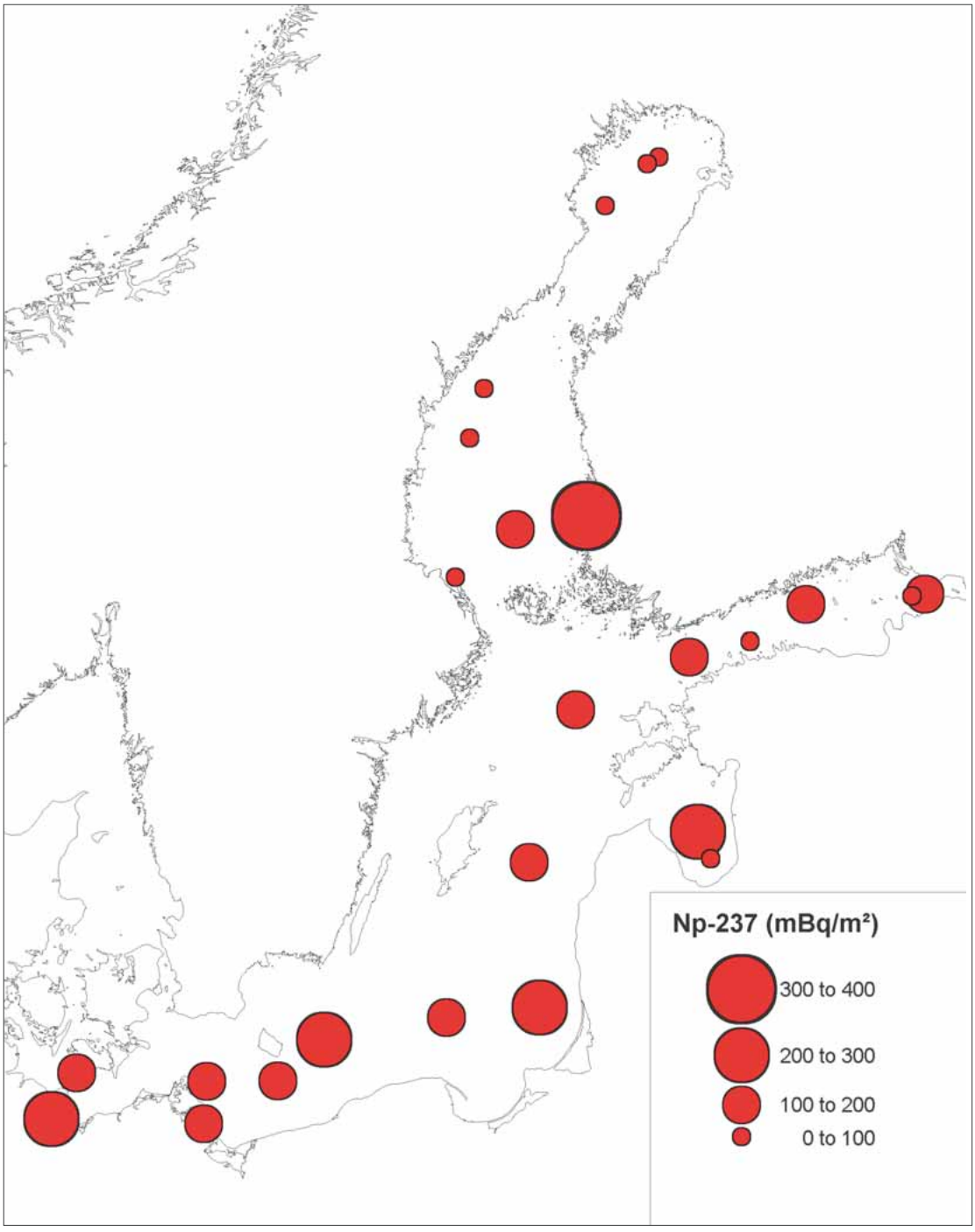


Figure 7.
Total amounts of Np-237 (mBq m^{-2}) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

northern Baltic Proper, the Gulf of Bothnia, and the Gulf of Finland, the total amounts were slightly higher (1.6–6.7 Bq m^{-2}) than generally in the southern Baltic Proper, as the activity concentrations ranged from 10 mBq kg^{-1} to 480 mBq kg^{-1} dry weight.

5.9 Pu-239,240

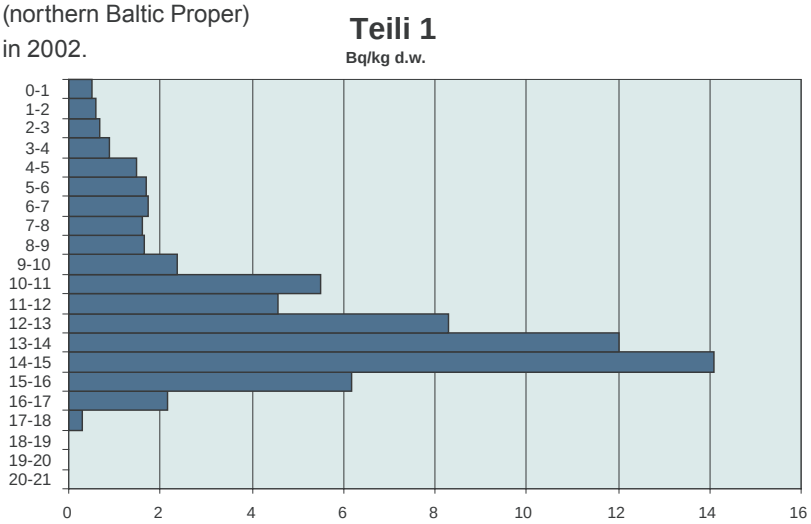
Plutonium is created from uranium in nuclear reactors. During the 1950s and 1960s, plutonium was dispersed worldwide from atmospheric testing of nuclear weapons. The fallout from these tests left low concentrations of plutonium in soils and sediments around the world. The fallout from the Chernobyl accident brought a small addition of plutonium to the aquatic environment of the Baltic Sea, but its contribution was minor compared to that of the weapons test fallout.

Data on Pu-239,240 were reported from 51 sediment cores taken from all sub-regions of the Baltic Sea, although most were from the southern Baltic Proper. The plutonium peak caused by the weapons test fallout provides an excellent time marker (1963), in addition to that of Cs-137 resulting from the Chernobyl accident, for the estimation of recent sediment accumulation rates in the Baltic Sea (Figures 8 and 9).

The total amounts of Pu-239,240 at different sampling stations in the Baltic Sea are shown in Figure 10. The inventories of Pu-239,240 in the sediments of different sub-regions are given in Table 3, as well as the maximum values and the median values (Bq m^{-2}) used in the inventory calculations. The ratio 1:10 was used in the estimation of Pu-239,240 values for hard bottoms versus soft bottoms, analogously with the earlier calculations published by Salo et al. (1986) from the beginning of the 1980s.

The total inventory of Pu-239,240 in the seabed of the Baltic Sea was estimated at 15.3 TBq. The portion for the Bothnian Sea was 45% of the total inventory and that for the Baltic Proper was 32%. If the ratios 1:20 or 1:5 were to be used for the total amounts of Pu-239,240 on hard bottoms compared to those on soft bottoms, the result would be 14.6 TBq or 16.8 TBq, respectively. The estimate is equal to that of Salo et al. (1986) from the beginning of the 1980s. This may be realistic taking into account the fact that the input of plutonium from Chernobyl into the Baltic Sea was relatively small, and on the other hand, the present estimation was based on a clearly more extensive study material.

Figure 8.
Vertical distribution of Pu-239,240 (Bq kg^{-1} d.w.) at the station Teili 1 (northern Baltic Proper) in 2002.



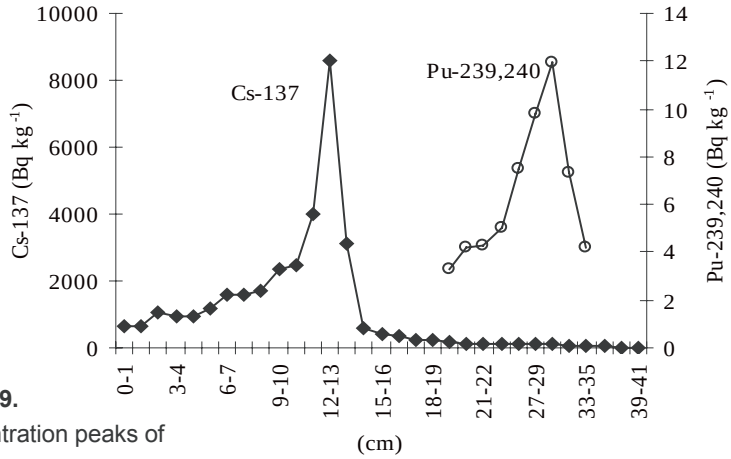


Figure 9.
Concentration peaks of Chernobyl-derived Cs-137 and weapons-tests-origin Pu-239,240 at the station K15 in the Gulf of Finland (Mattila et al., 2006).

5.10 Am-241

Americium is a man-made radionuclide produced in nuclear reactors and nuclear weapons detonations. Most americium-241 in the environment originates from the atmospheric testing of nuclear weapons during the 1950s and 1960s. Results on Am-241 were reported from three stations in the southern Baltic Proper and the Belt Sea and three stations in the Bothnian Bay and the Gulf of Finland. The activity concentrations in the samples ranged from 0.08 Bq kg^{-1} to 3.5 Bq kg^{-1} dry weight.

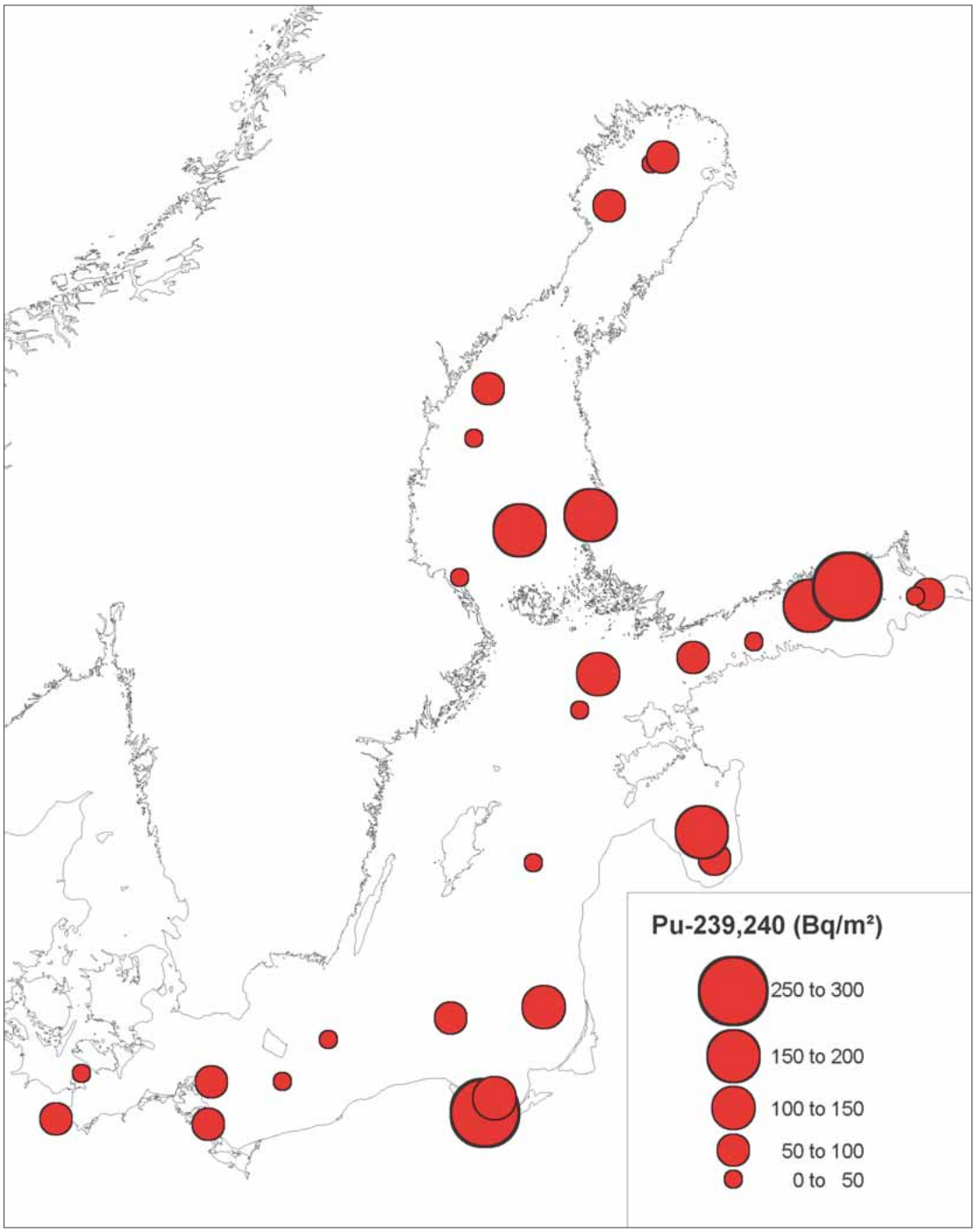


Figure 10.
Total amounts of Pu-239,240 (Bq m^{-2}) at different sampling stations in the Baltic Sea at the beginning of the 2000s.

Sub-region	Number of sediment cores	Maximum (Bq m^{-2})	Median (Bq m^{-2})	Total inventory (TBq)
Bothnian Bay	4	94	69	1.25
Bothnian Sea	8	178	156	6.86
Gulf of Finland	8	299	92	1.60
Baltic Proper + Gulf of Riga	29	265	47	5.60
Belt Sea	2	51		
Total	51			15.3

Table 3.
Total inventories and maximum and median values of Pu-239,240 in different sub-regions of the Baltic Sea.

6 Conclusions

The goal of this study was to complement the knowledge and inventories of long-lived radionuclides in the seabed of the Baltic Sea by providing additional data from so-called “white areas”, and from nuclides which were not recorded in the earlier surveys. According to the project plan, the sampling and analysis activities should mainly be based on the permanent HELCOM/MORS Monitoring Programme, but samples should also be taken from coastal areas and areas with hard bottoms, as well as from anoxic sediments. Especially in the case of Cs-137, the targets were fulfilled very well, although some parts of the western Baltic Proper still remained relatively poorly investigated. A large amount of data on certain other nuclides were compiled, but many gaps still remained concerning our knowledge on long-lived radionuclides in the seabed of the Baltic Sea as a whole.

The results show that about one half of the total input of Cs-137 from Chernobyl into the Baltic Sea area has accumulated in the seabed. The accumulation was strongest in the first 5–6 years after the fallout, but it is still in progress. Cs-137 is still transported from the drainage area to the sea, and in the sea from the water column to the bottom. In recent years, the accumulation of Cs-137 has become slower and the total inventories of this radionuclide in the seabed have stopped increasing, which means that the accumulation rate of Cs-137 and its radioecological half-life in the sediments are essentially attaining balance at present.

Chernobyl-derived Cs-137 was very unevenly distributed in the seabed of the Baltic Sea. The largest amounts were measured in the bottom sediments of the northern parts of the Bothnian Sea, the southern parts of the Bothnian Bay, and the eastern parts of the Gulf of Finland. In addition to the patchy nature of the atmospheric deposition, the distribution and accumulation in the sediments was affected by the character of the bottom, the sedimentation conditions, and the sedimentation rate in different areas. The maximum value in Baltic Sea sediments (125 000 Bq m^{-2}) was recorded in the northernmost part of the Bothnian Sea and was clearly higher than the highest deposition values recorded, e.g., from the Finnish territory in 1987. The dominant role of the Bothnian Sea as an accumulation basin of Chernobyl caesium is clear; its portion was 73% of the total inventory of the entire Baltic Sea.

The total inventories of certain long-lived radionuclides in the Baltic Sea sediments were estimated as follows:

Man-made radionuclides TBq		Natural radionuclides TBq	
Cs-137	2 100–2 400	K-40	8 500 [a,b]
Sr-90	26 [c]	Ra-226	420 [a,b]
Pu-239,240	15.3 [c]		
Np-237	0.02 [c]		

[a] in the surface (0–10 cm) sediment layer.
[b] supposing that the amounts are equal on hard/soft bottoms.
[c] rough estimate based on relatively few data.

The Sediment Baseline Study results show that the concentrations of naturally occurring radionuclides in Baltic Sea sediments remain at background levels. However, the concentrations of man-made radionuclides in 2000–2005 remained

higher than the target of the HELCOM ecological objective “radioactivity at pre-Chernobyl level”. This is particularly true for the Bothnian Sea and the Gulf of Finland, which received the largest amounts of the fallout in the Baltic Sea from the Chernobyl accident in 1986. Nonetheless, in 2000–2005 the concentrations of man-made radionuclides in sediments were generally at or below the concentrations of naturally occurring radionuclides and are not expected to cause harmful effects to the Baltic Sea wildlife.

In the course of time, the long-lived fallout nuclides, such as Cs-137 originating from the nuclear weapons tests and the Chernobyl accident, will continue to sink from the water phase to the seabed and then become buried into deeper sediment layers. Simultaneously, they slowly lose their availability for consumption by pelagic organisms and become available for consumption by benthic organisms; ultimately over time, they are buried deep enough in the sediment that they are no longer available to the biota. At the same time, the amounts of radionuclides such as Cs-137 with a half-life of 30 years continue to decay according to their radiological half-life.

After the Chernobyl accident, a large amount of data has been collected concerning the quantities of Cs-137 (and other gamma-emitting radionuclides) in the Baltic Sea sediments. Consequently, the present estimation of the total inventory of Cs-137 in the seabed of the Baltic Sea was based on an exceptionally large number of observations. To achieve a more comprehensive view of the total amount of all radioactive substances bound in the Baltic Sea sediments, however, it is recommended to continue the acquisition of additional data also on the more difficult to analyse radionuclides, such as Sr-90 and transuranic elements, and the naturally occurring radionuclides, many of which were not included in this consideration.

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Appendix

An Intercomparison of Radionuclide Analyses in a Baltic Sea Sediment Sample

Results of the HELCOM MORS-PRO intercomparison on Baltic Sea Sediment 2000

Tarja K. Ikäheimonen and Vesa-Pekka Vartti
STUK - Radiation and Nuclear Safety Authority
Helsinki, Finland

Introduction

As a part of the HELCOM MORS-PRO work, the MORS Group arranged an intercomparison exercise of radionuclide analyses using a sediment sample taken from the Baltic Sea. The sediment sample was taken by STUK on 21 June 1999 at the Station EB1 in the Bothnian Sea (latitude 61º04.00' N, longitude 19º43.78' E, at a depth of 130 m). The sampling was carried out with an Aquarius Box Corer (surface area of 18 cm x 18 cm). Fourteen parallel sediment cores were taken to achieve the target amount of 60 kg wet weight. The top layer (0–10 cm) of each core was sliced and used for the exercise. The material was sent to IAEA-MEL, which contracted with EUROLYO Ltd. for the freeze-drying and homogenization of the sample.

Aliquots of the homogenized sediment sample, *Baltic Sea Sediment 2000*, were delivered to the MORS Group in June 2000. Gamma-emitting nuclides, natural radionuclides, and transuranic elements were requested to be analysed.

Participants

Altogether eight laboratories from Denmark, Estonia, Finland, Germany, Latvia, Poland, and Sweden participated in the intercomparison as listed in Table A1.

Analytical results

All laboratories analysed the sediment sample for K-40 and Cs-137. Nine other gamma-emitting nuclides were also analysed, and of these, Cs-134, Pb-210, Ra-226, Th-228, Th-232, U-238, and Co-60 were analysed by more than one labora-

Laboratory	Country	Code
Risø National Laboratory, Roskilde	Denmark	RISO/DK
Estonian Radiation Protection Centre, Tallinn	Estonia	ERPC/ES
STUK – Radiation and Nuclear Safety Authority, Helsinki	Finland	STUK/FI
Federal Maritime and Hydrographic Agency, Hamburg	Germany	DHIG/GE
Federal Research Centre for Fisheries Institute of Fisheries Ecology, Hamburg	Germany	BFFG/GE
Ministry of Environmental Protection and Regional Development of Latvia, Riga	Latvia	LVEA/LV
Central Laboratory for Radiological Protection, Warsaw	Poland	CLOR/PO
Swedish Radiation Protection Institute, Stockholm	Sweden	SSI/SW

Table A1. Laboratories participating in the intercomparison exercise.

Lab.code	K-40	Uncert.	Cs-137	Uncert.	Cs-134	Uncert.
RISO/DK	998	100	1020	102	9.2	1.0
ERPC/ES	1100	80	1220	53	9.2	0.9
STUK/FI	980	59	1060	53	9.8	1.0
DHIG/GE	991	8.4	1140	5.82	9.97	0.47
BFFG/GE	1000	24	1100	27	9.95	0.28
LVEA/LV	795	24	988	20	9.30	1.58
CLOR/PO	1050	48	1060	21	-	-
SSI/SW	1130	17	1200	20	11.9	1.9
Mean	*1006*	*101*	*1098*	*82*	*9.9*	*0.94*

Table A2. Results for artificial gamma-emitting radionuclides in Bq kg^{-1} d.w. with an uncertainty of 1 sigma and the arithmetic means with the standard deviation (in the Uncert. column).

tory. Results for Sb-125, Ac-227, Rn-222, U-235, and Am-241 (with gamma) were each reported by one laboratory only (but not the same one). The results reported by only one laboratory and the results for Co-60 analysed by two laboratories are not considered in this report because the amount of Co-60 in the sample was very small and the results of the two laboratories differed by a factor of five from each other. Pu-239,240 was analysed by six laboratories, Pu-238 by three, and Am-241 (with alpha) by four laboratories. One laboratory reported Sr-90 and one Po-210 but these results are also not discussed here. The analytical results for radionuclides with more than one reported result are shown in Tables A2–A4, which also provide uncertainties of one sigma and arithmetic means of the results. In addition, the results are shown in column charts for each radionuclide in Figures A1–A11.

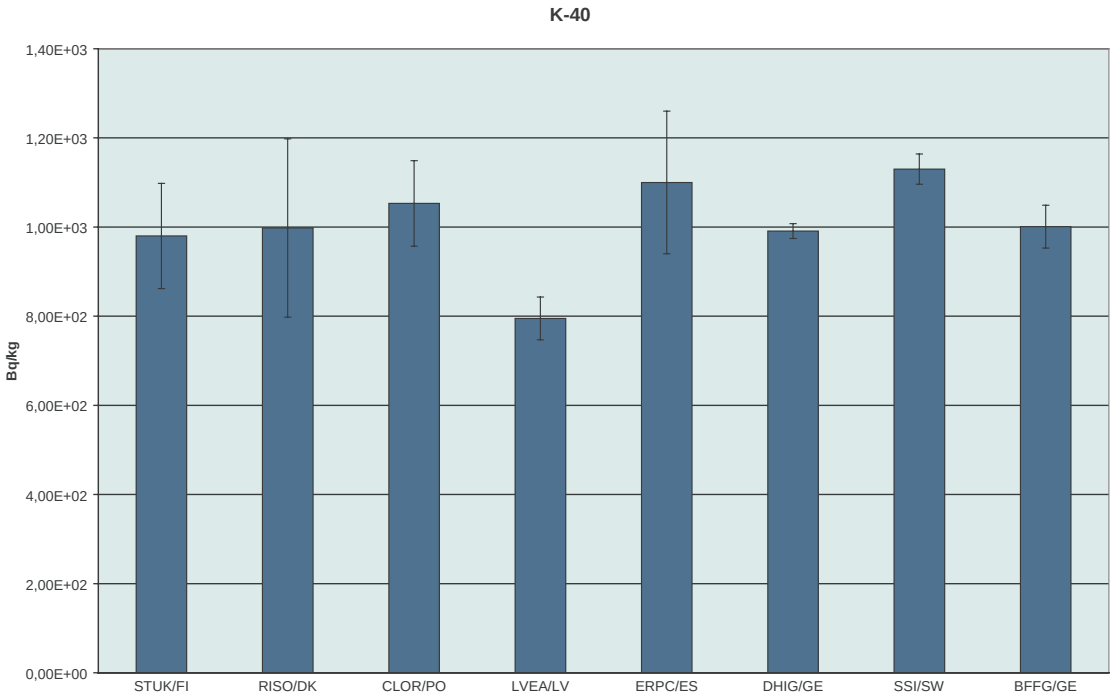
Table A3.
Results for natural radionuclides in Bq kg^{-1} d.w. with an uncertainty of 1 sigma and the arithmetic means with the standard deviation (in the Unc. column).

Lab.code	Pb-210	Unc.	Ra-226	Unc.	Th-228	Unc.	Th-232	Unc.	U-238	Unc.
RISO/DK	327	33	55	5.5	-	-	64	6.4	-	-
ERPC/ES	-	-	-	-	-	-	-	-	-	-
STUK/FI	329	23	54	2.0	72	5	69.4	4.9	82	5
DHIG/GE	360	11	6.26	0.69	-	-	-	-	72.6	3
BFFG/GE	361	12	8.31	4.6	65.9	1.2	-	-	63	2.6
LVEA/LV	-	-	-	-	-	-	-	-	-	-
CLOR/PO	-	-	-	-	-	-	-	-	-	-
SSI/SW	-	-	-	-	-	-	-	-	-	-
Mean	*344*	*19*	*63.7*	*13.5*	*69.0*	*4.3*	*65.9*	*3.1*	*72.5*	*9.5*

Table A4.
Results for transuranic elements in Bq kg^{-1} d.w. with an uncertainty of 1 sigma and the arithmetic means with the standard deviation (in the Unc. column).

Lab.code	Pu-239,240	Unc.	Pu-238	Unc.	Am-241	Unc.
RISO/DK	2.88	0.29	-	-	1.25	0.13
ERPC/ES	-	-	-	-	-	-
STUK/FI	3.04	0.18	0.12	0.019	1.17	0.09
DHIG/GE	2.87	0.075	0.13	0.014	1.32	0.014
BFFG/GE	3.05	0.12	0.138	0.016	1.30	0.044
LVEA/LV	-	-	-	-	-	-
CLOR/PO	3.15	0.26	-	-	-	-
SSI/SW	2.96	0.08	-	-	-	-
Mean	*2.99*	*0.11*	*0.13*	*0.009*	*1.26*	*0.07*

Figure A1.
Results for K-40 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



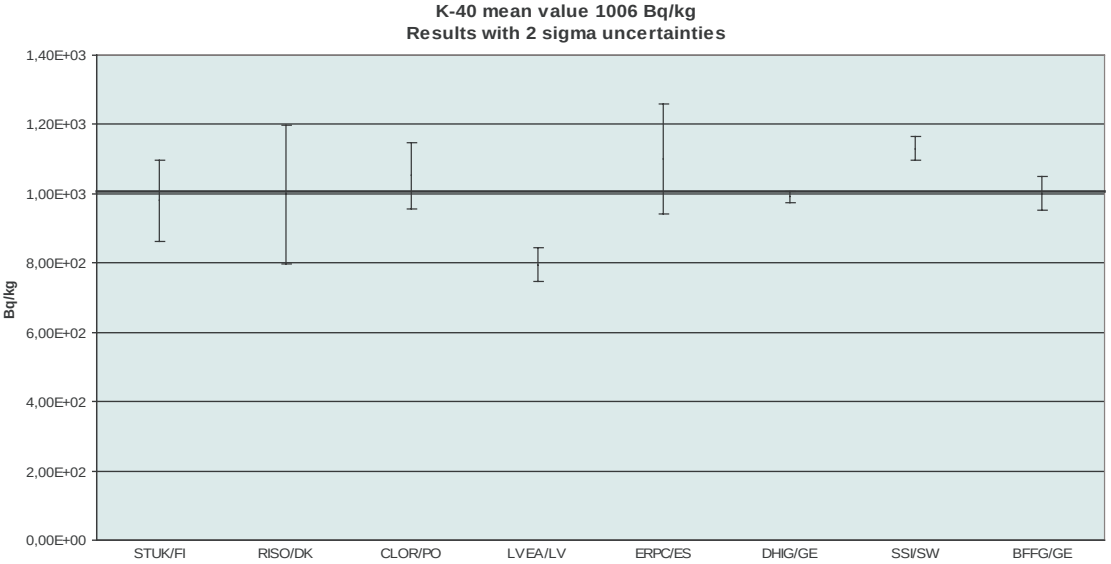
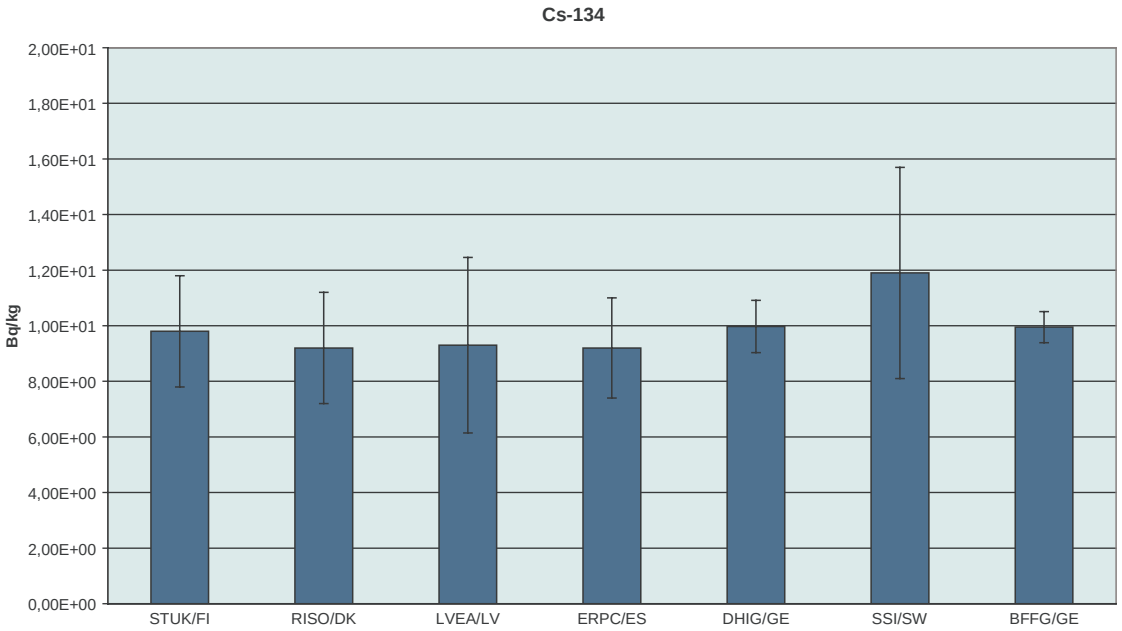


Figure A2.
Results for Cs-134 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



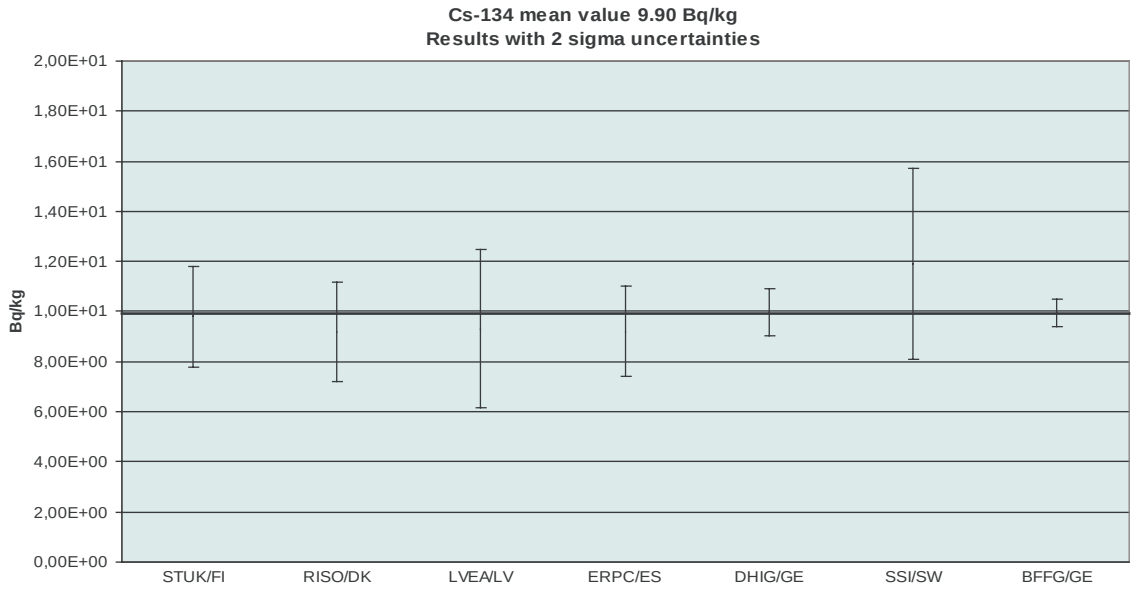
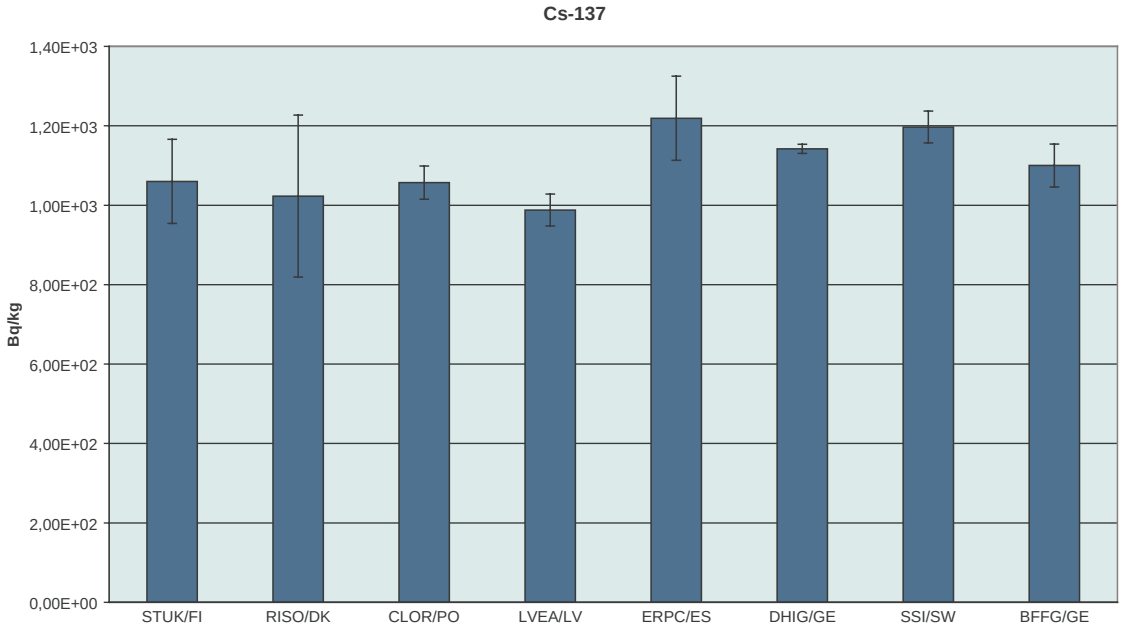


Figure A3.
Results for Cs-137 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



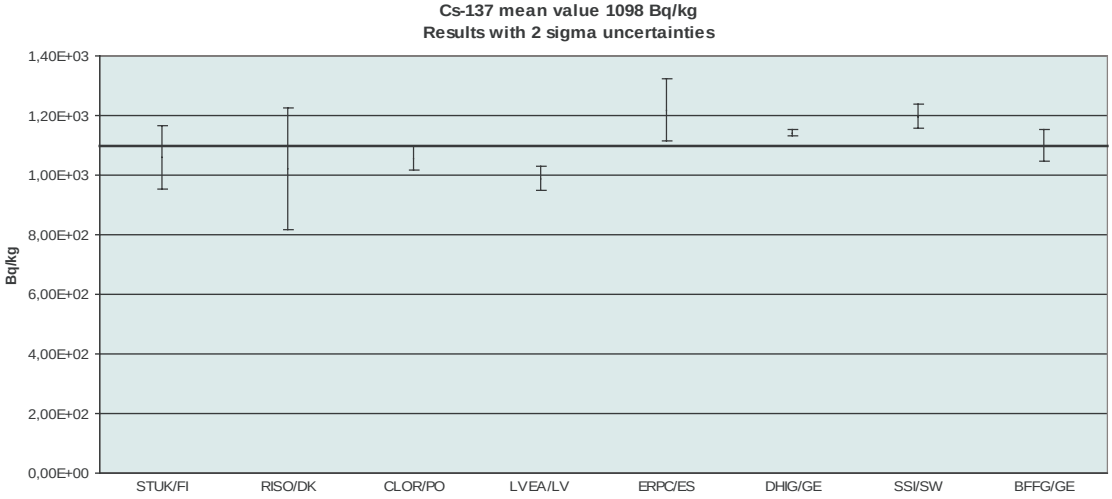
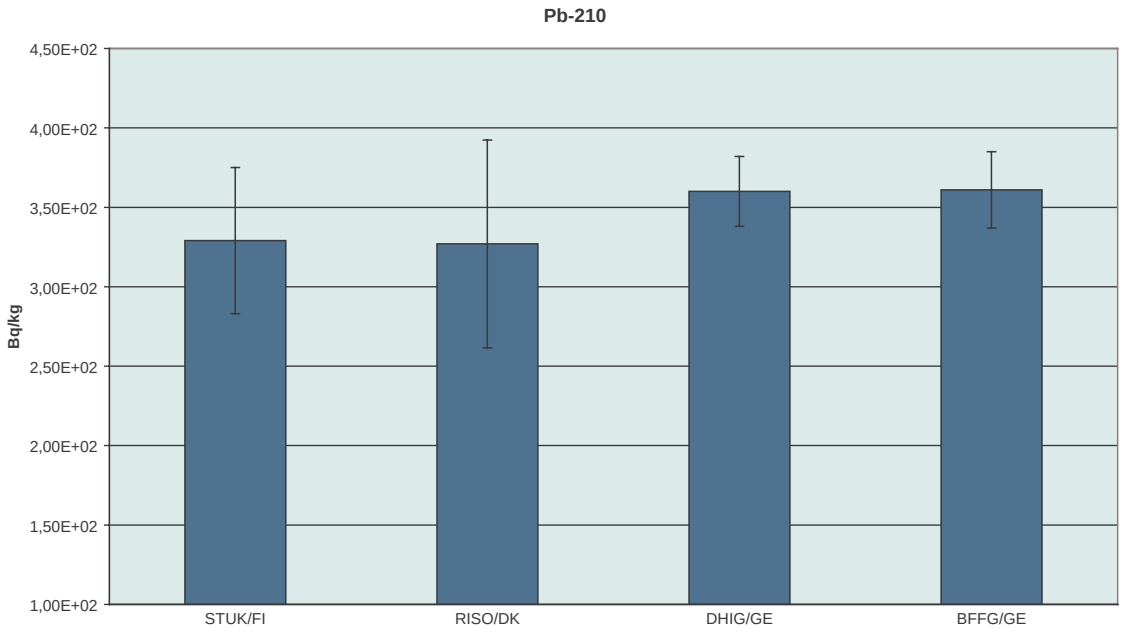


Figure A4.
Results for Pb-210 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



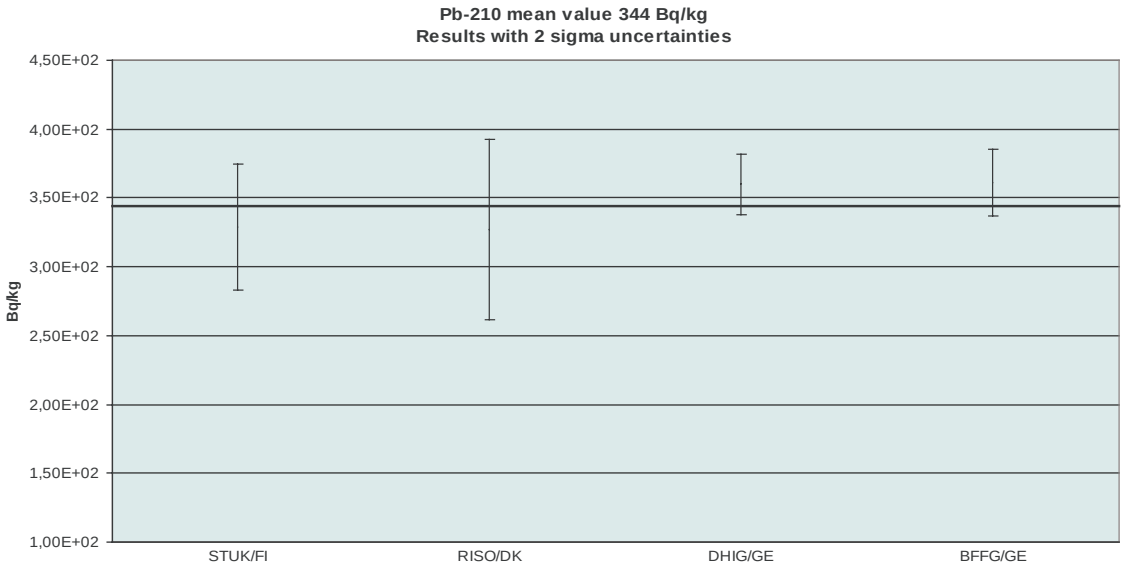
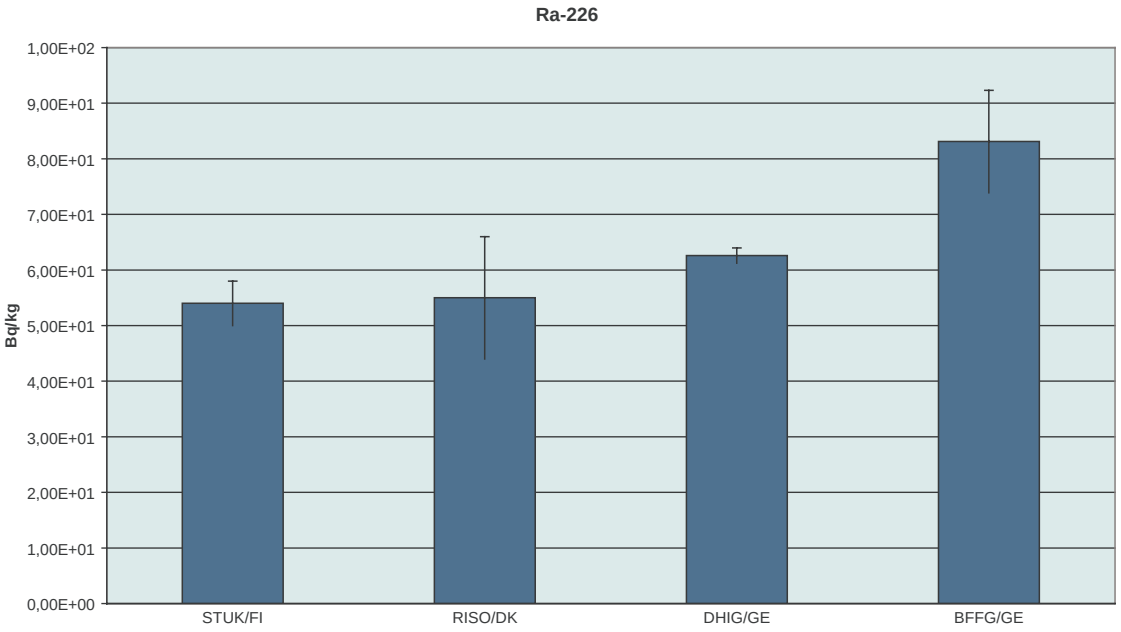


Figure A5.
Results for Ra-226 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



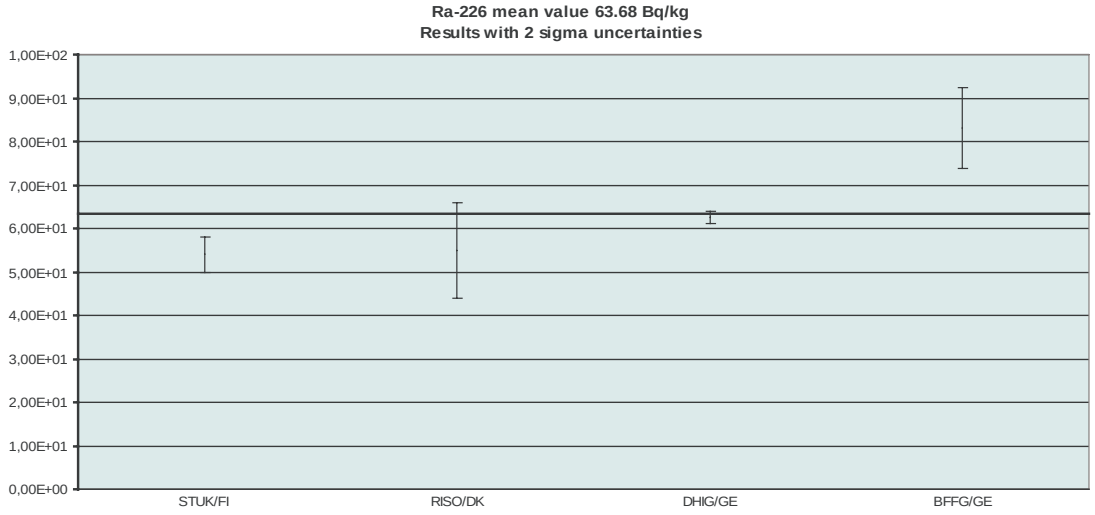
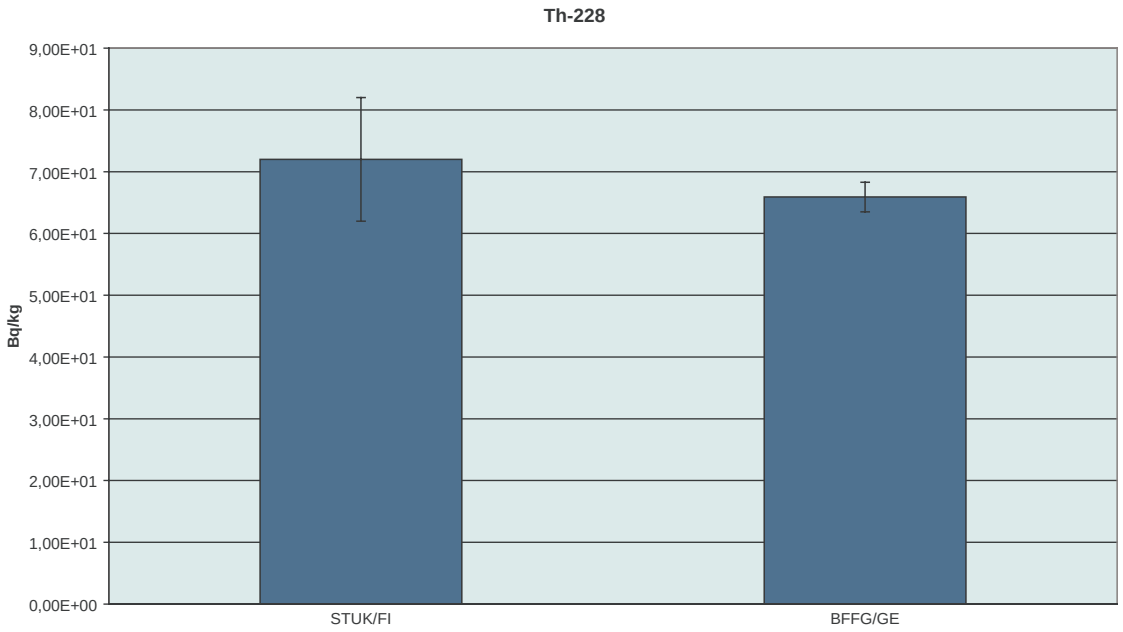


Figure A6.
Results for Th-228 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



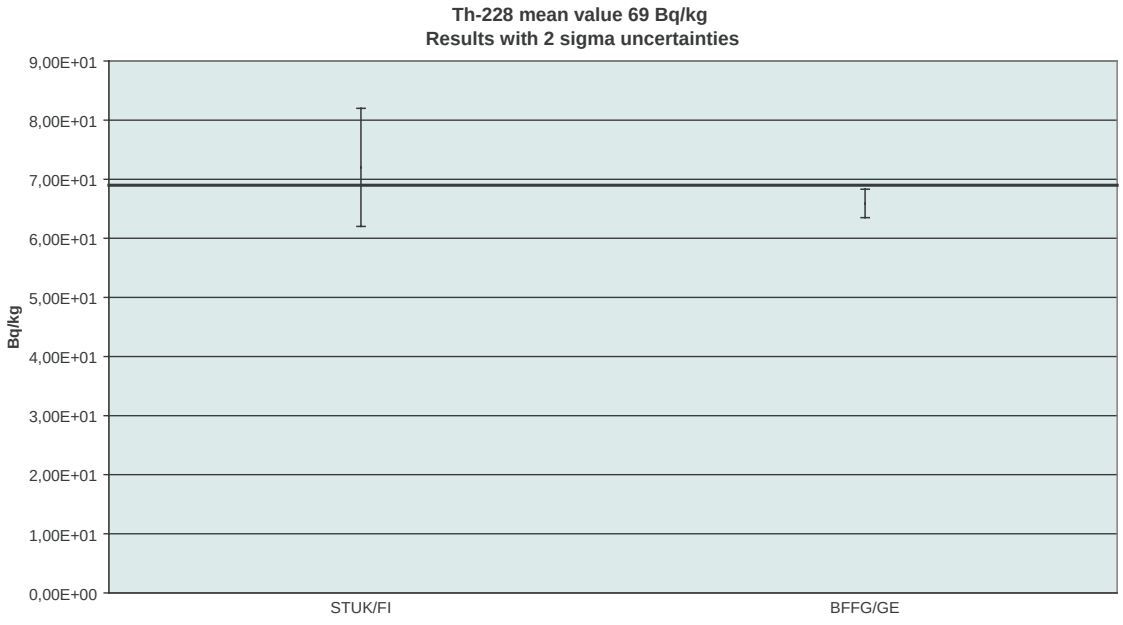
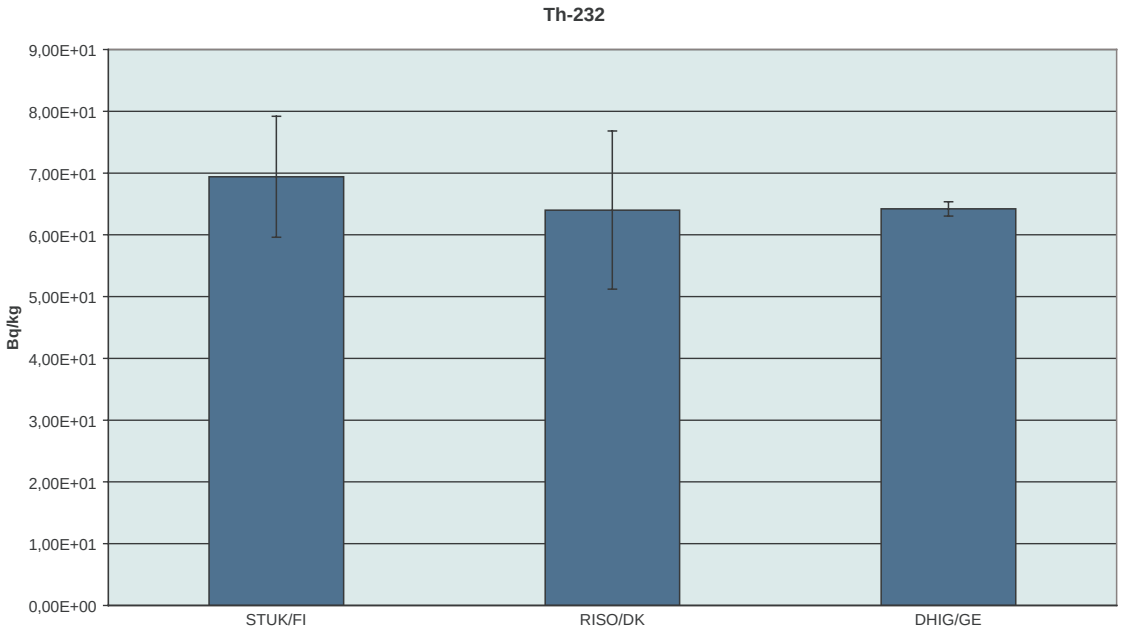


Figure A7.
Results for Th-232 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



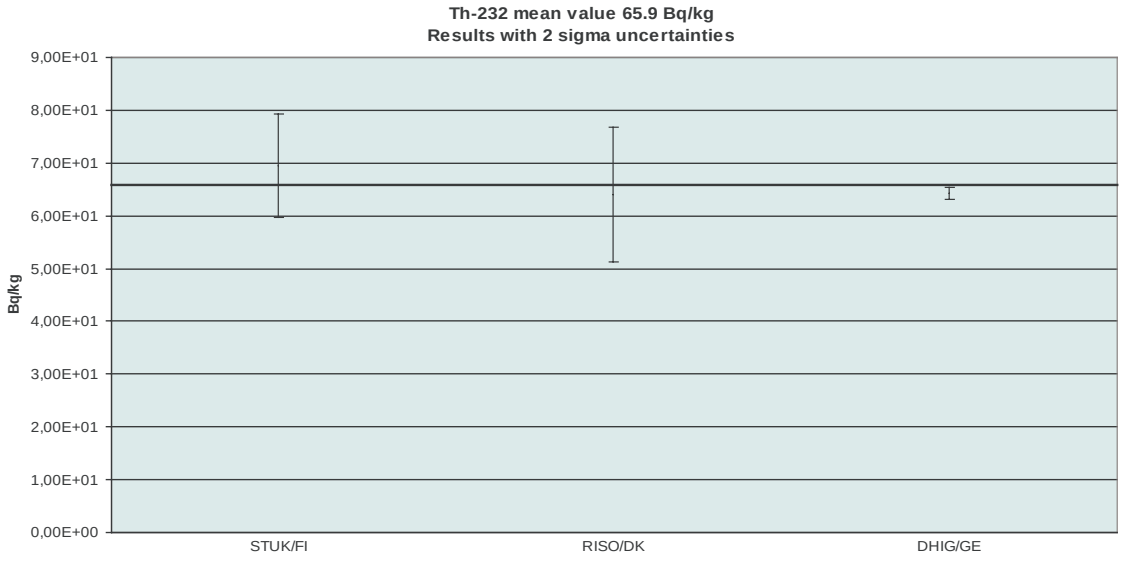
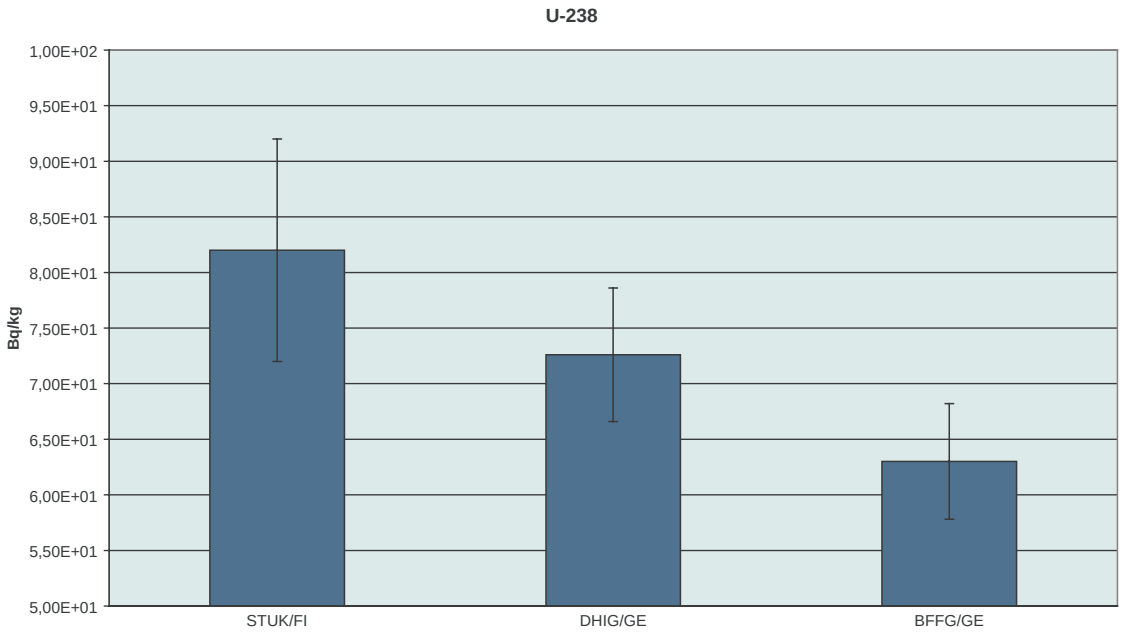


Figure A8.
Results for U-238 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



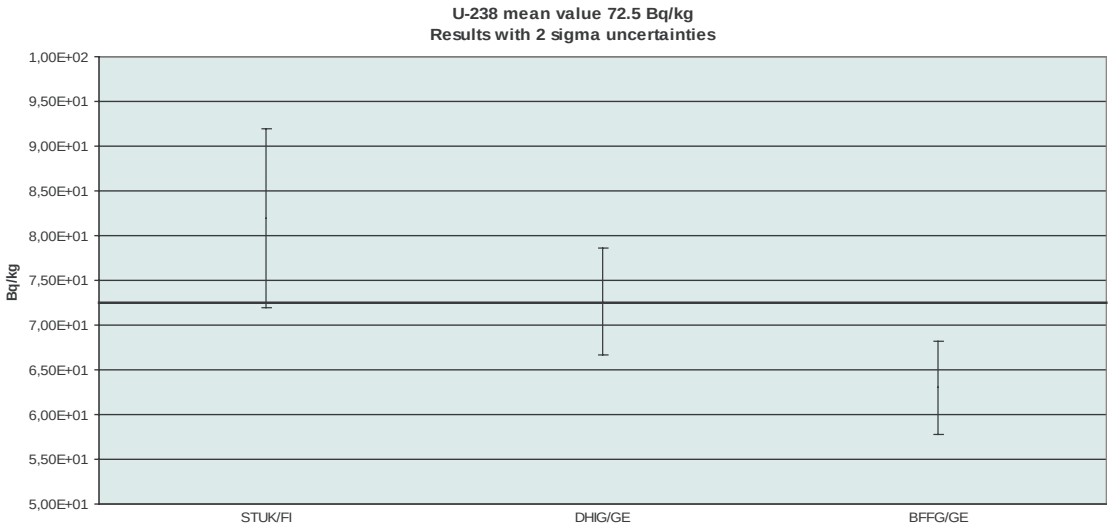
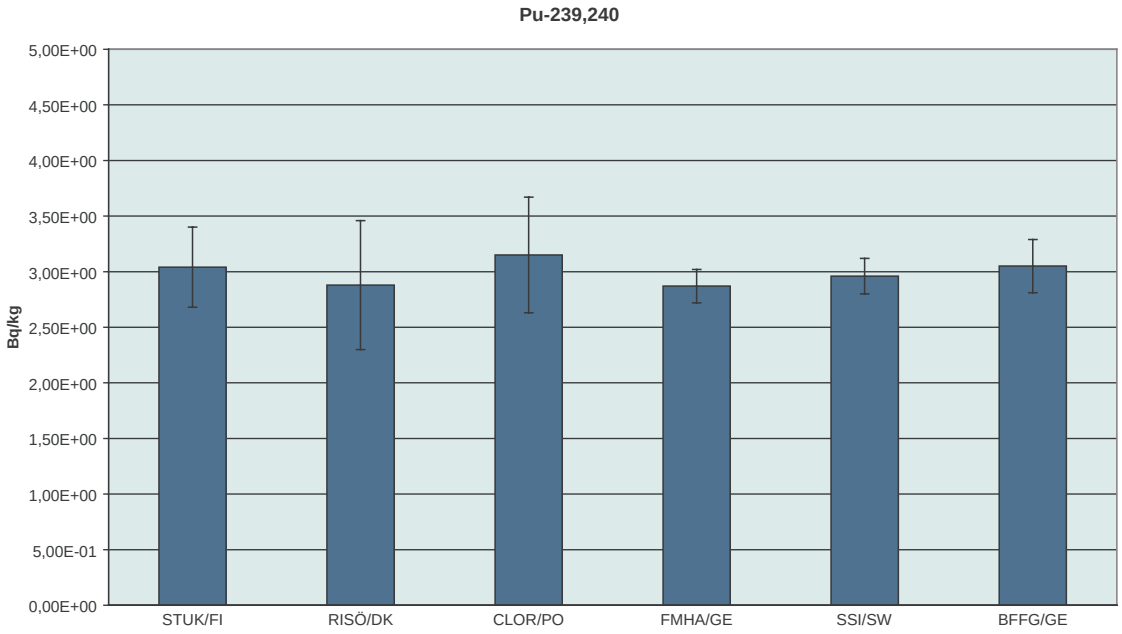


Figure A9.
Results for Pu-239,240 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



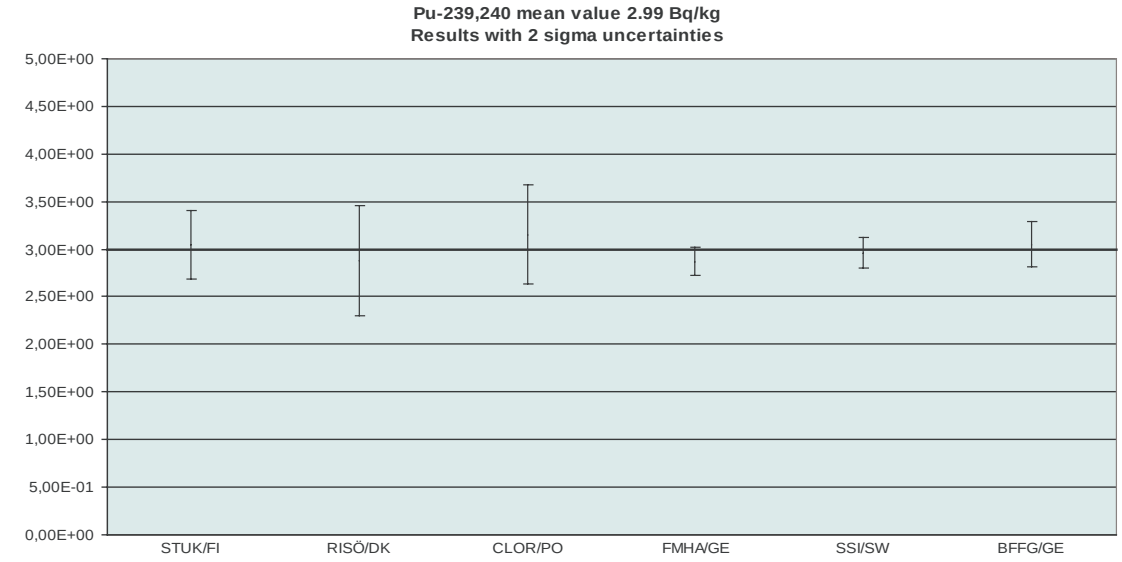
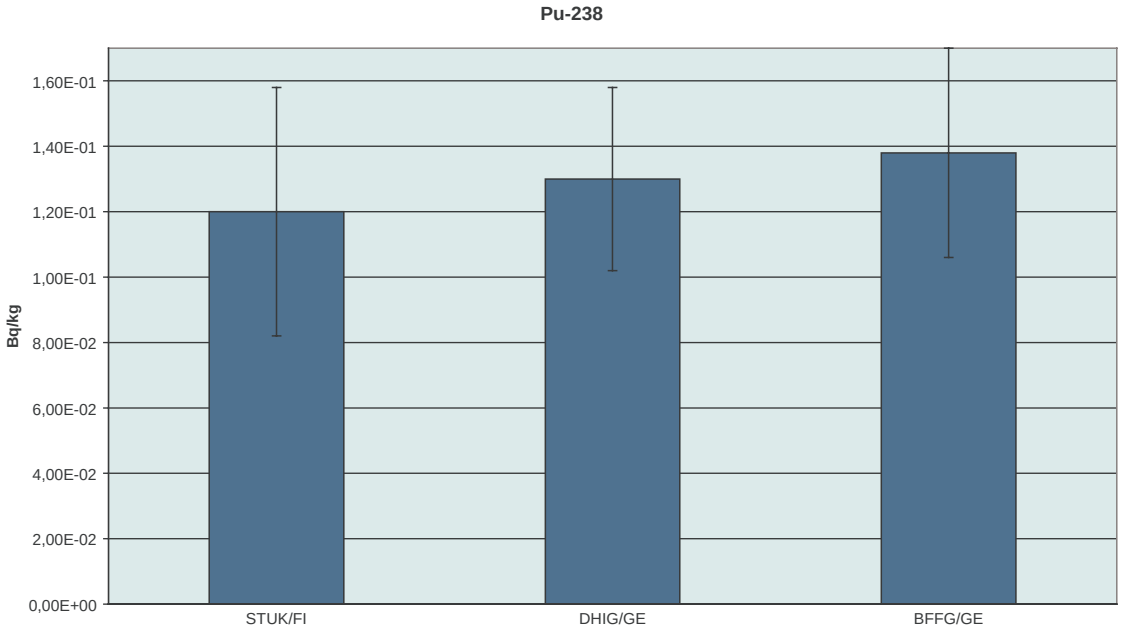


Figure A10.
Results for Pu-238 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).



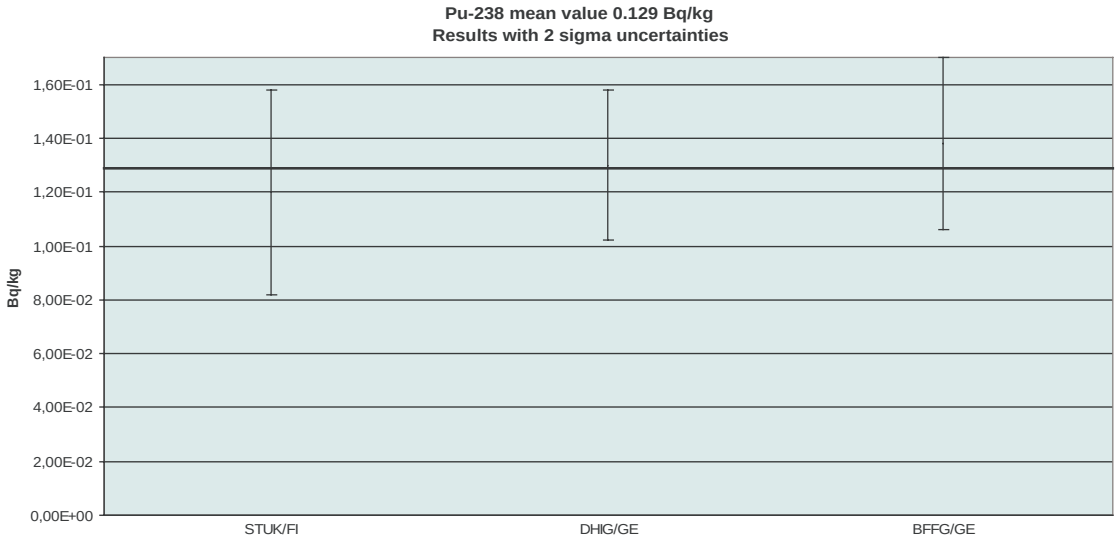
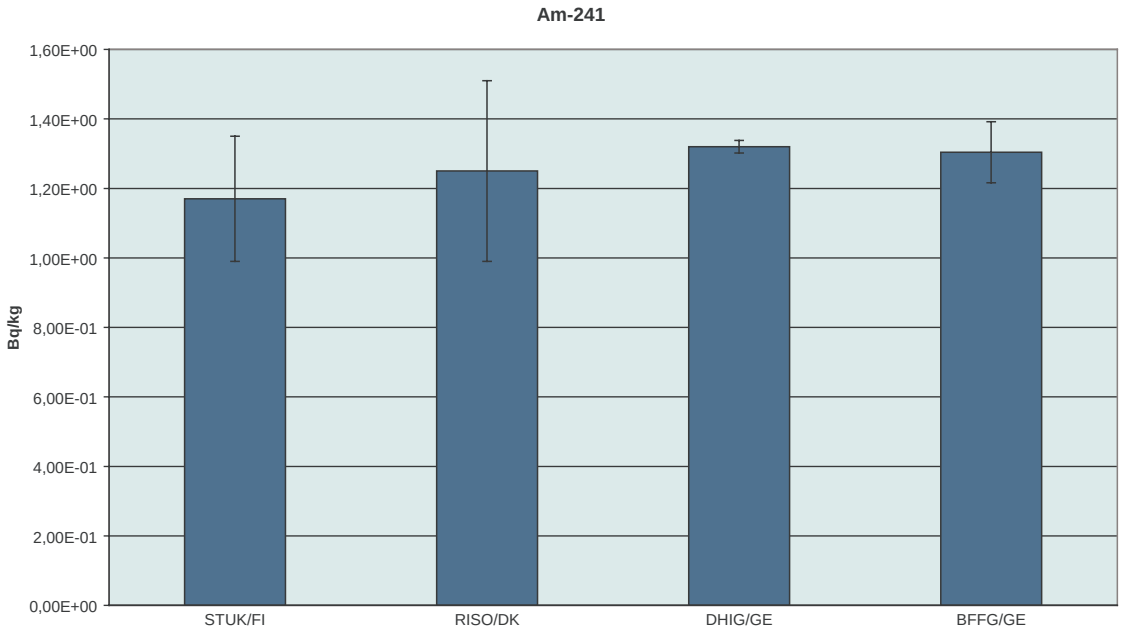
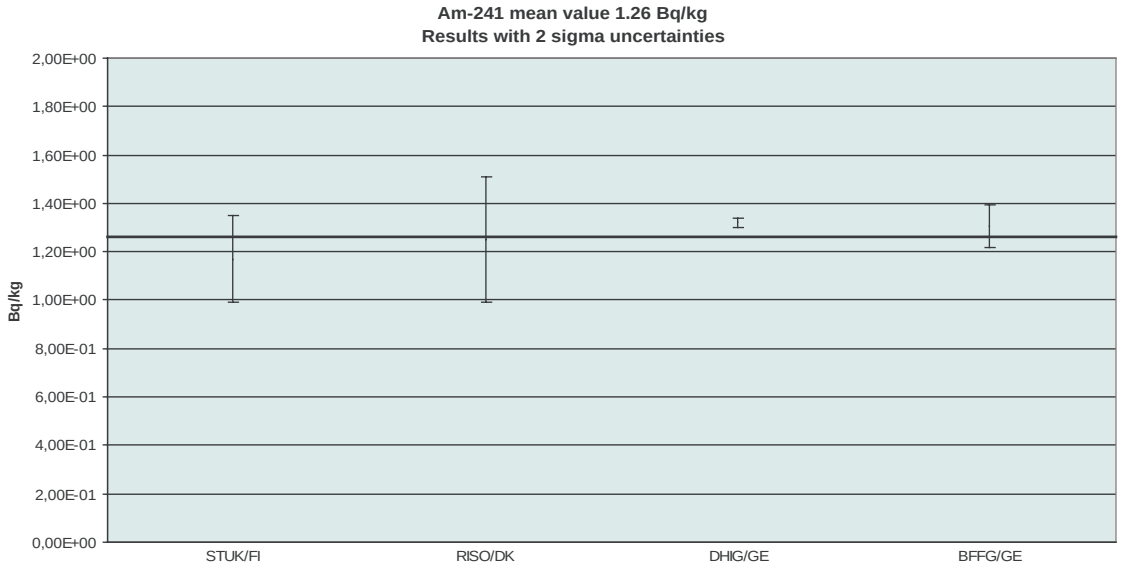


Figure A11.
Results for Am-241 with 2 sigma uncertainties (upper) and the arithmetic mean with variations of the result within 2 sigma uncertainties (lower).





Evaluation of the results

The evaluation procedure has been conducted for nuclides with four or more results (K-40, Cs-134, Cs-137, Pb-210, Ra-226, Pu-239,240, and Am-241).

The Z-score for an item indicates how far and in what direction the item deviates from the mean of its distribution, expressed in units of its distribution's standard deviation. The Z-score value for an item is calculated according to:

$$z_i = \frac{x_i - \bar{x}}{\sigma},$$

where

$\bar{x}$ = arithmetic mean of the values,

σ = standard deviation of the values.

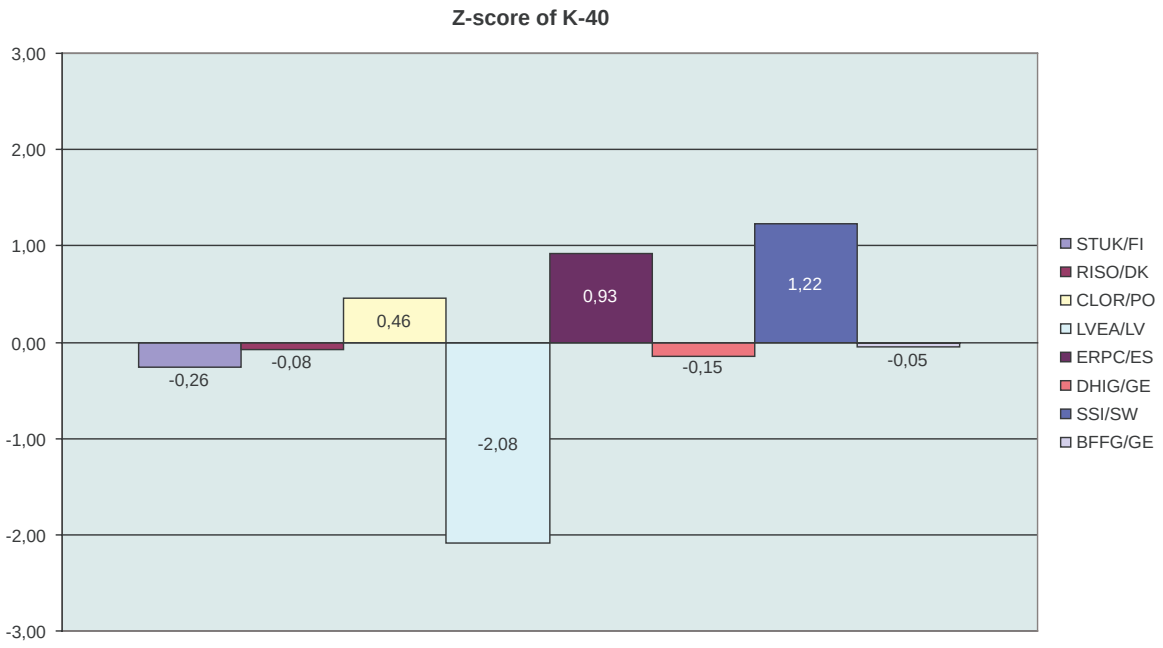


Figure A12. Column diagram of Z-scores for K-40.

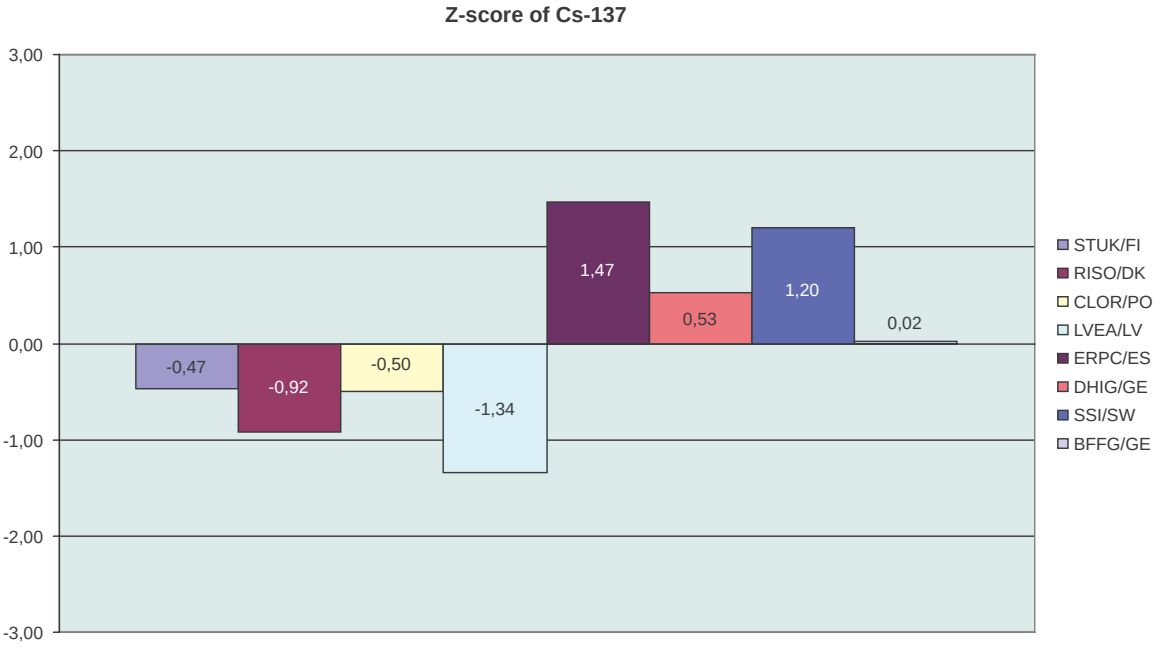


Figure A13. Column diagram of Z-scores for Cs-137.

If the values are Normally distributed, 68% of the Z-scores fall between −1 and 1, 95% between −2 and 2, and 99% between −3 and 3.

The results of the tests are presented for each radionuclide in Figures A12–A18.

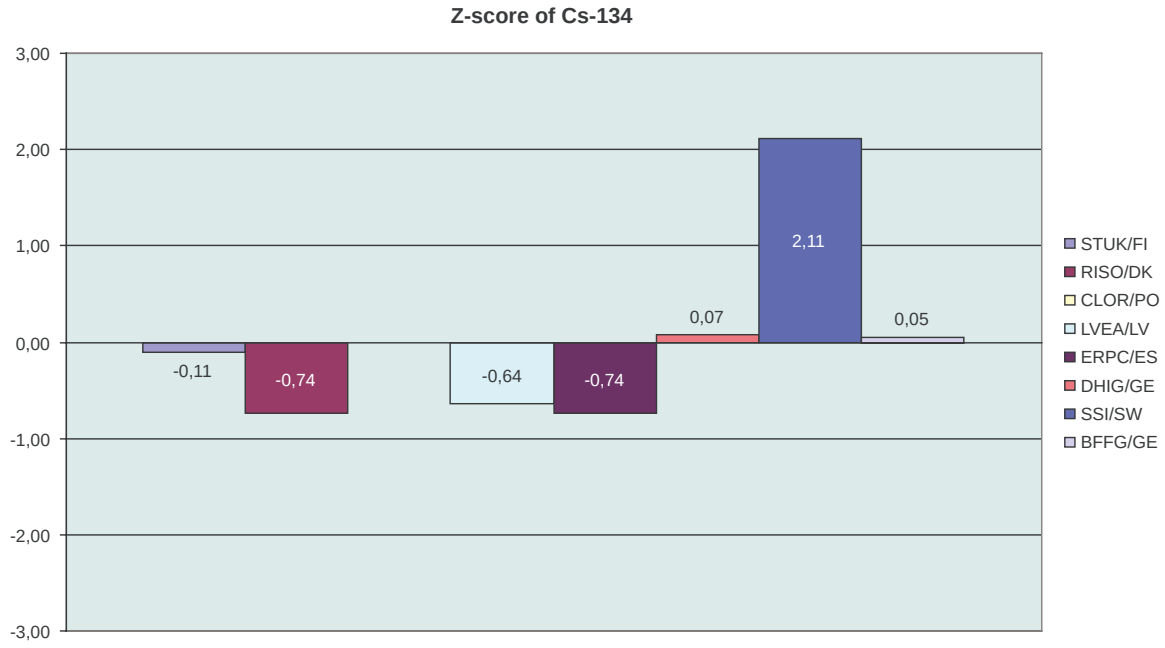


Figure A14. Column diagram of Z-scores for Cs-134.

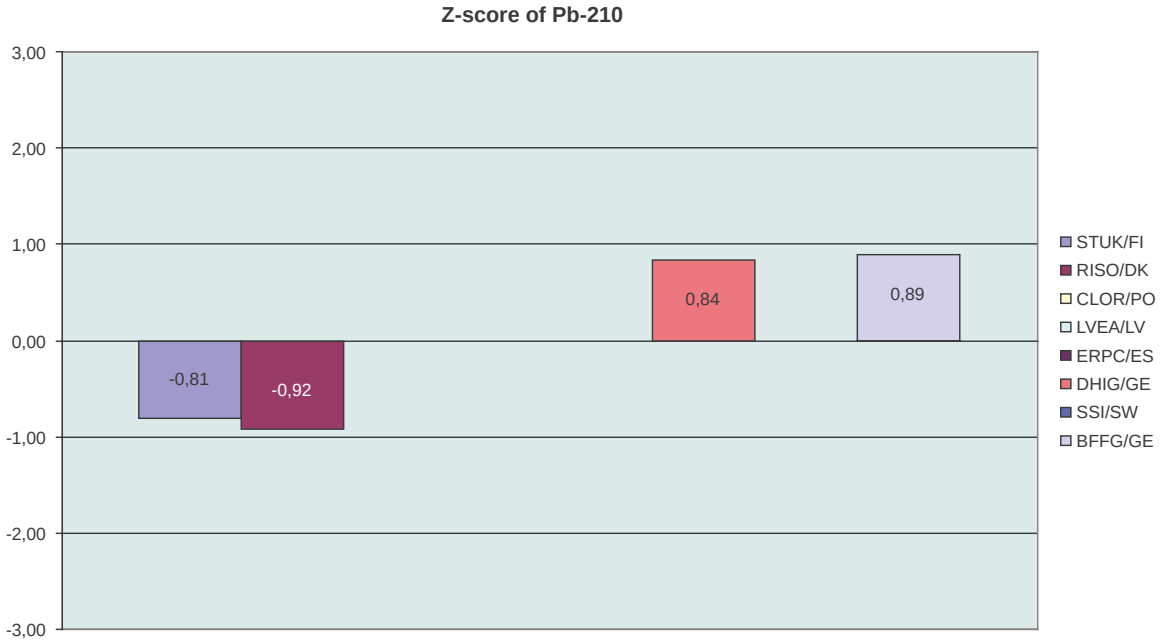


Figure A15. Column diagram of Z-scores for Pb-210.

Figure A16.
Column diagram of Z-scores for Ra-226.

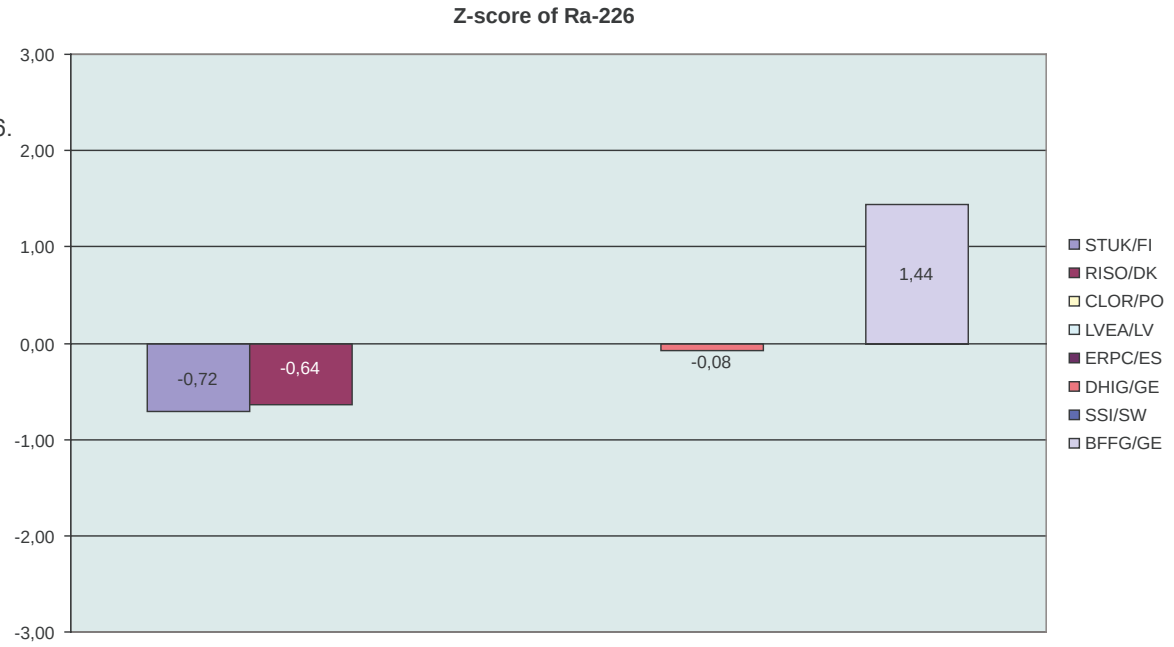
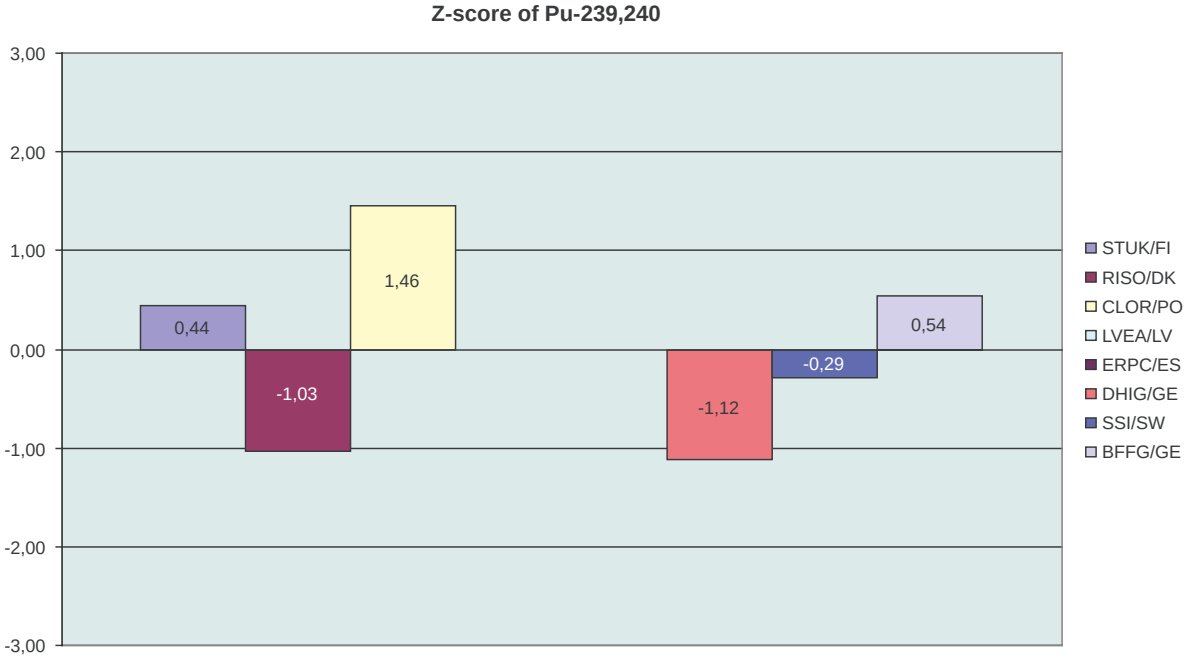


Figure A17.
Column diagram of Z-scores for Pu-239,240.



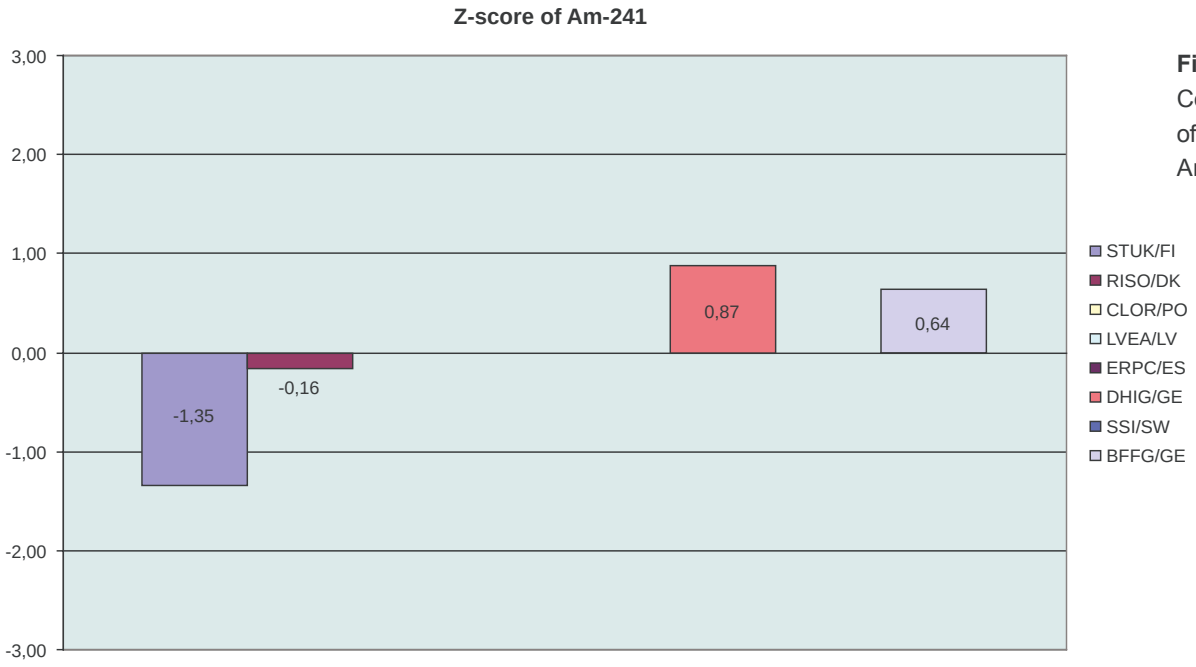


Figure A18. Column diagram of Z-scores for Am-241.

Conclusions

Eight laboratories from seven Baltic Sea countries participated in an intercomparison exercise carried out during the years 2000–2003 concerning laboratory analyses of artificial and natural radionuclides in a sediment sample. All the participating laboratories analysed K-40 and Cs-137 in the sample. In addition, results for several other gamma-emitting artificial and natural nuclides, as well as alpha-emitting transuranic elements were analysed.

The intercomparison showed that the analytical capability in the Baltic Sea countries is on a high level; the laboratories are competent to analyse a large number of important radionuclides (e.g., Cs-137, Cs-134, Pb-210) in the marine environment, if needed. In addition, there exists a capability for difficult radiochemical analyses of alpha-emitting nuclides (Pu and Am isotopes). Sr-90 analyses were performed only by one laboratory, which was mainly due to the lack of the sample material. However, the capability for Sr-90 measurements has been demonstrated in other intercomparisons (e.g., seawater).

The results indicate that measurements performed by these laboratories are in quite good agreement with each other. When comparing the Z-scores (Figures A12–A18), the difference from the mean value can be considered to be statistically significant ($|z|>1.96$) in only two cases.

The uncertainties reported with the results varied a great deal from each other. Harmonization of the uncertainty calculations in the laboratories could be a common target in the HELCOM MORS-PRO Group.

Acknowledgement

The first part of this work was organized by Sandor Mulsow, IAEA-MEL.

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