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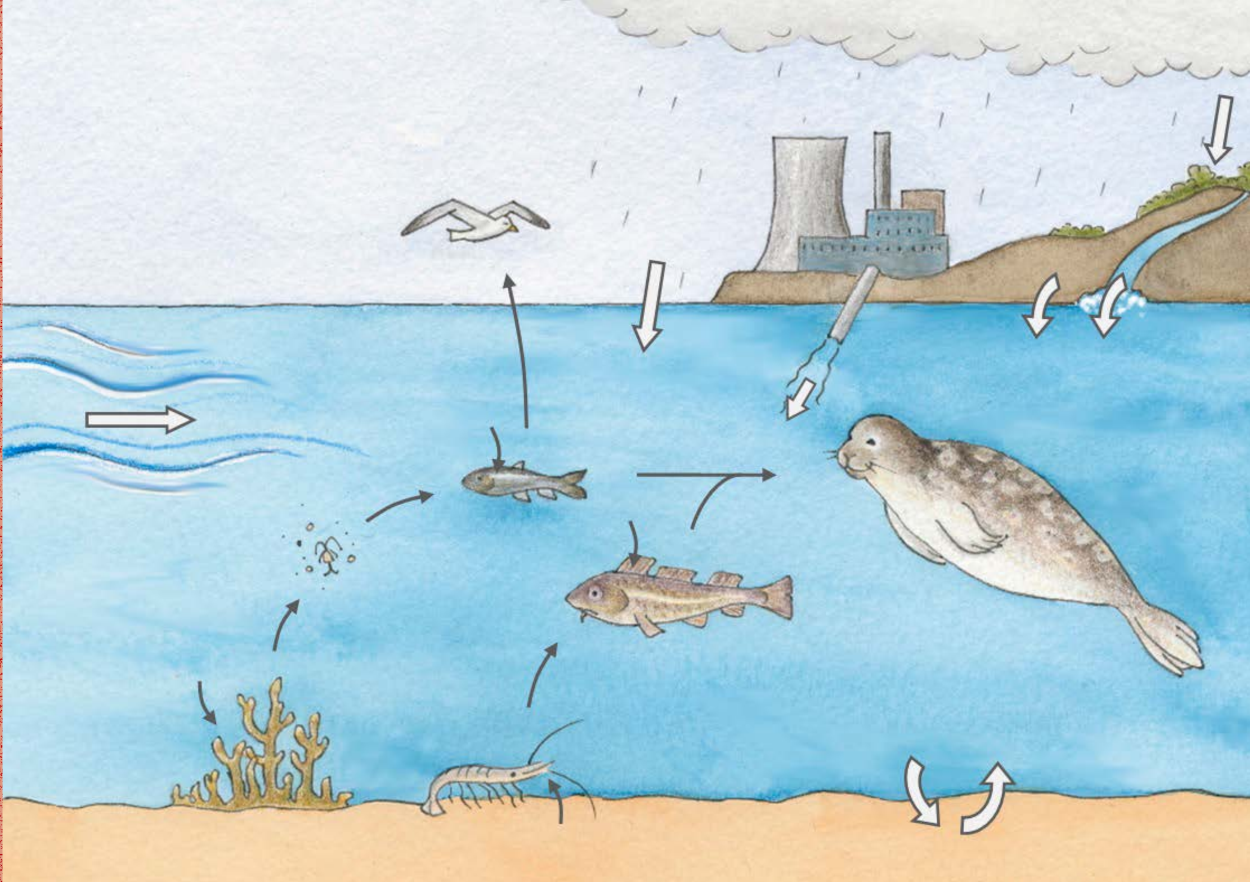
StrålevernRapport 2015:3



Radioactivity in the Marine Environment 2011

Results from the Norwegian Marine Monitoring Programme (RAME)

Reference:
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[1]Norwegian Radiation Protection Authority, [2]Institute of Marine Research, [3]Institute of Energy Technology
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Key words:
Radioactivity, marine environment, RAME, monitoring, Norway.

Abstract:
This report presents results of monitoring of radioactivity in sea water, sediment and biota collected in Norwegian waters in 2011.

Referanse:
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Rapporten inneholder resultater fra overvåkningen av radioaktivitet i sjøvann, sedimenter og biota i norske havområder i 2011.

Head of project: Hilde Skjerdal
Approved:

Unn H. Refseth

Unn Hilde Refseth, director, Department of Monitoring and Research

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Norwegian Radiation Protection Authority, P.O. Box 55, N-1332 Østerås, Norway.
Telephone +47 67 16 25 00, fax + 47 67 14 74 07.
E-mail: nrpa@nrpa.no
www.nrpa.no
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Radioactivity in the Marine Environment 2011

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Protection Authority
Østerås, 2015

Havforskningsinstituttet

Contents

1 Introduction

The issue of present and potential radioactive contamination in the marine environment has received considerable attention in Norway. In the late 1980s several accidents and incidents involving nuclear-powered submarines demonstrated that the risk of the release of radionuclides into the Barents Sea should be considered more carefully. In particular, it became evident that better documentation concerning the radioactivity levels in fish and other seafood was important for the seafood export industries. Furthermore, in the early 1990s, information concerning the dumping of nuclear waste emerged through bilateral environmental cooperation between Norway and Russia. In the years that followed, concern grew regarding the safety of military and civil nuclear installations in the northwest of Russia. This concern was associated not only with possible reactor accidents, but also with the prolonged or sudden release of radionuclides from radioactive waste facilities.

In addition to the potential threats outlined above, radionuclides originating from nuclear weapons fallout, the Chernobyl accident, and waste discharged from European reprocessing facilities have been detected in the Norwegian marine environment. In 1994 and 1995, the discharge of ^{99}Tc from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced, it continued at a high level up to 2003. There has been much public concern about the consequences of such kinds of releases, as radionuclides discharged to the Irish Sea are transported by ocean currents via the North Sea to the Norwegian coastal current and further northwards. In response to this concern, programmes for the monitoring of radioactivity in the marine environment have been established. Due to the economic importance of the fishing industry and its vulnerability to contamination, as well as any rumours of radioactive contamination, one of the main objectives of these programmes is to document levels and trends of radionuclides in the Norwegian marine environment.

Other industrial activities, such as mining and oil production, may change the distribution of naturally occurring radionuclides in the environment. The discharge of radium from water produced by oil installations is one area that has received special attention.

Recently, there have been an increased number of transports involving spent nuclear fuel in Norwegian marine waters. Spent nuclear fuel is shipped to Murmansk for further transport and processing at Mayak. Accidents during these transports may lead to releases of radioactivity in the marine environment.

During 2011, samples for monitoring radioactivity in the marine environment were collected mainly in the Skagerrak, the Norwegian Sea and the Jan Mayen area, and at permanent coastal stations along the Norwegian coastline. Results from the analysis of these samples are presented in Chapters 4 and 5. In Chapter 6, a summary of the findings and the conclusions is given. In the Appendix, technical information regarding sample preparation techniques and analytical methods employed in the laboratories are presented.

2 Sources of radionuclides in the marine environment

2.1 Long-range transport of radionuclides

Due to the prevailing ocean currents, a large part of discharged, dumped or deposited radioactive wastes in northern European marine waters will reach the Norwegian coastal current and be further transported into the Barents Sea region. Important sources that have affected the levels of anthropogenic radioactivity in the Norwegian marine environment during the last decades are global fallout from nuclear weapons testing in the 1950s and 1960s, authorised liquid discharges from the reprocessing plants at Sellafield and Cap de la Hague, and the Chernobyl accident in 1986.

Even though the Chernobyl accident occurred more than 25 years ago, the Baltic Sea still acts as a secondary source of ^{137}Cs due to outflowing brackish water from the Baltic Sea into the Skagerrak. In 2000, the annual net outflow from the Baltic was estimated at 39 TBq. This value shows an exponential decrease 1991-2000, with an observed half-life of 13.4 years. The "stable" caesium net outflow may be described by an exponential decrease with a half-life of 24 years. (Dahlgaard, 2002).

The discharges from Sellafield have decreased in recent years, but contaminated sediments in the Irish Sea from the large discharges in the 1970s and 1980s still continues to remobilise ^{137}Cs and plutonium isotopes, and has been estimated to do so for a long time (Cook *et al.*, 1997 and Leonard *et al.*, 1999). Discharge figures for Sellafield and Cap de la Hague are published annually by OSPAR.

From a global point of view, atmospheric nuclear weapons tests, conducted between 1945 and 1980, are the largest source of radioactive contamination in the environment. The most intensive test period was between 1952 and 1962, during which the USSR and USA were responsible for most of the tests. The last atmospheric nuclear detonation was conducted in China in 1980. In total, it has been estimated that 948 PBq ^{137}Cs, 622 PBq ^{90}Sr, 6.52 PBq ^{239}Pu and 4.35 PBq ^{240}Pu have been released to the environment (UNSCEAR, 2000a), mainly in the northern hemisphere, where most of the tests took place.

2.2 Norwegian sources

2.2.1 *The Institute for Energy Technology*

The Institute for Energy Technology (IFE) at Kjeller is located about 20 km east of Oslo. The facilities include a heavy-water-cooled and moderated research reactor with a thermal effect of 2 MW, called JEEP II, a metallurgical laboratory, production facilities for medical radioactive isotopes, and a radioactive waste treatment plant for low-level and intermediate-level waste (LLW and ILW). The liquid LLW originates from these facilities. Liquid effluent is discharged through a designated pipeline to the Nitelva River about 100 km from the sea. The river empties into Lake Øyeren, where the water is mixed with the water from the Glomma River. The Glomma River empties into the Oslofjord at the town of Fredrikstad.

The Halden Boiling Water Reactor (HBWR) is located in the town of Halden, in the southeast of Norway, close to the Swedish border. The HBWR is heavy-water-cooled and moderated with a thermal effect of 20 MW. The reactor was commissioned in 1959. IFE Halden is authorised to discharge a limited amount of radioactivity from the research reactor to the Tista River, which empties into the Iddefjord.

Discharge limits for IFE Kjeller and IFE Halden are based on a dose limit of 1 µSv per year to a hypothetical critical group. Discharges from IFE Kjeller and IFE Halden are reported annually to the authorities. In 2011, the annual dose from liquid discharges from IFE Kjeller and IFE Halden were estimated to 0.0123 µSv and 0.017 µSv, respectively.

2.2.2 *Unsealed radioactive substances in the medical sector*

Unsealed radioactive substances used in medicine dominate the anthropogenic radioactive discharges to the sewage system. Radioactive solutions are given to patients for diagnostic and therapeutic purposes, and enter the sewage systems mainly by the excretion of urine and faeces. The two most commonly used radionuclides in nuclear medicine for diagnostic and therapeutic purposes are ^{99m}Tc, which decays to ^{99}Tc with a half-life of 6 h, and ^{131}I ($T_{1/2}$ = 8.04 d). The most important radionuclide concerning dose to the public after discharge is ^{131}I. The discharge of ^{131}I has been estimated according instructions published by OSPAR. In 2011, an estimated total of 1113 GBq ^{131}I was discharged to the sewage system from the medical sector in Norway.

2.2.3 *Discharge of produced water containing ^{226}Ra and ^{228}Ra*

Large volumes of produced water, containing dissolved ^{226}Ra and ^{228}Ra, are discharged into the sea during oil exploitation. Produced water may be formation water trapped in the reservoirs for millions of years, or a mixture of formation water and sea water, if sea water has been injected in order to maintain pressure in the reservoirs during oil and gas production.

Although the levels of ^{226}Ra and ^{228}Ra are not technologically enhanced, the natural concentrations of these isotopes in the saline formation water are generally much higher (about 3 orders of magnitude) than the background concentration in sea water (a few Bq m^{-3}). The concentration of radium in the produced water may change over the lifetime of the well. Injection of large volumes of sea water to maintain the pressure within the well, may lead to dilution of the radium in the produced water, but at the cost of greater water-to-oil ratios later in the production process. Reported discharges of ^{226}Ra and ^{228}Ra from the Norwegian oil and gas industry in 2011 were 468 GBq and 371 GBq, respectively. In addition, 265 GBq ^{226}Ra and 270 GBq ^{228}Ra were re-injected into reservoirs.

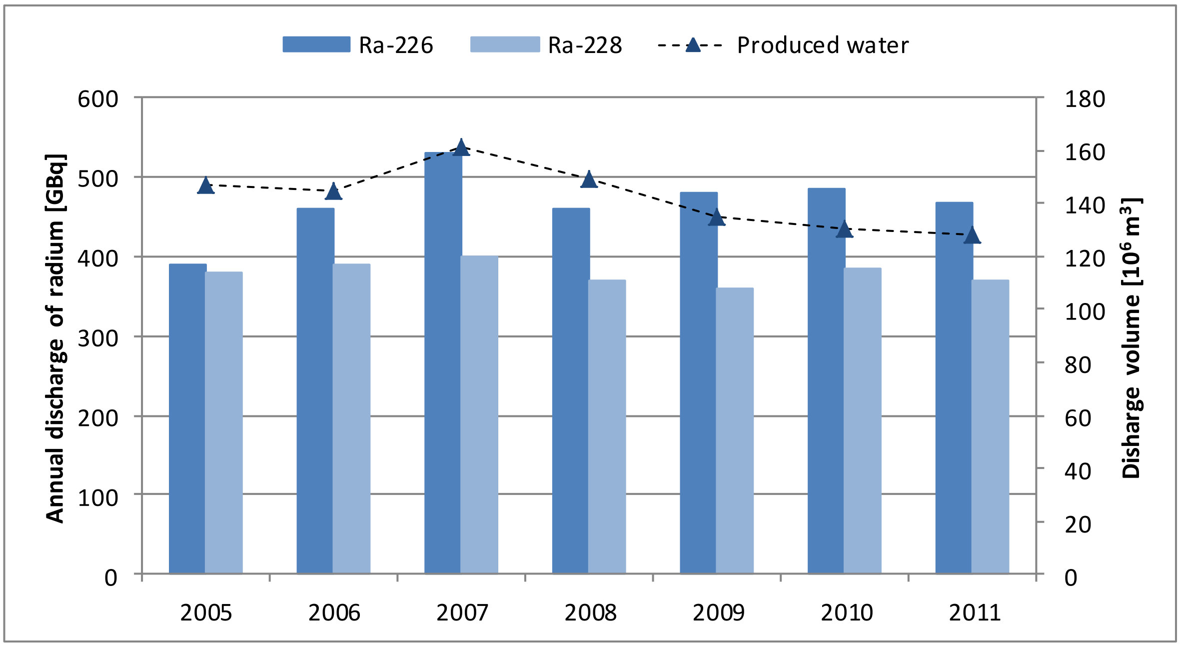

Figure 2.1: *Annual discharge of ^{226}Ra and ^{228}Ra to the marine environment via produced water, and discharged volume of produced water from the Norwegian oil and gas industry in the period 2005 to 2011.*

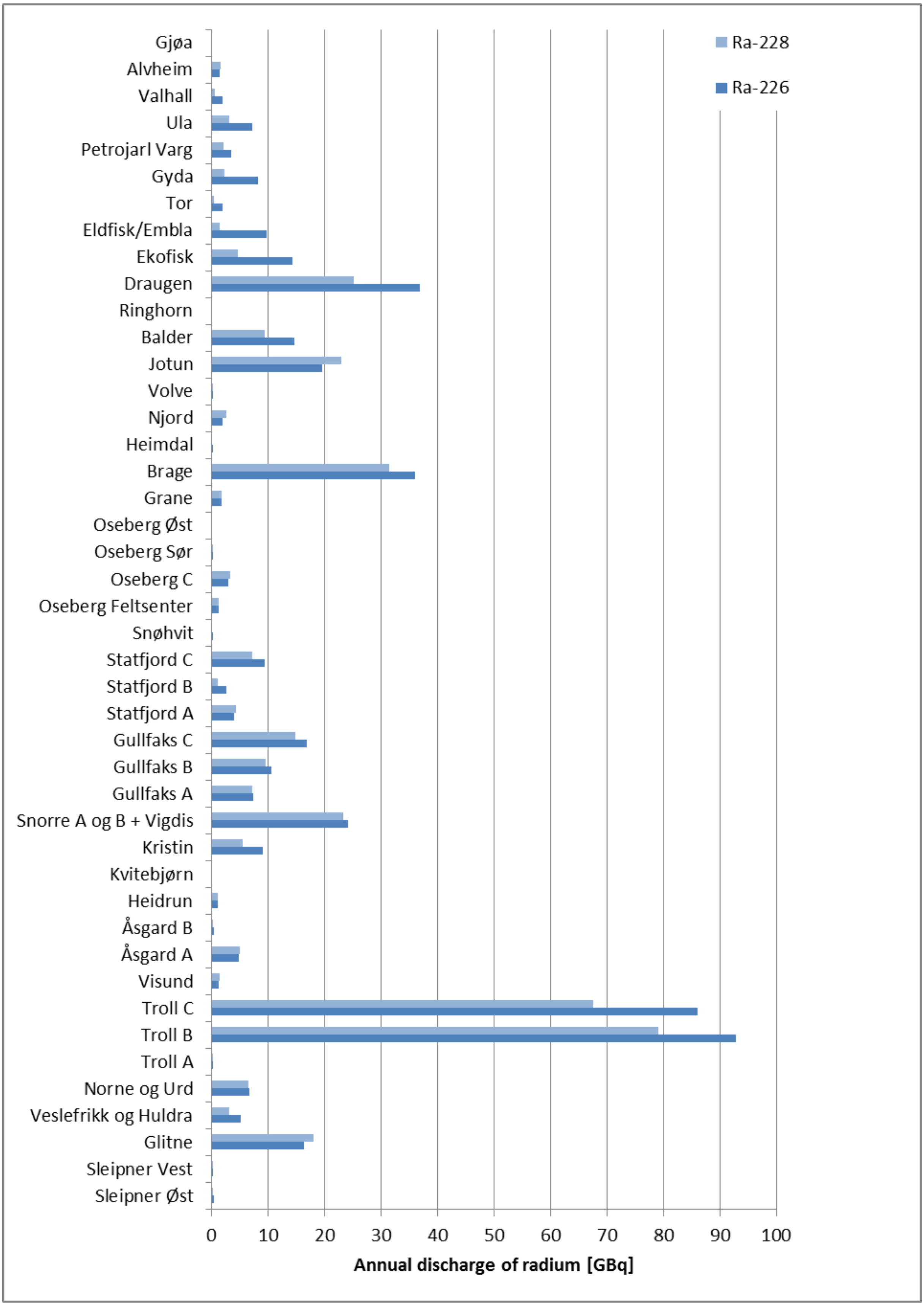

Figure 2.2: *Discharged activity of* ^{226}*Ra and* ^{228}*Ra from Norwegian oil and gas fields in 2011.*

2.3 Other present and potential sources

In addition to discharges from the western European reprocessing plants, northern marine waters have also received an input of various radionuclides from Russian reprocessing plants situated on the tributaries of the Russian rivers Ob and Yenisey. These are the Mayak Production Association in the southern Urals, the Siberian Chemical Combine (Tomsk-7) north of the town of Tomsk, and the Krasnoyarsk Mining and Chemical Combine (KMCC). Uncertainty exists regarding the magnitude of radionuclide contributions from these sources, although estimates have been made. For example, between 1958 and 1993, routine discharges from the KMCC are estimated to have led to an input of 30-100 TBq of ^{137}Cs into the Kara Sea, a level which is of a similar order of magnitude to inputs from Sellafield-derived ^{137}Cs to the area (Vakulovsky *et al*., 1995). However, the fact that the prevailing surface currents tend to transport contamination away from the Kara Sea in predominantly easterly and northerly directions, suggests that the resultant inputs to the Barents Sea have been relatively low. Nonetheless, there is a potential for significant radionuclide contamination following large accidental releases of radioactivity from these nuclear complexes.

Several other sources exist which also represent potential sources of radionuclides in the Barents and Norwegian Seas. Among these are radioactive waste containers dumped in the Barents and Kara Seas by the Former Soviet Union and the sunken submarines: the Komsomolets in the Norwegian Sea and the K-159 in the Barents Sea.

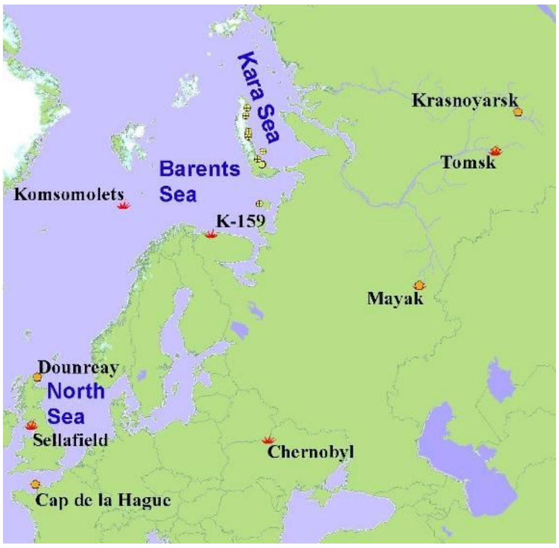

Figure 2.3. *Sources of radionuclides in the northern marine environment.*

Following an accident on April 7, 1989, the Russian nuclear submarine Komsomolets, lies at a depth of 1700 metres, 180 km southwest of Bear Island in the Norwegian Sea. The submarine contains a nuclear reactor and two torpedoes with nuclear warheads. Samples of water and sediments collected around the submarine by the Institute of Marine Research are analysed with respect to ^{137}Cs, and are presented in Figure 2.4.

Furthermore, the Russian submarine K-159 sank off the coast of the Kola Peninsula on 30 August, 2003, while under tow for scrapping. The submarine contains spent nuclear fuel, but no nuclear warheads.

3 Collection of samples

In 2011, samples were mainly collected in the Skagerrak, the Norwegian Sea, in selected fjords, and at permanent coastal stations. A geographic overview of the sampling area covered by the marine monitoring programme, with the fixed coastal stations marked, is shown in Figure 3.1.

Figure 3.1. *Geographic overview of the sampling stations and the sampling area covered by the marine monitoring programme.*

Sampling and analysis were carried out by the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR), and the Institute for Energy Technology (IFE). Samples from the Norwegian Sea were collected in October 2011 by IMR and NRPA from the vessel R/V "G. O. Sars" (Figure 3.2). During the expeditions, samples of surface water were collected, which were later analysed for ^{99}Tc, ^{137}Cs, ^{90}Sr, ^{241}Am and plutonium isotopes. Sediment was also sampled and later analysed for ^{137}Cs. Samples from Jan Mayen were collected by IMR, and also analysed for naturally occurring radionuclides.

Figure 3.2. *R/V G.O. Sars (Photo:*Hege Iren Svensen /imr.no)

3.1 Seawater

For ^{99}Tc, 100 litres of unfiltered seawater were collected. The samples were passed through anion exchange columns (Amberlite IRA-400) on board, according to the method described by Harvey *et al*. (1992).

For ^{90}Sr, 50-100 litres of unfiltered water were collected at each sampling station and later transported to IFE for analysis.

For the analysis of ^{137}Cs from the Norwegian Sea, $Cu_2[Fe(CN)_6]$-impregnated cotton filters are used as sorbents (Roos *et al.,* 1994). The system consists of a prefilter (1 micron) and two $Cu_2[Fe(CN)_6]$-impregnated cotton filters connected in series (Figure 3.3). Assuming identical collection efficiency for the $Cu_2[Fe(CN)_6]$-impregnated cotton filters, the amount of caesium passing through the prefilter can be calculated by considering the two filters as the first two terms in a geometrical series, which has the sum:

$$A(^{137}Cs) = A_1/(1-A_2/A_1)$$

where A_1 and A_2 are the ^{137}Cs activities on the first and second impregnated filters, respectively.

Figure 3.3. *Filter system used for analysis of* ^{137}*Cs in sea water (Photo: NRPA).*

Samples of 200 liters (unfiltered) were collected for the determination of the activity concentrations of $^{239+240}$Pu and ^{241}Am. To collect deep water samples, a CTD/Rosette multi-bottle sampler was used (Figure 3.4). Plutonium-242 and ^{243}Am were used as chemical yield determinants. The precipitation of plutonium and americium from the water was performed *in situ* according to the analytical procedure described by Chen *et al*. (1991).

Figure 3.4. *Sea water sampling with a CTD/Rosette multi-bottle sampler (Photo: Hilde Skjerdal, NRPA).*

3.2 Sediment

Sediment samples were retrieved from the seabed using a Smøgen box corer. Profiles were collected from the box by slowly inserting PVC tubes into the sediment sample. Surface sediment samples were obtained from the upper 2 cm layer of the sediment remaining in the box corer.

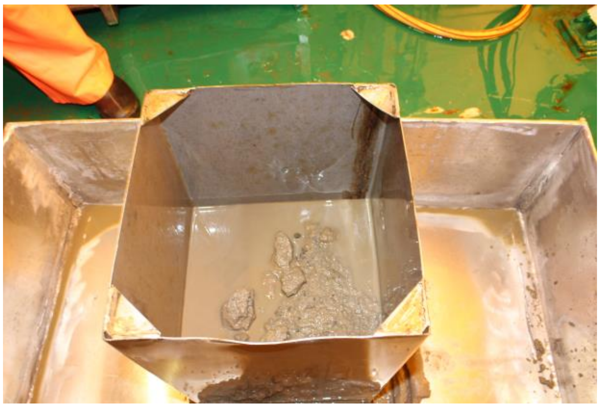

Figure 3.5. *Sediment sampling with a Smøgen box corer (Photo: Hilde Skjerdal, NRPA)*

Figure 3.6. *Sediment sampling (Photo: Hilde Skjerdal, NRPA)*

3.3 Seaweed

NRPA collected seaweed (*Fucus vesiculosus*) samples from three stations along the Norwegian coastline. At Hillesøy in northern Norway, seaweed is collected every month and at the other locations once a year. In addition, IFE performs monthly or annual seaweed sampling at ten locations along the coastline. IMR collected seaweed at Værlandet.

3.4 Fish and crustaceans

IMR collected samples of different fish species from Norwegian waters by combining muscle samples of 50-100 grams from about 25 individuals. 100 samples of cod were also collected from four locations in the Barents Sea in the spring and autumn by the Norwegian Reference Fleet. At IMR, 5-50 grams of muscle from each individual cod was thereafter combined to a pooled sample. Eight lobsters were collected as part of cooperation between IMR and Bulandet School at Værlandet (Sogn og Fjordane). The samples were thereafter prepared at IMR.

4 Radioactivity in sea water and sediments

4.1 Technetium-99 in sea water

Technetium-99 has a physical half-life of $2.13 \cdot 10^5$ years, and originates from the decay of ^{99}Mo, which is formed either as a fission product from ^{235}U or ^{239}Pu or by neutron activation of ^{98}Mo. The main source of ^{99}Tc in Norwegian waters is liquid discharge from the reprocessing plant at Sellafield. In oxygenated sea water, ^{99}Tc is present as the highly soluble pertechnetate ion (TcO_4^-). Due to its conservative behaviour in sea water, TcO_4^- has the ability to be transported by ocean currents without being significantly affected by sedimentation processes. From the Irish Sea, ^{99}Tc is transported by ocean currents to the North Sea and via the Norwegian Coastal Current up to the Barents Sea. The transit time (the time between a specific discharge and the maximum activity concentration from that discharge reaching the sampling location) for ^{99}Tc to reach this area (Hillesøy) from the Irish Sea has been estimated to 3-4 years (Brown *et al.*, 2002; Dahlgaard, 1995).

In 2011, sea water samples were collected in the Norwegian Sea and analysed for ^{99}Tc. The results are presented in Figure 4.1. The activity concentrations ranged from 0.05 Bq m^{-3} to 0.31 Bq m^{-3}.

The activity concentrations found in 2011 are generally lower than, or at the same level as, previous observations (NRPA, 2005; NRPA, 2007; NRPA 2009). The reason for the decreasing concentrations is the reduced discharge of ^{99}Tc from Sellafield.

Monthly samples have also been collected at Hillesøy (Figure 4.2). The results show that the activity concentration of ^{99}Tc in sea water continues to slowly decrease since the peak concentrations in 1998/1999.

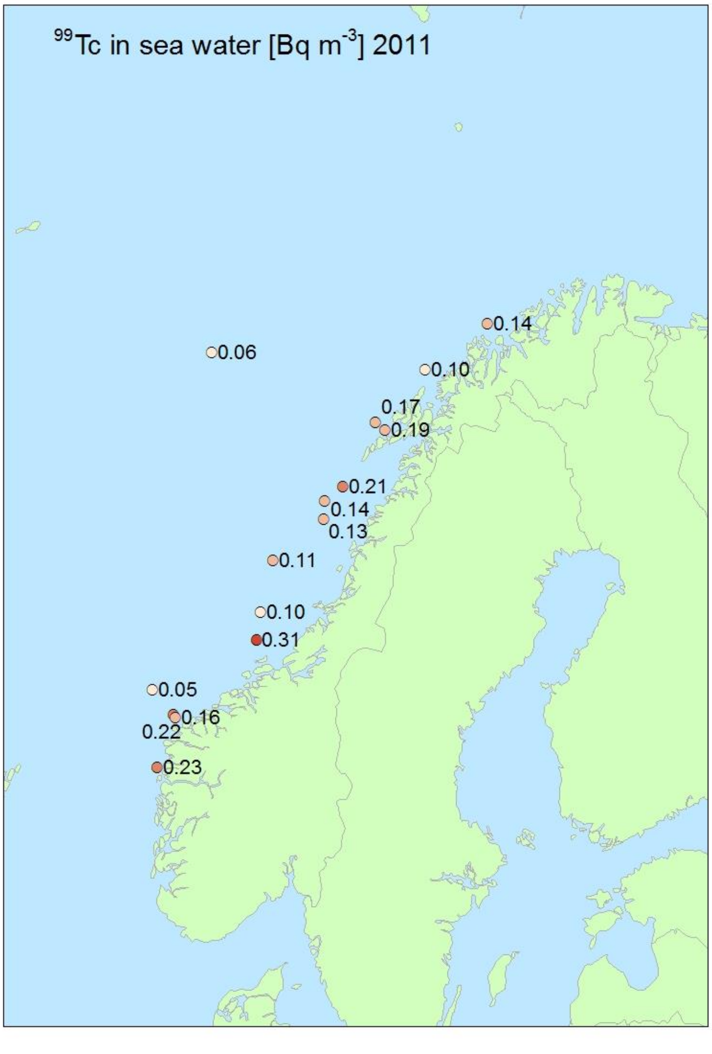

Figure 4.1. *Activity concentration (Bq m^{-3}) of ^{99}Tc in sea water samples collected in the Norwegian Sea in 2011.*

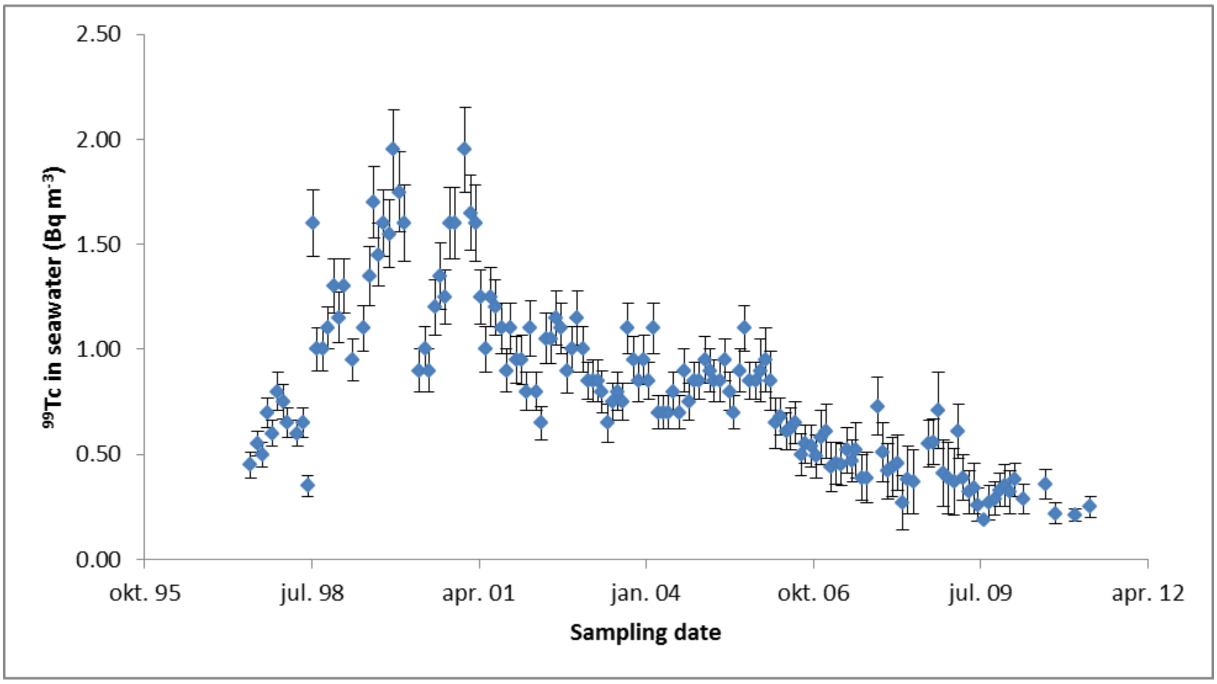

Figure 4.2. *Activity concentration (Bq m^{-3}) of ^{99}Tc in sea water at Hillesøy from monthly sampling.*

4.2 Strontium-90 in sea water

Strontium-90 is a fission product with a physical half-life of 29 years. Similar to ^{99}Tc, ^{90}Sr is a conservatively behaving element in the marine environment.

Sea water has been sampled mainly in the Norwegian Sea and later analysed for ^{90}Sr. The main sources of ^{90}Sr in these waters are discharge of liquid waste from reprocessing plants (mainly Sellafield), fallout from atmospheric nuclear weapons tests conducted mainly in the 1950s and 1960s, and outflow of water from the Baltic Sea. The results for 2011 are presented in Figure 4.3. In the Norwegian Sea, the activity concentration in surface water ranged from 0.6 Bq m^{-3} to 1.9 Bq m^{-3}.

Results from sampling at Hillesøy show that the activity concentration in sea water in the Norwegian coastal current is slowly decreasing (Figure 4.4). Explanations for this are the reduced discharges from Sellafield over the last 10 years and the physical decay of ^{90}Sr from global fallout.

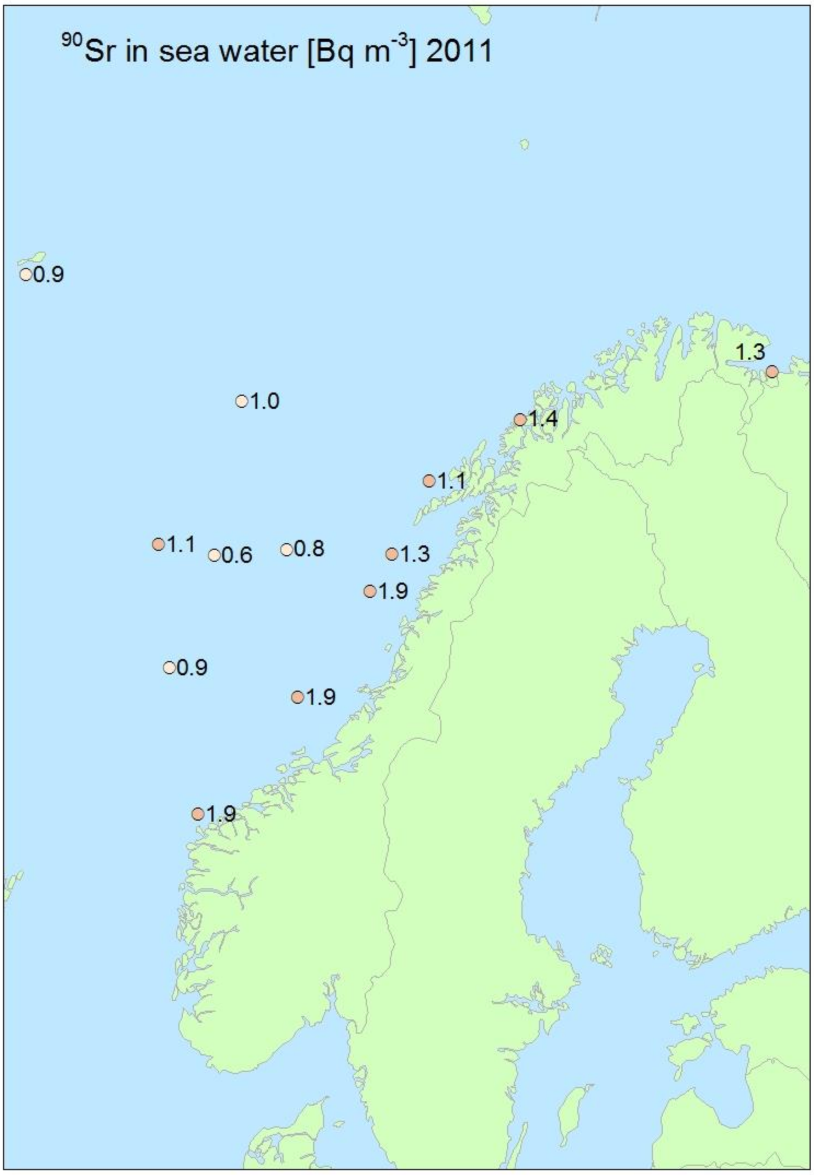

Figure 4.3. *Activity concentration (Bq m^{-3}) of ^{90}Sr in surface water samples collected in the Norwegian Sea in 2011.*

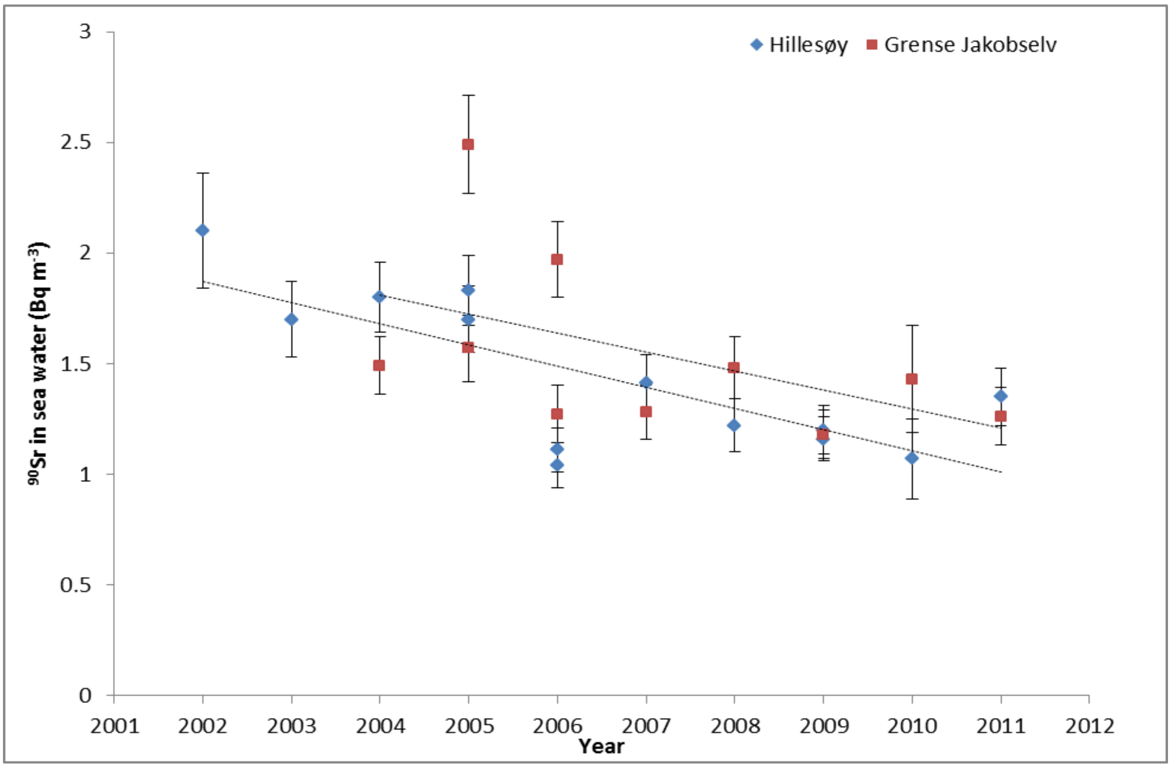

Figure 4.4. *Activity concentration (Bq m^{-3}) of ^{90}Sr in sea water at Hillesøy (2002-2011) and Grense Jakobselv (2004-2011).*

4.3 Caesium-137 in sea water and sediment

Caesium-137 is a fission product with a half-life of 30 years. The main sources of ^{137}Cs in Norwegian waters are fallout from atmospheric nuclear weapons tests in the 1950s and 1960s, outflowing water from the Baltic Sea, and ^{137}Cs remobilised from Irish Sea sediments. Runoff from land from the areas with the highest Chernobyl fallout, can also contribute locally in coastal water. Like ^{99}Tc and ^{90}Sr, ^{137}Cs is also a conservatively behaving radionuclide in sea water.

Observed levels of ^{137}Cs in the Norwegian Sea and the Skagerrak in 2011 are shown in Figure 4.5 and 4.6. The activity concentration in surface water sampled in the Norwegian Sea and Skagerrak ranged from 1.1 to 9.6 Bq m^{-3}, where the highest levels were found in the Skagerrak.

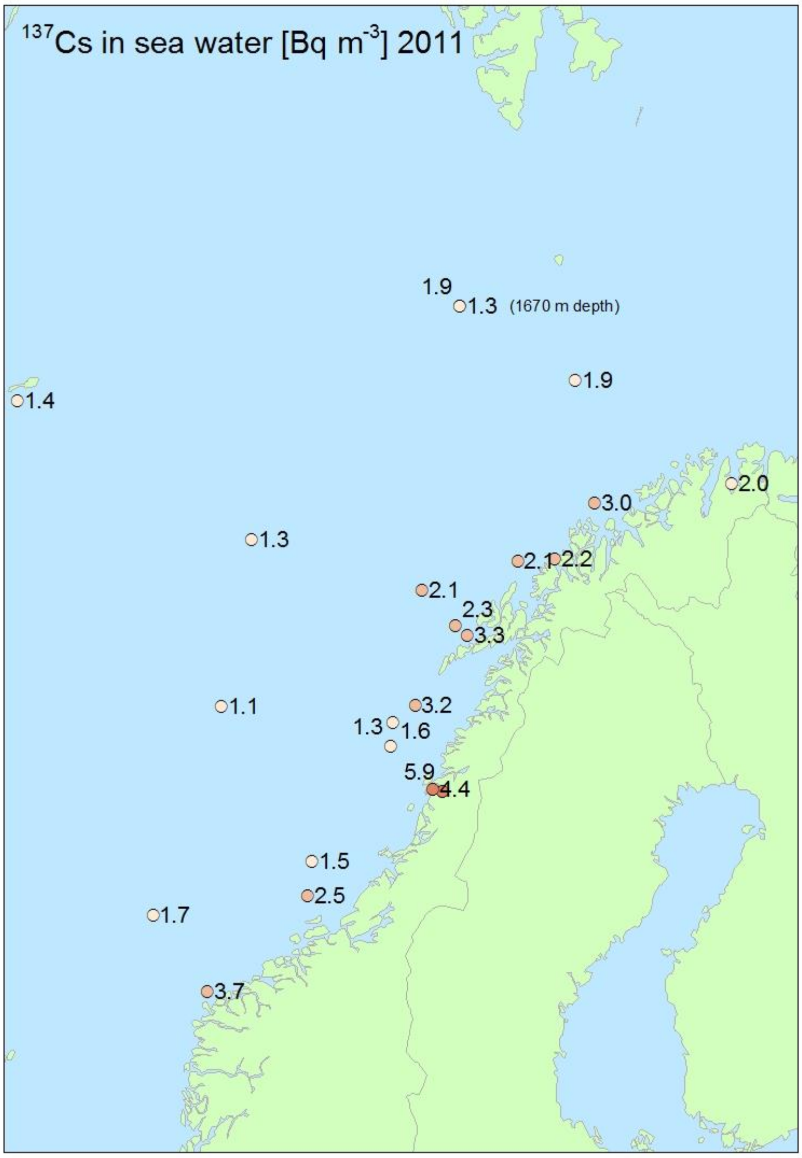

Figure 4.5. Activity concentration (Bq m^{-3}) of ^{137}Cs in sea water samples (surface water) collected in the Norwegian Sea in 2011.

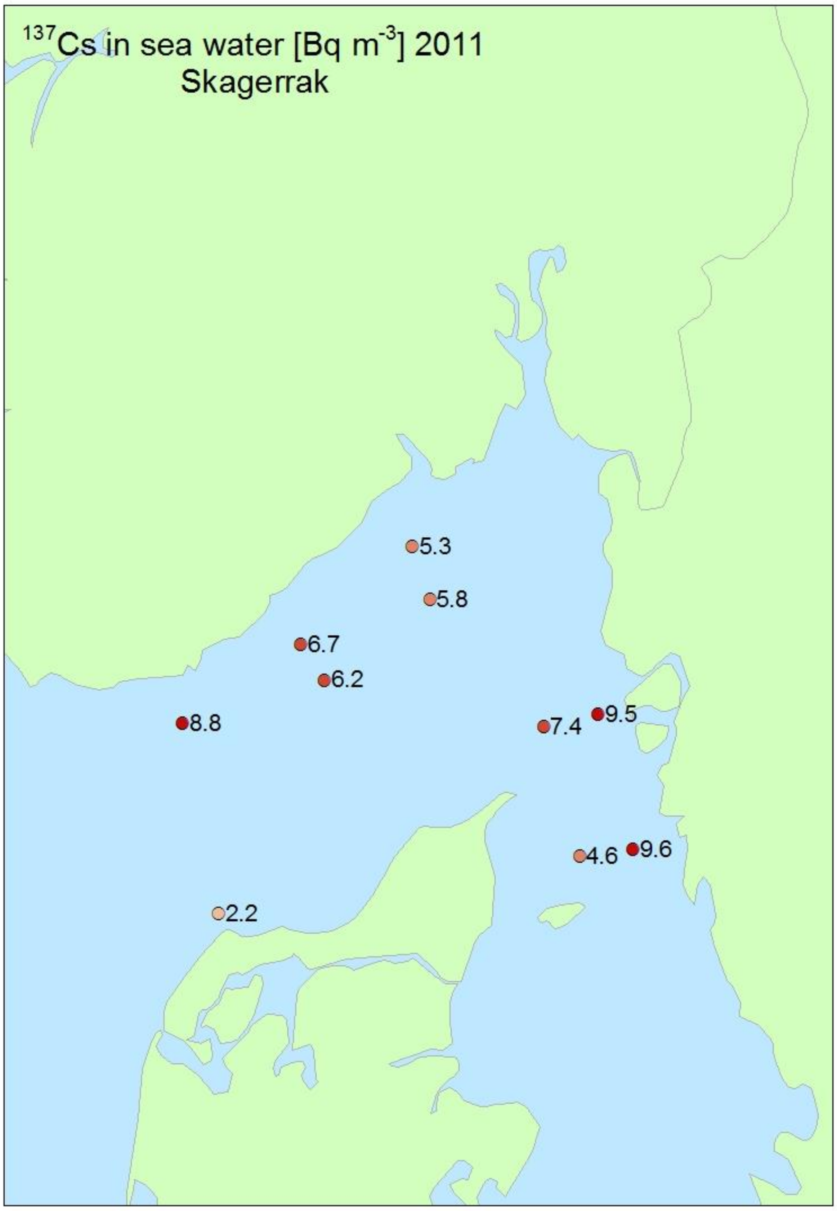

Figure 4.6. *Activity concentration (Bq m^{-3}) of ^{137}Cs in sea water samples (surface water) collected in the Skagerrak in 2011.*

Data from Hillesøy (2002-2011) and Grense Jakobselv (2005-2011) (Figure 4.7) show that the levels of ^{137}Cs in the Norwegian coastal current are slowly decreasing. The effective half-life of ^{137}Cs in the Baltic Sea, which is one of the main sources of ^{137}Cs in this region, has been estimated to 10 years (Ikäheimonen *et al*. 2009).

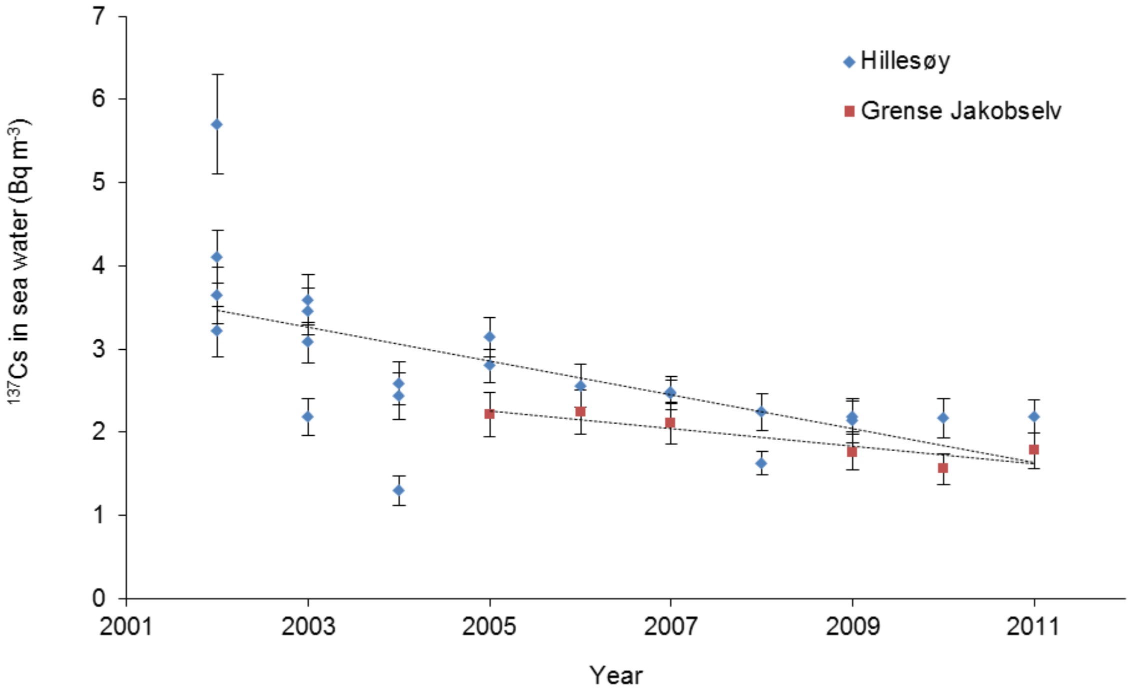

Figure 4.7. *Activity concentration (Bq m^{-3}) of ^{137}Cs in sea water at Hillesøy and Grense Jakobselv in the period 2002-2011.*

Caesium-137 has also been analysed in surface sediments (upper 2 cm layer) from the Norwegian Sea, selected fjords and close to the position of the sunken nuclear submarine Komsomolets.

The results are presented in Figure 4.8 and 4.9, and range from 1 to 12 Bq kg^{-1} (d.w.) in the Norwegian Sea.

The activity concentration ^{137}Cs in sediments sampled in the fjords ranged from 6 to 272 Bq kg^{-1} (d.w.). The highest activity concentrations were found in samples from the Vefsnfjord.

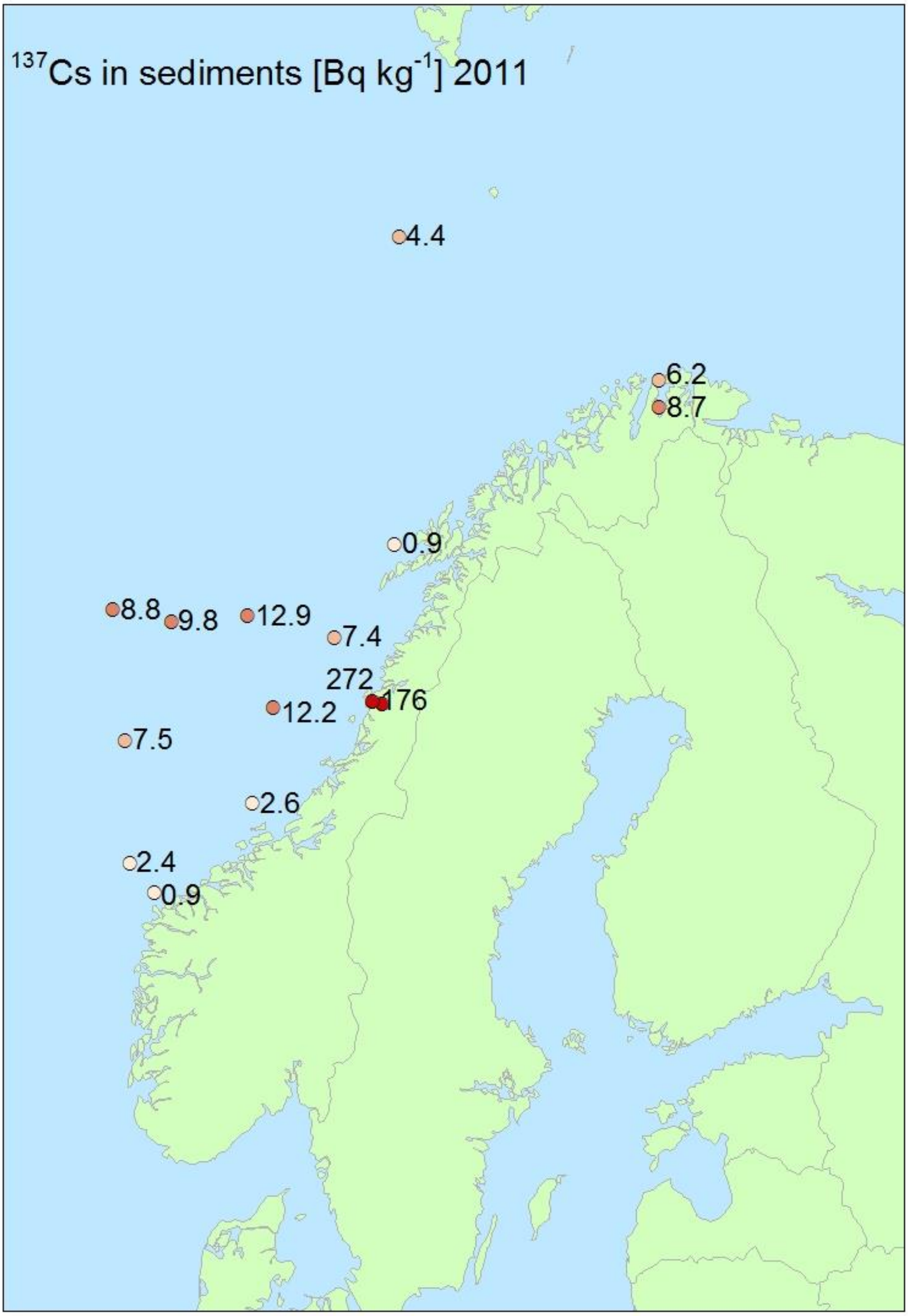

Figure 4.8. *Activity concentration (Bq kg^{-1} d.w.) of ^{137}Cs in surface sediment from the Norwegian and selected fjords in 2011. The sample south west of Bjørnøya has been collected close to the position of the sunken submarine Komsomolets.*

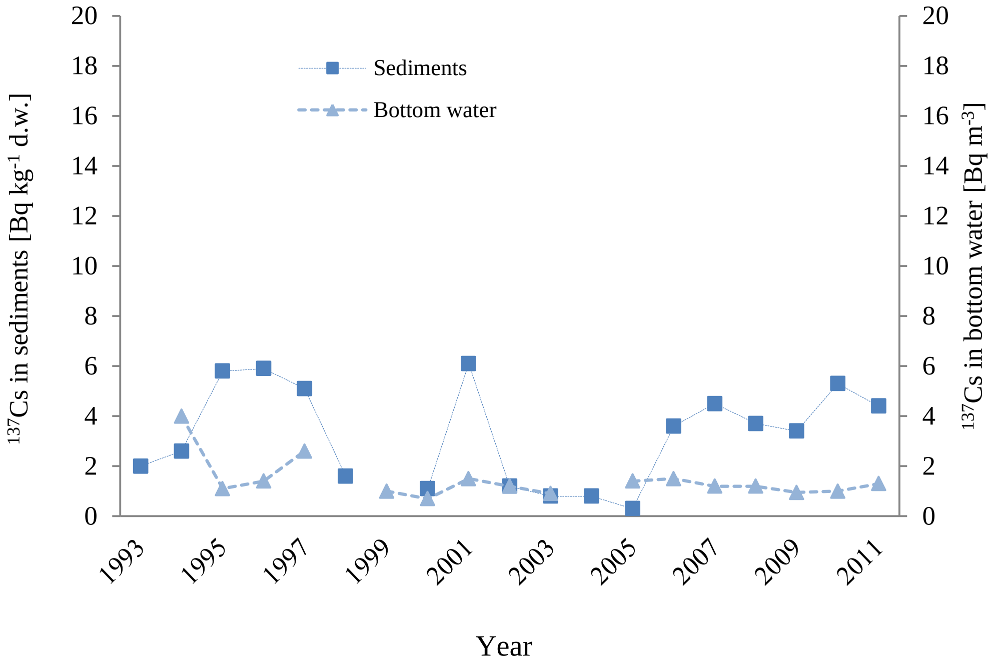

Figure 4.9. *^{137}Cs in samples close to the position of the sunken nuclear submarine Komsomolets. No indication of leakage from the submarine.*

4.4 Plutonium-239 and -240 in sea water

^{239}Pu ($T_{1/2}$ = 24 110 y) and ^{240}Pu ($T_{1/2}$ = 6 563 y) belong to the transuranium elements and are mainly produced by neutron capture, with subsequent beta decay, by ^{238}U and ^{239}Pu, respectively. The behaviour of plutonium in the marine environment is complex due to its different possible oxidation states. Under oxidizing conditions, plutonium is generally found in the more soluble +V and +VI states, while in reducing environments, the more particle-reactive +III and +IV states dominate.

The main sources of $^{239+240}$Pu in northern Norwegian marine waters are global fallout from atmospheric nuclear weapons tests in the 1950s and 1960s and remobilised plutonium from Irish Sea sediments.

Observed levels in the Norwegian Sea in 2011 are presented in Figure 4.10 and range from 2 to 10 mBq m^{-3}. The levels of $^{239+240}$Pu in the Norwegian Sea in 2011 are similar to the levels observed in 2001, 2004 and 2007 (NRPA, 2003; NRPA, 2006; NRPA 2009).

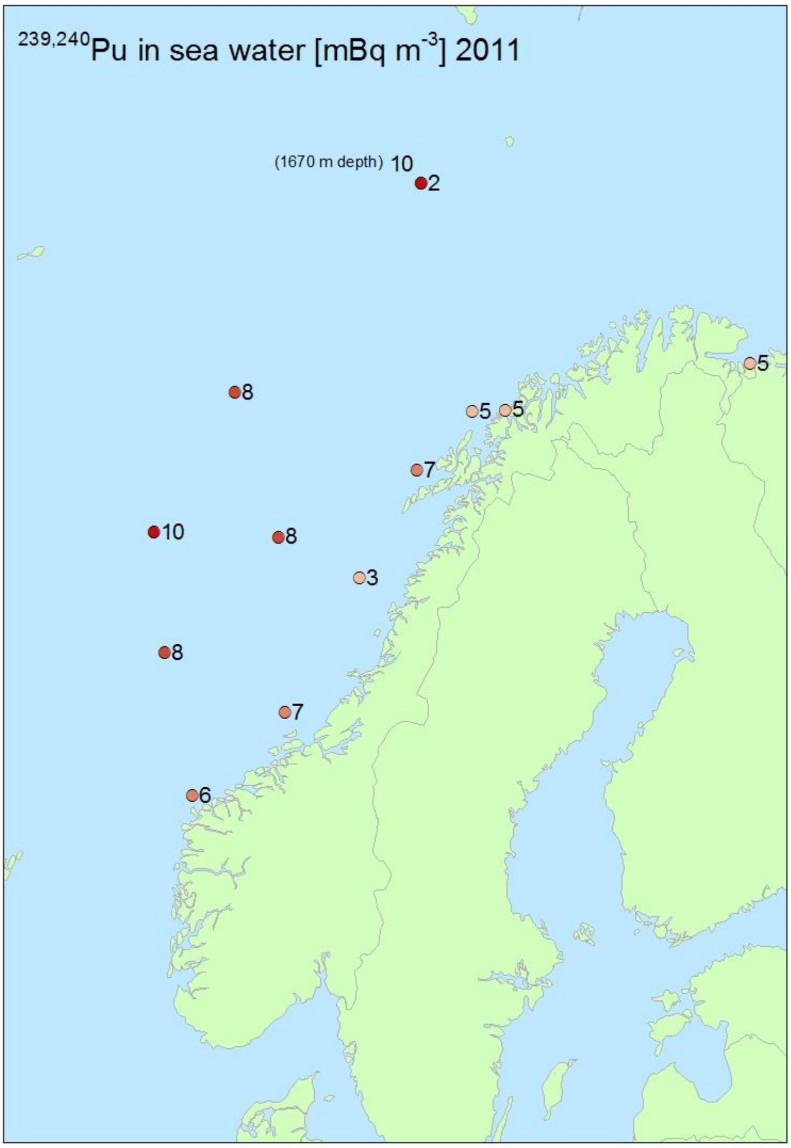

Figure 4.10. Activity concentration (mBq m^{-3}) of $^{239+240}$Pu in surface water samples collected in the Norwegian Sea in 2011.

4.5 Americium-241 in sea water

^{241}Am belongs to the transuranium elements and has a physical half-life of 432 years. It is produced, for example, in nuclear reactors by successive neutron capture by ^{239}Pu. Americium-241 is finally formed by beta-decay of ^{241}Pu ($T_{1/2}$ = 14.35 y).

Main sources of ^{241}Am in the environment are fallout of ^{241}Pu from nuclear weapon tests in the 1950s and 1960s and the discharge of ^{241}Am and ^{241}Pu from reprocessing plants.

Measured activity concentrations of ^{241}Am in the Norwegian Sea in 2011 are presented in Figure 4.11 and range from 1.2 to 10.7 mBq m^{-3}.

The observed levels of ^{241}Am are similar to the levels observed in 2002, 2005 and 2007 (NRPA, 2003; NRPA, 2006; NRPA, 2009).

The concentration of americium-241 in the Jan Mayen area was 2.5 mBq m^{-3}.

These are low levels and comparable to concentrations found in the Norwegian and Barents Seas (see for example NRPA, 2011 and Gwynn et al., 2012).

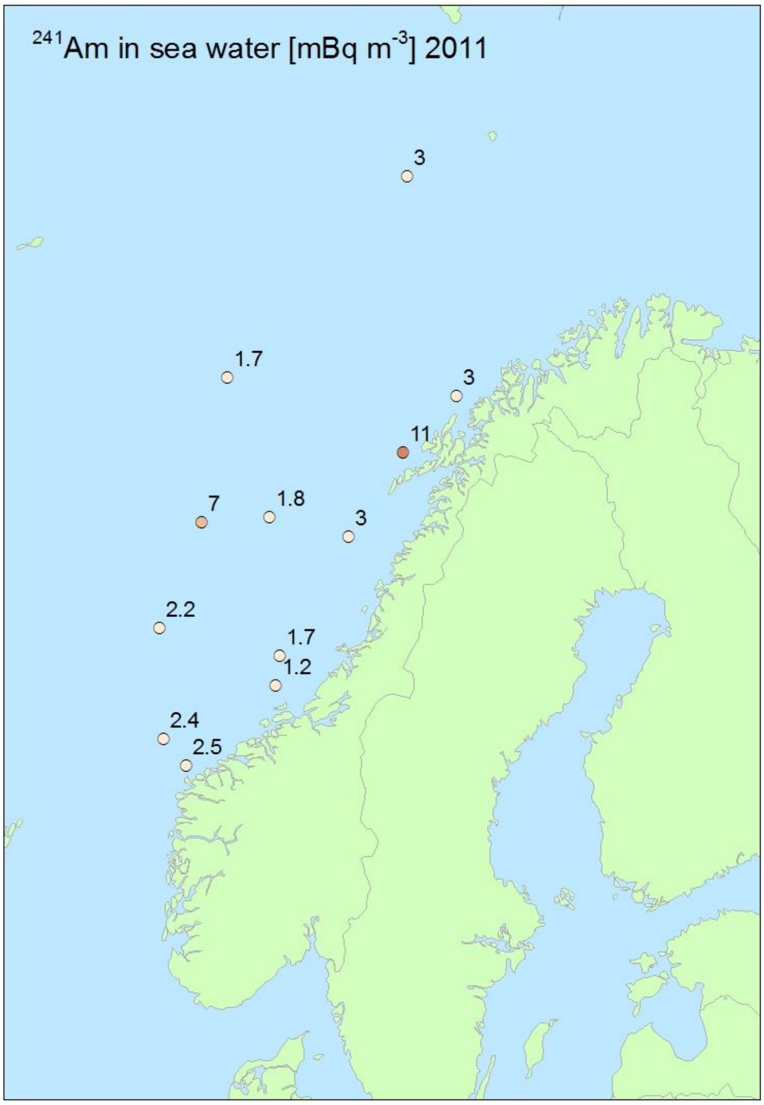

Figure 4.11. *Activity concentration (mBq m^{-3}) of ^{241}Am in sea water from the Norwegian Sea in 2011.*

4.6 Radioactive contamination in the Jan Mayen area 2011

When the management plan for the Norwegian Sea was adopted by the Norwegian Parliament in 2009, it was indicated that the waters off Jan Mayen could be opened for petroleum activities. During the work with a proposal for a study program, it became clear that there was a need for a better environmental description of the waters around Jan Mayen. The Institute of Marine Research (IMR) was commissioned by the Ministry of Petroleum and Energy to describe the environment and marine resources in the waters around Jan Mayen. The investigation also included an assessment of the levels of radioactive pollution.

During a cruise with the research vessel "G. O. Sars" to the Jan Mayen Ridge in October 2011, five sediment samples were taken, one sample of surface seawater and one pooled sample of cod.

Concentrations of cesium-137 in the sediment samples from Jan Mayen ranged from 3.2 to 5.6 Bq/kg dry weight (Fossum et al., 2011). The levels are low and comparable to those found in nearby waters (see for example NRPA, 2011 and Gwynn et al., 2012).

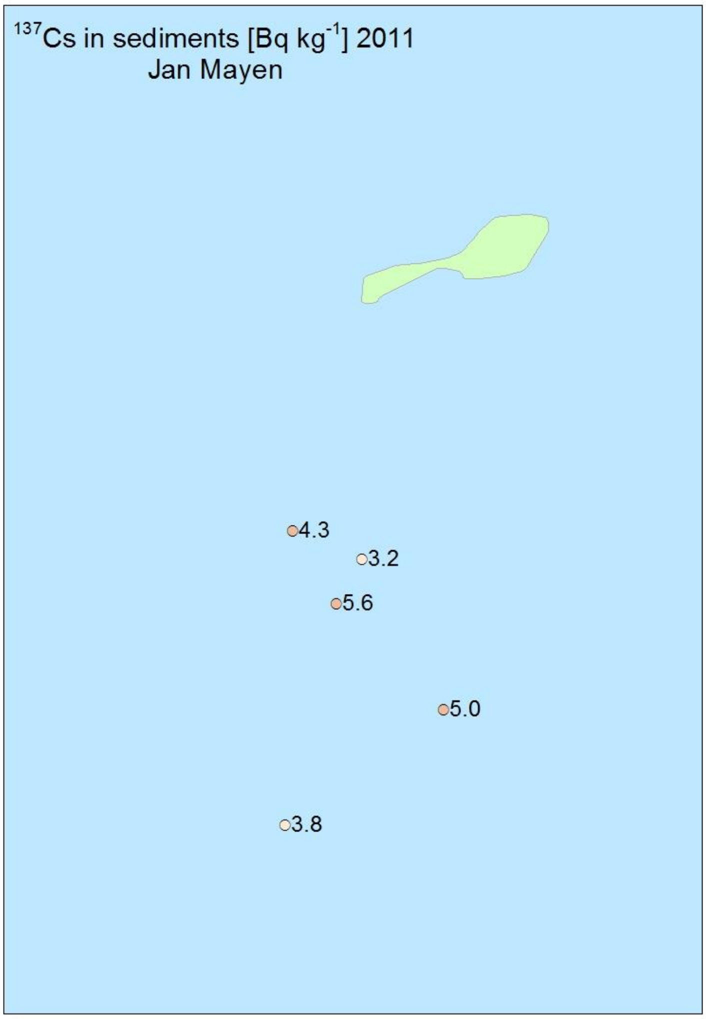

Figure 4.12. *Activity concentration (Bq kg^{-1} d.w.) of ^{137}Cs in surface sediment from the area south of Jan Mayen in 2011.*

4.7 Lead-210, polonium-210, radium-226 and radium-228 in sea water and sediments in the Jan Mayen area

The concentrations of lead-210, radium-226 and radium-228 are recently measured in the five sediment samples, and the results ranged from 166 to 234, 29.4 to 43.1 and 15.0 to 27.1 Bq kg^{-1} dry weight, respectively. The results are presented in Figures 4.13-4.15.

The concentrations of polonium-210 and ^{226}Ra in the seawater sample were 0.3 and 1.3 Bq m^{-3}, respectively.

Relatively few data sets on natural radionuclides in sediments in Norwegian waters have been reported. The concentrations found in the present study are, however, comparable to those given in a review article by Hosseini et al. (2012). The latter study reports typical ^{210}Pb concentrations in marine sediments to be around 150 Bq kg^{-1} dry weight, which is slightly lower than those found in the present study. Elevated levels of ^{210}Pb in surface sediments may be due to unsupported ^{210}Pb caused by sedimentation processes. The results from measurements of natural radionuclides in sediments in the present study are an important documentation of the levels before any opening of petroleum activities in these areas.

Figure 4.15. *Activity concentration (Bq kg^{-1} d.w.) of ^{210}Pb in surface sediment from the area south of Jan Mayen in 2011.*

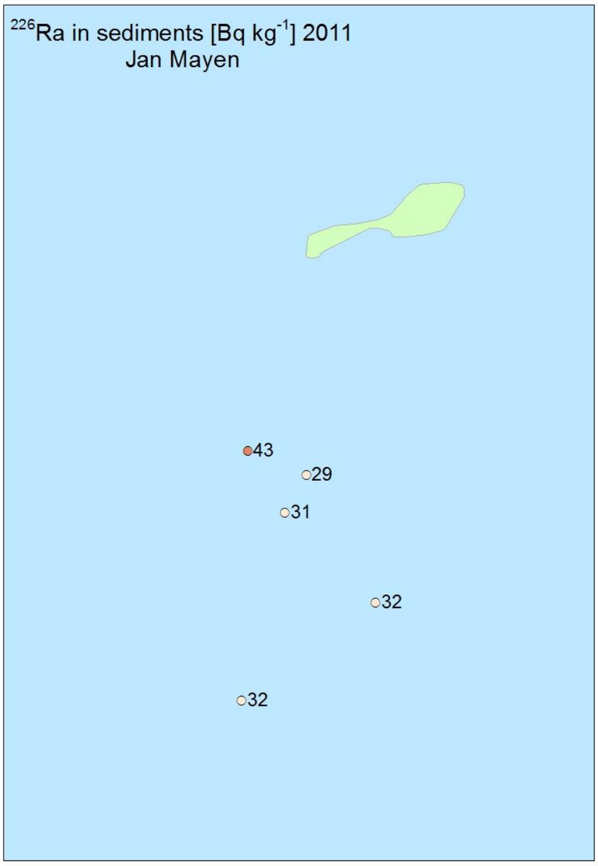

Figure 4.13. Activity concentration (Bq kg^{-1} d.w.) of ^{226}Ra in surface sediment from the area south of Jan Mayen in 2011.

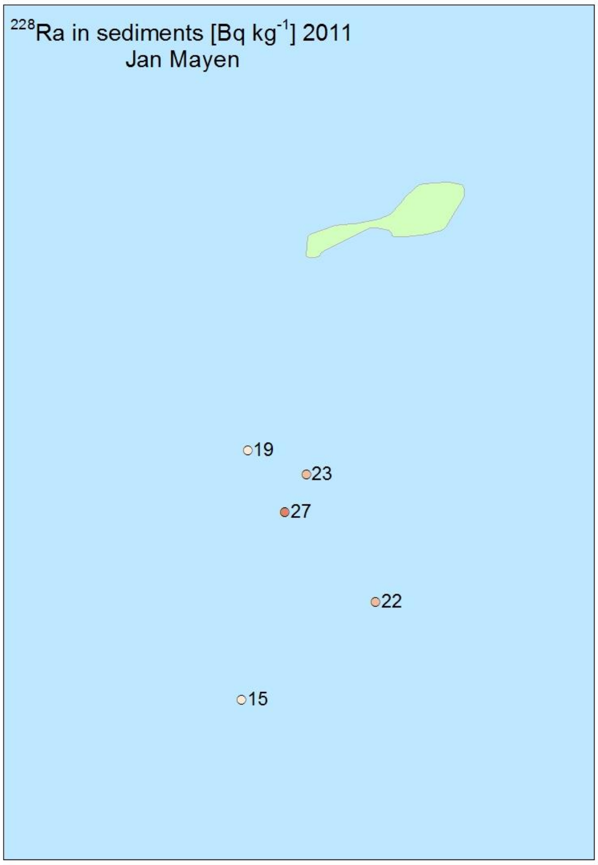

Figure 4.14. Activity concentration (Bq kg^{-1} d.w.) of ^{228}Ra in surface sediment from the area south of Jan Mayen in 2011.

5 Radioactivity in biota

5.1 Technetium-99 in seaweed

Figure 5.1. *Brown seaweed (*Fucus vesiculosus*) (Photo: NRPA).*

Seaweed is a useful bioindicator for ^{99}Tc in the marine environment. It has a high ability to concentrate ^{99}Tc from sea water and is easily accessible in most coastal areas. During 2011, seaweed (*Fucus vesiculosus,* Figure 5.1) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for ^{99}Tc.

At Hillesøy sampling was performed monthly, and sampling was performed six times at Utsira in 2011. At the remaining sites, sampling was conducted in August or September. The results of the analyses are presented in Figure 5.2 and range from 23 to 64 Bq kg^{-1} (d.w.), where the highest activity concentration (126 Bq kg^{-1} d.w.) was found in one of the monthly samples collected at Hillesøy. Compared with the results from 1999-2001 (Rudjord *et al.*, 2001; Gäfvert *et al*., 2003), the levels of ^{99}Tc have decreased at most sampling sites, due to the reduced discharge of ^{99}Tc from Sellafield. The trend can also be seen in Figure 5.3, which shows the annual average activity concentration of ^{99}Tc in *Fucus vesiculosus* at Utsira and Hillesøy, together with the annual discharge of ^{99}Tc from Sellafield.

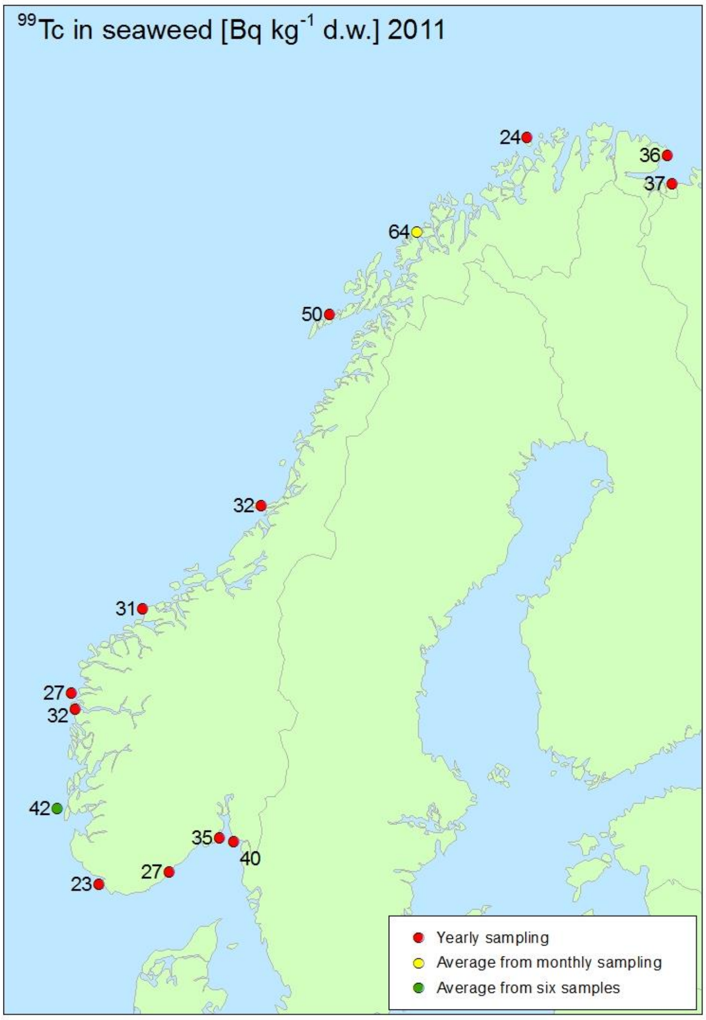

Figure 5.2. *Levels of* ^{99}*Tc in* Fucus vesiculosus *sampled along the Norwegian coastline in 2011.*

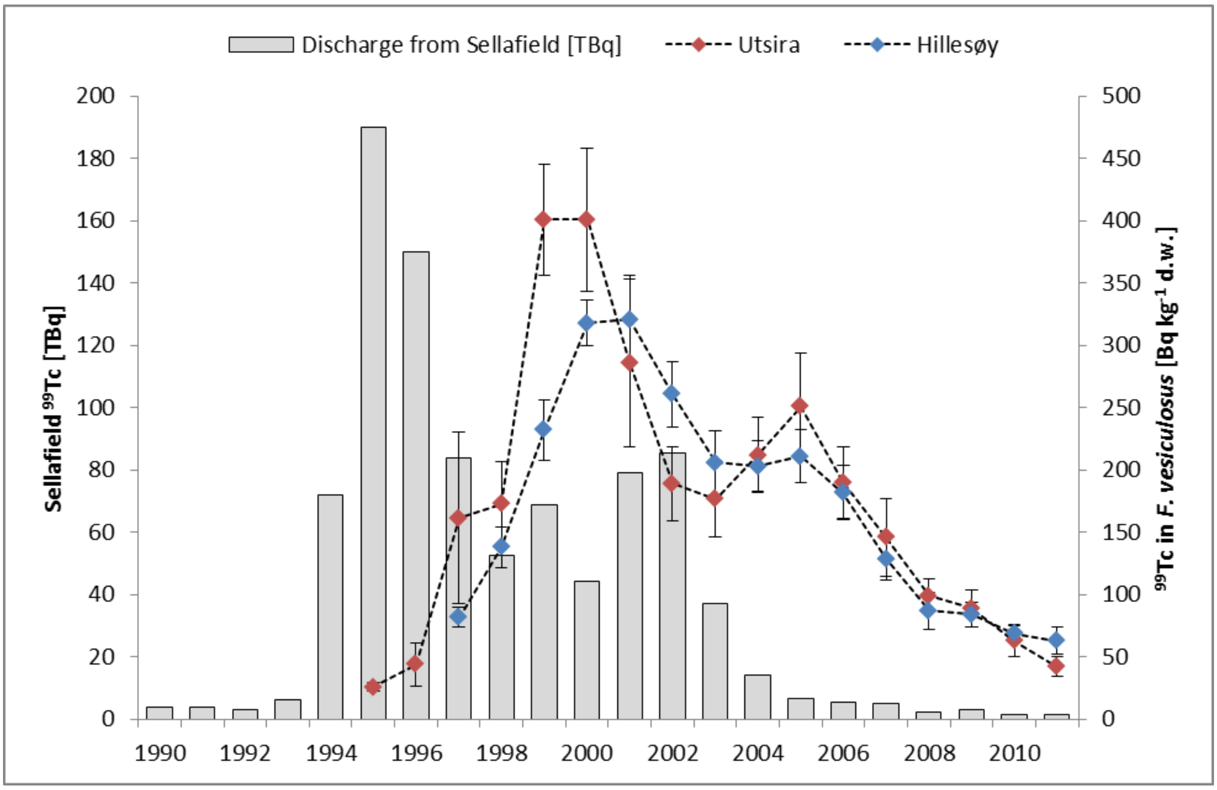

Figure 5.3. *Annual liquid discharge of ^{99}Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) ^{99}Tc activity concentration (secondary axis) in brown algae (*Fucus vesiculosus*) sampled at Utsira (data provided by IFE) in the period 1995-2011 and Hillesøy in the period 1997-2011.*

5.2 Plutonium-239 and -240 in seaweed

Fucus vesiculosus has been collected and analysed for ^{239,240}Pu at Utsira since 1980. The results from the period 1980 to 2011 are presented in Figure 5.4. The activity concentrations in these samples were in the range of 23 to 201 mBq kg^{-1}, with relatively large fluctuations from year to year. A slowly decreasing trend in the activity concentration of ^{239,240}Pu in the seaweed samples collected in the period from 1980 to 2011 is observed. The activity concentration in the sample from 2011, 0.14 ± 0.03, is somewhat higher than previous years, but with considerable uncertainty.

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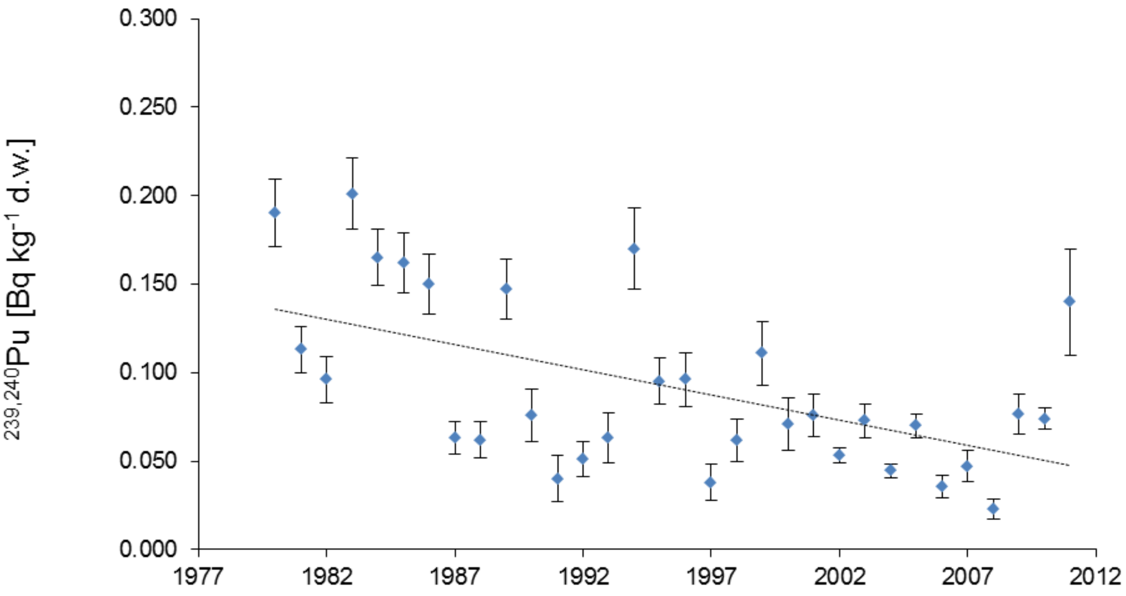

Figure 5.4. *$^{239+240}$Pu levels (Bq kg^{-1} d.w.) in* Fucus vesiculosus *at Utsira in the period 1980 to 2011 (data provided by IFE).*

5.3 Caesium-137 in seaweed

Fucus vesiculosus has also been widely used as a bioindicator for ^{137}Cs. The accumulation of ^{137}Cs in brown algae is, however, not as pronounced as for ^{99}Tc. The uptake of ^{137}Cs also depends on the salinity of the surrounding sea water, with higher uptake at lower salinities (Carlsson and Erlandsson, 1991).

In 2011, samples of *Fucus vesiculosus* from the permanent coastal stations were analysed with respect to ^{137}Cs. The results are presented in Figure 5.5, and ranged from 0.1 Bq kg^{-1} (d.w.) in the north to 2.6 Bq kg^{-1} (d.w.) at Hvaler. The peak in the ^{137}Cs levels in seaweed collected at Vikna can be explained by run-off of ^{137}Cs from land, since this area was affected by fallout from the Chernobyl accident in 1986.

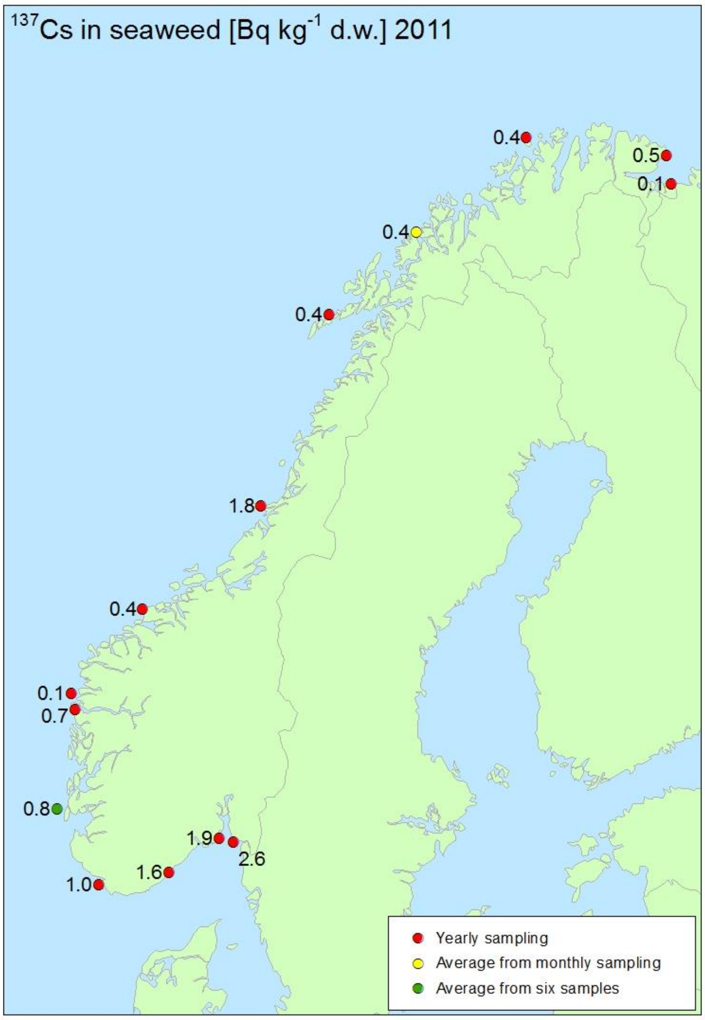

Figure 5.5. *Levels of ^{137}Cs (Bq kg^{-1} d.w.) in* Fucus vesiculosus *sampled along the Norwegian coastline in 2011.*

The reason for the higher activity concentration in *Fucus vesiculosus* in the southern part of Norway is a combination of two effects. First, the salinity in the water is lower and secondly, the activity concentration of ^{137}Cs in the sea water is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident.

Comparing the results with the levels found at the coastal sampling stations in the period 2000-2010 (NRPA, 2003; NRPA, 2004; NRPA 2005; NRPA 2006; NRPA 2007; NRPA 2008; NRPA, 2009 and NRPA, 2011), the activity concentration of ^{137}Cs in *Fucus vesiculosus* has been relatively stable in recent years. However, data from monthly sampling at Utsira (Figure 5.6) show that the activity concentration of ^{137}Cs has been slowly decreasing in seaweed. This is in agreement with the reported temporal trend of ^{137}Cs in Baltic Sea sea water (Ikäheimonen *et al*. 2009).

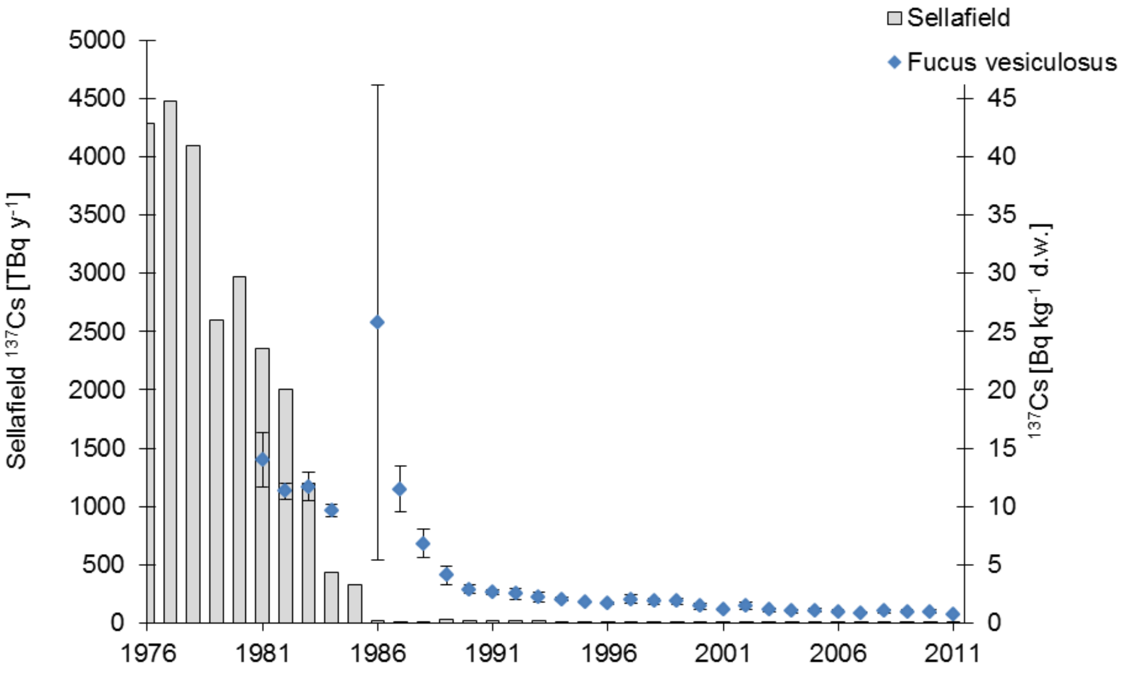

Figure 5.6. *Annual liquid discharge of* ^{137}*Cs from Sellafield and average activity concentration (Bq kg*$^{-1}$ *d.w.) from monthly sampling in seaweed (*Fucus vesiculosus*) from Utsira in the period 1980-2011 (data from IFE).The large variability in 1986 is due to the Chernobyl accident the same year.*

5.4 Caesium-137 in fish and crustaceans

Samples of cod from the Barents Sea have been analysed for ^{137}Cs since the early 1990s. In Figure 5.7 the activity concentration of ^{137}Cs in muscle tissue from cod caught in the Barents Sea is shown. All samples are below 1 Bq kg^{-1} (w.w.), and in recent years below 0.5 Bq kg^{-1} (w.w.). The results show a slightly decreasing trend in the period 1992-2011. The pooled sample of cod from the Jan Mayen area contained 0.15 and 0.39 Bq/kg fresh weight cesium-137 and polonium-210, respectively (Green et al., 2012). This is as expected, and comparable to levels found in the Norwegian and Barents Seas in 2008 and 2009. (e.g. NRPA, 2011 and Gwynn et al, 2012).

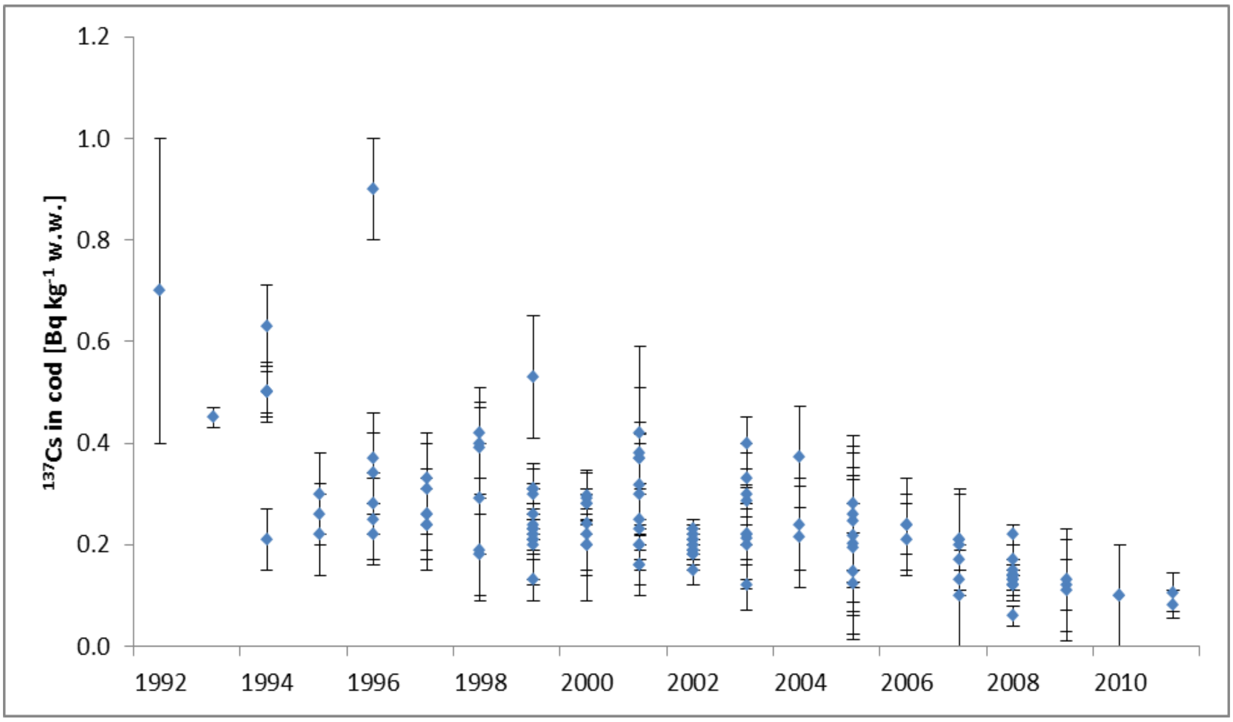

Figure 5.7. *Activity concentration of ^{137}Cs (Bq kg^{-1} w.w.) in cod from the Barents Sea (the area around Bjørnøya) sampled in the period 1992 to 2011.*

Caesium-137 levels in fish species and crustaceans caught in the Barents Sea, the Norwegian Sea and the North Sea in 2011 are shown in Table 5.1. All obtained results were below 0.3 ± 0.1 Bq kg^{-1} (w.w.).

Table 5.1. *Activity concentrations (Bq kg^{-1} w.w.) of ^{137}Cs in fish and crustaceans caught in the Barents, Norwegian and North Seas and in the Skagerrak in 2011.*

Species	Location	No. of samples (total no. of fish muscles)	^{137}Cs in muscle tissue (Bq kg^{-1} w.w.)
Cod	North Sea	2(57)	0.22±0.07 and 0.3±0.1
Haddock		2(42)	0.07±0.03 and 0.08±0.05
Cod	Coastal waters from Stad to Tromsø	2 (42)	0.23±0.06 and 0.2±0.1
Cod	Norwegian Sea, Jan Mayen		0.15±0.05
Saithe	Norwegian Sea	3 (75)	0.15±0.04 to 0.2±0.1
Norway Pout		3 (whole)	0.07±0.03 to 0.08±0.03
Haddock		3 (75)	0.08±0.04 to 0.11±0.03
Mackerel		3 (61)	0.08±0.04 to 0.2±0.1
Atlantic herring		2 (50)	0.1±0.05 and 0.12±0.03
Blue whiting		3 (66)	0.2±0.1
Grater argentine		1 (whole)	0.06±0.02
Redfish		1(25)	0.2±0.1
Deepwater Redfish		1(15)	0.13±0.04
Shrimp		(whole)	0.07±0.03
Cod	Barents Sea	6(450)	0.08±0.03 to 0.15±0.05
Polar Cod		(whole)	<0.03
Capelin		(whole)	0.06±0.06
Shrimp		(whole)	0.05±0.02

Technetium-99 has been analysed in lobster (*Hommarus gammarus*) tail muscle from Værlandet since 2002. The results for female lobsters and male lobsters are presented in Figures 5.8 and 5.9. In the period from 2002 to 2011 the levels of ^{99}Tc in lobster have decreased due to the reduced discharge of ^{99}Tc from Sellafield.

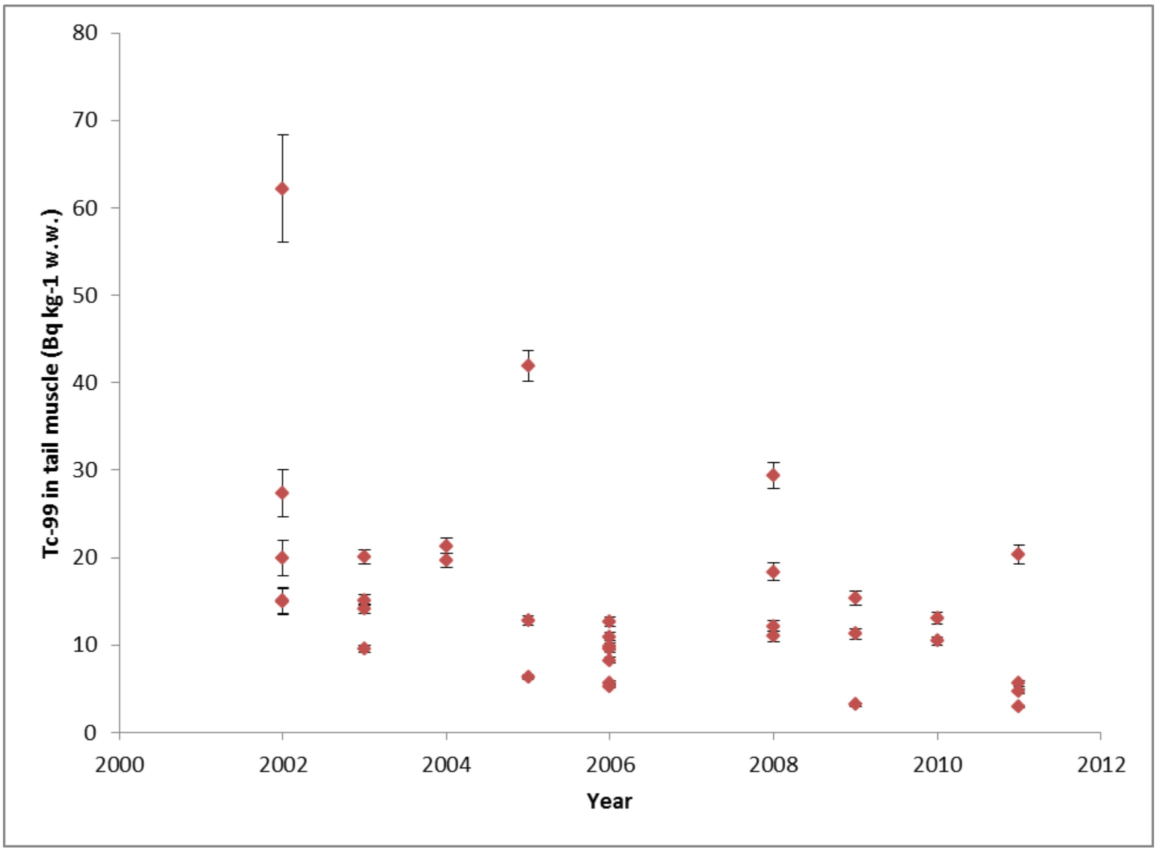

*Figure 5.8. Technetium-99 concentration in female lobster (*Hommarus gammarus*) from Værlandet (2002-2011).*

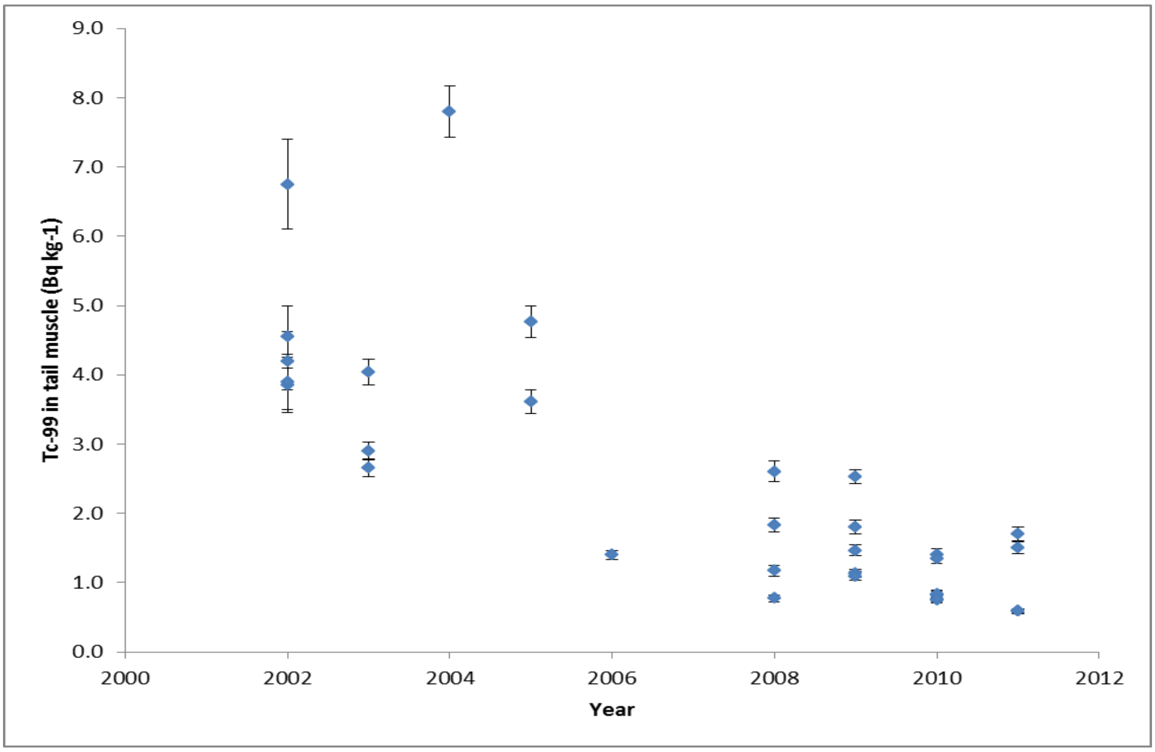

Figure 5.9. Techentium-99 concentration in male lobster (Hommarus gammarus) from Værlandet (2002-2011). Summary and conclusions

In 2011, samples of sea water, sediment, and biota were mainly collected in the Norwegian Sea, the Skagerrak and at a number of coastal stations and fjords. Data on radioactivity levels and trends in these areas are summarised in this report.

5.5 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2011 are within authorised limits according to the estimates made by the operator. The effective dose to a member of the critical group from liquid radioactive discharge at Kjeller was estimated to be 0.0123 µSv, which correspond to 1.23 % of the dose limit. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be 0.017 µSv, corresponding to 1.7 % the dose limit. The discharge of anthropogenic radionuclides from Norwegian sources is only detectable in the environment close to each discharge point and has no significant impact on the large-scale distribution of these radionuclides in the marine environment.

Produced water from offshore oil production may contain enhanced levels of naturally occurring radium isotopes. In 2011, the discharged activity of ^{226}Ra and ^{228}Ra from the Norwegian oil and gas industry were reported to 468 GBq and 371 GBq, respectively.

The long-range transports of radionuclides originating from nuclear weapons fallout, the Chernobyl accident, and from reprocessing of spent nuclear fuel are still the main contributors to anthropogenic radionuclides found in Norwegian waters. In addition to direct discharges from reprocessing, water from the Baltic Sea and remobilisation of ^{137}Cs and plutonium from contaminated Irish Sea sediments act as secondary sources of radionuclides to the Norwegian marine environment.

5.6 Radioactivity in sea water and sediment

In 2011, samples of sea water and sediment were collected mainly in the Norwegian Sea and analysed with respect to ^{137}Cs, ^{99}Tc, ^{90}Sr, ^{239,240}Pu, ^{241}Am, ^{210}Pb, ^{226}Ra and ^{228}Ra. A general trend seen in most samples is that the levels of radioactivity are similar to or slightly lower than have been observed in recent years. This can be explained by reduced discharges, radioactive decay, and other processes such as sedimentation and dilution. Discharge of produced water is at the same level as previous years.

5.6.1 Technetium-99 in sea water

Samples of sea water were collected in the Norwegian Sea. Levels of ^{99}Tc in surface sea water ranged from 0.05 Bq m^{-3} to 0.31 Bq m^{-3}. The average activity concentration from quarterly sampling at Hillesøy in 2011 was 0.25 ± 0.05, which is significantly lower than the peak values observed in 1999/2000. The concentration of ^{99}Tc is approaching the same level as before the increase in discharges in 1994/1995.

5.6.2 Strontium-90 in sea water

The activity concentration in surface sea water sampled in the Barents Sea and in the Skagerrak in 2011 ranged from 0.6 Bq m^{-3} to 1.9 Bq m^{-3}. The highest concentrations were found south in the Norwegian Sea. Data from Hillesøy and from Grense Jakobselv in the period 2002/2004 to 2011 indicates that the levels of ^{90}Sr are slowly decreasing in the Norwegian coastal current.

5.6.3 Caesium-137 in sea water

Observed levels of ^{137}Cs in surface water in the Norwegian Sea and the Skagerrak in 2011 ranged from 1.1 to 9.6 Bq m^{-3}. The highest levels were found in brackish water in the Skagerrak. This is generally similar to the activity concentrations observed in the same area in 2010. Data from Hillesøy and Grense Jakobselv in the period 2002 to 2011 also show that the levels of ^{137}Cs are slowly decreasing in the Norwegian coastal current.

5.6.4 Plutonium-239, -240 and americium-241 in sea water

Observed levels of $^{239,240}Pu$ in the Norwegian Sea in 2011 ranged from 1.5 to 10.4 mBq m^{-3}. The activity concentration of ^{241}Am in sea water from the Norwegian Sea ranged from 1.2 to 10.7 mBq m^{-3}. This is generally similar to levels observed in the same area previous years.

5.6.5 Radioactive contamination in the Jan Mayen area 2011

The concentrations of lead-210, radium-226 and radium-228 are recently measured in the five sediment samples, and the results ranged from 166 to 234, 29.4 to 43.1 and 15.0 to 27.1 Bq/kg (d.w.), respectively. The concentrations of polonium-210 and radium-226 in the seawater sample were 0.3 and 1.3 Bq/m^3, respectively.

Relatively few data sets on natural radionuclides in sediments in Norwegian waters have been reported. The concentrations found in the present study are, however, comparable to those given in a review article by Hosseini et al. (2012).

5.7 Radioactivity in biota

5.7.1 Technetium-99 in seaweed

Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 23 to 126 Bq kg^{-1} (d.w.). For most stations the levels were lower in 2011 compared to observed levels in the period 2002-2007. The levels in 2011 are significantly lower than the peak values observed in the period 1999-2001. Monthly sampling at Hillesøy and Utsira show that the levels of ^{99}Tc in seaweed have decreased since 2005 due to the reduced discharge of ^{99}Tc from Sellafield.

5.7.2 Caesium-137 in seaweed

The activity concentration of ^{137}Cs in *Fucus vesiculosus* sampled at the coastal stations in 2011 was in the range of <0.2 to 2.6 Bq kg^{-1} (d.w.), where the highest levels were found in the Skagerrak. Data from monthly sampling at Utsira indicates that the levels of ^{137}Cs in seaweed are slowly decreasing and have been relatively stable the last years.

5.7.3 Caesium-137 in fish and crustaceans

Different species of fish and crustaceans, commercially important and others, have been sampled in the Barents Sea, the Norwegian Sea and the North Sea. The activity concentration of ^{137}Cs in fish from Norwegian marine waters is generally low. All analysed samples were below 0.3 ± 0.1 Bq kg^{-1} (w.w.).

Samples of cod from the Barents Sea have been analysed for ^{137}Cs since the early 1990s. These results show a slightly decreasing trend of ^{137}Cs in this period.

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7 Appendix

7.1 Analytical methods

During recent years, laboratories at the Norwegian Radiation Protection Authority (NRPA), and the Institute of Marine Research (IMR) have both been accredited according to the requirements of NS-EN ISO/IEC 17025. Laboratories and organisations that are not named here have not been accredited according to the requirements of NS-EN ISO/IEC 17025. The scope and conditions governing the accreditation of the institutes mentioned above are quite different, but they have all been accredited for gamma spectrometric measurements. For example, the NRPA is accredited for gamma spectrometric measurements in the energy interval 100-1800 keV (except ^{226}Ra), while the other organisations are accredited for gamma spectrometric measurements of ^{137}Cs. The analytical techniques employed at each institution are described below. The results in this report are not claimed to be accredited results as not all results are accredited.

7.2 The Norwegian Radiation Protection Authority (NRPA)

NRPA analyses alpha and gamma emitters, using the procedures described in this Appendix.

7.2.1 *Detection of gamma emitters*

At NRPA analyses of gamma-emitting nuclides are performed with HPGe detectors. All the detectors are situated in a low-background laboratory to ensure low detection limits. The HPGe detectors have relative efficiencies in the range of 23 % to 50 %. The resolution of the detectors, the full width at half maximum (FWHM) at 1332 keV, was less than 1.2 keV. Three of the detectors cover the energy interval 50-2000 keV, and two cover the interval 20-2000 keV.

Caesium-absorbing filters (sometimes also the prefilter) from the filtering system for sea water samples were dried separately at 105°C and ashed at 550°C before the activity was determined with an HPGe detector. The counting time varied from 1 to 4 days.

Samples of seaweed were dried at 105°C and homogenized, and placed in containers prior to gamma counting. Sediment samples were freeze-dried and placed in containers prior to gamma counting. Samples were counted for a minimum of 2 days. At NRPA, the detection limits for ^{137}Cs and ^{134}Cs in the sediment samples ranged from approximately 0.3-2 Bq kg^{-1} (d.w.) Uncertainties in the analysis include uncertainties from counting statistics, calibration and sample preparation and are normally in the range 5 % to 20 %.

7.2.2 *Determination of ^{239,240}Pu and ^{241}Am*

The concentrations of ^{239,240}Pu and ^{241}Am were measured in samples of 200 litres of sea water or 10-20 g of sediment. ^{242}Pu and ^{243}Am were added for chemical yield determination. Precipitation from the water samples was achieved using the analytical procedure described by Chen *et al*., (1991). After separation, americium and plutonium fractions were electrodeposited on stainless steel discs and the activity measured in semiconductor silicon detectors. The resolution of the detectors, the full width at half maximum (FWHM), was approximately 20 keV at 5486 keV (^{241}Am). Chemical yields obtained from the ^{242}Pu and ^{243}Am yield monitor were in the range 40 % to 80 %.

7.3 The Institute of Marine Research (IMR)

7.3.1 *Detection of gamma emitters*

Sediment samples were transported deep-frozen to IMR, where they were subsequently ground, freeze-dried, homogenized and transferred to polyethylene containers of appropriate size prior to analysis. The gamma counting for ^{137}Cs detection was performed in IMR's low-background laboratory using a HPGe detector with 30 % relative efficiency and an HPGe detector with 60 % relative efficiency. Both detectors have electric cryostat cooling systems, and 10 cm lead shielding.

7.3.2 *Determination of ^{99}Tc*

Before the activity concentration of ^{99}Tc can be determined, a radiochemical separation is necessary. Briefly, rhenium is added as a yield monitor in the form of $KReO_4$, and a preliminary extraction of ^{99}Tc (and Re) based on anion-exchange separation is performed. After iron hydroxide scavenging, ^{99}Tc and Re are further extracted by a second anion-exchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts are isolated. ^{99}Tc is beta-counted using a low-background anti-coincidence beta counter (Model Risø GM-25-5), and the yield of the rhenium tetraphenyl arsonium salt is determined gravimetrically. See Harvey *et al*. (1992) for a detailed description of the method.

7.4 Institute for Energy Technology (IFE)

7.4.1 *Determination of ^{99}Tc*

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to ^{99}Tc content. The pretreatment was performed at IFE Kjeller and the analysis at the Department of Radiation Physics at Lund University, Sweden. The following analytical procedure was used: Technetium was extracted into tri-butyl-phosphate (TBP) from sulphuric acid-hydrogen fluoride solution. Technetium was then back--extracted from the organic phase with a sodium hydroxide solution from which the technetium was electrodeposited onto stainless steel discs, and ^{99m}Tc was used as a radiochemical yield determinant. After decay of the yield determinant, ^{99}Tc was measured with an anti-coincidence-shielded GM counter (Holm *et al*., 1984).

7.4.2 *Determination of ^{90}Sr*

For ^{90}Sr, the standard method using fuming nitric acid was used. The recovery of ^{90}Sr in the analytical process was monitored by adding ^{85}Sr as a yield determinant, and the recovery of the daughter nuclide ^{90}Y was determined by titration with EDTA (Varskog *et al*., 1997). Finally, ^{90}Y was measured with an anti-coincidence-shielded GM counter.

7.4.3 *Detection of gamma emitters*

At IFE, analyses of gamma-emitting nuclides are performed with low-background HPGe detectors. Samples of seaweed were dried at 105°C and then homogenized, and placed in suitable containers prior to gamma counting.

7.4.4 *Determination of ^{239,240}Pu*

Samples of dried and homogenized *Fucus vesiculosus* were ashed and treated with *aqua regia* before separation of plutonium isotopes. Pu-242 was used as a tracer for radiochemical yield determination. The separation process involved extraction chromatography with TRU-resins (Sidhu *et al*. 2004). The plutonium fraction was finally coprecipitated with CeF_3 on a filter and counted using alpha spectrometry.

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