



## Radioactivity in the Marine Environment 2010

Results from the Norwegian Marine Monitoring Programme (RAME)

**Reference:**

NRPA. Radioactivity in the Marine Environment 2010. Results from the Norwegian National Monitoring Programme (RAME). StrålevernRapport 2012:10. Østerås: Norwegian Radiation Protection Authority, 2012.

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**Key words:**

Radioactivity, marine environment, RAME, monitoring, Norway

**Abstract:**

This report presents results of monitoring of radioactivity in seawater, sediment and biota collected in the North Sea, the Skagerrak and the Barents Sea in 2010.

**Referanse:**

Statens strålevern. Radioaktivitet i det marine miljø 2010. Resultater fra det nasjonale overvåknings-programmet (RAME). StrålevernRapport 2012:12. Østerås: Statens strålevern, 2012. Språk: engelsk.

**Forfattere:**

Gäfvert T<sup>1</sup>, Haldal H E<sup>2</sup>, Brungot A L<sup>1</sup>, Gwynn J<sup>1</sup>, Strålberg E<sup>3</sup>, Sværen I<sup>2</sup>, Strømsnes H<sup>2</sup>, Kolstad A K<sup>1</sup>, Møller B<sup>1</sup>, Komperød M<sup>1</sup>, Lind B<sup>1</sup>, Rudjord A L<sup>1</sup>

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**Emneord:**

Radioaktivitet, marin miljø, RAME, overvåking, Norge

**Resymé:**

Rapporten inneholder resultater fra overvåkingen av radioaktivitet i sjøvann og biota i 2010 i Nordsjøen, Skagerrak og Barentshavet.

Head of project: Torbjörn Gäfvert

*Approved:*



Per Strand, director, Department for Emergency Preparedness and Environmental Radioactivity.

36 pages.

Published 2012-12-31

Printed number 100 (12-12)

Cover design: 07 Oslo

Printed by 07 Oslo

Cover: Sampling of seawater from the water column (such as bottom water near the wreck of «Komsomolets»), is conducted with a Seabird 911 plus CTD with a carousel of 10 liter water samplers. .Photo: IMR

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www.nrpa.no

ISSN 0804-4910 (print)

ISSN 1891-5191 (online)

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## Results from the Norwegian Marine Monitoring Programme (RAME)

**Statens strålevern**

Norwegian Radiation  
Protection Authority  
Østerås, 2012

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# 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  $^{99}\text{Tc}$  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 2010, samples for monitoring radioactivity in the marine environment were collected mainly in the North Sea, the Skagerrak 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 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  $^{137}\text{Cs}$  due to outflowing brackish water from the Baltic Sea into the Skagerrak. In 2000, the annual net outflow from the Baltic was estimated at 40 TBq, with a half-life of 13.4 y (Dahlgard, 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  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$ , 622 PBq  $^{90}\text{Sr}$ , 6.52 PBq  $^{239}\text{Pu}$  and 4.35 PBq  $^{240}\text{Pu}$  have been released to the environment (UNSCEAR,

2000a), mainly in the northern hemisphere, where most of the tests took place.

### 2.2 Norwegian sources

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  $\mu\text{Sv}$  per year to a hypothetical critical group. Discharges from IFE Kjeller and IFE Halden are reported annually to the authorities. In 2010, the annual dose from liquid discharges from IFE Kjeller and IFE Halden were estimated to 0.03  $\mu\text{Sv}$  and 0.012  $\mu\text{Sv}$ , respectively.

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  $^{99\text{m}}\text{Tc}$  (which decays to  $^{99}\text{Tc}$  with a half-life of 6 h) and  $^{131}\text{I}$  ( $T_{1/2} = 8.04$  d). The most important radionuclide concerning dose to the public after discharge is  $^{131}\text{I}$ . In 2010, a total of 1297

GBq was discharged to the sewage system from the medical sector in Norway.

Large volumes of produced water, containing dissolved  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , 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  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  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  $\text{Bq m}^{-3}$ ). 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  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  from the Norwegian oil and gas industry in 2010 were 486 GBq and 385 GBq, respectively.

### 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  $^{137}\text{Cs}$  into the Kara Sea; a level which is of a similar order of magnitude to inputs from Sellafield-derived  $^{137}\text{Cs}$  to the area

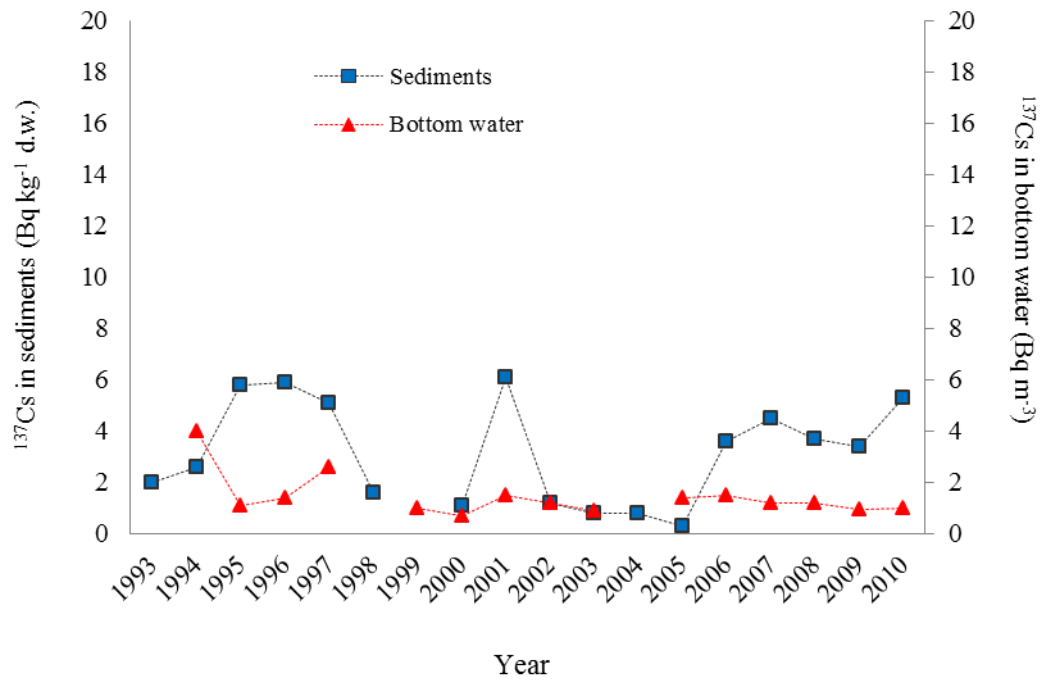
(Vakulovsky *et al.*, 1995). However, the fact that the prevailing surface currents tend to transport contamination away from the Kara Sea in predominantly easterly and northerly directions, suggests that the resultant inputs to the Barents Sea has been relatively low. Nonetheless, there is a potential 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 and the K-159 in the Norwegian Sea and the Barents Sea. 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 and analysed with respect to  $^{137}\text{Cs}$  are presented in Figure 2.2. In 2010 the activity concentration of  $^{137}\text{Cs}$  in sea water was below detection limit of  $1.0 \text{ Bq m}^{-3}$ .



Figure 2.1. Sources of radionuclides in the northern marine environment.





**Figure 2.2.** Cs-137 in samples close to the position of the sunken nuclear submarine Komsomolets.

### 3 Collection of samples

In 2010, samples were mainly collected in the North Sea, in the Skagerrak, 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 North Sea were collected in July-August 2010 by IMR and NRPA from the vessel R/V “Johan Hjort” (Figure 3.2). During the expeditions, samples of surface water were collected which were later analysed for  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and plutonium isotopes. Sediment was also sampled and later analysed for  $^{137}\text{Cs}$ .



**Figure 3.2.** R/V Johan Hjort (Photo: H.P. Tysnes).

### 3.1 Seawater

For  $^{99}\text{Tc}$ , 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  $^{90}\text{Sr}$ , 50-100 litres of unfiltered water were collected at each sampling station and later transported to IFE for analysis.

For the analysis of  $^{137}\text{Cs}$ ,  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters are used as sorbents (Roos *et al.*, 1994). The system consists of a prefilter (1 micron) and two  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters connected in series (Figure 3.3). Assuming identical collection efficiency for the  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -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:

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

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



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

Samples of 200 liters (unfiltered) were collected for the determination of the activity concentrations of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$ . To collect deep water samples, a CTD/Rosette multi-bottle sampler was used (Figure 3.4). Plutonium-242 and  $^{243}\text{Am}$  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.** CTD/Rosette multi-bottle sampler (Photo: 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:IMR)

### 3.3 Seaweed

NRPA collected seaweed (*Fucus vesiculosus*) samples from four 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 eleven locations along the coastline, from the Russian border in the north to the Swedish border in the south.

### 3.4 Fish and crustaceans

IMR collected samples of different fish species from the North Sea by combining muscle samples of 50-100 grams from 25 individuals. 100 samples of cod were also collected from two 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. At the request of NRPA, Labora A/S in Salten collected different samples of fish species by combining muscle samples of from 25

individual fish, and crustaceans from the northern part of Norway.

## 4 Radioactivity in sea water and sediments

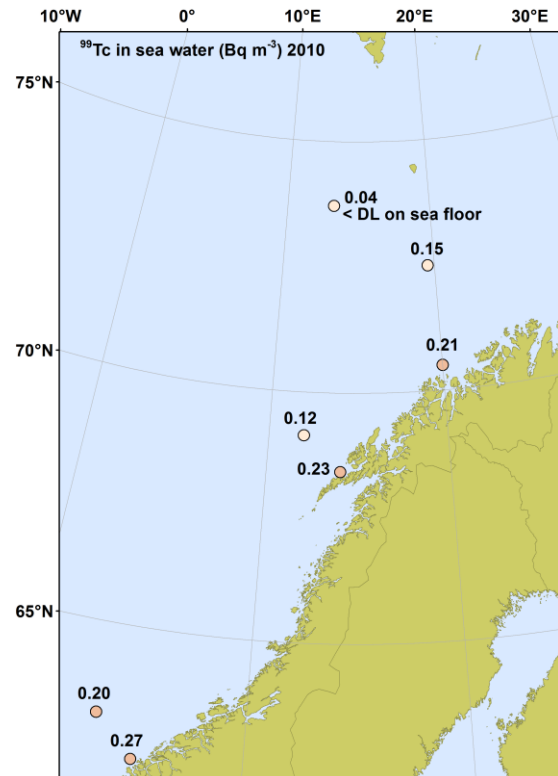
### 4.1 Technetium-99 in sea water

Technetium-99 has a physical half-life of  $2.13 \cdot 10^5$  years, and originates from the decay of  $^{99}\text{Mo}$ , which is formed either as a fission product from  $^{235}\text{U}$  or  $^{239}\text{Pu}$  or by neutron activation of  $^{98}\text{Mo}$ . The main source of  $^{99}\text{Tc}$  in Norwegian waters is liquid discharge from the reprocessing plant at Sellafield. In oxygenated sea water,  $^{99}\text{Tc}$  is present as the highly soluble pertechnetate ion ( $\text{TcO}_4^-$ ). Due to its conservative behavior in sea water,  $\text{TcO}_4^-$  has the ability to be transported by ocean currents without being significantly affected by sedimentation processes. From the Irish Sea,  $^{99}\text{Tc}$  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  $^{99}\text{Tc}$  to reach this area (Hillesøy) from the Irish Sea has been estimated to 3-4 years (Brown *et al.*, 2002; Dahlgard, 1995).

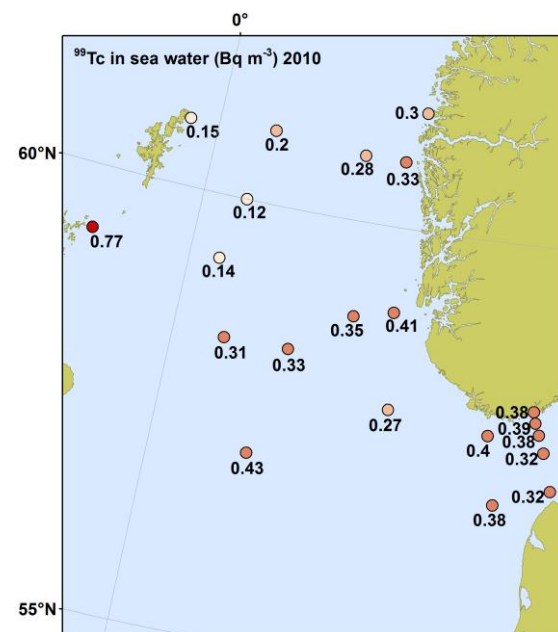
In 2010, sea water samples were collected in the North Sea and at IMR's standard hydrographic sections Svinøy, Gimsøy and Fugløya-Bjørnøya, and analysed for  $^{99}\text{Tc}$ . The results are presented in Figure 4.1 and 4.2. The activity concentrations ranged from 0.12 to 0.77  $\text{Bq m}^{-3}$ . The highest concentration was found off the coast of Scotland.

The activity concentrations found in 2010 are generally lower than those observed in 2002, 2003, 2005 and 2007 (NRPA, 2005; NRPA, 2007; NRPA 2009). One reason for this is the reduced discharge of  $^{99}\text{Tc}$  from Sellafield.

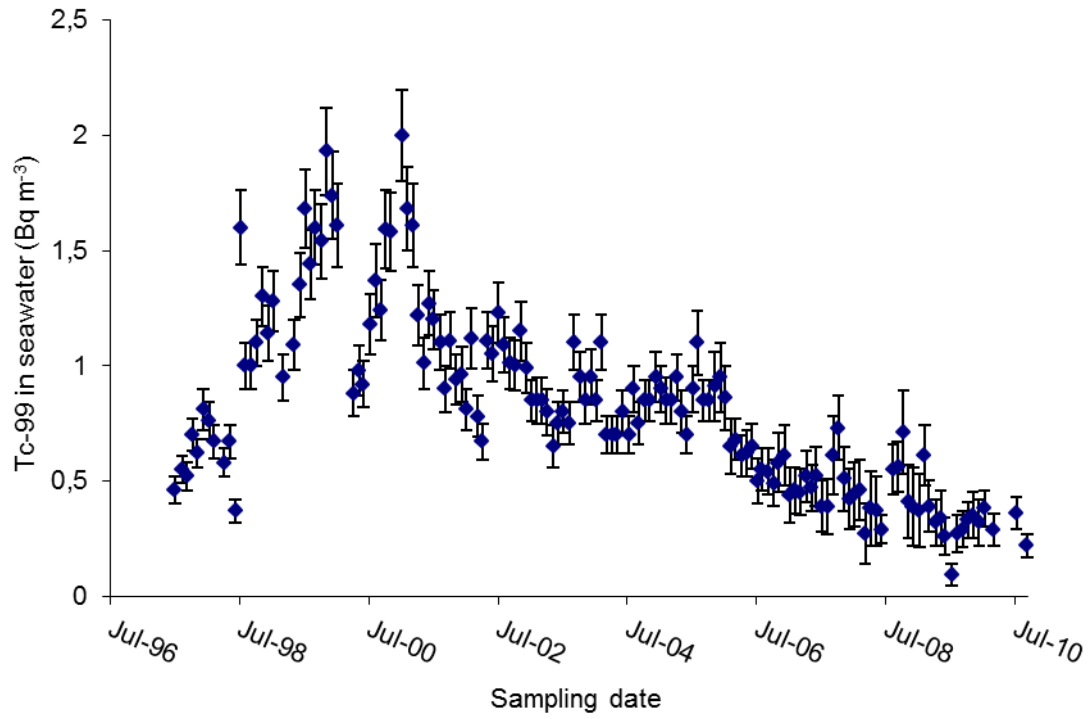
Monthly samples have also been collected at Hillesøy (Figure 4.3). The results show that the activity concentration of  $^{99}\text{Tc}$  in sea water continues to slowly decrease since the peak concentrations in 1998/1999.



**Figure 4.1.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{99}\text{Tc}$  in sea water samples collected in the Norwegian Sea and Barents Sea in 2010.



**Figure 4.2.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{99}\text{Tc}$  in sea water samples collected in the North Sea in 2010.

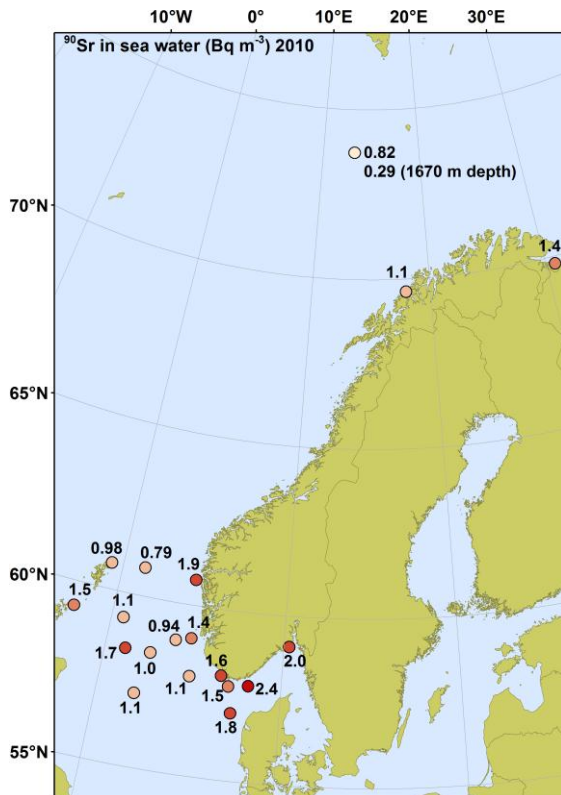


**Figure 4.3.** Activity concentration (Bq m<sup>-3</sup>) of <sup>99</sup>Tc in sea water at Hillesøy from monthly sampling.

## 4.2 Strontium-90 in sea water

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

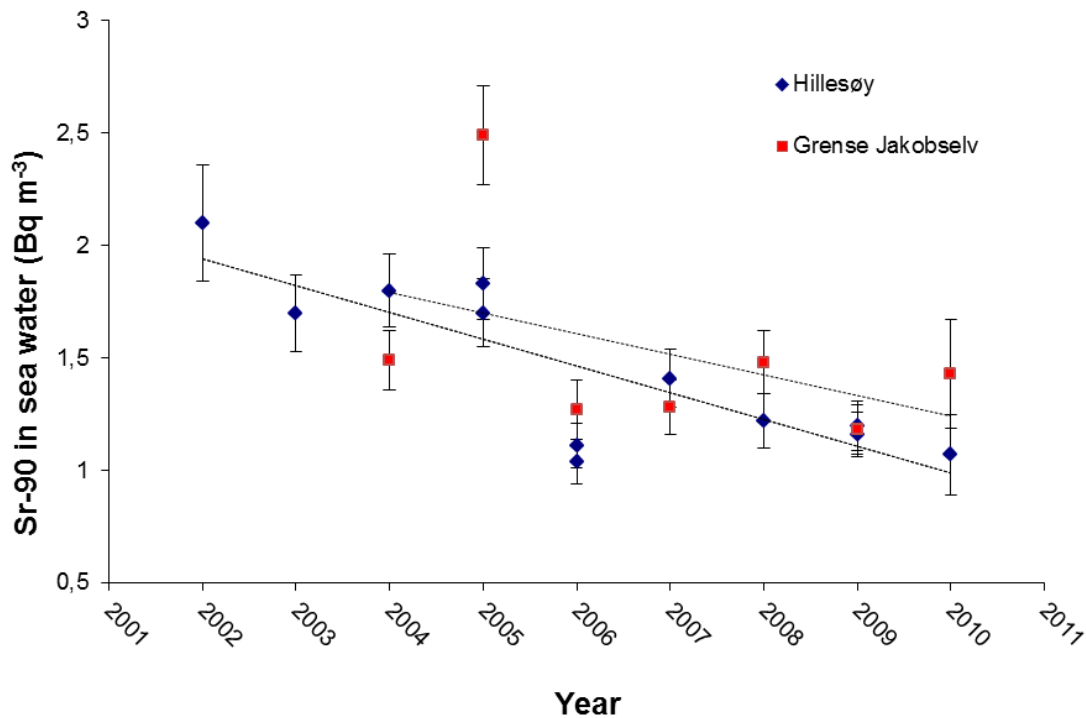
Sea water has been sampled mainly in the North Sea and later analysed for  $^{90}\text{Sr}$ . The main sources of  $^{90}\text{Sr}$  in these waters are discharge of liquid waste from reprocessing plants (mainly Sellafield), fallout from atmospheric nuclear weapons tests conducted mainly in the 1950s and 1960s, and outflow of water from the Baltic Sea. The results for 2010 are presented in Figure 4.4.



**Figure 4.4.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{90}\text{Sr}$  in surface water samples collected in the North Sea and the Barents Sea in 2010. The sample south west of Bjørnøya has been collected close to the position of the sunken submarine Komsomolets.

In the Barents Sea, the activity concentration in surface water ranged from  $0.3 \text{ Bq m}^{-3}$  to  $1.4 \text{ Bq m}^{-3}$ . The highest concentrations were found in the Skagerrak. Typical activity concentration of  $^{90}\text{Sr}$  in sea water from the southern Baltic Sea in 2008 has been reported to be around  $6 \text{ Bq m}^{-3}$  (Zalewska and Saniewski, 2009).

Results from annual or biannual sampling at Hillesøy show that the activity concentration in sea water in the Norwegian coastal current is slowly decreasing (Figure 4.5). Explanations for this are the reduced discharges from Sellafield over the last 10 years and the physical decay of  $^{90}\text{Sr}$  from global fallout.

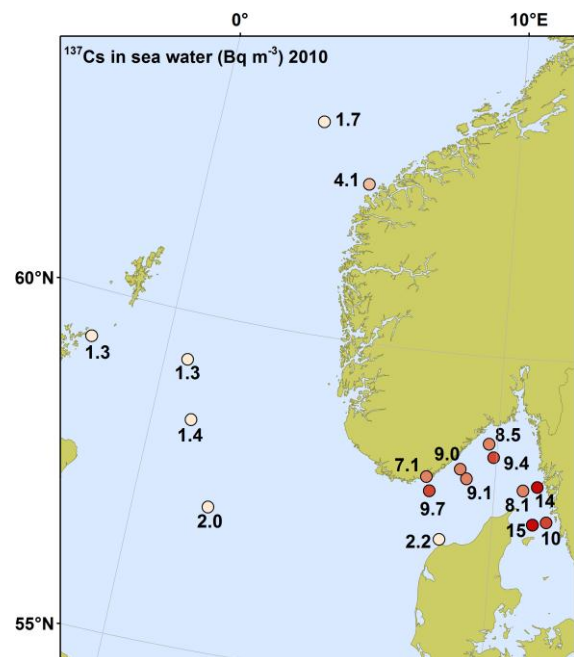


**Figure 4.5.** Activity concentration ( $Bq\ m^{-3}$ ) of  $^{90}Sr$  in sea water at Hillesøy (2002-2010) and Grense Jakobselv (2004-2010).

### 4.3 Caesium-137 in sea water and sediment

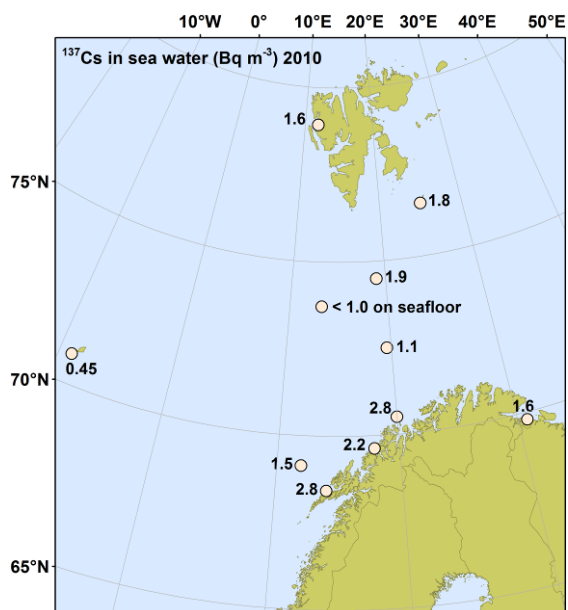
Caesium-137 is a fission product with a half-life of 30 years. The main sources of  $^{137}Cs$  in the North Sea and the Skagerrak are fallout from atmospheric nuclear weapons tests in the 1950s and 1960s, outflowing water from the Baltic Sea, and  $^{137}Cs$  remobilised from Irish Sea sediments. Runoff 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 North Sea and the Skagerrak in 2010 are shown in Figure 4.6. The activity concentration in surface water sampled in the North Sea and Skagerrak ranged from 1.3 to 15  $Bq\ m^{-3}$ , where the highest levels were found in the Skagerrak.



**Figure 4.6.** Activity concentration ( $Bq\ m^{-3}$ ) of  $^{137}Cs$  in sea water samples (surface water) collected in the North Sea and the Skagerrak in 2010.





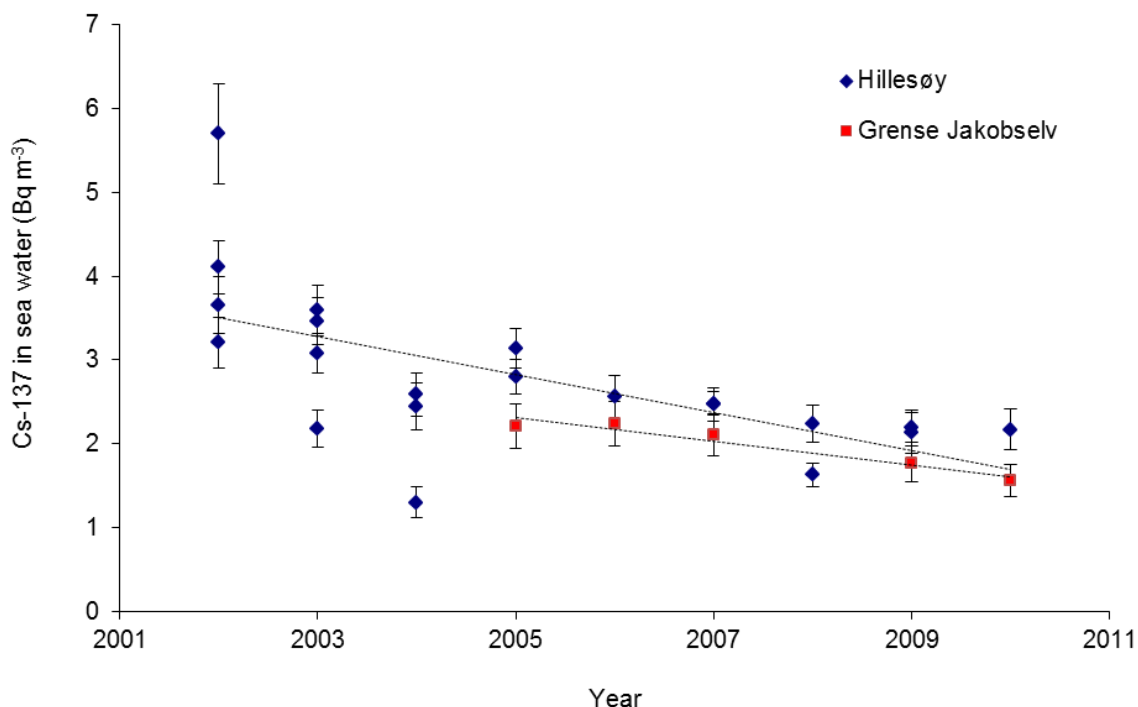
**Figure 4.7.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{137}\text{Cs}$  in sea water samples (surface water) collected in the Norwegian Sea and the Barents Sea in 2010.

The activity concentration of  $^{137}\text{Cs}$  in the Barents Sea in 2010 ranged from  $<1.0$  to  $2.8 \text{ Bq m}^{-3}$  (Figure 4.7). This is similar to or lower than the observed levels in 2002, 2003, 2005,

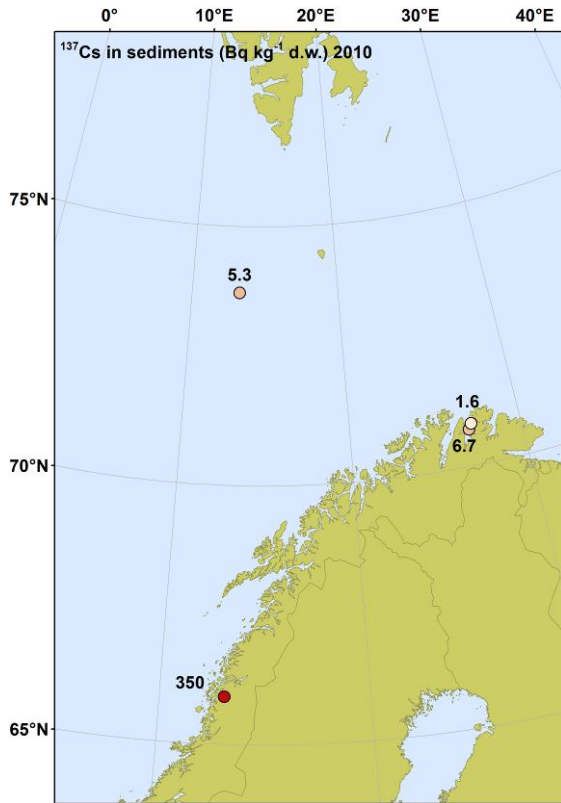
2006 and 2009 (NRPA, 2004; NRPA, 2005; NRPA, 2007; NRPA 2008 and NRPA 2011).

Data from Hillesøy (2002-2010) and Grense Jakobselv (2005-2010) (Figure 4.8) show that the levels of  $^{137}\text{Cs}$  in the Norwegian coastal current are slowly decreasing. The effective half-life of  $^{137}\text{Cs}$  in the Baltic Sea, which is one of the main sources of  $^{137}\text{Cs}$  in this region, has been estimated to 10 years (Ikäheimonen *et al.* 2009).

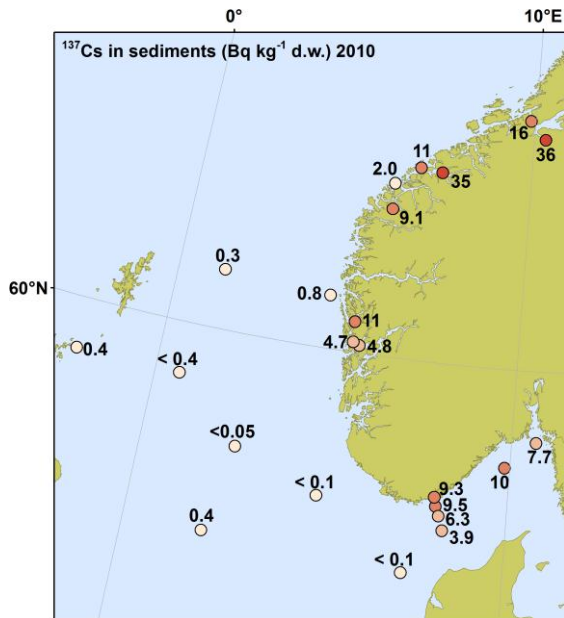
Caesium-137 has also been analysed in surface sediments (upper 2 cm layer) from the Barents Sea, the North Sea, and selected fjords. The results are presented in Figure 4.9 and Figure 4.10 and range from  $< 0.1$  to  $10 \text{ Bq kg}^{-1}$  (d.w.) in the Barents Sea and the North Sea. The activity concentration  $^{137}\text{Cs}$  in sediments sampled in the fjords ranged from 1.6 to  $350 \text{ Bq kg}^{-1}$  (d.w.). The highest activity concentrations were found in samples from the Vefsnfjord.



**Figure 4.8.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{137}\text{Cs}$  in sea water at Hillesøy and Grense Jakobselv in the period 2002-2010.



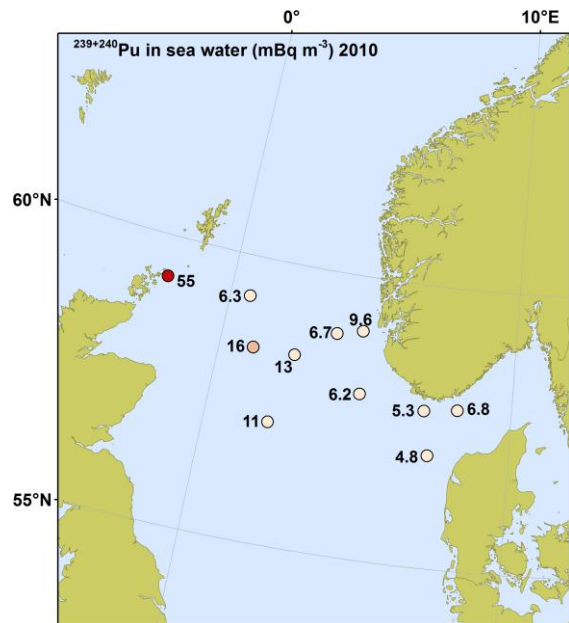
**Figure 4.9.** Activity concentration ( $\text{Bq kg}^{-1}$  d.w.) of  $^{137}\text{Cs}$  in surface sediment from the Barents Sea and selected fjords in 2010. The sample south west of Bjørnøya has been collected close to the position of the sunken submarine Komsomolets.



**Figure 4.10.** Activity concentration ( $\text{Bq kg}^{-1}$  d.w.) of  $^{137}\text{Cs}$  in surface sediment from the North Sea and selected fjords in 2010.

#### 4.4 Plutonium-239+240 in sea water

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

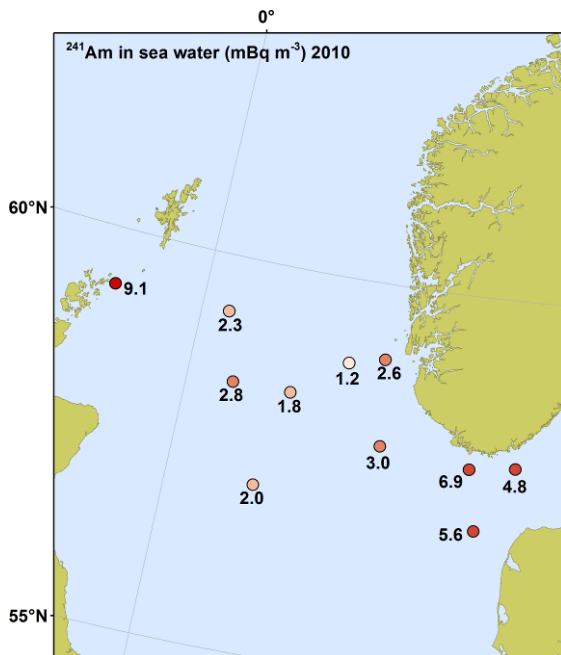


**Figure 4.11.** Activity concentration ( $\text{mBq m}^{-3}$ ) of  $^{239+240}\text{Pu}$  in surface water samples collected in the North Sea in 2010.

Observed levels in the North Sea in 2010 are presented in Figure 4.11 and range from 4.8 to 55  $\text{mBq m}^{-3}$ . The highest activity concentration was found in the western part of the North Sea, off the coast of Scotland. The levels of  $^{239+240}\text{Pu}$  in the North Sea in 2010 are similar to the levels observed in 2001, 2004 and 2007 (NRPA, 2003; NRPA, 2006; NRPA 2009).

## 4.5 Americium-241 in sea water

Americium-241 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  $^{239}\text{Pu}$ . Americium-241 is finally formed by beta-decay of  $^{241}\text{Pu}$  ( $T_{1/2} = 14.35$  y). Main sources of  $^{241}\text{Am}$  in the environment are fallout of  $^{241}\text{Pu}$  from nuclear weapon tests in the 1950s and 1960s and the discharge of  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  from reprocessing plants.



**Figure 4.12.** Activity concentration ( $\text{mBq m}^{-3}$ ) of  $^{241}\text{Am}$  in sea water from the North Sea in 2010.

Measured activity concentrations of  $^{241}\text{Am}$  in the North Sea in 2010 are presented in Figure 4.12 and range from 1.2 to  $9.1 \text{ mBq m}^{-3}$ . The observed levels of  $^{241}\text{Am}$  are similar to the levels observed in 2002, 2005 and 2007 (NRPA, 2003; NRPA, 2006; NRPA, 2009).

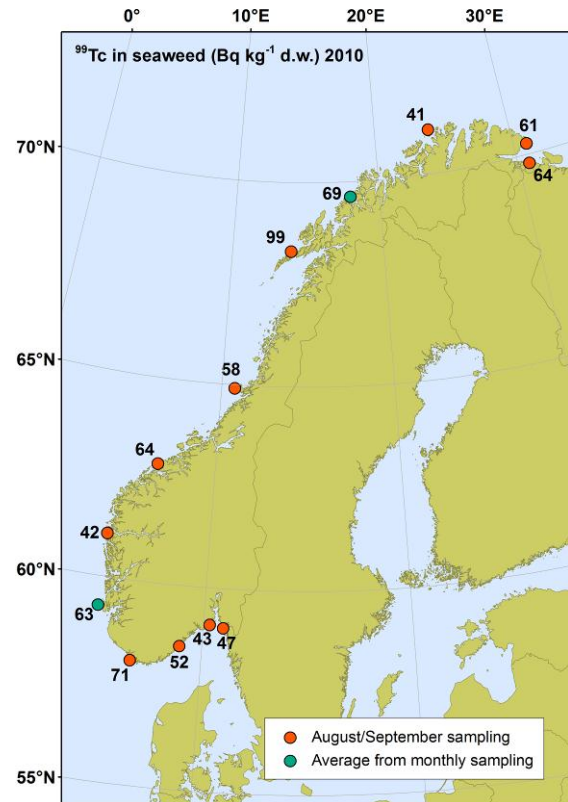
## 5 Radioactivity in biota

### 5.1 Technetium-99 in seaweed

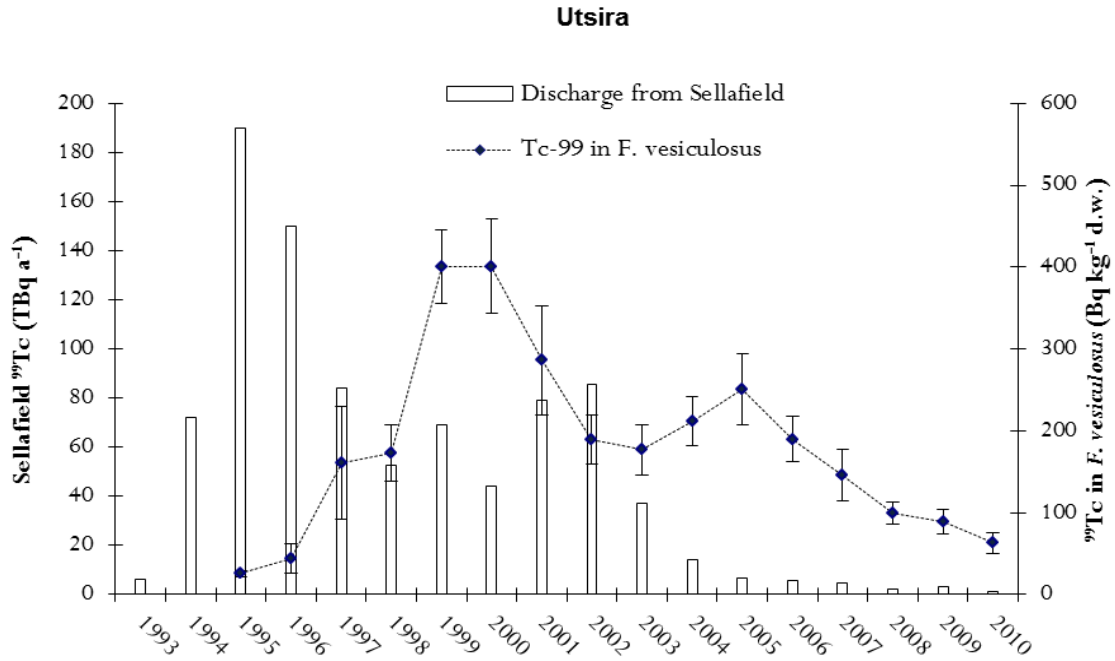


**Figure 5.1.** Brown seaweed (*Fucus vesiculosus*) (Photo: NRPA).

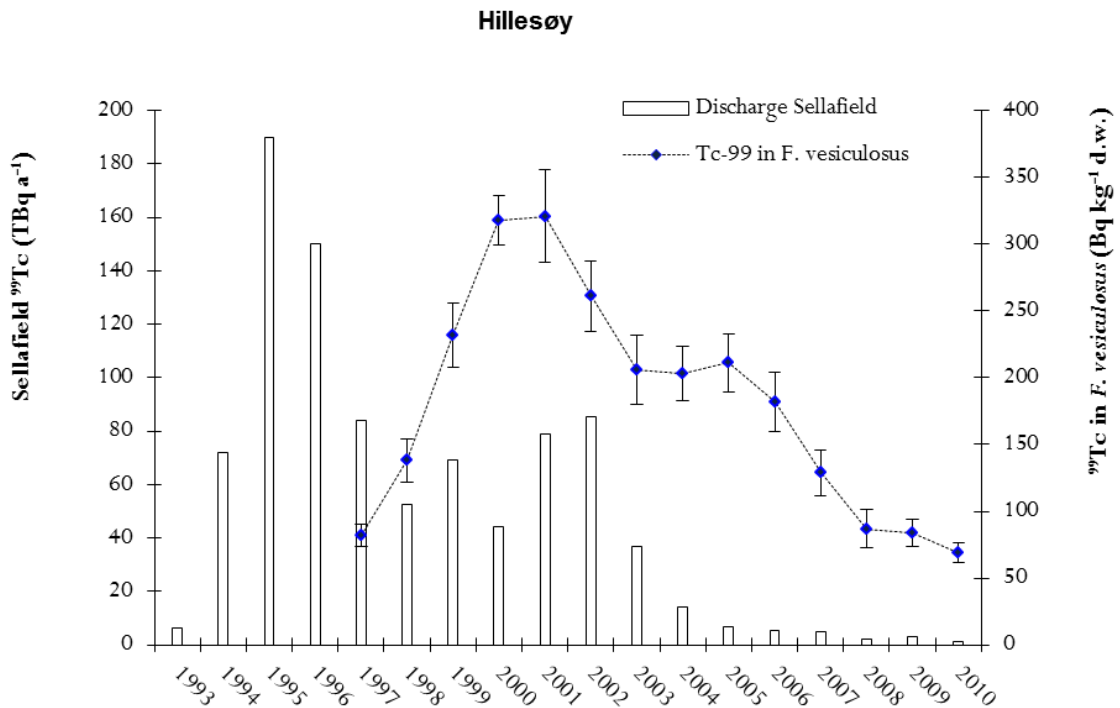
Seaweed is a useful bioindicator for  $^{99}\text{Tc}$  in the marine environment. It has a high ability to concentrate  $^{99}\text{Tc}$  from sea water and is easily accessible in most coastal areas. During 2010, seaweed (*Fucus vesiculosus*, Figure 5.1) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for  $^{99}\text{Tc}$ . At two locations, Hillesøy and Utsira, sampling was performed monthly. 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 41 to 99  $\text{Bq kg}^{-1}$  (d.w.), where the highest activity concentration (104  $\text{Bq kg}^{-1}$  d.w.) was found in one of the monthly samples collected at Utsira. Compared with the results from 1999-2001 (Rudjord *et al.*, 2001; Gäfvert *et al.*, 2003), the levels of  $^{99}\text{Tc}$  have decreased at most sampling sites, due to the reduced discharge of  $^{99}\text{Tc}$  from Sellafield. The trend can also be seen in Figures 5.3 and 5.4, which show the annual average activity concentration of  $^{99}\text{Tc}$  in *Fucus vesiculosus* at Utsira and Hillesøy, together with the annual discharge of  $^{99}\text{Tc}$  from Sellafield.



**Figure 5.2.** Levels of  $^{99}\text{Tc}$  in *Fucus vesiculosus* sampled along the Norwegian coastline in 2010.



**Figure 5.3.** Annual liquid discharge of  $^{99}\text{Tc}$  from Sellafield (primary axis) and annual average (with 95 % confidence limits)  $^{99}\text{Tc}$  activity concentration in brown algae (*Fucus vesiculosus*) sampled at Utsira (data provided by IFE) in the period 1995-2010 (secondary axis).

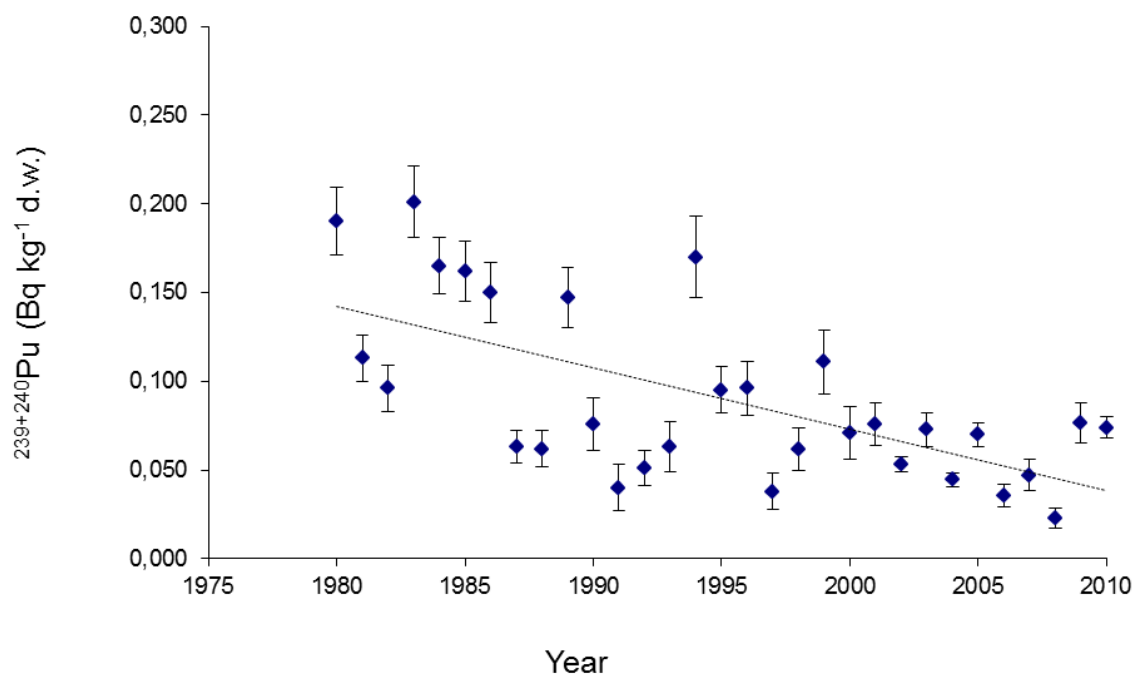


**Figure 5.4.** Annual liquid discharge of  $^{99}\text{Tc}$  from Sellafield (primary axis) and annual average (with 95 % confidence limits)  $^{99}\text{Tc}$  activity concentration in brown algae (*Fucus vesiculosus*) sampled at Hillesøy in the period 1997-2010 (secondary axis).

## 5.2 Plutonium-239+240 in seaweed

*Fucus vesiculosus* has been collected and analysed for  $^{239+240}\text{Pu}$  at Utsira since 1980. The results from the period 1980 to 2010 are presented in Figure 5.5. The activity

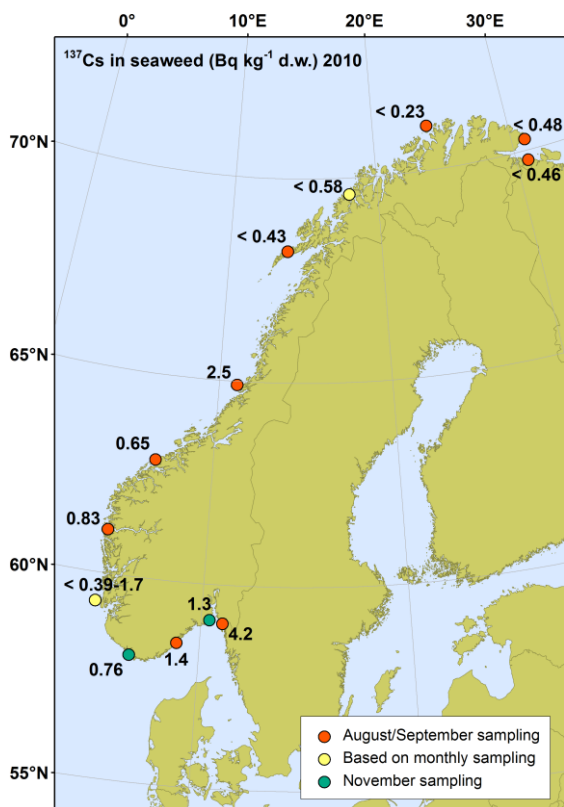
concentrations in these samples were in the range of 23 to 201  $\text{mBq kg}^{-1}$ , with relatively large fluctuations from year to year. One can see a slowly decreasing trend in the activity concentration of  $^{239+240}\text{Pu}$  in the seaweed samples collected in the period from 1980 to 2010.



**Figure 5.5.**  $^{239+240}\text{Pu}$  levels ( $\text{Bq kg}^{-1}$  d.w.) in *Fucus vesiculosus* at Utsira in the period 1980 to 2010 (data provided by IFE).

## 5.3 Caesium-137 in seaweed

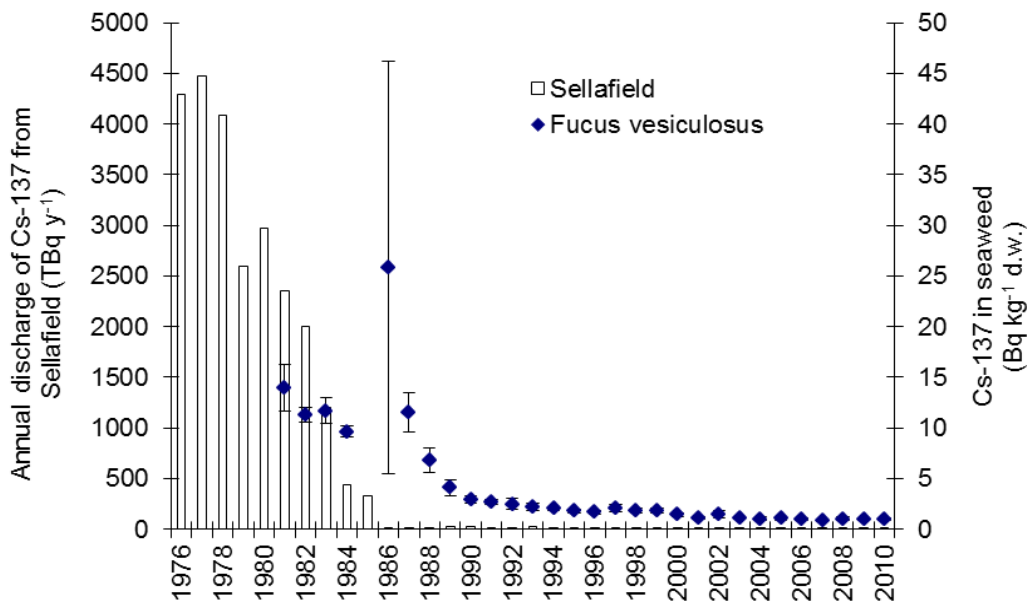
*Fucus vesiculosus* has also been widely used as a bioindicator for  $^{137}\text{Cs}$ . The accumulation of  $^{137}\text{Cs}$  in brown algae is, however, not as pronounced as for  $^{99}\text{Tc}$ . The uptake of  $^{137}\text{Cs}$  also depends on the salinity of the surrounding sea water, with higher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



**Figure 5.6.** Levels of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  d.w.) in *Fucus vesiculosus* sampled along the Norwegian coastline in 2010.

In 2010, samples of *Fucus vesiculosus* from the permanent coastal stations were analysed with respect to  $^{137}\text{Cs}$ . The results are presented in Figure 5.6, and ranged from  $< 0.2 \text{ Bq kg}^{-1}$

(d.w.) in the north to  $4.2 \text{ Bq kg}^{-1}$  (d.w.) at Hvaler. The peak in the  $^{137}\text{Cs}$  levels in seaweed collected at Vikna can be explained by run-off of  $^{137}\text{Cs}$  from land, since this area was affected by fallout from the Chernobyl accident in 1986. 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  $^{137}\text{Cs}$  in the sea water is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident. Comparing those results with the levels found at the coastal sampling stations in the period 2000-2007 (Gäfvert *et al.*, 2003; NRPA, 2004, NRPA 2005, NRPA 2006, NRPA 2007, NRPA 2008, NRPA, 2009 and NRPA, 2011), one can see that the activity concentration of  $^{137}\text{Cs}$  in *Fucus vesiculosus* has been relatively stable in recent years. However, data from monthly sampling at Utsira (Figure 5.7) show that the activity concentration of  $^{137}\text{Cs}$  is slowly decreasing in seaweed. This is in agreement with the reported temporal trend of  $^{137}\text{Cs}$  in Baltic Sea sea water (Ikäheimonen *et al.* 2009).

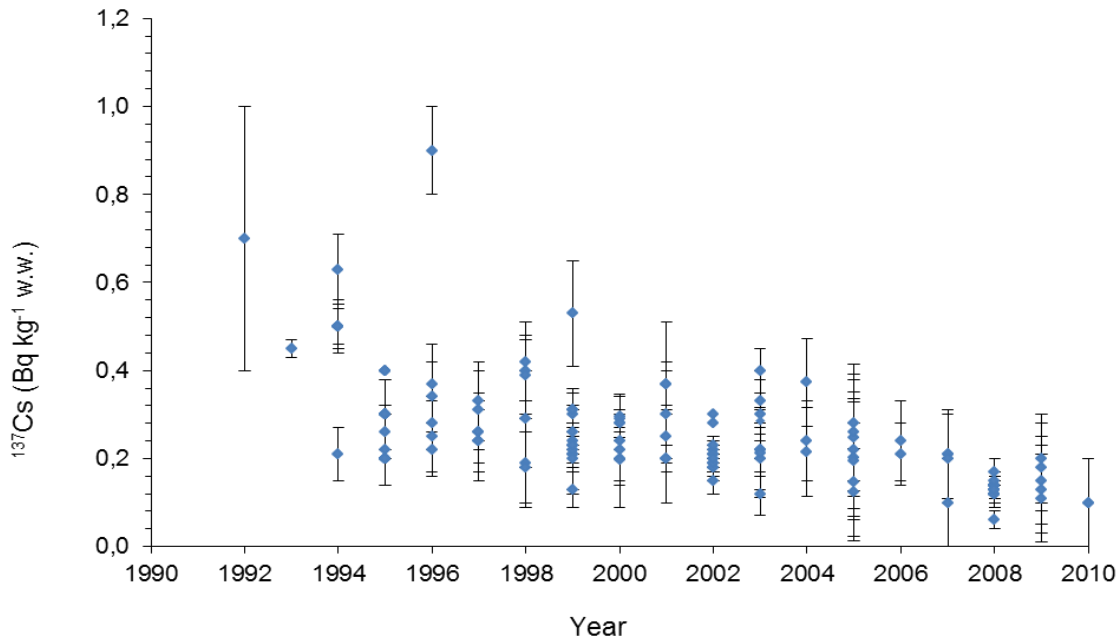


**Figure 5.7.** Average activity concentration ( $\text{Bq kg}^{-1}$  d.w.) from monthly sampling in seaweed (*Fucus vesiculosus*) from Utsira in the period 1980-2010 (data from IFE).

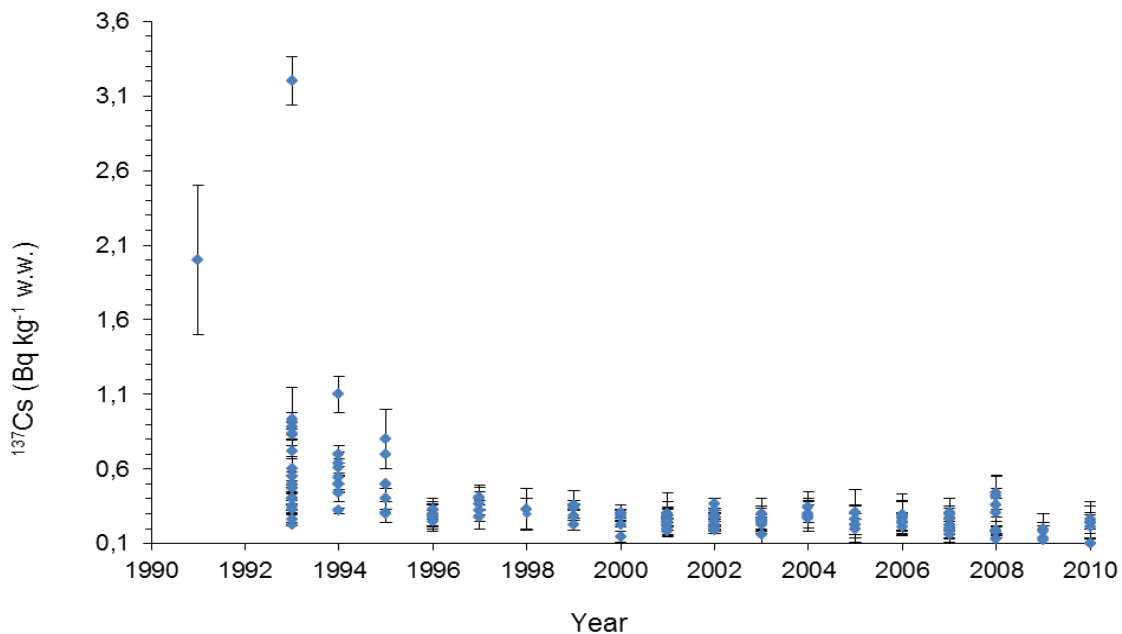
## 5.4 Caesium-137 in fish and crustaceans

Samples of cod from the Barents Sea have been analysed for  $^{137}\text{Cs}$  since the early 1990s. In Figures 5.8 and 5.9 the activity concentration of  $^{137}\text{Cs}$  in muscle tissue from

cod caught in two areas of the Barents Sea is shown. Almost all samples from both locations are below  $1 \text{ Bq kg}^{-1}$  (w.w.), and in recent years below  $0.5 \text{ Bq kg}^{-1}$  (w.w.). The results show a slightly decreasing trend in the period 1992-2010.



**Figure 5.8.** Activity concentration of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  w.w.) in cod from the Barents Sea (the area around Bjørnøya) sampled in the period 1992 to 2010.



**Figure 5.9.** Activity concentration of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  w.w.) in cod muscle tissue sampled in the period 1991 to 2010 off the coast of Finnmark.

Caesium-137 levels in fish species and crustaceans caught in the Barents Sea, the Norwegian Sea and the North Sea in 2010 are

shown in Table 5.1. All obtained results were below  $0.5 \text{ Bq kg}^{-1}$  (w.w.). Technetium-99 has been analysed in lobster (*Hommarus*

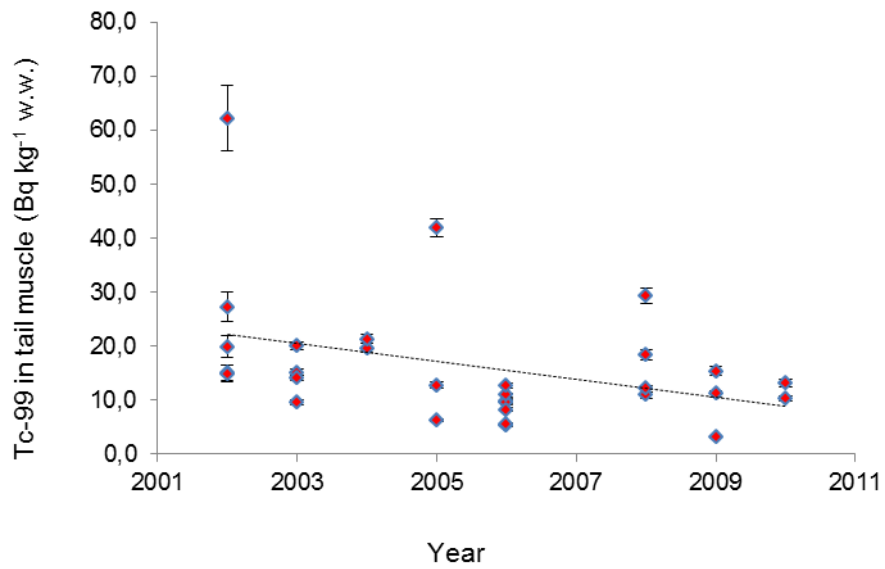


*gammarus*) tail muscle from Værlandet since 2002. The results for female lobsters and male lobsters are presented in Figures 5.10 and 5.11. In the period from 2002 to 2010 the levels of

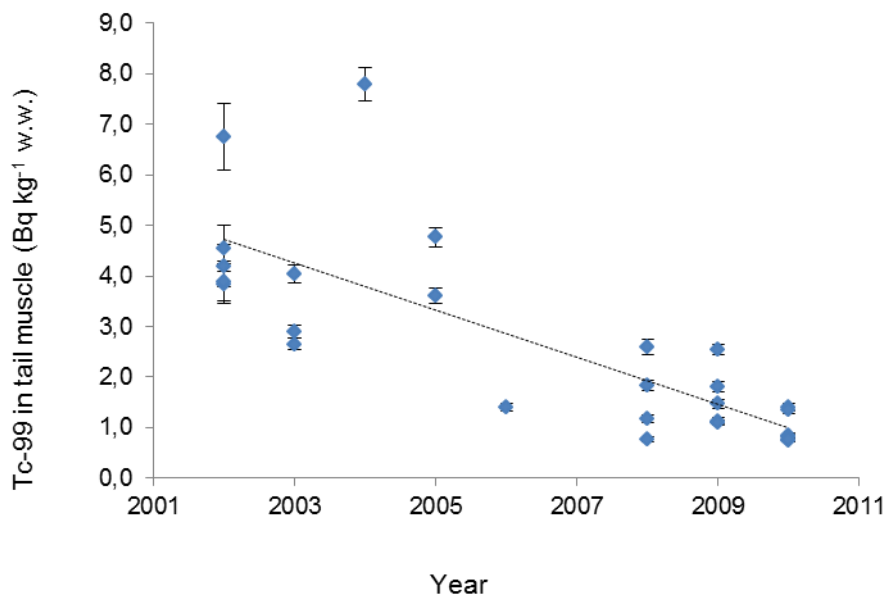
<sup>99</sup>Tc in lobster have decreased due to the reduced discharge of <sup>99</sup>Tc from Sellafeld.

**Table 5.1.** Activity concentrations (Bq kg<sup>-1</sup> w.w.) of <sup>137</sup>Cs in fish and crustaceans caught in the Barents, Norwegian and North Seas and in the Skagerrak in 2010.

Species	Location	No. of samples (total no. of fish muscles)	<sup>137</sup> Cs in muscle tissue (Bq kg <sup>-1</sup> w.w.)
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	2 (200)	0.10 ± 0.10 and 0.10 ± 0.10
Cod ( <i>Gadus morhua</i> L.)	Coastal waters of Finnmark and Troms	7 (325)	0.10 ± 0.10 to 0.26 ± 0.12
Norway pout ( <i>Triopterus esmarkii</i> )		1 (25)	0.06 ± 0.02
Norway redfish ( <i>Sebastes viviparus</i> )		1 (25)	0.15 ± 0.13
Farmed salmon ( <i>Salmo salar</i> )		1 (25)	0.22 ± 0.09
Atlantic herring ( <i>Clupea harengus</i> )		Norwegian Sea	1 (25)
Cod ( <i>Gadus morhua</i> L.)	Coastal waters of Nordland	5 (125)	0.19 ± 0.06 to 0.34 ± 0.10
Saithe ( <i>Pollachius virens</i> )		5 (125)	0.18 ± 0.08 to 0.26 ± 0.13
Farmed salmon ( <i>Salmo salar</i> )		1 (25)	0.17 ± 0.10
Cod ( <i>Gadus morhua</i> L.)	Coastal waters of Nord- Trøndelag, Sør- Trøndelag, and Møre og Romsdal	2 (50)	0.28 ± 0.08 and 0.29 ± 0.11
Saithe ( <i>Pollachius virens</i> )		2 (50)	0.28 ± 0.08 and 0.30 ± 0.10
Atlantic herring ( <i>Clupea harengus</i> )		1 (21)	0.26 ± 0.09
Blue whiting ( <i>Micromesistius poutassou</i> )		1 (10)	0.13 ± 0.09
Greater argentine ( <i>Argentina silus</i> )		1 (7)	<0.10
Roundnose grenadier ( <i>Coryphaenoides rupestris</i> Gunnerus)		1 (25)	0.06 ± 0.03
Cod ( <i>Gadus morhua</i> L.)	The North Sea including coastal waters from the southern and eastern part of Norway	2 (53)	0.20 ± 0.10 and 0.20 ± 0.10
Haddock ( <i>Melanogrammus aeglefinus</i> )		2 (49)	<0.10 and 0.16 ± 0.07
Saithe ( <i>Pollachius virens</i> )		2 (50)	0.23 ± 0.02 and 0.30 ± 0.07
Atlantic herring ( <i>Clupea harengus</i> )		1(25)	0.50 ± 0.10
Blue whiting ( <i>Micromesistius poutassou</i> )		1(25)	0.24 ± 0.06
Mackerel ( <i>Scomber scombrus</i> )		2 (50)	0.14 ± 0.05 and 0.17 ± 0.04
Norway pout ( <i>Triopterus esmarkii</i> )		2 (50)	0.05 ± 0.10 and 0.08 ± 0.10
European plaice ( <i>Pleuronectes platessa</i> )		1(13)	0.14 ± 0.07
Shrimp ( <i>Pandalus borealis</i> )		1 kilogram	0.06 ± 0.03



**Figure 5.10.** Technetium-99 concentration in female lobster (*Hommarus gammarus*) from Værlandet (2002-2010).



**Figure 5.11.** Technetium-99 concentration in male lobster (*Hommarus gammarus*) from Værlandet (2002-2010).

## 6 Summary and conclusions

In 2010, samples of sea water, sediment, and biota were collected in the Barents Sea, the North 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.

### 6.1 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2010 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 0.03  $\mu\text{Sv}$ , which correspond to 3 % of the dose limit. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be 0.012  $\mu\text{Sv}$ , corresponding to 1.2 % 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 2010, the discharged activity of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  from the Norwegian oil and gas industry were reported to 486 GBq and 385 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  $^{137}\text{Cs}$  and plutonium from contaminated Irish Sea sediments act as secondary sources of

radionuclides to the Norwegian marine environment.

### 6.2 Radioactivity in sea water and sediment

In 2010, samples of sea water and sediment were collected mainly in the North Sea and analysed with respect to  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ , and  $^{241}\text{Am}$ . 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.

#### 6.2.1 Caesium-137 in sea water

Observed levels of  $^{137}\text{Cs}$  in surface water in the North Sea, the Skagerrak and the Barents Sea in 2010 ranged from 0.5 to 14  $\text{Bq m}^{-3}$ . The highest levels were found in brackish water in the Skagerrak. Samples from the Barents Sea in 2010 showed activity concentrations in the range of 0.5 to 2.8  $\text{Bq m}^{-3}$ . This is generally similar to or lower than the activity concentrations observed in the same area in 2009. Data from Hillesøy in the period 2002 to 2010 also show that the levels of  $^{137}\text{Cs}$  are slowly decreasing in the Norwegian coastal current.

#### 6.2.2 Technetium-99 in sea water

Samples of sea water were collected mainly in the North Sea and the Barents Sea. Levels of  $^{99}\text{Tc}$  in surface sea water ranged from 0.12  $\text{Bq m}^{-3}$  to 0.8  $\text{Bq m}^{-3}$ , where the highest concentrations were found in the North Sea off the coast of Scotland. The average activity concentration from quarterly sampling at Hillesøy in 2010 was  $0.31 \pm 0.07$ , which is significantly lower than the peak values observed in 1999/2000.

#### 6.2.3 Strontium-90 in sea water

The activity concentration in surface sea water sampled in the Barents Sea and in the Skagerrak in 2010 ranged from 0.7  $\text{Bq m}^{-3}$  to

2.8 Bq m<sup>-3</sup>. The highest concentrations were found off the southern coast of Norway in the Skagerrak. Data from Hillesøy in the period 2002 to 2010 indicates that the levels of <sup>90</sup>Sr are slowly decreasing in the Norwegian coastal current.

#### 6.2.4 Plutonium-239+240 and americium-241 in sea water

Observed levels of <sup>239+240</sup>Pu in the North Sea in 2010 ranged from 4.8 to 55 mBq m<sup>-3</sup>. The activity concentrations of <sup>239+240</sup>Pu are similar to those found in the same area in 2001, 2005 and 2007 and 2005. The activity concentration of <sup>241</sup>Am in sea water from the Barents Sea and the Skagerrak ranged from 1.2 to 9.1 mBq m<sup>-3</sup>. This is similar to what was observed in 2005 in the same area.

### 6.3 Radioactivity in biota

#### 6.3.1 Technetium-99 in seaweed

Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 41 to 99 Bq kg<sup>-1</sup> (d.w.). For most stations the levels were lower in 2010 compared to observed levels in the period 2002-2007. The levels in 2010 are significantly lower than the peak values observed in the period 1999-2001. Monthly sampling at Hillesøy and Utsira show that the levels of <sup>99</sup>Tc in seaweed have decreased since 2005 due to the reduced discharge of <sup>99</sup>Tc from Sellafield.

#### 6.3.2 Caesium-137 in seaweed

The activity concentration of <sup>137</sup>Cs in *Fucus vesiculosus* sampled at the coastal stations in 2010 was in the range of <0.2 to 4.2 Bq kg<sup>-1</sup> (d.w.), where the highest levels were found in the Skagerrak. Data from monthly sampling at Utsira indicates that the levels of <sup>137</sup>Cs in seaweed are slowly decreasing.

#### 6.3.3 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 <sup>137</sup>Cs in fish from Norwegian marine waters is generally low. All analysed samples were below 1.0 Bq kg<sup>-1</sup> (w.w.).

Samples of cod from two locations in the Barents Sea have been analysed for <sup>137</sup>Cs since the early 1990s. These results show a slightly decreasing trend of <sup>137</sup>Cs in this period.

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## 8 Appendix

### 8.1 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  $^{226}\text{Ra}$ ), while the other organisations are accredited for gamma spectrometric measurements of  $^{137}\text{Cs}$ . 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.

### 8.2 The Norwegian Radiation Protection Authority (NRPA)

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

#### 8.2.1 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 40 %. The resolution of the detectors, the full width at half maximum (FWHM) at 1332 keV, was less than 1.9 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 450°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  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  in the sediment samples ranged from approximately 0.3-2 Bq  $\text{kg}^{-1}$  (d.w.) Uncertainties in the analysis include uncertainties from counting statistics, calibration and sample preparation and are normally in the range 5 % to 20 %.

#### 8.2.2 Determination of $^{239+240}\text{Pu}$ and $^{241}\text{Am}$

The concentrations of  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  were measured in samples of 200 litres of sea water or 10-20 g of sediment.  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  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 ( $^{241}\text{Am}$ ). Chemical yields obtained from the  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  yield monitor were in the range 40 % to 80 %.

### 8.3 The Institute of Marine Research (IMR)

#### 8.3.1 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  $^{137}\text{Cs}$  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.

### 8.3.2 Determination of $^{99}\text{Tc}$

Before the activity concentration of  $^{99}\text{Tc}$  can be determined, a radiochemical separation is necessary. Briefly, rhenium is added as a yield monitor in the form of  $\text{KReO}_4$ , and a preliminary extraction of  $^{99}\text{Tc}$  (and Re) based on anion-exchange separation is performed. After iron hydroxide scavenging,  $^{99}\text{Tc}$  and Re are further extracted by a second anion-exchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts are isolated.  $^{99}\text{Tc}$  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.

## 8.4 Institute for Energy Technology (IFE)

### 8.4.1 Determination of $^{99}\text{Tc}$

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to  $^{99}\text{Tc}$  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 tributyl-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  $^{99\text{m}}\text{Tc}$  was used as a radiochemical yield determinant. After decay of the yield determinant,  $^{99}\text{Tc}$  was measured with an anti-coincidence-shielded GM counter (Holm *et al.*, 1984).

### 8.4.2 Determination of $^{90}\text{Sr}$

For  $^{90}\text{Sr}$ , the standard method using fuming nitric acid was used. The recovery of  $^{90}\text{Sr}$  in the analytical process was monitored by adding  $^{85}\text{Sr}$  as a yield determinant, and the recovery of the daughter nuclide  $^{90}\text{Y}$  was determined by titration with EDTA (Varskog *et al.*, 1997).

Finally,  $^{90}\text{Y}$  was measured with an anti-coincidence-shielded GM counter.

### 8.4.3 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.

### 8.4.4 Determination of $^{239+240}\text{Pu}$

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  $\text{CeF}_3$  on a filter and counted using alpha spectrometry.











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