

Radioactivity in the Marine Environment 2003

Results from the Norwegian Marine Monitoring Programme (RAME)



INSTITUTE OF MARINE RESEARCH



Statens
strålevern

**Norwegian Radiation
Protection Authority**

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Abstract:

This report presents results of monitoring of radioactivity in water, sediment, fish, seaweed and crustaceans collected along the Norwegian coast and in the Norwegian Sea/ Barents Sea in 2003. An overview of discharges from Norwegian sources and data concerning the long-range transport of radionuclides from European nuclear facilities is included.

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Resymé:

Rapporten inneholder resultater fra overvåkingen av radioaktivitet i sjøvann, sedimenter, fisk og skalldyr i 2003 langs norskekysten, i Norskehavet/ Barentshavet. En oversikt over utslipp fra norske kilder og utslippsdata fra europeiske nukleære anlegg som er relevante for langtransport av radioaktivitet til norske havområder er inkludert i rapporten.

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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 recent years. 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 slightly, it has continued at a high level up to 2003. The discharge of ^{99}Tc was substantially reduced from 2004, but it will take 3-4 years before this is observable in Norwegian waters. There has been much public concern about the consequences of such kinds of release, as the radionuclides discharged to the Irish Sea are transported by ocean currents via the North Sea into the Norwegian coastal current and to the Barents Sea. 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 recently received special attention.

In Norway there are currently two monitoring programmes concerned with radioactivity in the marine environment, both coordinated by the Norwegian Radiation Protection Authority (NRPA). One is funded by the Ministry of the Environment and focuses on monitoring of radioactivity in the marine environment both in coastal areas and in the open seas, the other by the Ministry of Fisheries which focuses on monitoring of radioactivity in commercially important fish species. Results of both these programmes are presented in this report. In addition, data from the Food Control Authority's monitoring programme, which is concerned with radioactivity in marine fish, have been included.

The marine monitoring programmes include the compilation of discharge data from Norwegian sources, in addition to the collection of discharge data relevant for the long-range transport of radionuclides from various sources. Liquid discharge data for 2003 from nuclear installations and recent trends in such discharges are summarised in Chapter 2.

During 2003, samples for monitoring radioactivity in the marine environment were collected in the Norwegian Sea/Barents Sea, in selected fjords and at coastal sampling stations, including the islands of Hopen, Bjørnøya and

Jan Mayen. 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 are 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

This chapter provides updated information and an overview of radionuclide discharges and other sources that are relevant to the Norwegian marine environment. The collection and updating of this information is an integral part of the marine monitoring programme RAME (Radioactivity in the Marine Environment), the main purpose of which is to present recent trends in radionuclide discharges and other sources of radioactivity in the marine environment. Information on discharges from Norwegian sources and on long-range transport of radionuclides from various distant sources is included. Anthropogenic radionuclides are discharged from the nuclear facilities of the Institute for Energy Technology, IFE Kjeller and IFE Halden. Discharges from these facilities are authorised by the NRPA. Unsealed radioactive materials used in medicine and science will primarily be released to the marine environment via sewage treatment plants. The utilisation of such unsealed sources is regulated through guidelines issued by the NRPA.

In recent years, increased attention has been devoted to releases containing elevated levels of naturally occurring radionuclides. Such releases are the result of offshore oil production and, probably, leaching of mine tailings.

The discharge of radionuclides from Norwegian sources is only detectable in the local environment, near the discharge point. The long-range transport of radionuclides originating from fallout from atmospheric nuclear weapons tests (conducted mainly in the 1950s and 1960s), reprocessing of nuclear fuel and from the Chernobyl accident in 1986 are still the main contributors to the general levels of anthropogenic radionuclides found in Norwegian waters.

2.1 Discharges of anthropogenic radionuclides from Norwegian sources

2.1.1 IFE Kjeller and Amersham Health

IFE 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 River Nitelva 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 River Glomma empties into the Oslofjord at the city of Fredrikstad.

A plant manufacturing radiopharmaceutical products, managed by the private company Amersham Health, operates in close collaboration with IFE Kjeller. Authorisation for IFE Kjeller discharges also includes the discharge from this production facility.

Table 2.1. Liquid discharges (MBq) from IFE Kjeller (IFE, 2004a; IFE, 2003a; IFE 2002a; IFE, 2001a; IFE 2000a), 1999-2003 (including Amersham Health).

| Nuclide | 1999 | 2000 | 2001 | 2002 | 2003 |
|-----------------------|----------------------|---------------------|---------------------|---------------------|---------------------|
| ¹³⁷ Cs | 30 | 6.7 | 25.7 | 25 | 22 |
| ¹³⁴ Cs | 1.9 | 0.4 | 2.5 | 2.3 | 1.2 |
| ¹³¹ I | 1.35·10 ³ | 6.3 | 107 | 6.0 | 15 |
| ¹²⁵ I | 182 | 91 | 310 | 350 | 540 |
| ⁶⁵ Zn | 0.8 | 0.9 | 3.8 | <0.4 | 0.27 |
| ⁶⁰ Co | 6.9 | 8.7 | 74 | 60 | 55 |
| ³⁵ S | 18.5 | 142 | - | - | - |
| ⁹⁰ Sr | 161 | 6.5 | 1.2 | 0.6 | 0.33 |
| ²³⁹⁺²⁴⁰ Pu | 0.02 | 0.2 | 0.04 | 0.04 | 0.034 |
| ³ H | 0.9·10 ⁵ | 4.0·10 ⁵ | 1.5·10 ⁶ | 2.4·10 ⁶ | 2.8·10 ⁶ |

The discharge limit authorised by the NRPA is based on the annual dose to any member of a critical group of the population along the River Nitelva, and shall not exceed 1 μSv . Each year, IFE Kjeller reports discharge data, the results of their environmental monitoring programme and calculations of effective doses resulting from discharges, to the NRPA. The reported discharges for the period 1999-2003 are summarised in Table 2.1.

Table 2.2. Effective doses (μSv) to any member of the critical group from river water exposure pathways as reported by IFE Kjeller (IFE 2004a; IFE, 2003a; IFE 2002a; IFE, 2001a; IFE 2000a).

| 1999 | 2000 | 2001 | 2002 | 2003 |
|------|------|------|------|------|
| 0.53 | 0.04 | 0.21 | 0.18 | 0.17 |

Effective doses have been calculated by IFE for an individual in the hypothetical critical group exposed through the annual consumption of 20 kg of fish from the river and 100 hours per year presence on the riverbanks. In 2003, the calculated effective dose to this critical group was 0.17 μSv , corresponding to 17 % of the annual dose limit of 1 μSv . The effective dose to the critical group varies, as can be seen in Table 2.2, corresponding to between 4 and 53 % of the dose limit, with an average of 23 % for the years 1999-2003.

2.1.2 IFE Halden

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 River Tista (which empties into the Iddefjord). The discharge limit for liquid waste to the marine environment is based on a dose limit of 1 μSv per year to a hypothetical critical group, and the actual annual discharge is

typically 5-10 % of the authorised limit. Liquid discharges in the period 1999-2003, and the corresponding doses to the critical group are presented in Tables 2.3 and 2.4.

Table 2.3. Liquid discharge (MBq) from IFE Halden, 1999 - 2003 (IFE, 2004b; IFE, 2003b; IFE 2002b; IFE 2001b; IFE 2000b).

| Nuclide | 1999 | 2000 | 2001 | 2002 | 2003 |
|--------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| ¹³⁷ Cs | 530 | 289 | 58 | 110 | 130 |
| ¹³⁴ Cs | 29 | 14.5 | 2 | 18 | 8.8 |
| ¹³¹ I | 6.5 | 0.4 | 0.04 | 0.9 | 3.6 |
| ⁵⁸ Co | 19 | 13.9 | 49 | 5.1 | 3.3 |
| ⁹⁵ Zr | 14 | 21.4 | 16 | 5.4 | 3.3 |
| ⁶⁰ Co | 380 | 530 | 440 | 88 | 62 |
| ¹²⁵ Sb | 39 | 116 | 130 | 18 | 0.06 |
| ¹²⁴ Sb | - | 1.2 | - | 0.5 | - |
| ⁹⁵ Nb | 60 | 38 | 40 | 12 | 8.5 |
| ⁵⁴ Mn | 5.6 | 5 | 7 | 0.5 | 0.18 |
| ⁵¹ Cr | 240 | 610 | 290 | 150 | 130 |
| ^{110m} Ag | - | 3.4 | 0.5 | 0.005 | 0.17 |
| ¹⁴⁴ Ce | 8.3 | 15.5 | 14 | 5.7 | 3.2 |
| ³ H | 6.7·10 ⁵ | 5.2·10 ⁵ | 2.4·10 ⁵ | 1.1·10 ⁶ | 2.7·10 ⁵ |

The calculation of the effective dose to the critical group is based on the following assumptions:

- An annual consumption of 30 kg of fish from the Iddefjord,
- 200 hours per year exposure on the shore of the fjord,
- 50 hours per year bathing in the fjord, and
- 1000 hours per year boating on the fjord.

Table 2.4. Effective doses (μSv) to the critical group from marine exposure pathways as reported by IFE Halden, 1999 - 2003 (IFE, 2004b; IFE, 2003b; IFE 2002b; IFE 2001b; IFE 2000b).

| 1999 | 2000 | 2001 | 2002 | 2003 |
|------|------|------|------|-------|
| 0.08 | 0.11 | 0.09 | 0.02 | 0.014 |

2.2 Unsealed radioactive substances in medical research and industry

Unsealed radioactive substances are used in hospitals, research laboratories and various industrial activities. According to regulations from 1981, laboratories etc. handling unsealed radioactive substances must be authorised by the NRPA. With this authorisation, laboratories and hospitals are allowed to discharge activity into the sewage system according to predefined limits. On January 1, 2004, new regulations came into force, describing requirements for handling and discharge of unsealed radioactive sources. At the moment all discharges require authorisation.

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.

In some tracer experiments in the offshore oil industry, tritium (^3H) is injected into rock formations or drilled wells, a fraction of which is recovered. Water-based material is discharged to the sea while oil-containing mud must be stored. However, for the consequence assessments required by the NRPA, it is conservatively assumed that all the ^3H is discharged to the open sea. The release of radionuclides used in research laboratories is in general considered less important, partly because only small quantities of activity are used and partly because some of the waste is collected and sent to the IFE (Kjeller) for storage as radioactive waste. Furthermore, most of these radionuclides have short half-lives and the activity is rapidly reduced during storage. Exact data on the nuclides and amounts discharged from the above sources are not available. However, the manufacturers of unsealed radioactive sources are required to report their sales to NRPA monthly, listing customer, type of radionuclide and quantity (activity). In Table 2.5 the sales of unsealed radioactive sources in Norway in 2003 are

listed. Regarding sales of $^{99\text{m}}\text{Tc}$, it is important to consider the short half-life of this radionuclide, as a large fraction will decay before it is even used.

Table 2.5. Unsealed radioactive sources (isotopes where total activity exceeds 1 GBq) sold in Norway in 2003.

| Nuclide | Half-life | Activity (GBq) |
|---|-----------|----------------|
| $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ | 66 h/6 h | 25 784 |
| ^{131}I | 8 d | 1 876 |
| ^{111}In | 2.8 d | 28.3 |
| ^{51}Cr | 27.7 d | 6.5 |
| ^{67}Ga | 3.3 d | 1.0 |
| ^{133}Xe | 5.3 d | 300 |
| ^{125}I | 60 d | 10.4 |
| ^{32}P | 14.3 d | 31.3 |
| ^{33}P | 25.4 d | 4.4 |
| ^3H | 12.3 y | 16 090 |
| ^{35}S | 87 d | 44.7 |
| ^{18}F | 110 min | 171 |
| ^{123}I | 13.1 h | 54.3 |
| ^{153}Sm | 46.7 h | 53.0 |
| ^{89}Sr | 50.6 d | 3.7 |
| ^{201}Tl | 73.1 h | 59.3 |
| ^{90}Y | 64.1 h | 18.4 |

2.3 Naturally occurring radioactive material

All minerals and rocks in the earth's crust contain small, but measurable amounts of naturally occurring radioactive material, often referred to as NORM. NORM includes long-lived isotopes (primordials), such as potassium-40 (^{40}K), as well as isotopes of the uranium decay chain and the thorium decay chain (^{238}U and ^{232}Th are the parent radionuclides of these two natural radioactive decay series). Naturally occurring radionuclides are released into the marine environment by natural processes such as erosion and leaching from marine sediments. The material is carried in river sediments, or dissolved in river water and finally reaches the sea.

In certain industrial or other technological processes naturally occurring radionuclides can be concentrated to levels orders of magnitude higher than those normally found in nature. In Norway, there are two main processes where technologically enhanced concentrations of naturally occurring radioactive material (TENORM) can be regarded as a discharge or leakage of radionuclides into the marine environment: oil and gas production in the North Sea and runoff from mine tailings.

2.3.1 TENORM/NORM in oil and gas production in Norway



Photo: NRPA

Two isotopes in the uranium and thorium series are the most important in relation to the

discharge of produced water and radioactive deposits in oil and gas production, namely ^{226}Ra and ^{228}Ra . The occurrence of natural radionuclides in North Sea oil and gas production was first detected in 1981, and enhanced levels of radioactivity are now found in the production systems of several North Sea oil fields (Strand *et al.*, 1997). The activity concentrations in scale deposited inside tubes and pipes range from background levels to several hundred Bq g^{-1} of ^{226}Ra (Smith, 1987). Doses to workers involved in handling contaminated equipment or waste are usually low, and the main problems related to radioactive deposits are waste disposal and the discharge of produced water.

2.3.2 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. 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 mBq per litre). A minor fraction, about 10 % of the total volume produced by Norwegian platforms, is currently reinjected into the reservoirs.

The concentration of radium in the produced water is assumed to 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.

In 2003 produced water from all 41 Norwegian platforms discharging produced water was collected monthly during 5 consecutive months and analysed for ^{226}Ra , ^{228}Ra and ^{210}Pb . The activity concentration of ^{226}Ra and ^{228}Ra ranged from below detection limit (about 1 Bq l^{-1}) up to 16 and 21 Bq l^{-1} , respectively. Average activity concentrations of ^{226}Ra and ^{228}Ra were found to be 3.3 and 2.8 Bq l^{-1} , respectively. The

total discharged activities of ^{226}Ra and ^{228}Ra with produced water were estimated to 440 GBq and 380 GBq, respectively. For most platforms the discharge of ^{226}Ra in 2003 was below a few tens of GBq, while a higher discharge was observed from the Troll B and C platforms (Fig. 2.1 and 2.2). The discharged activity from these platforms corresponded to about 40 % of the total discharged activity. For ^{210}Pb , all analysed samples of produced water, except one, were below the detection limit of about 1 Bq l^{-1} . Further details about discharges of natural radioactivity from the Norwegian oil and gas industry can be found in StrålevernRapport 2005:2 (NRPA, 2005).

There has also been some concern that the use of so called scale inhibitors and scale solvers may change the behavior of the radium isotopes released to the sea. These compounds are used to prevent scale to precipitate inside the production system and occasionally to dissolve scale in plugged wells and may form complexes with radium ions. If the radium isotopes are discharged in a complexed form they may be more available for marine organisms than if they were released as ions.

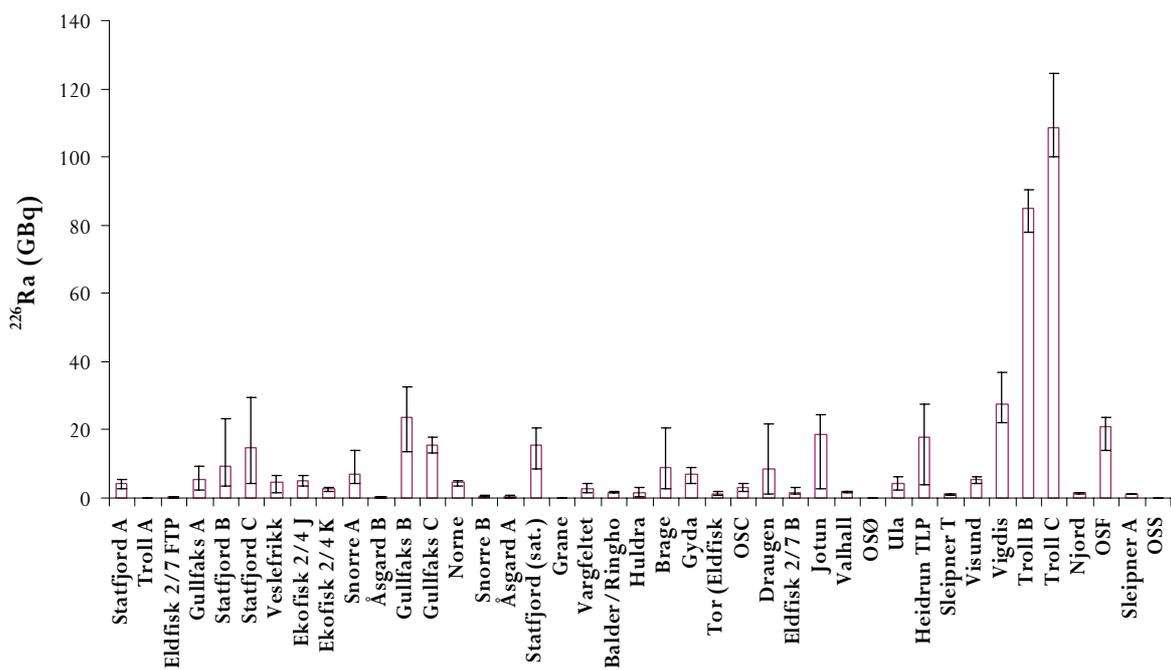


Figure 2.1. Estimated total discharged activity of ^{226}Ra for each platform during 2003.

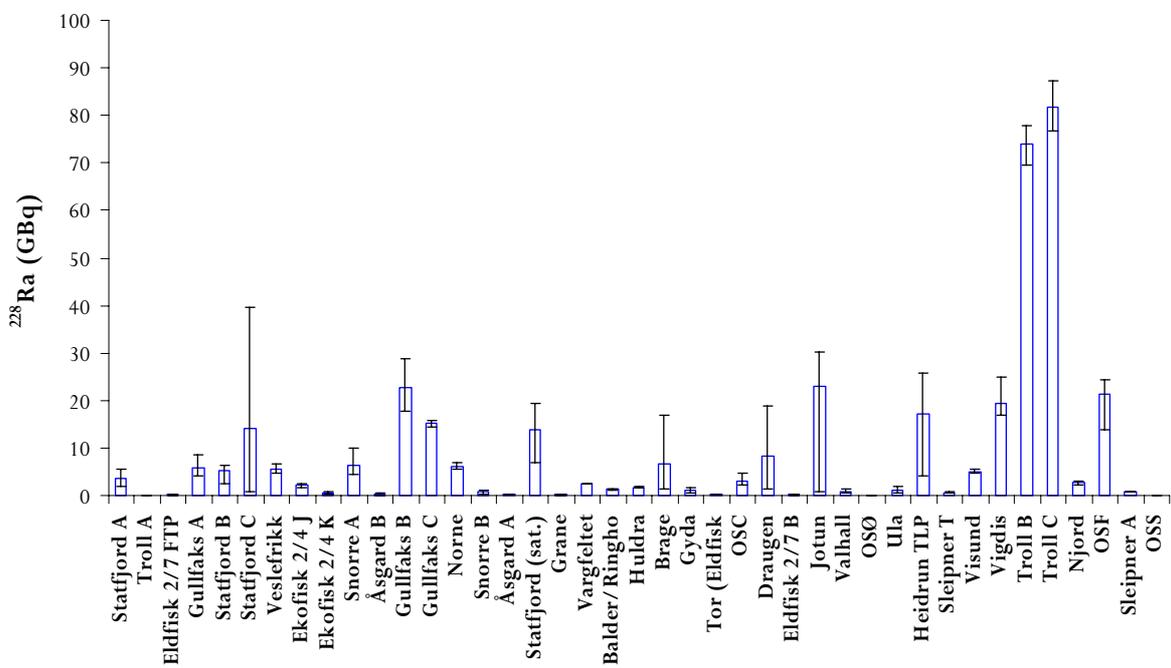


Figure 2.2. Estimated total discharge of ^{228}Ra for each platform during 2003.

2.4 Long-range transport of radionuclides

There are several sources of radioactive contamination of the northeast Atlantic and the Arctic marine environment, as shown in Figure 2.3.



Figure 2.3. Sources of radionuclides in the northern marine environment: Chernobyl in the Ukraine, the reprocessing plants at Sellafield, Cap de la Hague and Dounreay, the dumping sites for nuclear waste in the Kara Sea, the sites of the sunken submarines Komsomolets and K-159 and Russian nuclear installations (Mayak, Tomsk and Krasnoyarsk) releasing radionuclides to the Russian rivers Ob and Yenisey.

The primary sources are fallout from atmospheric nuclear weapons testing during the 1950s and 1960s, discharged radionuclides from reprocessing plants and fallout from the Chernobyl accident.

Radioactive contaminants discharged into northern European marine waters can reach Norwegian waters by ocean current transportation. The main ocean currents of the area are presented in Figure 2.4. Of importance is also the interaction between the radionuclides and particulate material in the water, which controls the residence time of a radionuclide in the water column. Some elements, such as Pu and Am, are particle-reactive and will to a large extent end up in the sediments. From the sediments the radionuclides can later be

remobilised and be transported away by local currents. Remobilised plutonium and ^{137}Cs from Irish Sea sediments contaminated by previous Sellafield discharges are one of the main sources of these elements in Norwegian marine waters. Other elements, such as ^{99}Tc and ^{90}Sr , are not particle-reactive and will follow currents and can be transported large distances away from the discharge point.

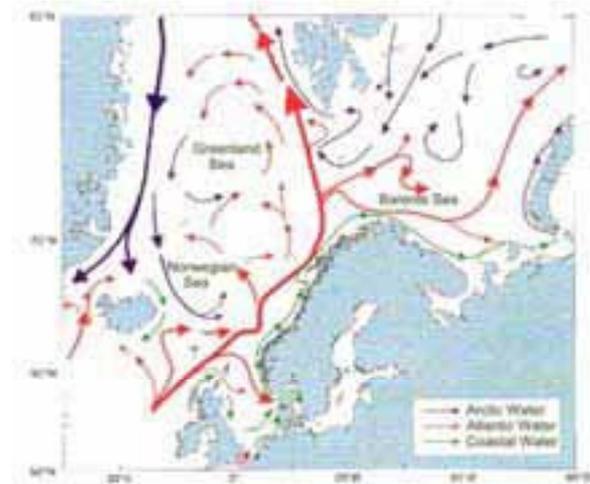


Figure 2.4. Overview of the main current system in the North Sea, Norwegian Sea, Greenland Sea and Barents Sea (Aure et al., 1998).

2.4.1 Discharge of radionuclides from European nuclear installations

Spent nuclear fuel can be managed in different ways. In a few countries, for instance in France and the UK, spent nuclear fuel is reprocessed. The major part of the waste from these facilities, such as fission products, is stored pending final management and disposal, but a small part is discharged as liquid waste to the marine environment. Discharge limits are set by the relevant national authority.

Sellafield (formerly Windscale) is located on the west coast of Cumbria in the UK. Further information about this reprocessing plant can be found in Amundsen *et al.*, (2003).

The other reprocessing plant in Europe affecting Norwegian marine waters is located at Cap de la Hague in France. Except for tritium (^3H), the discharge of liquid radioactive waste

has generally been lower here than from Sellafield.

Springfields is located on the west coast of the United Kingdom, and liquid waste is discharged to the Ribble estuary. This facility is mainly involved in the manufacture of fuel elements for nuclear reactors and the production of uranium hexafluoride. The Dounreay facilities on the northern coast of Scotland were established in 1955, and have mainly been used in the development of technology for fast breeder reactors. Liquid discharge of radioactive waste from Cap de la Hague, Sellafield, Springfields and Dounreay in 2003 is presented in Table 2.6.

Table 2.6. Liquid discharge of radionuclides (TBq) from Sellafield, Cap de la Hague, Springfields and Dounreay to the marine environment in 2003 (OSPAR, 2005).

| Nuclide | Cap de la Hague | Sellafield | Springfields | Dounreay |
|-------------------|----------------------|------------|---------------------|----------------------|
| ^3H | $1.19 \cdot 10^{-4}$ | 3900 | - | |
| Total- α | $2.3 \cdot 10^{-2}$ | 0.407 | 0.18 | $2.75 \cdot 10^{-3}$ |
| Total- β | 14 | 83.3 | 97.0 | 0.367 |
| ^{14}C | 8.7 | 17.0 | - | - |
| ^{60}Co | 0.36 | 0.43 | - | $2.49 \cdot 10^{-4}$ |
| ^{90}Sr | 0.52 | 14.0 | - | 0.129 |
| ^{99}Tc | 0.18 | 37.0 | 0.052 | - |
| ^{106}Ru | 7 | 11.5 | | $5.48 \cdot 10^{-4}$ |
| ^{129}I | 1.3 | 0.554 | - | - |
| ^{134}Cs | $4.2 \cdot 10^{-2}$ | 0.392 | - | - |
| ^{137}Cs | 0.76 | 6.24 | - | 0.0126 |
| ^{144}Ce | - | 0.885 | | $4.46 \cdot 10^{-4}$ |
| Pu- α | $6.9 \cdot 10^{-3}$ | 0.358 | - | - |
| ^{241}Pu | 0.15 | 10.1 | - | $2.05 \cdot 10^{-4}$ |
| ^{241}Am | $5.7 \cdot 10^{-3}$ | 0.059 | - | - |
| ^{237}Np | $9.2 \cdot 10^{-5}$ | - | 0.0018 | - |
| ^{230}Th | - | - | 0.067 | - |
| ^{232}Th | - | - | $6.0 \cdot 10^{-4}$ | - |
| U- α | - | - | 0.056 | - |
| U (kg) | 325* | 484 | - | - |

*Reported as $4.4 \cdot 10^{-3}$ TBq and converted to kg by assuming a specific activity for U of 13.7 MBq kg^{-1} .

In addition to the direct discharges from re-processing, the remobilisation of ^{137}Cs and plutonium from contaminated sediments in the Irish Sea acts as a secondary source of radionuclides in the marine environment. Substantial discharges of ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am and ^{137}Cs and other radionuclides in the 1970s and early 1980s resulted in widespread contamination of Irish Sea sediments. It has been shown that these radionuclides can be redissolved and transported out of the Irish Sea. An annual loss of $1.2 \text{ TBq } ^{239+240}\text{Pu}$ and $86 \text{ TBq } ^{137}\text{Cs}$ from the Irish Sea has been estimated by Cook *et al.*, (1997), while Leonard *et al.*, (1999) estimated annual losses of plutonium based on remobilisation from surface sediments to be 0.6 TBq . The half-lives of plutonium and americium in Irish Sea sediments were estimated to be 58 years and ~ 1000 years, respectively. This implies that remobilisation of radionuclides from Irish Sea sediments is a significant source of anthropogenic radionuclides in the North Sea and Norwegian coastal areas, and will remain so for a long time.

2.4.2 Global fallout from nuclear weapons testing

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 $948 \text{ PBq } ^{137}\text{Cs}$, $622 \text{ PBq } ^{90}\text{Sr}$, $6.52 \text{ PBq } ^{239}\text{Pu}$ and $4.35 \text{ PBq } ^{240}\text{Pu}$ have been released to the environment (UNSCEAR, 2000a), the major part in the northern hemisphere, where most of the tests took place.

Fallout from atmospheric nuclear weapons tests can still be found in the marine environment. About 60 % of the released activity was deposited in the world's oceans. The distribution of the fallout today depends on parameters such as, ocean currents, diffusion and sedimentation rates. Earlier expeditions in

the Atlantic have shown that the concentration of ^{137}Cs is about a few Bq m^{-3} (about twice as much as ^{90}Sr), between 45°S and 45°N , with a slight increase to the north (Bourlat *et al.*, 1996). The activity concentration of $^{239+240}\text{Pu}$ shows the same trend, but the activity concentrations are about 1000 times lower. Reasons for this are that less plutonium was released, but also that plutonium is more particle-reactive and has a higher sedimentation rate, which leads to a shorter residence time in the water.

2.4.3 Outflow of Chernobyl-contaminated water from the Baltic Sea

The worst nuclear accident in history occurred on April 26, 1986 in Chernobyl, Ukraine. In the initial explosion and the following fire, about 3-4 % of the nuclide inventory in the core was released to the atmosphere. The major part consisted of volatile radionuclides such as inert gases, iodine isotopes and ^{134}Cs and ^{137}Cs . Many of the released nuclides had short half-lives and can not be observed in the environment today. The radionuclide that will affect the environment in a long-term perspective is ^{137}Cs , with a physical half-life of 30 years. In total about 85 PBq of ^{137}Cs was released to the environment (UNSCEAR, 2000b). The Baltic Sea was the most affected marine area, where about 5 PBq was deposited (Evans, 1991). Shortly after the accident the activity concentration of ^{137}Cs was extremely high in part of the Baltic Sea. Due to low water exchange and a constant supply from rivers it will take a relatively long time for the levels to decrease. The estimated mean activity concentration of ^{137}Cs in the Baltic Sea surface water in 2000 was $50 \pm 20 \text{ Bq m}^{-3}$, with an effective half-life of 13 ± 2 years (Povinec *et al.*, 2003). Due to the water exchange through the Danish Straits it has been estimated that about 40 TBq flows from the Baltic Sea into the Kattegat annually (Dahlgaard, 2002). The ^{137}Cs will subsequently be transported by the Baltic current which continues as the Norwegian coastal current. This is one of the major sources of ^{137}Cs in the Norwegian marine environment,

together with remobilised ^{137}Cs from the seabed of the Irish Sea.

2.5 Other present and potential sources of radioactivity in the northern marine environment

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 (see Figure 2.2). These are the Mayak Production Association in the southern Urals and 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 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 (FSU) and the sunken submarines the Komsomolets and the K-159¹ in the Norwegian Sea and the Barents Sea. The dumping areas and the locations of the sunken submarines are given in Figure 2.2. Several investigations have been conducted to detect possible leakage from these sources.

As part of the bilateral environmental cooperation with the Russian Federation, a joint Russian-Norwegian expert group has been investigating the condition of the contained dumped objects and has analysed sediment and water samples at these sites.

According to Strand *et al.* (1998), elevated levels of radionuclides in sediments collected in the vicinity of the dumped objects demonstrated that leakage had occurred, but no significant inputs of radionuclides to the open sea from this source have been detected. Following an accident on April 7, 1989, the Russian nuclear submarine the 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. According to Kolstad (1995) a very small leakage of radiocaesium isotopes may have occurred. Samples of water and sediments collected around the submarine by the Institute of Marine Research and analysed with respect to ¹³⁷Cs are presented in Figure 2.5.

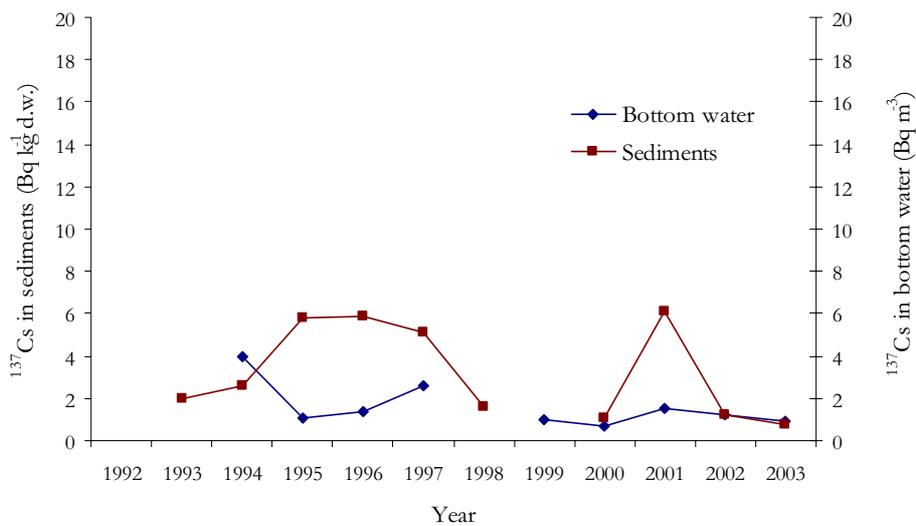


Figure 2.5. ¹³⁷Cs in samples from the position of the sunken nuclear submarine "Komsomolets".

¹ On August 29, 2003, the Russian nuclear submarine K-159 sank in the Barents Sea (5-6 km NW of Kildin Island) while it was being towed to a scrap yard. The submarine was not carrying nuclear weapons, but the reactors onboard were loaded with fuel.

3 Collection of samples

In 2003 samples were collected in the Norwegian Sea/Barents Sea, in selected fjords and at permanent coastal stations including the island of Svalbard. A geographic overview of the sampling area covered by the marine monitoring programmes, with the permanent coastal stations marked, is shown in Figure 3.1.

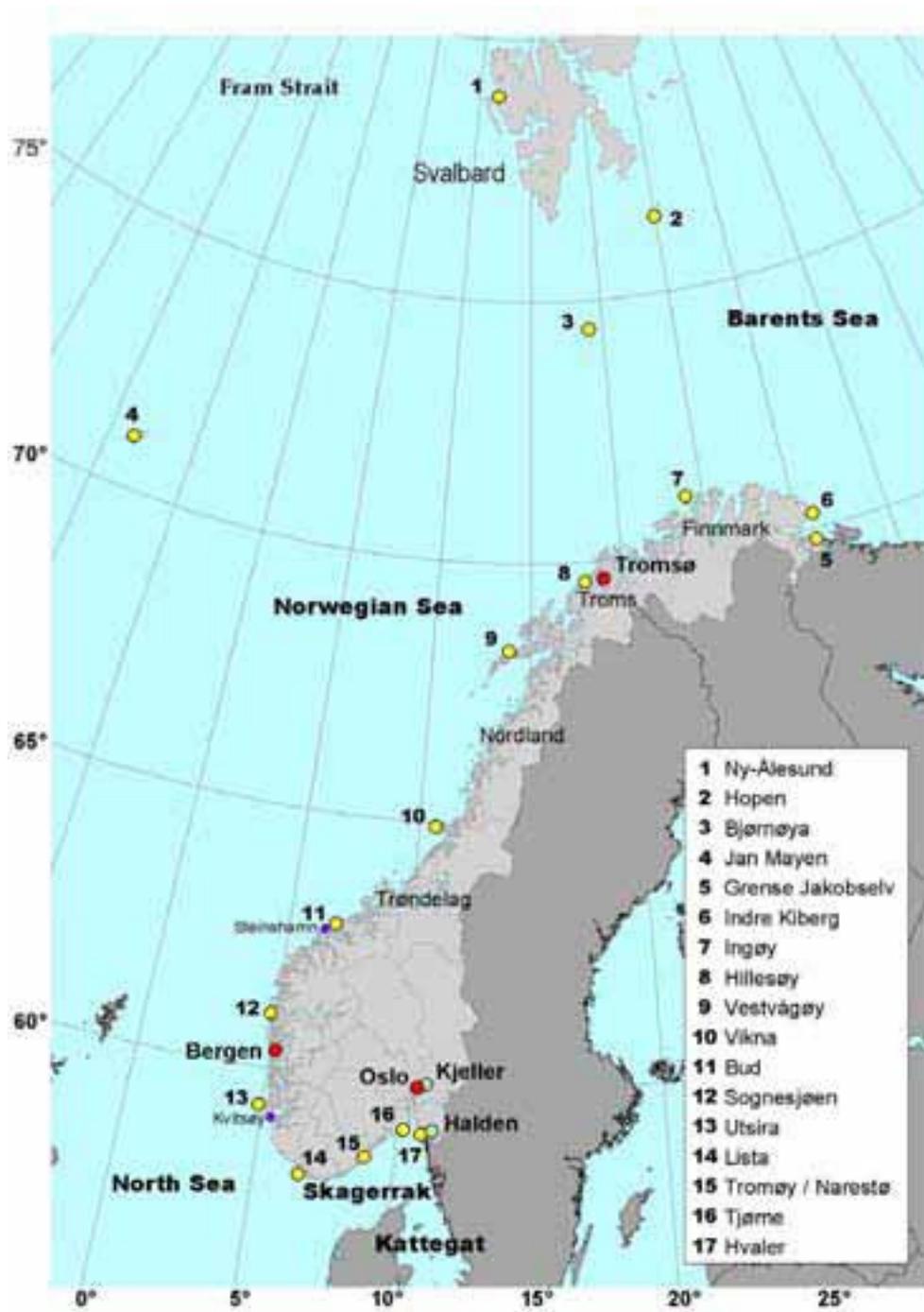


Figure 3.1. Geographic overview of the sampling area covered by the marine monitoring programmes. (Permanent coastal sampling stations are indicated by yellow dots).

Organisations participating in the monitoring programmes are the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR), the Directorate of Fisheries (Fdir), the Institute for Energy Technology (IFE), the Norwegian University of Life Sciences (UMB) and the National Institute of Nutrition and Seafood Research (NIFES).

One expedition to the Norwegian Sea took place between October 10 and November 11, 2003, with the R/V “Johan Hjort”, and another one, in the Norwegian Sea/Barent Sea, between May 27 and June 16, 2003, with the R/V “Johan Hjort”. During the expeditions samples of surface water were collected which were later analysed for ^{228}Ra , ^{226}Ra , ^{99}Tc , ^{137}Cs , ^{90}Sr , ^{241}Am and plutonium isotopes. Sediment was also sampled and later analysed for plutonium and ^{137}Cs . Fish samples collected were mainly analysed for ^{137}Cs and, to some extent, ^{99}Tc and ^{210}Po .

3.1 Sea water

For ^{99}Tc , ^{90}Sr and ^{226}Ra 50-100 litres of unfiltered water was collected at each sampling station and later transported to various laboratories for analysis.

For the analysis of ^{137}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 (Fig 3.2). 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}Cs activities on the first and second impregnated filters, respectively.



Figure 3.2. Filter system used for analysis of ^{137}Cs in sea water.

Sea water samples of 200 litres (unfiltered) were collected for the determination of the activity concentrations of ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am . 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).

3.2 Sediments

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.

3.3 Fish, crustaceans and molluscs

The Directorate of Fisheries collected fish and shrimp samples from commercial fishing locations in the Barents Sea. Samples of cod were prepared four times per year by combining muscle samples of 50 grams from 100 individual fish. Twenty-five individual cod fish were also analysed each month with respect to ^{137}Cs . Additionally, NIFES sampled different fish species, lobsters, crabs and fish food from different locations along the Norwegian coastline which were analysed for ^{137}Cs and ^{99}Tc . During expeditions to the Barents Sea/Norwegian Sea fish were also collected by

the IMR and analysed for ^{137}Cs . At the request of the NRPA, Labora A/S in Salten was sampling different species of fish and crustaceans from the northern part of Norway.

Lobsters (*Homarus gammarus*) from three sites in Norway have been analysed for ^{99}Tc and ^{137}Cs . From one region (Værlandet) samples were delivered By IMR. From the two other sites (Tisler and Arendal), local fishermen delivered the lobsters to the NRPA immediately after they had been caught.

3.4 Seaweed and other biota

The NRPA collected seaweed (*Fucus vesiculosus*) samples from eight stations along the Norwegian coastline. At Hillesøy in northern Norway, seaweed is collected every month and at the other locations once per 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. Samples of the algae *Laminaria hyperborea* have been collected at two different locations on the Norwegian west coast, Kvitsøy and Steinshamn by the UMB. At Svalbard, samples of both *Fucus distichus* and *Laminaria digitata* were collected by the NRPA.

3.5 Marine mammals

All marine mammal samples were provided by the Norwegian Polar Institute. Muscle, kidney, liver, brain and bone samples of seal species were collected by the Norwegian Polar Institute as part of their 2003 scientific field campaigns, whilst muscle samples of polar bears from Svalbard were obtained from animals killed during 2003. All muscle samples were analysed for gamma-emitting nuclides. A number of seal samples were also analysed with respect to ^{210}Po and ^{90}Sr .

4 Radioactivity in sea water and sediments

4.1 Technetium-99 in sea water

Tc-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 far away from its discharge point without being significantly affected by sedimentation processes. From the Irish Sea, ^{99}Tc is transported by 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 by Brown *et al.* (2002) to about 3 ½ years.

During 2003, sea water samples were collected in the Norwegian Sea, along the Norwegian coast and NW of Svalbard and at the Islands of Hopen, Bjørnøya and Jan Mayen, and analysed for ^{99}Tc . The results are presented in Figure 4.1 and Table 4.1 and range from 0.07 to 1.7 Bq m^{-3} , where the highest levels were found off the southern coast of Norway. The levels observed NW of Svalbard is significantly lower due to further dilution with Arctic and Atlantic sea water containing low levels of ^{99}Tc . A survey was also conducted in the Norwegian Sea in 2000 (Gäfvert *et al.*, 2003). The levels found in 2003 in the Norwegian Sea are generally lower than those observed in the same area in 2000. Monthly sampling at Hillesøy also reveals that the average activity concentration in sea water at this site has decreased from $1.5 \pm 0.3 \text{ Bq m}^{-3}$ in 1999 and $1.0 \pm 0.2 \text{ Bq m}^{-3}$ in 2002 to 0.85 ± 0.1 in 2003.

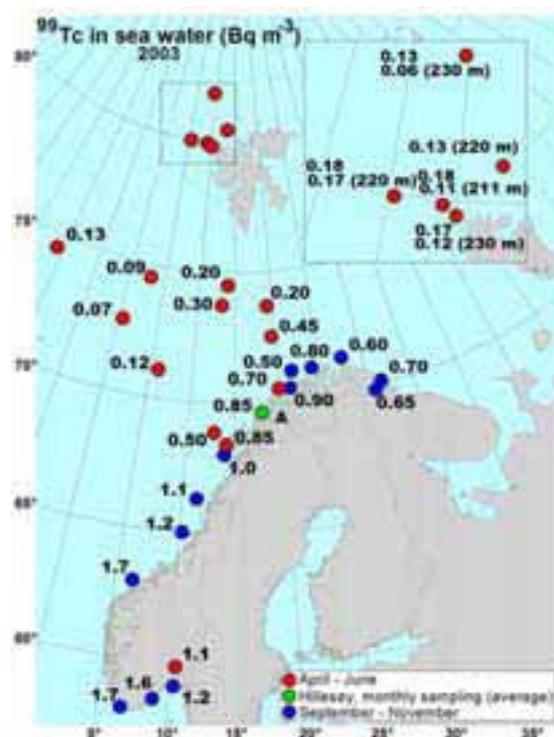


Figure 4.1. Activity concentration (Bq m^{-3}) of ^{99}Tc in sea water samples collected in the Norwegian Sea and at coastal stations in 2003. For Hillesøy, the average from monthly sampling is presented.

Samples collected at the islands of Hopen, Bjørnøya and Jan Mayen show activity concentrations in the range from 0.08 to 0.20 Bq m^{-3} (Table 4.1), which is similar to what was found both 2001 and 2002 at these sampling stations (Gäfvert *et al.*, 2003; NRPA 2004).

Table 4.1. Activity concentration of ^{99}Tc in surface water at the Islands of Hopen, Bjørnøya and Jan Mayen in 2003.

| Location | ^{99}Tc (Bq m^{-3}) |
|-----------|---|
| Hopen | 0.19 |
| Bjørnøya | 0.20 |
| Jan Mayen | 0.08 |

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 considered as a conservatively behaving element in the marine environment.

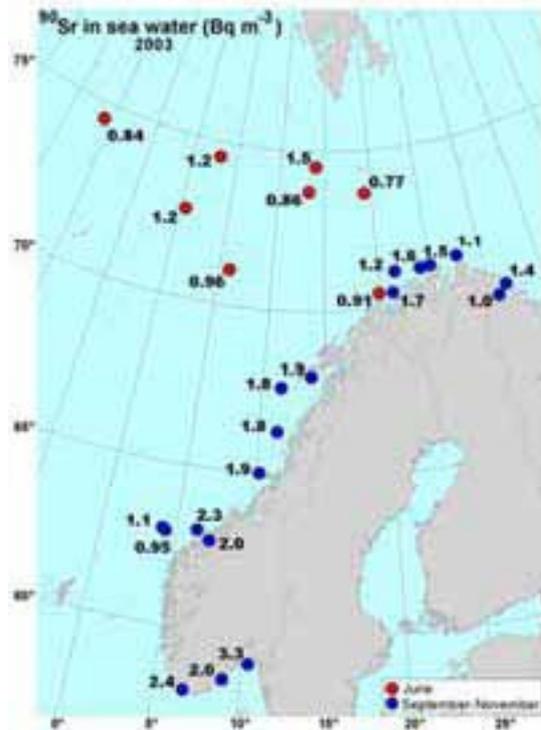


Figure 4.2. Activity concentration (Bq m^{-3}) of ^{90}Sr in surface water samples collected in the Norwegian Sea and at coastal stations in 2003.

Sea water samples were collected in the Norwegian Sea and along the Norwegian coast and 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 annual discharge of ^{90}Sr from Sellafield between 1998 and 2003 has been in the range of 14 to 31 TBq. The results for 2003 are presented in Figure 4.2 and Table 4.2.

In the northern marine waters Sea and along the Norwegian coast, the activity concentration ranged from 0.77 Bq m^{-3} to 3.3 Bq m^{-3} . The highest concentrations were found at the coastal stations along the southern coast of Norway.

Comparing the activity concentrations observed in 2001, 2002 and 2003 (Gäfvert *et al.*, 2003; NRPA 2004) in the Skagerrak and Barents Sea/Norwegian Sea, one can see that the level of ^{90}Sr have been relatively stable in recent years.

Table 4.2. Activity concentration of ^{90}Sr and salinity in sea water off the southern coast of Norway in September 2003.

| Location | ^{90}Sr (Bq m^{-3}) | Salinity (‰) |
|----------|---|--------------|
| Lista | 2.4 | 30.2 |
| Narestø | 2.0 | 31.4 |
| Tjøme | 3.3 | 28.2 |

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 Barents Sea and the Norwegian Sea are fallout from atmospheric nuclear weapons tests in the 1950s and 60s, 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.



Figure 4.3. Activity concentration (Bq m^{-3}) of ^{137}Cs in sea water samples collected in northern marine waters in 2002.

Observed levels of ^{137}Cs 2003 in northern marine waters and along the Norwegian coast are presented in Figure 4.3. The activity concentration in surface water ranged from 0.8 to 7.5 Bq m^{-3} , where the highest concentrations were found off the southern and western coast of Norway. The higher concentration at some sites along the coast is due to long-range transportation of ^{137}Cs with the Norwegian Coastal Current and runoff of Chernobyl

related ^{137}Cs from land. The general level in the open sea can be considered as low. Samples collected south of Svalbard (NRPA, 2004) showed similar levels of ^{137}Cs as those observed in 2003.

Caesium-137 has also been analysed in surface sediment from the Barents Sea/Norwegian Sea and along the coast. The results are presented in Figure 4.4. The results range from below detection limit up to 15.9 Bq kg^{-1} (d.w.), where the highest levels are observed in coastal areas. This can be explained by a higher sedimentation rate and run-off of ^{137}Cs from land. The activity concentration in samples collected in the open sea shows relatively low variation and range between 1.0 and 4.4 Bq kg^{-1} (d.w.).

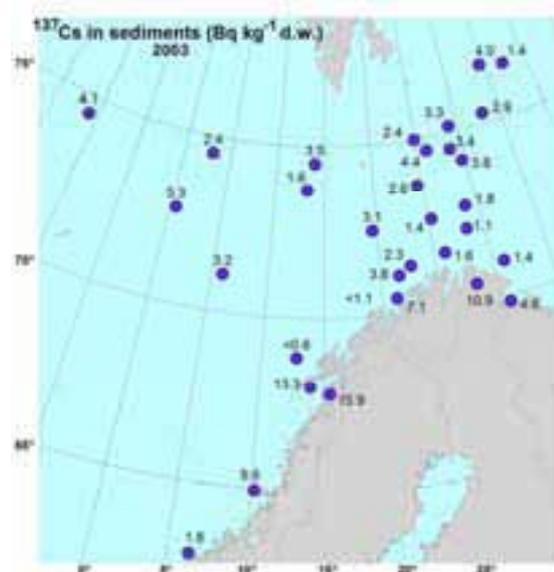


Figure 4.4. Activity concentration (Bq kg^{-1} d.w.) of ^{137}Cs in surface sediment.

4.4 Plutonium-239+240 in sea water and sediment

Plutonium-239 ($T_{1/2} = 24\,110 \text{ y}$) and ^{240}Pu ($T_{1/2} = 6\,563 \text{ y}$) belong to the transuranium elements and are mainly produced by neutron capture (with subsequent beta decay) by ^{238}U and ^{239}Pu , respectively. The 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 state, while in reducing environments the more particle-reactive +III and +IV states dominate. The main source of $^{239+240}\text{Pu}$ in northern Norwegian

marine waters is global fallout from atmospheric nuclear weapons tests in the 1950s and '60s. Another possible source is remobilised plutonium from Irish Sea sediments.



Figure 4.5. Activity concentration (mBq m^{-3}) of $^{239+240}\text{Pu}$ in surface water samples collected in the Norwegian Sea and along the coast in 2003.

Observed levels in samples collected in 2003 are presented in Figure 4.5 and range from 3.1 to 19.9 mBq m^{-3} . The highest level was observed in water collected from the Norwegian Coastal Current, having a $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio of 0.07 (slightly higher than 0.03 found for nuclear weapon fallout). This indicates that other sources, of which remobilised Pu from Irish Sea sediments is the most probable, have contributed. Levels above average were also observed in a sample collected southwest of Svalbard at a depth of 1655 m, in the vicinity of the sunken submarine Comsomolets. This does not necessarily indicate leakage of plutonium from the submarine. Pu onboard the submarine (nuclear warheads and a nuclear reactor) has a very low solubility, and the levels of more soluble radionuclides present onboard Comsomolets, such as ^{137}Cs (see Fig. 4.3), are not elevated. A

similar level of $^{239+240}\text{Pu}$ in bottom water (15 mBq m^{-3}) was also observed in 1993 (Kolstad, 1995). Oceanic studies of the behaviour of Pu have previously shown that subsurface maximum of Pu concentrations and significant increases in Pu concentrations near the sea-floor are common. A large part of the Pu is sedimenting with organic material that is exposed to oxidation and bacterial degradation during the descent to the sea-floor. As a result a large fraction of the Pu will be released before it reaches the sea-floor and form a subsurface concentration maximum (IAEA, 2005).

Surface sediments (upper 2 cm layer) sampled in coastal waters in northern Norway have also been analysed with respect to $^{239+240}\text{Pu}$. The results are presented in Figure 4.6 and range from 0.13 to 1.4 Bq kg^{-1} (d.w.).

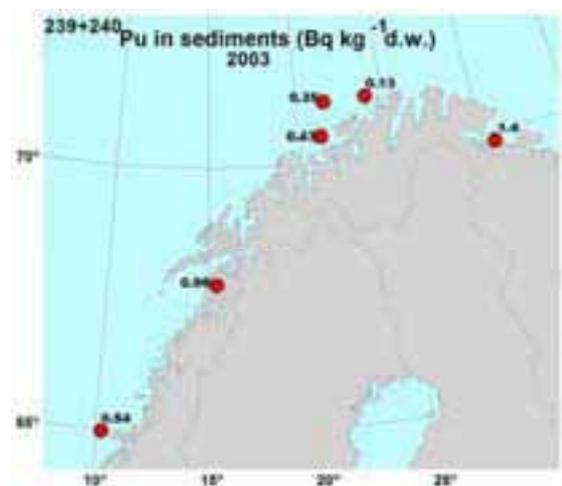


Figure 4.6. Activity concentration of $^{239+240}\text{Pu}$ (Bq kg^{-1} d.w.) in surface sediment.

5 Radioactivity in biota

5.1 Technetium-99 in seaweed

During 2003, seaweed (*Fucus vesiculosus*) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for ^{99}Tc . The advantage of using *Fucus vesiculosus* (see Fig. 5.1) as a bioindicator for ^{99}Tc is that it has a high concentration factor (CF) for this element and that *Fucus vesiculosus* is readily accessible in Norwegian waters. At two locations, Hillesøy and Utsira, sampling was performed monthly. At the remaining sites sampling was conducted in the period August to October. The results of the analyses are presented in Figure 5.2 and range from 100 to 340 Bq kg^{-1} (d.w.), where the highest level was found in a sample collected in the Oslo fjord. 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. The trend can also be seen in Figures 5.3 and 5.4 which show 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. The maximum levels in Figures 5.3 and 5.4 are the response to the increased discharge from Sellafield from 1994 and onwards. Due to advection and dispersion, pulsed discharges from Sellafield will be smoothed at distances far away from the point of discharge, and give broader peaks in ^{99}Tc levels in the seaweed. From Figures 5.3 and 5.4, one can see that the peak in ^{99}Tc levels in *F. vesiculosus* occurred after about 3-4 years at Utsira and about 4-5 years at Hillesøy further down-stream of the discharge.



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

In 2003, seaweed was also sampled in Kongsfjorden, Svalbard, showing ^{99}Tc activity concentrations in samples of *Fucus distichus* and *Laminaria saccharina* of $37 \pm 4 \text{ Bq kg}^{-1}$ (d.w.) and $2.3 \pm 0.6 \text{ Bq kg}^{-1}$ (d.w.), respectively. For *F. distichus* this value is similar to those reported for this species from the Svalbard area in 2000, 2001 and 2002 (Gäfvert *et al.*, 2003 and NRPA, 2003).

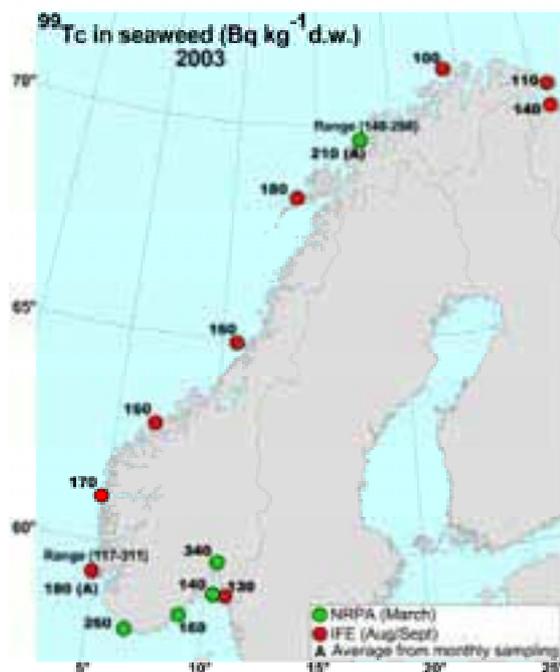


Figure 5.2. Levels of ^{99}Tc in *Fucus vesiculosus* sampled along the Norwegian coastline in 2003.

At Hillesøy both sea water and *F. vesiculosus* have been analysed monthly with respect to ^{99}Tc since 1997. In Figure 5.5 one can see that the activity concentration in the seaweed responded

rapidly with the increased ^{99}Tc levels in the sea water up to mid 2001. From mid 2001 and onwards, a decreasing trend is observed for the ^{99}Tc level in the water, while the activity concentration in the seaweed has decreased at a slightly slower rate, and shows relatively large fluctuations. Laboratory experiments have shown that the elimination of Tc from macroalgae generally has an initial rapid component (biological $T_{1/2}$ of 1-3 days) followed by a slower component with a biological $T_{1/2}$ of 20-200 days (Beasley and Lorz, 1986; Topocuoiglo and Fowler, 1984).

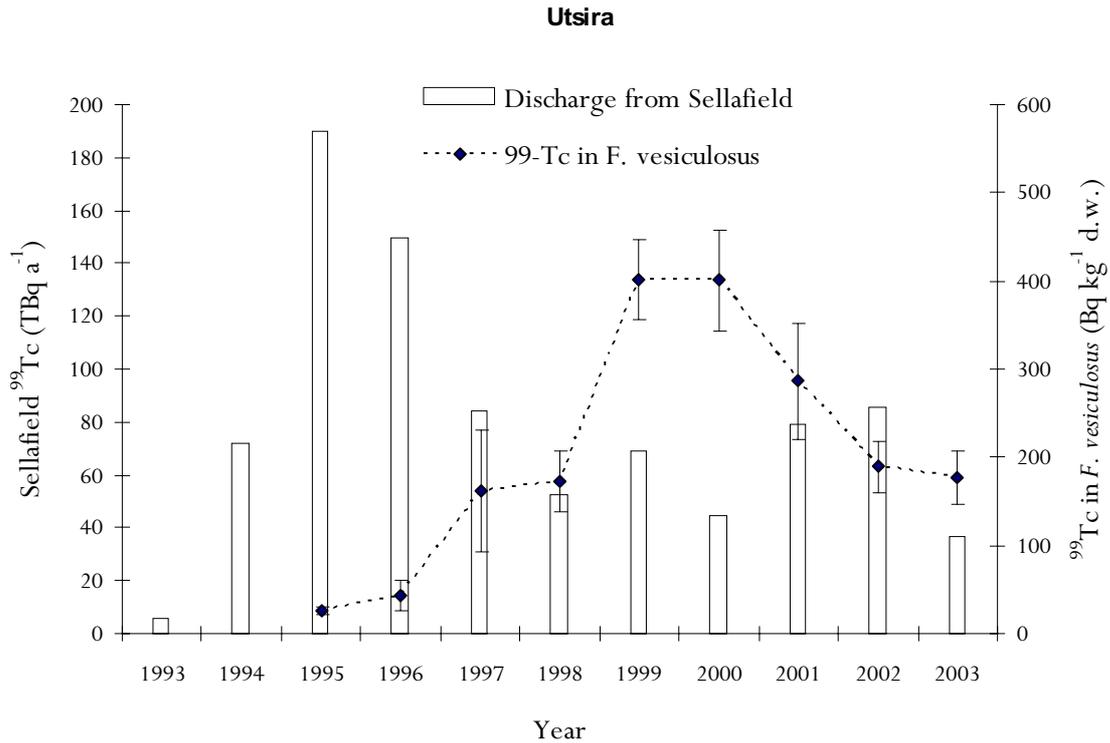


Figure 5.3. Annual liquid discharge of ^{99}Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) ^{99}Tc activity concentration in brown algae (*Fucus vesiculosus*) sampled at Utsira (data provided by IFE) in the period 1995-2003 (secondary axis).

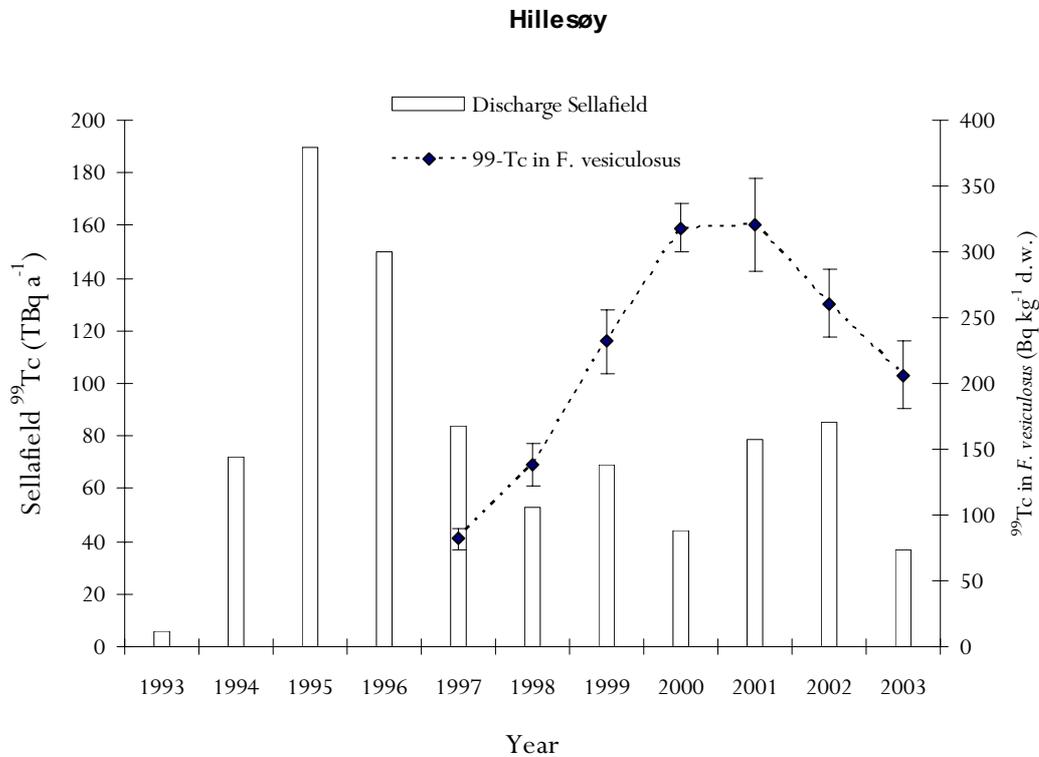


Figure 5.4. Annual liquid discharge of ^{99}Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) ^{99}Tc activity concentration in brown algae (*Fucus vesiculosus*) sampled at Hillesøy in the period 1997-2003 (secondary axis).

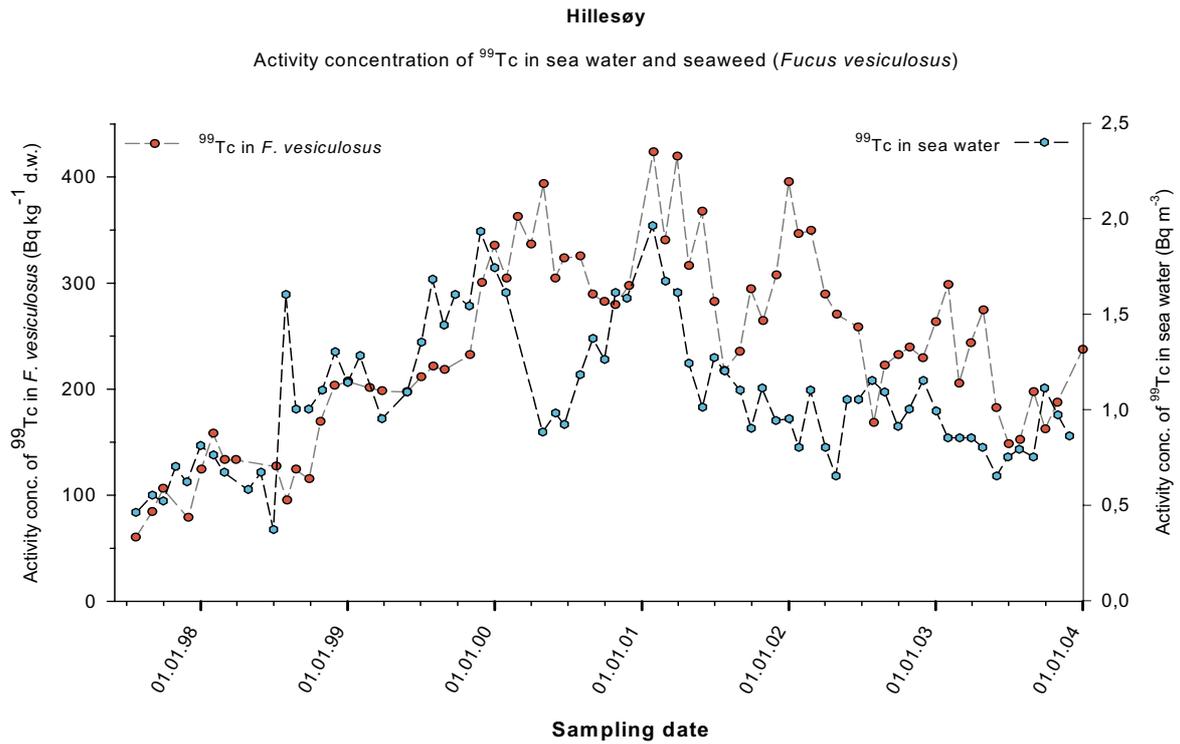


Figure 5.5. Activity concentration of ^{99}Tc in sea water and seaweed sampled at Hillesøy in the period 1997 to 2003.

Samples of the commercially used algae *Laminaria hyperborea* have been collected and analysed with regard to ^{99}Tc by the UMB at two different sites on the Norwegian west coast, Kvitøy and Steinshamn (see Figure 3.1) in the periods 1997 to 2001, and 1999 to 2003, respectively (Mobbs and Salbu, 2002; Jerpetjøn *et al.*, 2005). This alga is routinely harvested along the coastline and used in the alginate industry. *Laminaria hyperborea* is a large brown alga which grows on rocky substrata up to 30 m below the surface. It consists of a stiff, tapering stipe (constituting about 60 % of the biomass), up to 2 m tall, and a broad, deeply divided blade. During the growing season the old blades are shed and replaced with new ones. It can live for up to 15 years. The algae samples were divided into blades and stipes and the results are presented in Figure 5.6.

⁹⁹Tc in stipes and blades of *Laminaria hyperborea* sampled at Kvitsoy and Steinshamn in the period 1997 to 2003

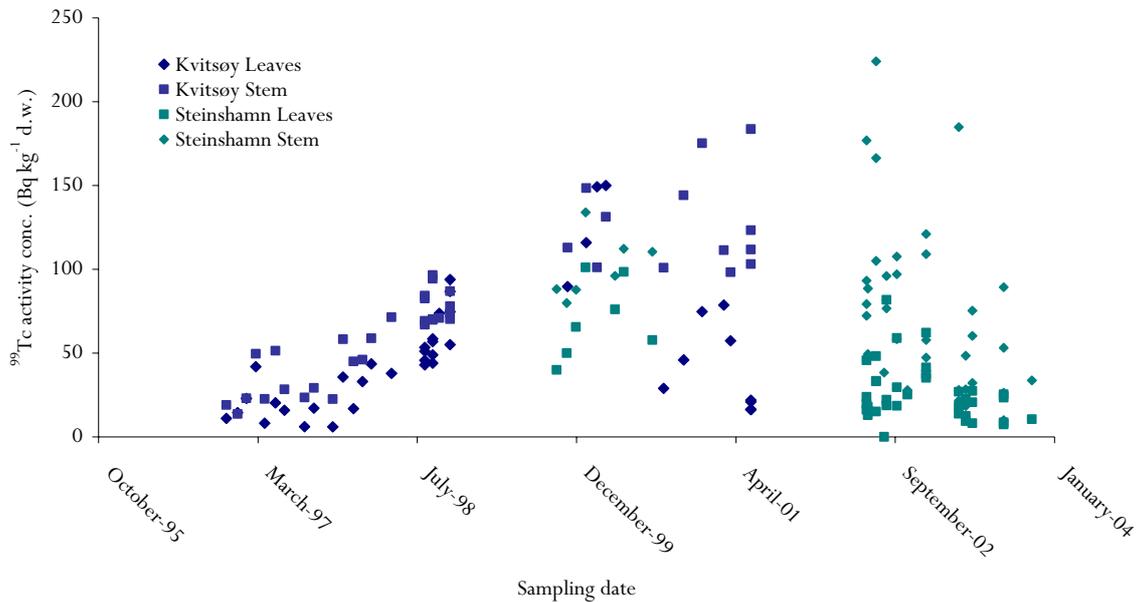


Figure 5.6. Activity concentration of ⁹⁹Tc in stipes and blades of *Laminaria hyperborea* sampled at Kvitsoy and Steinshamn (data provided by the UMB).

The results show that the activity of ⁹⁹Tc increased in both blades and stipes in the period 1997 to early 2000, with a slightly higher CF for the stipes compared with the blades. Since then, the activity has decreased in the blades, which are replaced annually, while the activity in the stipes has remained at an elevated level. The pattern of activity in the blades suggests that the peak of the discharge plume from Sellafield has reached these sites. However, the higher uptake and slower elimination of ⁹⁹Tc in the stipes shows that the levels in the stipes will remain at an elevated level after the plume of ⁹⁹Tc-contaminated water has passed these sites.

5.2 Caesium-137 and plutonium-239+240 in seaweed

5.2.1 Caesium-137 in seaweed

Fucus vesiculosus has also been widely used as a bioindicator for ¹³⁷Cs. The accumulation of ¹³⁷Cs in brown algae is, however, not as

pronounced as for ⁹⁹Tc. The uptake of ¹³⁷Cs also depends on the salinity of the surrounding sea water, with higher uptake at lower salinities (Carlsson and Erlandsson, 1991).

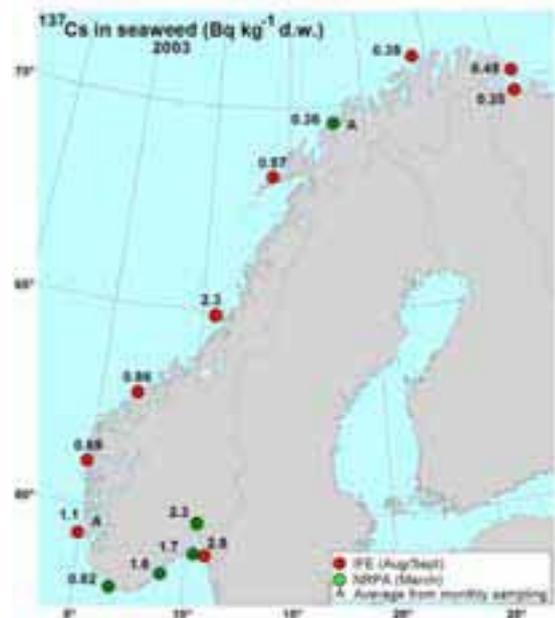


Figure 5.7. Levels of ¹³⁷Cs (Bq kg⁻¹ d.w.) in *Fucus vesiculosus* sampled along the Norwegian coastline in 2003.

In 2003, samples of *Fucus vesiculosus* from the permanent coastal stations (see Figure 3.1) were analysed with respect to ^{137}Cs . The results are presented in Figure 5.7, and range from 0.35 in the north to 2.8 Bq kg⁻¹ (d.w.) close to the Swedish border. 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, that the activity concentration of ^{137}Cs in the sea water is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident. In Figure 5.8 the activity concentration of ^{137}Cs in the seaweed is plotted together with the salinity of the water for 11 of the permanent coastal stations. 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.

Comparing those results with the levels found at the coastal sampling stations in 2001 and 2002 (Gäfvert *et al.*, 2003; NRPA, 2004), one can see that the activity concentration of ^{137}Cs in *F. vesiculosus* has been relatively stable during recent years. In the southern part of Norway the levels have been in the range 1.2 to 6.0 Bq kg⁻¹ (d.w.), while in the northern part all results have been below 1 Bq kg⁻¹ (d.w.).

5.2.2 The Svalbard region

Three types of seaweed, *Fucus distichus*, *Laminaria digitata* and *Laminaria saccharina*, have been sampled in the waters around Svalbard and analysed for ^{137}Cs (Table 5.1). The activity concentration of ^{137}Cs in *Fucus distichus* samples from Kongsfjorden and Ny-Ålesund, Svalbard were all below 2.0 Bq kg⁻¹ (d.w.). Samples of *Laminaria digitata* collected in Kongsfjorden and *Laminaria saccharina* collected in Kongsfjorden and Ny-Ålesund also showed low activity concentration, all below 2.4 Bq kg⁻¹ (d.w.).

Table 5.1. Activity concentration of ^{137}Cs in different types of seaweed collected in Svalbard 2003.

| Sample type | Location | Activity conc. of ^{137}Cs (Bq kg ⁻¹ d.w.) |
|-----------------------------|--------------|--|
| <i>Fucus distichus</i> | Kongsfjorden | <1.0 |
| <i>Fucus distichus</i> | Kongsfjorden | <2.0 |
| <i>Fucus distichus</i> | Kongsfjorden | 0.4 |
| <i>Fucus distichus</i> | Ny-Ålesund | <0.9 |
| <i>Laminaria digitata</i> | Kongsfjorden | <1.2 |
| <i>Laminaria digitata</i> | Kongsfjorden | <2.4 |
| <i>Laminaria saccharina</i> | Ny-Ålesund | <0.9 |
| <i>Laminaria saccharina</i> | Ny-Ålesund | 0.6 |
| <i>Laminaria saccharina</i> | Kongsfjorden | <1.4 |

¹³⁷Cs in *Fucus vesiculosus* and sea water salinity

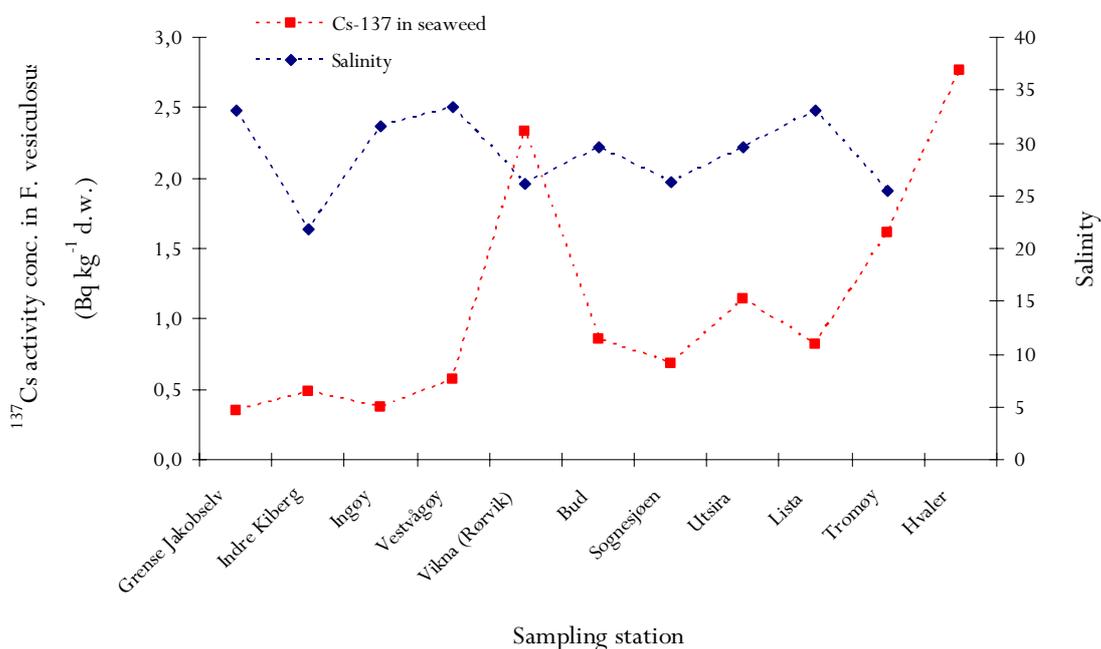


Figure 5.8. Activity concentration ($Bq\ kg^{-1}\ d.w.$) in *Fucus vesiculosus* versus salinity in sea water sampled along the Norwegian coastline in 2003 (data provided by IFE).

5.2.3 Plutonium 239+240 in seaweed

Fucus vesiculosus has been collected and analysed for ²³⁹⁺²⁴⁰Pu at Utsira since 1980. The results from the period 1980 to 2003 are presented in Figure 5.9. The activity concentrations in these samples were in the range 40 to 201 mBq kg⁻¹, with relatively large fluctuations from year to year. Two samples collected at Hillesøy in Aug./Sept. 2003 showed an average activity concentration of 60 mBq kg⁻¹ (d.w.). In Figure 5.10 the activity concentration of ²³⁹⁺²⁴⁰Pu in *Fucus vesiculosus* at Hillesøy since 1998 is presented. One can also see that the levels of ²³⁹⁺²⁴⁰Pu has remained at the same level in recent years. Since the activity of ²³⁸Pu was very low, it has not been possible to use the ²³⁸Pu/²³⁹⁺²⁴⁰Pu ratio for source identification (the use of the ²³⁸Pu/²³⁹⁺²⁴⁰Pu ratio in this area may also be complicated by the relatively low discharge of ²³⁸Pu and ²³⁹⁺²⁴⁰Pu, with a very high ²³⁸Pu/²³⁹⁺²⁴⁰Pu ratio (IRSN, 1999) from the reprocessing plant at Cap de la Hague)). The

levels are similar to those found by Roos *et al.* (1993) in the late 1980s and early 1990s off the Swedish west coast. At this site (Särdal) levels up to around 600 mBq kg⁻¹ (d.w.) were found in *Fucus vesiculosus* in the 1970s due to the higher discharge of plutonium from Sellafield during the 1960s and 1970s.

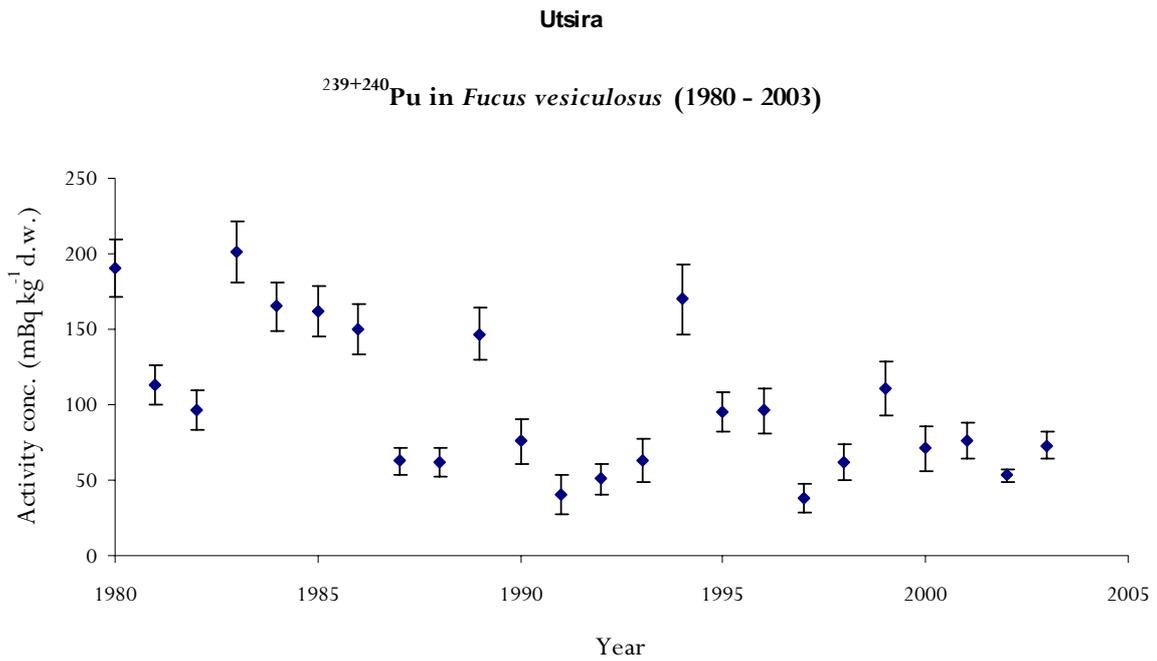


Figure 5.9. ²³⁹⁺²⁴⁰Pu levels (mBq kg⁻¹ d.w.) in *Fucus vesiculosus* at Utsira in the period 1980 to 2003 (data provided by IFE).

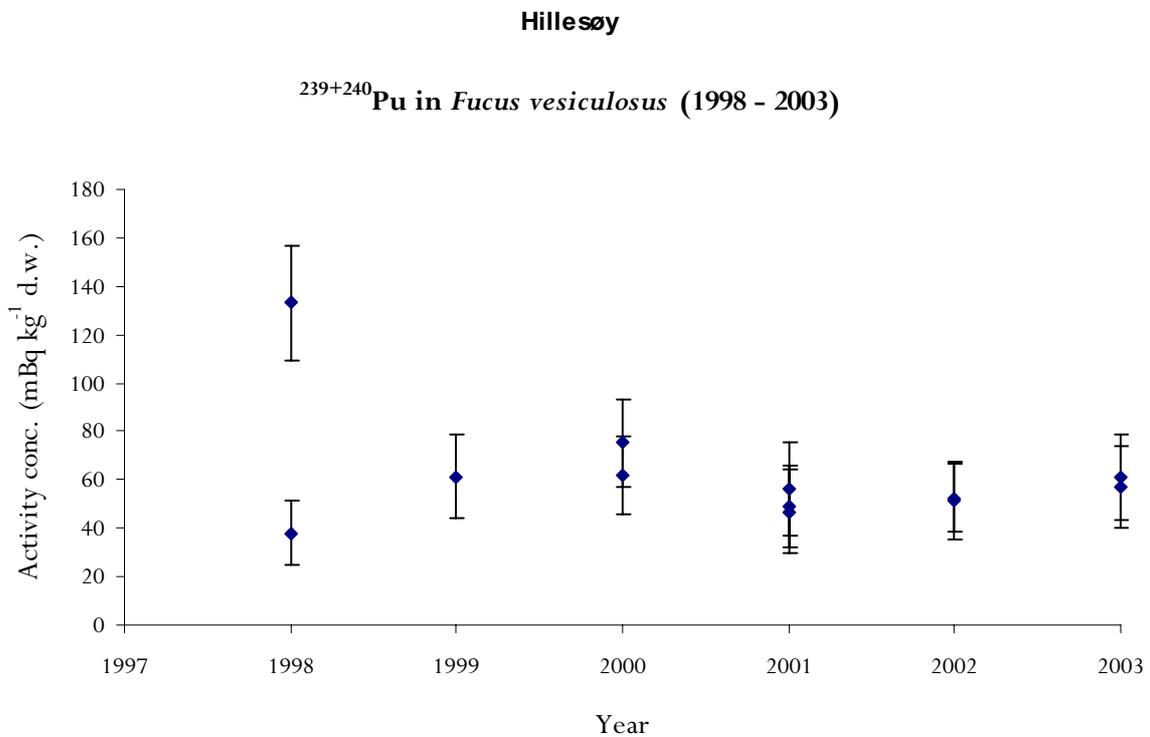


Figure 5.10. ²³⁹⁺²⁴⁰Pu levels (mBq kg⁻¹ d.w.) in *Fucus vesiculosus* at Hillesøy in the period 1998 to 2003.

5.3 Radioactivity in marine mammals

5.3.1 Caesium-137 in Marine Mammals

In 2003, muscle samples collected by the Norwegian Polar Institute (NPI) were taken from ringed seals (*Phoca hispida*) from the west coast of Svalbard and from harp seals (*Phoca groenlandica*) from the South Barents Seas. Additionally, muscle samples of polar bears that were killed on Svalbard during 2003 were given to the Norwegian Radiation Protection Authority (NRPA) by the NPI. All samples were analysed by the NRPA.

Activity concentrations of ^{137}Cs in muscle samples of all marine mammals (Table 5.2) were relatively low and of the order of $< 1 \text{ Bq kg}^{-1}$ (w.w). Concentration factors (Bq kg^{-1} dry weight biota / Bq l^{-1} sea water) for ringed seals

ranged from 201 to 305, while concentration factors for harp seals ranged from 73 to 155. These ranges of activity concentrations and concentration factors are comparable to those previously reported for these seal species from North East Svalbard (Carroll *et al.*, 2002) and the White Sea (Rissanen *et al.*, 1997) and reflect the current low levels of ^{137}Cs in the Svalbard and Barents marine environments (NRPA, 2004).

Additionally, ^{137}Cs activity concentrations were determined in organs of three ringed seals from the west coast of Svalbard (Table 5.3). Activity concentrations of ^{137}Cs in all organs were lower than those in muscle from the same animal and in general followed the trend muscle > liver = kidney > brain. A similar trend has been reported for liver and kidneys of ringed seals from NE Svalbard in 1999 and from Alaska and Canada in 1995 to 1997 (Cooper *et al.*, 2000; Carroll *et al.*, 2002).

Table 5.2. Activity concentrations (Bq/kg w.w.) of ^{137}Cs in muscle of polar bears, ringed seals and harp seals collected in 2003.

| Species | Location | ^{137}Cs in Muscle (Bq kg^{-1} w.w.) | No. of Samples |
|--|----------------|---|----------------|
| Polar bear (<i>Ursus maritimus</i>) | Svalbard | 0.45-1.50 | 3 |
| Ringed Seal (<i>Phoca hispida</i>) | W. Spitsbergen | 0.40-0.61 | 15 |
| Harp Seal (<i>Phoca groenlandica</i>) | S. Barents sea | < 0.20-0.53 | 14 |

Table 5.3. Activity concentrations (Bq/kg w.w.) of ^{137}Cs in organs of ringed seals from the west coast of Svalbard from 2003 compared to muscle. Adults are older than 4 years.

| Tissue | ^{137}Cs (Bq/kg w.w.) | | |
|--------|---|-----------------|-----------------|
| | Male Adult | Male Adult | Female Adult |
| Muscle | 0.40 ± 0.03 | 0.57 ± 0.06 | 0.48 ± 0.06 |
| Liver | 0.38 ± 0.04 | 0.40 ± 0.05 | 0.27 ± 0.05 |
| Kidney | 0.34 ± 0.05 | 0.50 ± 0.07 | 0.23 ± 0.04 |
| Brain | 0.27 ± 0.07 | 0.13 ± 0.05 | <0.24 |

5.3.2 *Sr-90 in Marine Mammals*

Activity concentrations of ^{90}Sr in the leg bones of three ringed seals from the west coast of Svalbard in 2003 were all below the limits of detection. Previously reported activity concentrations of ^{90}Sr in seal species are limited, with only a range of 0.10 to 0.18 Bq/kg (w.w.) reported for a variety of skeletal bones in Greenland seals from the White Sea area in 1996 (Rissanen *et al.*, 1999). At present there are no recommended values for concentration factors for ^{90}Sr in marine mammals. Current levels of ^{90}Sr in the Svalbard marine environment are low, with activity concentrations of $\sim 1 \text{ Bq/m}^3$ reported for 2002 (NRPA, 2004).

5.3.3 *Po-210 in Marine Mammals*

Activity concentrations of ^{210}Po in muscle of ringed seals from the west coast of Svalbard in 2003 ranged from 12 ± 4 to 32 ± 4 Bq/kg (w.w.) This range of values is similar to a previously reported ^{210}Po activity concentration range for ringed seals from Central West Greenland and the Baltic Sea (Holm and Leisvik, 2001; Dahlgard *et al.*, 2004). Concentration factors for ^{210}Po in muscle of ringed seals ranged from 1.2×10^4 to 3.2×10^4 , a magnitude higher than the ^{210}Po concentration factor recommended for fish, but similar to those for other marine organisms (IAEA, 2004).

5.4 Technetium-99 and caesium-137 in crustaceans and molluscs

Samples of lobster (*Homarus gammarus*) were collected at three coastal locations; Tisler, Arendal and Vaerlandet (see Figure 5.11) in 2003, and analysed for ^{99}Tc and ^{137}Cs . Lobsters have previously been identified as a marine organism with a high ability to accumulate ^{99}Tc from sea water (Swift, 1985; Smith *et al.*, 1998; Smith *et al.*, 2001). The observed levels are presented in Table 5.4, and range from 0.5 to 44.0 Bq kg⁻¹ w.w. (0.5 to 44.0 Bq kg⁻¹ (w.w.) for female lobsters and 0.7 to 20.0 Bq kg⁻¹ (w.w.) for male lobsters). The levels are comparable with those found in 2001 and 2002 in lobster samples from Rogaland (Kolstad and Lind, 2002) and Tisler, Arendal and Vaerlandet in 2002 (NRPA, 2004). The results show, however, a relatively large variation, even for samples collected in the same area. The activity concentration of ^{99}Tc in the sea water in the different sampling areas is in the range 1.2 to 1.6 Bq m⁻³. One can, furthermore, see a clear difference between female and male lobsters; female lobsters having a significantly higher uptake of ^{99}Tc .



Figure 5.11. Geographic overview of the sampling areas.

Table 5.4. Average activity concentration and range of ^{99}Tc in lobster (*Homarus gammarus*) tail muscle in 2003.

| Location | Gender (F/M) | n | ^{99}Tc Average (range) (Bq kg ⁻¹ w.w.) |
|--------------------------------|--------------|----|---|
| Tisler (Outer Oslofjord) | F | 7 | 19.7 (9.3 - 36.5) |
| | M | 4 | 7.5 (2.5 - 20.0) |
| Arendal | F | 15 | 17.1 (2.6 - 44.0) |
| | M | 15 | 4.3 (2.2 - 9.7) |
| Vaerlandet | F | 2 | 12.4 (9.6 - 15.1) |
| | M | 1 | 2.9 |

Some lobster samples in were also analysed for ^{137}Cs . The levels were considerably lower than for ^{99}Tc and ranged from < 0.1 to 0.32 Bq kg⁻¹ (w.w.).

Mussels (*Mytilus edulis*) were collected in 2003 in coastal regions from Nordland (see Figure 5.11) in the north, down to the southern coast of Norway. From each region about 0.5 kg (w.w.) of the mussel soft tissue was collected, homogenized and later analysed for ^{99}Tc and ^{137}Cs .

Table 5.5. Activity concentration of ^{99}Tc and ^{137}Cs in Blue mussels and Common limpet (*Mytilus edulis* and *Patella vulgata*)

| Location/Species | ^{99}Tc (Bq kg ⁻¹ w.w.) | ^{137}Cs (Bq kg ⁻¹ w.w.) |
|-----------------------------|--|---|
| Vestfold/ Blue mussels | 0.45 ± 0.05 | 0.16 ± 0.01 |
| Øst-Agder/ Blue mussels | 0.50 ± 0.06 | - |
| Vest-Agder/ Blue mussels | 0.30 ± 0.05 | 0.49 ± 0.15 |
| Nordland/ Common Limpet | - | 0.23 ± 0.18 |
| Blue mussels | - | - |

The results are presented in Table 5.5 The activity concentration range from 0.30 to 0.50 Bq kg⁻¹ (w.w.) for ^{99}Tc , and from 0.16 to 0.49 Bq kg⁻¹ (w.w.) for ^{137}Cs .

Samples of Kamtchatca crab (*Paralithodes camtschaticus*), Periwinkles (*Littorinidae*) and shrimps (*Pandalus borealis*) from Norwegian and adjacent marine waters have been collected and analysed for ^{99}Tc and ^{137}Cs . The results and the sampling location are presented in Table 5.6.

In addition samples of Amphipods is analysed for $^{239+240}\text{Pu}$, with the result $0.009 \pm 0.006 \text{ Bq kg}^{-1}$ (d.w.).

Table 5.6. Activity concentration (Bq kg^{-1} w.w.) in various marine species 2003.

| Species | Location | ^{99}Tc (Bq kg^{-1} w.w) | ^{137}Cs (Bq kg^{-1} w.w) |
|---|-----------------------------------|---|--|
| Periwinkles (<i>Littorinidae</i>) | Outer Oslofjord Vestfold | 0.75 ± 0.10 | - |
| Periwinkles (<i>Littorinidae</i>) | Lista Vest-Agder | 1.00 ± 0.13 | - |
| Shrimps (<i>Pandalus borealis</i>) | Landegode- fjorden Nordland | 0.23 ± 0.13 | 0.23 ± 0.13 |
| Kamtchatca crab (<i>Paralithodes camtschaticus</i>) | Holmengrå Finmark | - | < 0.10 |

5.5 Caesium-137 and technetium-99 in fish

Commercially important, as well as other species of fish have been collected from Norwegian marine waters and analysed with respect to ^{137}Cs and ^{99}Tc . Farmed salmon has also been collected at different locations along the coast and analysed for ^{137}Cs .

The activity concentrations of ^{137}Cs in fish caught in the Barents Sea, in Norwegian fjords and in the coastal waters of Finnmark (see Figure 3.1) are presented in Table 5.7. The levels of ^{137}Cs in all sampled fish species from this area range from 0.1 to 0.3 Bq kg⁻¹ (w.w.). The activity concentrations of ^{137}Cs in fish from the Norwegian Sea, the North Sea and the fjords and coastal waters of central and northern Norway are given in Table 5.8. The results from this area range from <0.20 to 0.50 Bq kg⁻¹ (w.w.).

The activity concentrations of ^{99}Tc in fish from the Barents Sea, Norwegian Sea, the North Sea and the coastal waters of northern, central and western Norway are given in Table 5.9. The results from this area range from < 0.007 to 0.088 Bq kg⁻¹ (w.w.), where the highest levels were found in Mackerel from the North Sea.

5.5.1 Farmed fish

In Table 5.10 the results from the analyses of ^{137}Cs in farmed salmon (*Salmo salar*) from different locations along the Norwegian coast are presented. The activity concentration of ^{137}Cs ranges from 0.17 ± 0.07 to 0.90 ± 0.10 Bq kg⁻¹ (w.w.), where the highest levels were found in the coastal waters of Hordaland.

Technetium-99 was also analysed in different types of fish food and ensilage used in the fish farming industry (Table 5.11). All samples showed activity concentrations below 0.5 Bq kg⁻¹ (d.w.)

Generally, the levels of ^{137}Cs in Norwegian marine waters can be considered low. In Figure 5.12, typical activity concentrations of ^{137}Cs found in cod in northern European waters are presented. In the Baltic Sea, the levels of ^{137}Cs in fish are higher due to higher levels of ^{137}Cs in the water, but also due to the lower salinity of the water, resulting in a higher uptake. In the Irish Sea, the levels are influenced by past and present discharges of ^{137}Cs from the reprocessing plant at Sellafield.

Table 5.7. Levels of ^{137}Cs in common fish species from the Barents Sea, the North Sea and the fjords and coastal waters of the northern part of Norway, in 2003.

| Species | Location | No. of samples (total no. of fish) | ^{137}Cs in muscle tissue (Bq kg ⁻¹ w.w.) |
|--|--|---------------------------------------|--|
| Cod (<i>Gadus morhua</i> L.) | Barents Sea | 10 (850) | 0.10 ± 0.04 to 0.30 ± 0.02 |
| | | 300 (300) | < 5.5 |
| Polar Cod (<i>Boreogadus saida</i>) | | 2 (50) | 0.10 ± 0.10 |
| Haddock (<i>Mellangorammus aeglefinus</i>) | | 2 (50) | 0.20 ± 0.10 |
| Long Rough Dab (<i>Hippoglossoides Platessoides</i>) | | 2 (50) | 0.20 ± 0.10 |
| Capelin (<i>Mallotus Villosus</i>) | | 3(75) | 0.10 ± 0.10 |
| Greenland Halibut (<i>Reinhardtius Hippoglossoides</i>) | | 2 (50) | 0.30 ± 0.10 |
| Golden Redfish (<i>Sebastes Marinus</i>) | | 2 (50) | 0.10 ± 0.10 and 0.20 ± 0.10 |
| Cod (<i>Gadus morhua</i> L.) | Coastal waters of Finnmark, and fjords | 5 (125) | 0.20 ± 0.10 to 0.30 ± 0.10 |
| Haddock (<i>Mellangorammus aeglefinus</i>) | | 1 (25) | 0.20 ± 0.10 |
| Saithe (<i>Pollachius virens</i>) | | 2 (50) | 0.30 ± 0.10 |
| Rockfishes (<i>Anarhichadidae</i>) | | 1 (5) | 0.10 ± 0.10 |

Table 5.8. Levels of ^{137}Cs in common fish species from the Norwegian Sea and the coastal waters and fjords in the northern and central parts of Norway, in 2003.

| Species | Location | No. of samples (total no. of fish) | ^{137}Cs in muscle tissue (Bq kg ⁻¹ w.w.) |
|---|---|---------------------------------------|--|
| Cod (<i>Gadus morhua</i> L.) | Norwegian Sea | 1 (25) | 0.4 ± 0.1 |
| Atlantic herring (<i>Clupea harengus</i>) | | 1 (25) | 0.2 ± 0.1 |
| Cod (<i>Gadus morhua</i> L.) | Coastal waters of Troms, Nordland and Trøndelag, and fjords | 6 (150) | 0.20 ± 0.08 to 0.48 ± 0.10 |
| Saithe (<i>Pollachius virens</i>) | | 3 (55) | 0.30 ± 0.10 to 0.32 ± 0.10 |
| Atlantic herring (<i>Clupea harengus</i>) | | 1 (25) | 0.40 ± 0.10 |
| Rockfishes (<i>Anarhichadidae</i>) | | 2 (3) | 0.20 ± 0.10 |
| Halibut (<i>Hippoglossus hippoglossus</i>) | | 2 (50) | 0.20 ± 0.10 and 0.40 ± 0.10 |
| Atlantic herring (<i>Clupea harengus</i>) | | North Sea | 1 (25) |
| Mackerel (<i>Scomber scombrus</i>) | 1 (25) | | < 0.20 |
| Anglerfish (<i>Lophius piscatorius</i>) | 2 (50) | | 0.20 ± 0.10 and 0.30 ± 0.10 |
| Cat-fish (<i>Anarhichas lupus</i>) | 1 (25) | | 0.30 ± 0.10 |
| Halibut (<i>Hippoglossus hippoglossus</i>) | 1 (25) | | 0.50 ± 0.10 |

Table 5.9. Levels of ^{99}Tc in common fish species from the Barents Sea, Norwegian Sea, the North Sea and the coastal waters and fjords in the northern, central and western parts of Norway, in 2003.

| Species | Location | No. of samples (total no. of fish) | ^{99}Tc in muscle tissue (Bq kg^{-1} w.w.) ¹ |
|---|---|---------------------------------------|---|
| Cod (<i>Gadus morhua</i> L.) | Barents Sea and coastal waters of Finnmark. | 2 (50) | < 0.007 and 0.008 ± 0.002 |
| Halibut (<i>Hippoglossus hippoglossus</i>) | Norwegian Sea and coastal waters of | 2 (50) | < 0.016 and 0.119 ± 0.030 |
| Atlantic herring (<i>Clupea harengus</i>) | Troms, Nordland and Trondelag | 1 (25) | 0.053 ± 0.003 |
| Halibut (<i>Hippoglossus hippoglossus</i>) | North Sea | 1 (25) | 0.037 ± 0.001 |
| Atlantic herring (<i>Clupea harengus</i>) | | 1 (25) | 0.077 ± 0.017 |
| Mackerel (<i>Scomber scombrus</i>) | | 1 (25) | 0.088 ± 0.005 |
| Cat-fish (<i>Anarhichas lupus</i>) | | 1 (25) | < 0.007 |
| Anglerfish (<i>Lophius piscatorius</i>) | | 2 (50) | < 0.007 and 0.021 ± 0.010 |

¹Analysed by the UMB

Table 5.10. Levels of ^{137}Cs in farmed salmon along the Norwegian coastline, from Troms in the north to Rogaland in the south, in 2003.

| Location | ^{137}Cs in muscle tissue (Bq kg^{-1} w.w.) |
|------------------|---|
| Troms | 0.17 ± 0.07 |
| Sør-Trondelag | 0.40 ± 0.10 |
| Møre og Romsdal | 0.20 ± 0.10 |
| Hordaland | 0.90 ± 0.10 |
| Sogn og Fjordane | 0.20 ± 0.10 |
| Rogaland | 0.30 ± 0.10 |

Table 5.11. Levels of ^{99}Tc in different types of fish food and ensilage used in the fish farming industry.

| Location | ^{99}Tc (Bq kg^{-1} d.w.) ¹ |
|---------------------------|--|
| Fish meal (8 samples) | < 0.09 to 0.35 |
| Fish ensilage (2 samples) | 0.42 and 0.44 |

¹Analysed by the UMB

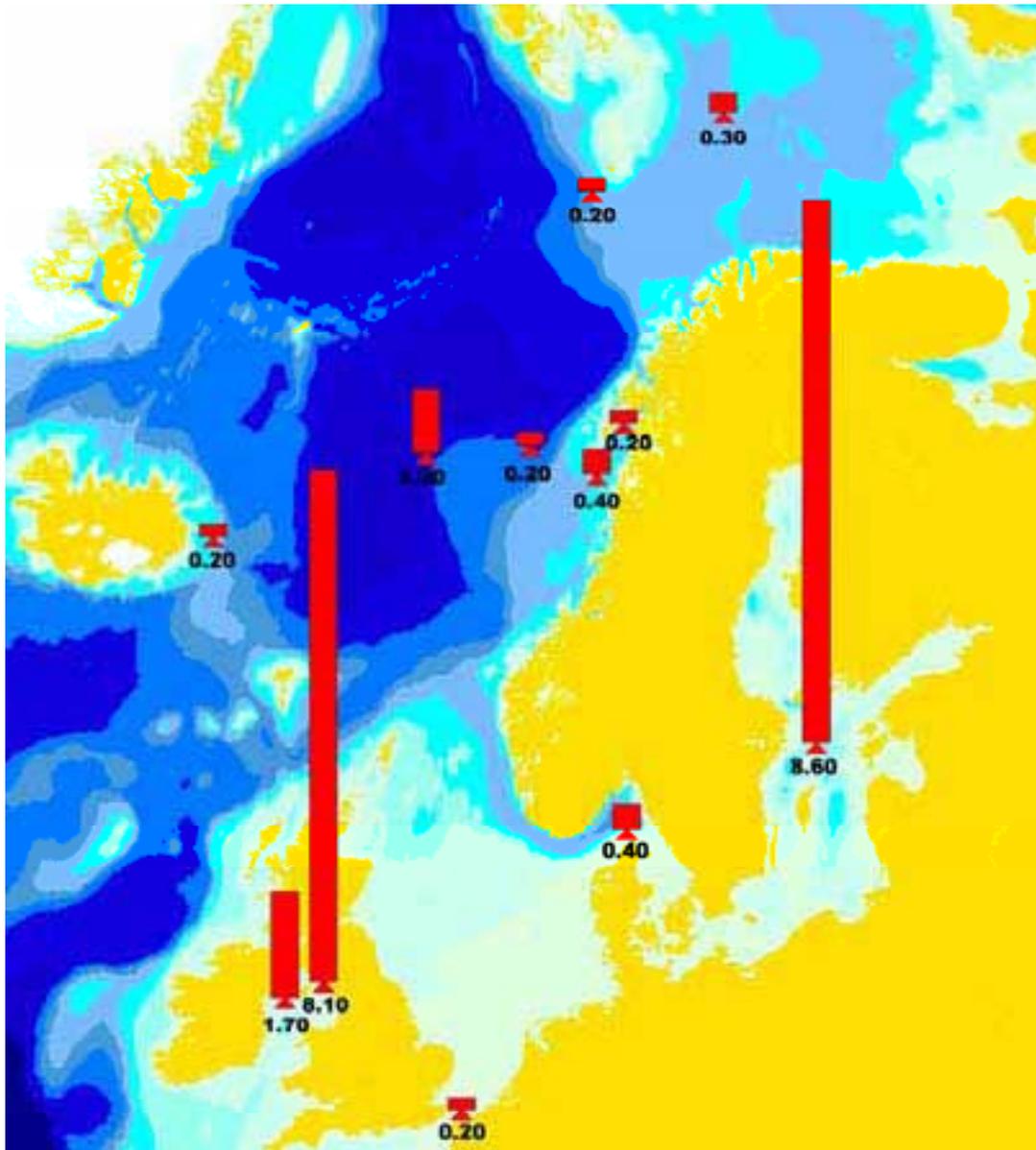


Figure 5.12. Levels of ^{137}Cs in cod from different marine area (data from EA, EHS, FSA & SEPA (2004) and this report) in 2003.

6 Summary and conclusions

In Norway there are currently two monitoring programmes concerned with radioactivity in the marine environment coordinated by the Norwegian Radiation Protection Authority (NRPA). One is funded by the Ministry of the Environment, which focuses on monitoring of radioactivity in the marine environment both in coastal areas and in the open seas, and the other by the Ministry of Fisheries, which focuses on monitoring radioactivity in commercially important fish species. Results of both these programmes are presented in this report. In addition, results obtained by the Food Control Authority from a monitoring programme concerned with radioactivity in marine fish are included.

The collection and updating of discharge data from Norwegian sources and data concerned with the long-range transport of radionuclides from various sources are included in the marine monitoring programme. Liquid discharge data for 2003 from nuclear installations and recent trends in such discharges are summarised, together with the available information concerning nuclear weapons fallout and the outflow of ^{137}Cs of Chernobyl origin from the Baltic Sea. In addition, data regarding the discharges of ^{226}Ra and ^{228}Ra in 2003 from produced water from the North Sea oil and gas industry are included.

In 2003 samples of sea water, sediment, and/or other biota were collected in the Norwegian Sea/Barents Sea, in selected fjords and at a number of coastal stations, including those off the islands of Svalbard, Bjørnøya, Hopen and Jan Mayen. Different species of fish were collected in the Barents Sea, the Norwegian Sea and in coastal areas along the entire Norwegian coast.

6.1 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2003 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 $0.17 \mu\text{Sv}$ in 2003, which correspond to 17 % of the dose limit. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be $0.014 \mu\text{Sv}$ in 2003, corresponding to 1.4 % of 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.

Unsealed radioactive substances are used in hospitals, research laboratories and various industrial activities. Among the most abundant radionuclides are $^{99\text{m}}\text{Tc}$, ^{131}I and ^3H . Regarding the radiological impact on the public of the discharge of these substances, ^{131}I is one of the most important radionuclides. The amount of ^{131}I sold in Norway in 2003 was 1.876 TBq.

Produced water from offshore oil production may contain enhanced levels of naturally occurring radium isotopes. A survey of ^{226}Ra and ^{228}Ra in produced water from 41 Norwegian platforms in 2003 showed that the average activity concentration of these radionuclides were 3.3 Bq l^{-1} and 2.8 Bq l^{-1} . The total activity of ^{226}Ra and ^{228}Ra discharged from all 41 production platforms in 2003 have been estimated to be 440 GBq and 380 GBq, respectively, where about 40 % was discharged from only 2 of the 41 platforms, Troll B and Troll C.

The long-range transport of radionuclides originating from nuclear weapons fallout, reprocessing of spent nuclear fuel and from the Chernobyl accident 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.

6.2 Radioactivity in sea water and sediment

In 2003, samples of sea water and sediment were collected in the Norwegian Sea and the Barents Sea, and at coastal sampling stations, and analysed with respect to ^{137}Cs , ^{99}Tc , ^{90}Sr , ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am . A general trend seen in most samples is that the levels of radioactivity are similar, or slightly lower than have been observed in recent years.

6.2.1 Caesium-137 in sea water and sediment

Observed levels of ^{137}Cs in surface water in the Norwegian Sea/Barents Sea and along the Norwegian coast in 2003 ranged from 0.8 to 7.5 Bq m⁻³. The highest activity concentrations were found in the coastal waters of southern and central Norway. The levels are similar to those found in the same area in 2002.

Samples of surface sediment were collected in the Barents Sea/Norwegian Sea area and in fjords along the Norwegian coast. Activity concentrations in sediment samples collected in the open sea were all relatively low, up to a few Bg kg⁻¹ (d.w.). The highest levels were found in coastal areas. These levels are however much lower than those encountered in 2002 in fjords affected by Chernobyl fallout.

6.2.2 Technetium-99 in sea water

Samples of sea water were collected in the Norwegian Sea, along the Norwegian coast and north west of Svalbard. Levels of ^{99}Tc in surface water ranged from 0.07 to 1.7 Bq m⁻³. The highest concentrations were found off the southern and central coast of Norway. The levels are similar to those found in 2001 and 2002 in the same areas. The annual average obtained from monthly sampling at Hillesøy

shows a slightly lower activity concentration in 2003 than in 2002.

6.2.3 Strontium-90 in sea water

Sea water samples collected in the Barents Sea showed activity concentrations in the range 0.8 to 3.3 Bq m⁻³. The highest levels were found in samples from the coastal stations at Lista, Narestø and Tjøme in the southern part of Norway.

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

Observed levels of $^{239+240}\text{Pu}$ in the the Norwegian Sea and in samples collected along the Norwegian coast in 2003 ranged from 3.1 to 19.9 mBq m⁻³, where the highest level was found in water off the coast in central Norway. The activity concentrations of ^{241}Am in sea water samples from the same area were generally lower and ranged from 1.5 to 5.0 mBq m⁻³. Samples collected at the coastal stations at Narestø and Tjøme showed activity concentrations of $^{239+240}\text{Pu}$ of 4.8 and 4.9 mBq m⁻³, which are similar to the levels found in 2002.

6.3 Radioactivity in biota

Samples of biota included different species of seaweed, crustaceans, molluscs and fish analysed, foremost, for ^{137}Cs and ^{99}Tc . A number of samples of marine mammals, such as polar bear and seal, were also analysed. For seaweed samples the levels of ^{99}Tc are still relatively high, but have shown a slightly decreasing trend, while the ^{137}Cs levels have been relatively stable in recent years.

6.3.1 Technetium-99 in seaweed

Samples of seaweed were collected along the Norwegian coast and in the Svalbard region, and analysed for ^{99}Tc . Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 100 to 340 Bq kg⁻¹ (d.w.). For most stations the levels were of similar magnitude in

2003 compared to 2002. Monthly sampling at Hillesøy, Utsira, and Steinshamn all indicate that the peak of ^{99}Tc has passed these sites and that the levels are declining. They are, however, still expected to be elevated the next few years. In contrast, samples of seaweed (*F. distichus* and *L. digitata*) from Svalbard have not yet shown decreasing levels compared with samples collected in 2001 and 2002.

6.3.2 Caesium-137 in seaweed

The activity concentration of ^{137}Cs in *F. vesiculosus* sampled at the coastal stations was in the range 0.35 to 2.8 Bq kg⁻¹ (d.w.), where the highest levels were found in the southern part of Norway, where the impact from outflowing Baltic Sea water is highest. Apart from a higher activity concentration of ^{127}Cs in the water, lower salinity also leads to a higher uptake of ^{137}Cs in the seaweed. A comparison with results from 2000 and 2001 shows that the levels have been relatively stable in recent years.

6.3.3 Plutonium-239+240 in seaweed

At two sites, Hillesøy and Utsira, seaweed (*F. vesiculosus*) has also been analysed for $^{239+240}\text{Pu}$. The activity concentration of $^{239+240}\text{Pu}$ in seaweed at these locations has in the period 1998 - 2003 been in the range from 40 to 130 mBq kg⁻¹ (d.w.). Annual sampling at Utsira since 1980 reveals relatively large fluctuations of $^{239+240}\text{Pu}$ in seaweed samples.

6.3.4 Technetium-99 and caesium-137 in crustaceans and molluscs

Samples of lobster (*Homarus gammarus*), mussels (*Mytilus edulis*) and other species have been sampled at different locations and analysed for ^{99}Tc and ^{137}Cs . The observed levels of ^{137}Cs were below 1 Bq kg⁻¹ (w.w.) in all samples. For ^{99}Tc , the activity concentrations were also low, generally below 1 Bq kg⁻¹ (w.w.), except for lobster, where levels up to 44 Bq kg⁻¹ (w.w.) were found.

6.3.5 Technetium-99 and caesium-137 in fish

Different species of fish, commercially important and others, have been sampled in the Barents Sea, Norwegian Sea, the North Sea and in coastal areas around Norway. Analyses of ^{137}Cs show low activity concentrations. The level of ^{137}Cs was generally below 0.5 Bq kg⁻¹ (w.w.). The highest measured activity concentration of ^{137}Cs , 1.2 Bq kg⁻¹ (w.w.), was found in Saithe (*Pollachius virens*) caught in More og Romsdal. The levels of ^{99}Tc in various fish species were also low and ranged from <0.01 to 0.09 Bq kg⁻¹ (w.w.).

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Appendix

Analytical methods

During recent years, laboratories at the Norwegian Radiation Protection Authority, the local office of the Directorate of Fisheries in Tromsø, Labora A/S (previously the Norwegian Food Control Authority in Salten) and the Institute of Marine Research have all 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.

The Norwegian Radiation Protection Authority (NRPA)

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

Determination of ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am activity

The concentrations of ^{238}Pu , $^{239+240}\text{Pu}$ 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). Different radiochemical separation techniques were

applied to separate plutonium and americium from other nuclides using solvent extraction with 10 % TIOA/xylene solution and ion-exchange chromatography with a BIO-RAD AG1-X4 (100-200 mesh) column. Purified americium and plutonium fractions were electrodeposited on stainless steel discs and the activity measured in semiconductor silicon detectors. As alpha spectrometry is not able to distinguish between ^{239}Pu and ^{240}Pu , because the energies of their emitted alpha particles are too close to be resolved, these isotopes are measured and reported as the sum $^{239+240}\text{Pu}$. Relative efficiencies of the detectors were in the range 25 to 30 %. 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 75 %.

Determination of ^{99}Tc activity

To determine the activity concentration of ^{99}Tc in sea water, samples of 50 litres were filtered through a 1-micron polypropylene cartridge to remove suspended particulate matter. Seaweed and other biota samples were dried, milled and homogenized. A 10-20 g dried sample was transferred to a specially designed bottle and carbonised and then dissolved by adding concentrated H_2SO_4 followed by HNO_3 . $^{99\text{m}}\text{Tc}$ was added to all samples for chemical recovery determination.



Figure A.1. Fifty litres of sea water is passed through an ion-exchange column.

The analytical procedure used is a modified version of that developed by Risø National Laboratory, Denmark (Chen *et al.*, 2001). The procedure is outlined briefly below. Technetium is initially separated from the matrix by ion-exchange chromatography using a BIO-RAD AG1-X4 (100-200 mesh) column (Figure A.1) and separation techniques such as precipitation and solvent extraction are then applied before the technetium is electro-deposited onto stainless steel discs (Figure A.2). The chemical yields were determined by gamma counting of the ^{99m}Tc tracer in a NaI well-type detector. Typically, the radio-chemical yields varied between 70 % and 85 %. After one week, the ^{99}Tc activity was measured using a low-background anti-coincidence beta counter (Model Risø GM-25-5).



Figure A.2. Dried sample material in different containers ready for gamma measurements. To the right, ^{99}Tc preparations ready for counting.

The limits of detection for 10 g seaweed and 50 l sea water have been calculated to be approximately 0.5 Bq kg^{-1} (d.w.) and 0.10 Bq m^{-3} , respectively. The limit of detection may vary slightly owing to variations in chemical yield, counting efficiency and the mass of the sample. The total uncertainty in ^{99}Tc analysis is normally around 10 %.

Determination of ^{210}Po activity

The analytical procedure for the determination of ^{210}Po has been described by Chen *et al.* (2001). Samples of muscle tissue were wet ashed with nitric and hydrochloric acid (followed by conc. HCl and H_2O_2) after the

addition of ^{209}Po as a yield determinant. Finally, polonium was spontaneously deposited onto silver discs at 90°C for 2 hours from a dilute hydrochloric acid solution. After source preparation ^{210}Po activity was measured by alpha spectrometry using ion-implanted silicon detectors.

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 fish and seaweed were dried at 105°C and homogenized, and placed in containers prior to gamma counting. The activity from each fish sample was counted for a minimum of 2 days. In fish and shrimp samples the detection limit of ^{137}Cs at the NRPA is approximately 0.1 Bq kg^{-1} (w.w.).

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 $0.3\text{-}2 \text{ Bq kg}^{-1}$ (d.w.) Uncertainties in the analysis include uncertainties from counting statistics, calibration and sample preparation and are normally in the range 5% to 20%.

The Institute of Marine Research (IMR)

Detection of gamma emitters

Biota and 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.

Institute for Energy Technology (IFE)

Determination of ^{99}Tc

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to ^{99}Tc content. The pretreatment was performed at Kjeller and the analysis at the Department of Radiation Physics, at Lund University. The following analytical procedure was used at the Department of Radiation Physics at Lund University, Sweden. 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 $^{99\text{m}}\text{Tc}$ 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).

Determination of ^{90}Sr in bone samples

^{90}Sr determination was carried out by the Institute of Energy Technology (IFE, Norway) by chemical separation and analysis of the daughter product ^{90}Y . Bone samples were dried to constant weight at 105 °C, and dry ashed at 700 °C after addition of ^{85}Sr tracer and analysed using a modified HASL 300 procedure

(Erickson, 1997; Varskog *et al.* 1997). Following chemical separation of Sr using fuming HNO_3 and precipitation of hydroxide and chromate, the samples were left for in-growth of ^{90}Y . Y was precipitated as oxalate, and collected by filtration, and the ^{90}Y activity measured by low level anticoincidence beta counters. Chemical yield was determined by ^{85}Sr tracer and titration of Y with EDTA.

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.

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. ^{242}Pu was used as a tracer for radiochemical yield determination. The separation process involved solvent extraction with 10 % TIOA/Xylene followed by anion exchange on AG1-X4 columns. The plutonium fraction were then electrodeposited on a stainless steel disc and counted using alpha spectrometry.

Risø National Laboratory

Determination of ^{90}Sr in sea water

Sr was precipitated as strontium carbonate from the sea water. The carbonate was later dissolved in 4 M HNO_3 and Ca was separated by precipitating Sr and Ba nitrates by adding conc. nitric acid and finally fuming nitric acid. Ba was removed by precipitation of BaSO_4 followed by a $\text{Fe}(\text{OH})_3$ to remove yttrium. After a known amount of yttrium carrier had been added, the sample was stored for three weeks to ensure secular equilibrium between ^{90}Sr and ^{90}Y . Yttrium was separated by precipitation of $\text{Y}(\text{OH})_3$ and the activity measured by beta counting. The $\text{Y}(\text{OH})_3$ precipitate was then dissolved in 6 M HNO_3 and yttrium was

precipitated as an oxalate by adding $\text{H}_2\text{C}_2\text{O}_4$ to the solution. After the precipitate had been filtered from the solution and dried, the recovery of yttrium was measured by weighing the oxalate precipitate. The recovery of Sr was determined by using ^{85}Sr as a radiochemical tracer.

The Norwegian University of Life Sciences (UMB)

Determination of ^{99}Tc

Blades and stipes were separated, dried and ground before analysis. Fish was freeze dried, ashed and ground prior to analysis.

Tc-99 was extracted using microwave acid digestion followed by separation on a TEVA ion-exchange column. Tc-99m was used as yield monitor for the extraction. The concentration of Tc-99 in the extracts was determined using Inductively Coupled Plasma - Mass Spectrometry (ICP-MS).

Labora A/S in Salten (previously The Norwegian Food Control Authority (SNT) in Salten)

Detection of gamma emitters

The Laboratory uses an HPGe detector when analysing samples of fish and shrimp with low activity. The detector is an EG&G Ortec GEM (p-type) detector with 45 % relative efficiency. Dried muscle (500 g, pooled sample of 25 fish) was put in a Marinelli beaker and the activity counted for 2 to 3 days. The detection limit is estimated to be 0.1 Bq kg^{-1} (w.w.).

The Laboratory of the Local Office of the Directorate of Fisheries in Tromsø

Detection of gamma emitters

The laboratory of the local office of the Directorate of Fisheries in Tromsø is equipped with a Canberra series 10 portable NaI detector. The detection limit for fish reported by the Directorate is approximately 5.5 Bq kg^{-1} . Samples of 200 g fresh fish containing both muscle tissue and bone were analysed.

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