



## Radioactivity in the Marine Environment 2006

Results from the Norwegian Marine Monitoring Programme (RAME)

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**Key words:**

Radioactivity, marine environment, RAME, monitoring, Norway

**Abstract:**

This report presents results of monitoring of radioactivity in seawater and biota collected along the Norwegian coast, the Norwegian Sea and the Skagerrak in 2006. 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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**Emneord:**

Radioaktivitet, marin miljø, RAME, overvåking, Norge

**Resymé:**

Rapporten inneholder resultater fra overvåkingen av radioaktivitet i sjøvann og biota i 2006 langs norskekysten og i Norskehavet og Skagerrak. 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  $^{99}\text{Tc}$  from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced, it has continued at a high level up to 2003. The discharge of  $^{99}\text{Tc}$  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.

The marine monitoring programme (RAME) is funded by the Ministry of the Environment and focus on monitoring of radioactivity both in coastal areas and in the open sea. The marine monitoring programme also includes 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 2006 from nuclear installations and recent trends in such discharges are summarised in Chapter 2.

During 2006, samples for monitoring radioactivity in the marine environment were collected mainly in the Norwegian Sea and at permanent coastal stations along the Norwegian coastline. Results from the analysis of these samples are presented in Chapters 4 and 5. In Chapter 6, a summary of the findings and the conclusions is given. In the Appendix, technical information regarding sample preparation techniques and analytical methods employed in the laboratories are presented.

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## 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 may occur in connection with 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), the Chernobyl accident in 1986 and from reprocessing of nuclear fuel 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 GE Healthcare

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 GE Healthcare (former 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 2007a; IFE 2006a; IFE, 2005a; IFE, 2004a; IFE, 2003a), 2002-2006 (including GE Healthcare).

| Nuclide               | 2002                | 2003                | 2004                | 2005                | 2006                 |
|-----------------------|---------------------|---------------------|---------------------|---------------------|----------------------|
| <sup>241</sup> Am     | 0.7                 | 0.005               | 0.003               | 0.03                | 0.006                |
| <sup>124</sup> Sb     | <0.06               | <0.2                | <0.1                | <0.1                | <0.3                 |
| <sup>125</sup> Sb     | <0.11               | 2.7                 | 0.075               | <0.7                | 0.7                  |
| <sup>144</sup> Ce     | 1.6                 | 3.3                 | 1.1                 | 0.054               | 2.7                  |
| <sup>134</sup> Cs     | 2.3                 | 1.2                 | 0.35                | 0.08                | 5.3                  |
| <sup>137</sup> Cs     | 25                  | 22                  | 7.9                 | 0.51                | 53.6                 |
| <sup>3</sup> H        | 2.4·10 <sup>6</sup> | 2.8·10 <sup>6</sup> | 2.9·10 <sup>5</sup> | 4.1·10 <sup>5</sup> | 1.43·10 <sup>6</sup> |
| <sup>59</sup> Fe      | <0.15               | <0.2                | <0.3                | <0.2                | <0.5                 |
| <sup>125</sup> I      | 350                 | 540                 | 327                 | 50                  | 42.8                 |
| <sup>131</sup> I      | 6.0                 | 15                  | 27                  | 39                  | 4.2                  |
| <sup>58</sup> Co      | <0.10               | 0.26                | <0.1                | <0.1                | <0.3                 |
| <sup>60</sup> Co      | 60                  | 55                  | 24                  | 6.7                 | 74                   |
| <sup>51</sup> Cr      | <0.5                | <0.6                | <2                  | <1.3                | <0.9                 |
| <sup>54</sup> Mn      | <0.14               | <0.2                | <0.2                | <0.2                | <0.4                 |
| <sup>95</sup> Nb      | 0.17                | 0.42                | <0.06               | <0.1                | 0.04                 |
| <sup>238</sup> Pu     | 0.011               | 0.002               | 0.01                | 0.007               | 0.003                |
| <sup>239+240</sup> Pu | 0.04                | 0.034               | 0.15                | 0.15                | 0.04                 |
| <sup>103</sup> Ru     | <0.07               | 0.14                | 0.10                | <0.3                | 0.02                 |
| <sup>106</sup> Ru     | <0.9                | 2.0                 | <4                  | <2.2                | <2.6                 |
| <sup>90</sup> Sr      | 0.55                | 0.33                | 2.6                 | 0.34                | 1.2                  |
| <sup>110m</sup> Ag    | 1.2                 | 2.1                 | 0.089               | 1.2                 | 0.4                  |
| <sup>65</sup> Zn      | <0.4                | 0.27                | 0.1                 | 8.4                 | <0.9                 |
| <sup>95</sup> Zr      | 0.13                | 0.14                | <0.2                | <0.2                | <0.5                 |

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 NRPA. The reported discharges for the period 2002-2006 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 2007a; IFE 2006a; IFE, 2005a; IFE 2004a; IFE, 2003a).

| 2002 | 2003 | 2004 | 2005 | 2006 |
|------|------|------|------|------|
| 0.18 | 0.17 | 0.07 | 0.02 | 0.48 |

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 2006, the calculated effective dose to this critical group was 0.48 µSv,

corresponding to 48 % 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 2 and 48 % of the dose limit, with an average of 18 % for the period 2002-2006.

### 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 1.5-3 % of the authorised limit. Liquid discharges in the period 2002-2006, 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, 2002 - 2006 (IFE 2007b; IFE 2006b; IFE, 2005b; IFE, 2004b; IFE, 2003b).

| Nuclide            | 2002                | 2003                | 2004                | 2005                | 2006                |
|--------------------|---------------------|---------------------|---------------------|---------------------|---------------------|
| <sup>124</sup> Sb  | 0.53                | -                   | -                   | 0.003               | -                   |
| <sup>125</sup> Sb  | 18                  | 0.06                | 0.002               | 0.037               | 0.0064              |
| <sup>141</sup> Ce  | 0.82                | 0.29                | 0.68                | 0.61                | 0.25                |
| <sup>144</sup> Ce  | 5.7                 | 3.2                 | 16                  | 8.8                 | 3.0                 |
| <sup>134</sup> Cs  | 18                  | 8.8                 | 8.0                 | 18                  | 25                  |
| <sup>137</sup> Cs  | 110                 | 130                 | 70                  | 130                 | 190                 |
| <sup>3</sup> H     | 1.1·10 <sup>6</sup> | 2.7·10 <sup>5</sup> | 5.4·10 <sup>5</sup> | 5.3·10 <sup>5</sup> | 3.0·10 <sup>5</sup> |
| <sup>59</sup> Fe   | 0.32                | -                   | 0.75                | 0.88                | -                   |
| <sup>131</sup> I   | 0.9                 | 3.6                 | 0.68                | 0.42                | 21                  |
| <sup>109</sup> Cd  | 0.33                | 0.016               | -                   | -                   | -                   |
| <sup>58</sup> Co   | 5.1                 | 3.3                 | 8.7                 | 41                  | 4.0                 |
| <sup>60</sup> Co   | 88                  | 62                  | 68                  | 99                  | 86                  |
| <sup>51</sup> Cr   | 150                 | 130                 | 210                 | 180                 | 240                 |
| <sup>54</sup> Mn   | 0.5                 | 0.18                | 0.73                | 2.5                 | 1.1                 |
| <sup>56</sup> Mn   | -                   | -                   | 2.8                 | -                   | -                   |
| <sup>95</sup> Nb   | 12                  | 8.5                 | 11                  | 12                  | 8.3                 |
| <sup>103</sup> Ru  | 0.59                | 0.17                | 0.45                | 0.42                | 0.16                |
| <sup>106</sup> Ru  | 0.18                | -                   | -                   | -                   | -                   |
| <sup>90</sup> Sr   | -                   | 3.3                 | 2.1                 | 4.9                 | 4.4                 |
| <sup>110m</sup> Ag | 0.005               | 0.17                | 0.001               | 0.94                | 0.21                |
| <sup>95</sup> Zr   | 5.4                 | 3.3                 | 4.9                 | 5.3                 | 3.8                 |

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 ( $\mu\text{Sv}$ ) to the critical group from marine exposure pathways as reported by IFE Halden, 2002 - 2006 (IFE 2007b; IFE 2006b; IFE, 2005b; IFE, 2004b; IFE, 2003b).

| 2002 | 2003  | 2004  | 2005  | 2006 |
|------|-------|-------|-------|------|
| 0.02 | 0.014 | 0.016 | 0.025 | 0.02 |

## 2.2 Unsealed radioactive substances in the medical sector

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. The two most used radionuclides in nuclear medicine for diagnostic and therapeutic purposes are  $^{99\text{m}}\text{Tc}$  (which decays to  $^{99}\text{Tc}$  with a half-life of 6 h) and  $^{131}\text{I}$  ( $T_{1/2}$  8.04 d). The discharge of these radionuclides has been estimated according

instructions published by OSPAR. The percentage of the administered doses that is discharged as liquid waste is:

### $^{131}\text{I}$ Ablation therapy

100% of the administered dose.

### $^{131}\text{I}$ Thyrotoxicosis treatment

for in-patients, 50% of the administered dose; for out-patients, 30% of the administered dose; for patients who cannot be classified as in-patients or out-patients, 50% of the administered dose.

### $^{99\text{m}}\text{Tc}/^{99}\text{Tc}$

100% of the amount of  $^{99\text{m}}\text{Tc}$  administered has decayed to  $^{99}\text{Tc}$  and that it is all discharged.

Under these assumptions the total discharged activity of  $^{131}\text{I}$  and  $^{99}\text{Tc}$  in 2006 from the medical sector was 1456 GBq and 0.09 MBq, respectively.

## 2.3 Discharge of produced water containing $^{226}\text{Ra}$ and $^{228}\text{Ra}$

Large volumes of produced water, containing dissolved  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ , are discharged into the sea during oil exploitation. Produced water may be formation water trapped in the reservoirs for millions of years or a mixture of formation water and seawater, when seawater has been injected in order to maintain pressure in the reservoirs during oil and gas production. Although the levels of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  are not technologically enhanced, the natural concentrations of these isotopes in the saline formation water are generally much higher (about 3 orders of magnitude) than the background concentration in seawater (a few mBq per litre).

The concentration of radium in the produced water may change over the lifetime of the well. Injection of large volumes of seawater, 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 2006  $144 \cdot 10^6 \text{ m}^3$  of produced water was discharged to the marine environment from the Norwegian oil and gas industry, while  $29 \cdot 10^6 \text{ m}^3$  was reinjected in the reservoirs. The total activity of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  discharged was about 460 GBq and 390 GBq, respectively. (OLF, 2007). In Fig. 2.1 the activity of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  discharged from Norwegian oil fields are presented. About 40 % of the total discharged activity of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  was discharged from the Troll B and C platforms.



Photo: NRPA

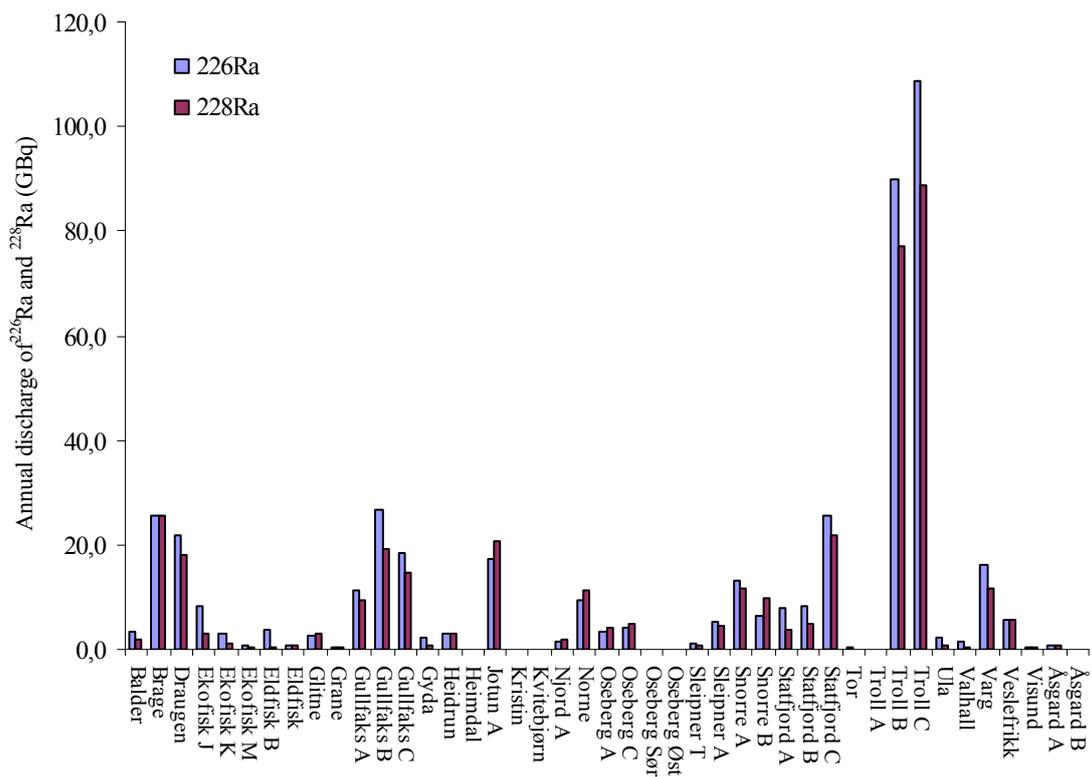


Figure 2.1. Estimated discharged activity of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  from Norwegian oil and gas fields in 2006 (OLF, 2007).

## 2.4 Long-range transport of radionuclides

There are several real and potential sources of radioactive contamination of the northeast Atlantic and the Arctic marine environment, as shown in Figure 2.2.

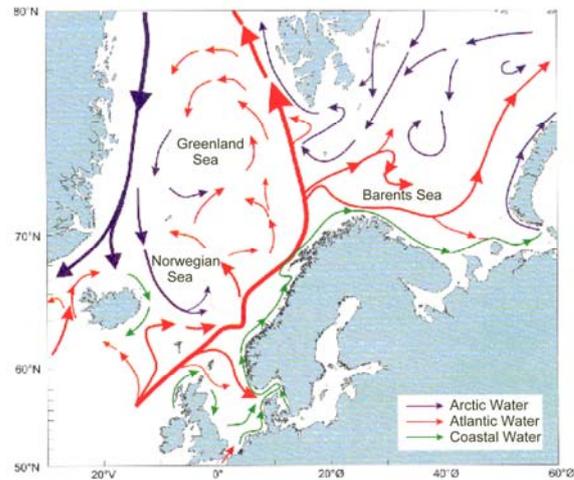


**Figure 2.2.** Sources of radionuclides in the northern marine environment: Chernobyl in the Ukraine, the reprocessing plants at Sellafield, Cap de la Hague and Dounreay, the dumping sites for nuclear waste in the Kara Sea, the sites of the sunken submarines Komsomolets and K-159 and Russian nuclear 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 2.3. Some elements, such as Pu and Am, are particle-reactive and will to a large extent sink and end up in the sediments. From the sediments the radionuclides can later be remobilised and transported away by local currents. Remobilised plutonium and  $^{137}\text{Cs}$  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  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ , are not particle-reactive and will follow currents and can be transported large distances away from the discharge point.



**Figure 2.3.** 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 ( $^3\text{H}$ ), the discharge of liquid radioactive waste has generally been lower here than from Sellafield. In 1994 and 1995, the discharge of  $^{99}\text{Tc}$  from the reprocessing facility at Sellafield in the UK increased sharply, and although this discharge has been reduced, it was continued at a high level up to 2003. The discharge of  $^{99}\text{Tc}$  has been substantially reduced since 2004, when a new chemical treatment process that removes  $^{99}\text{Tc}$  was implemented.

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 2005 is presented in Table 2.5.

In addition to the direct discharges from re-processing, the remobilisation of  $^{137}\text{Cs}$  and plutonium from contaminated sediments in the Irish Sea acts as a secondary source of radionuclides in the marine environment. Substantial discharges of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  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 TBq  $^{239+240}\text{Pu}$  and 86 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 radio-nuclides in the North Sea and Norwegian coastal areas, and will remain so for a long time.

**Table 2.5.** Liquid discharge of radionuclides (TBq) from Sellafield, Cap de la Hague, Springfields and Dounreay to the marine environment in 2006 (OSPAR, 2008 and Environment Agency *et al.*, 2007).

| Nuclide               | Cap de la Hague   | Sellafield | Springfields      | Dounreay          |
|-----------------------|-------------------|------------|-------------------|-------------------|
| $^3\text{H}$          | $1.11 \cdot 10^4$ | 1090       | -                 | $9.7 \cdot 10^4$  |
| Total- $\alpha$       | $2.50 \cdot 10^2$ | 0.205      | 0.08              | $4.17 \cdot 10^4$ |
| Total- $\beta$        | 7.55              | 29         | 20.7              | $3.2 \cdot 10^4$  |
| $^{14}\text{C}$       | 7.46              | 10.9       | -                 | -                 |
| $^{54}\text{Mn}$      | $7.54 \cdot 10^3$ | -          | -                 | -                 |
| $^{57}\text{Co}$      | $2.80 \cdot 10^4$ | -          | -                 | -                 |
| $^{58}\text{Co}$      | $4.48 \cdot 10^4$ | -          | -                 | -                 |
| $^{60}\text{Co}$      | 0.21              | 0.14       | -                 | -                 |
| $^{63}\text{Ni}$      | $6.18 \cdot 10^2$ | -          | -                 | -                 |
| $^{65}\text{Zn}$      | $4.24 \cdot 10^5$ | -          | -                 | -                 |
| $^{90}\text{Sr}$      | 0.216             | 4.96       | -                 | $9.63 \cdot 10^2$ |
| $^{99}\text{Tc}$      | $4.47 \cdot 10^2$ | 5.62       | 0.065             | -                 |
| $^{106}\text{Ru}$     | 4.80              | 3.51       | -                 | -                 |
| $^{125}\text{Sb}$     | $9.64 \cdot 10^2$ | -          | -                 | -                 |
| $^{129}\text{I}$      | 1.32              | 0.198      | -                 | -                 |
| $^{134}\text{Cs}$     | $6.05 \cdot 10^2$ | 0.154      | -                 | -                 |
| $^{137}\text{Cs}$     | 0.623             | 5.93       | -                 | $1.17 \cdot 10^2$ |
| $^{144}\text{Ce}$     | $5.45 \cdot 10^4$ | 0.553      | -                 | -                 |
| $^{154}\text{Eu}$     | $1.55 \cdot 10^3$ | -          | -                 | -                 |
| $^{155}\text{Eu}$     | $4.56 \cdot 10^4$ | -          | -                 | -                 |
| $^{239+240}\text{Pu}$ | $1.75 \cdot 10^3$ | 0.147      | -                 | -                 |
| $^{241}\text{Pu}$     | 0.148             | 3.64       | -                 | -                 |
| $^{241}\text{Am}$     | $3.03 \cdot 10^3$ | 0.0518     | -                 | -                 |
| $^{237}\text{Np}$     | $6.86 \cdot 10^5$ | 0.0548     | 0.00158           | -                 |
| $^{242}\text{Cm}$     | $2.42 \cdot 10^5$ | -          | -                 | -                 |
| $^{243+244}\text{Cm}$ | $2.55 \cdot 10^3$ | -          | -                 | -                 |
| $^{230}\text{Th}$     | -                 | -          | 0.0119            | -                 |
| $^{232}\text{Th}$     | -                 | -          | $3.10 \cdot 10^4$ | -                 |
| U                     | -                 | -          | 0.026             | -                 |
| U                     | 32.0 kg           | 439 kg     | -                 | -                 |

#### 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 PBq  $^{137}\text{Cs}$ , 622 PBq  $^{90}\text{Sr}$ , 6.52 PBq  $^{239}\text{Pu}$  and 4.35 PBq  $^{240}\text{Pu}$  have been released to the environment (UNSCEAR, 2000a), 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  $^{137}\text{Cs}$  in surface water is about a few Bq  $\text{m}^{-3}$  (about twice as much as  $^{90}\text{Sr}$ ), 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  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ . 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  $^{137}\text{Cs}$ , with a physical half-life of

30 years. In total about 85 PBq of  $^{137}\text{Cs}$  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  $^{137}\text{Cs}$  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 average activity concentration of  $^{137}\text{Cs}$  in the southern Baltic Sea surface water in 2006 was reported to 42 Bq  $\text{m}^{-3}$  (Zalewska, 2007). 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  $^{137}\text{Cs}$  will subsequently be transported by the Baltic current which continues as the Norwegian coastal current. This is one of the major sources of  $^{137}\text{Cs}$  in the Norwegian marine environment, together with remobilised  $^{137}\text{Cs}$  from the seabed of the Irish Sea.

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

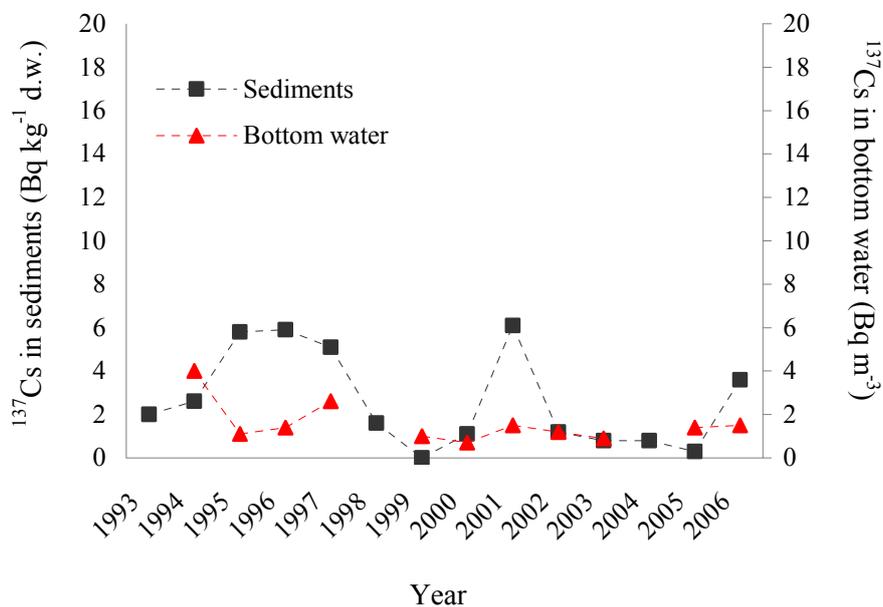
In addition to discharges from the western European reprocessing plants, northern marine waters have also received an input of various radionuclides from Russian reprocessing plants situated on the tributaries of the Russian rivers Ob and Yenisey (see Figure 2.2). These are the Mayak Production Association 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  $^{137}\text{Cs}$  into the Kara Sea; a level which is of a similar order of magnitude to inputs from Sellafield-derived  $^{137}\text{Cs}$  to the area (Vakulovsky *et al.*, 1995). However, the fact that the prevailing surface currents tend to transport contamination away from the Kara Sea in predominantly easterly and northerly directions, suggests that the resultant inputs to

the Barents Sea has been relatively low. Nonetheless, there is a potential for significant radionuclide contamination following large accidental releases of radioactivity from these nuclear complexes.

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

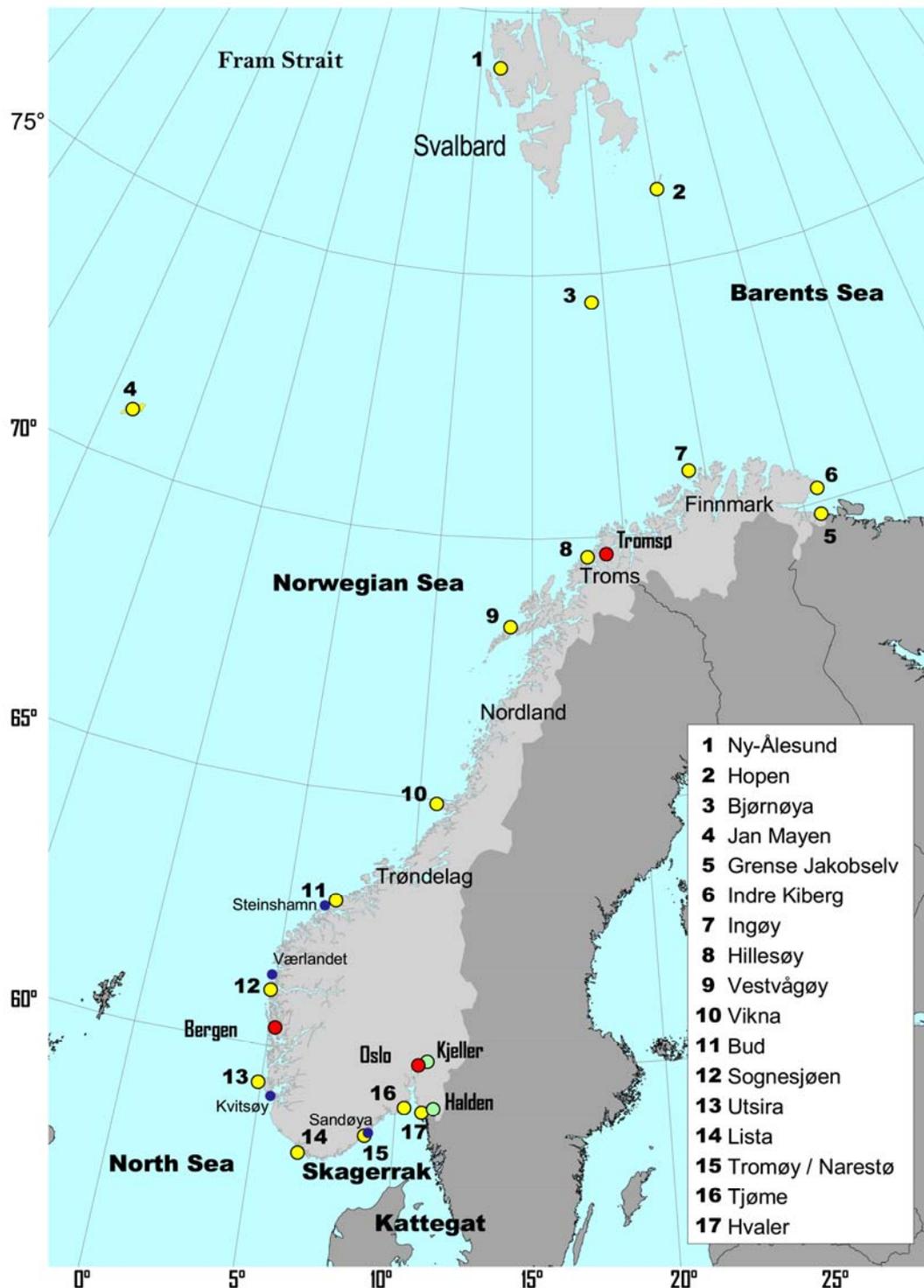
According to Strand *et al.* (1998), elevated levels of radionuclides in sediments collected in the vicinity of the dumped objects demonstrated that leakage had occurred, but no significant inputs of radionuclides to the open sea from this source have been detected. Following an accident on April 7, 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. Kolstad (1995) reported low activity concentrations of  $^{134}\text{Cs}$  in seawater samples collected near the submarine, showing that 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  $^{137}\text{Cs}$  are presented in Figure 2.4.



**Figure 2.4.**  $^{137}\text{Cs}$  in samples from the position of the sunken nuclear submarine "Komsomolets".

### 3 Collection of samples

In 2006 samples were collected mainly in the Norwegian Sea, in selected fjords and at permanent coastal stations. A geographic overview of the sampling area covered by the marine monitoring programme, with the fixed coastal stations marked, is shown in Figure 3.1.



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

Sampling and analysis were carried out by the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR) and the Institute for Energy Technology (IFE).

In 2006 samples from the Norwegian Sea, were collected in the period April 27 – May 10 by IMR and NRPA from the vessels R/V “G.O. Sars” (Fig. 3.2). During the expeditions samples of surface water were collected which were later analysed for  $^{226}\text{Ra}$ ,  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{241}\text{Am}$  and plutonium isotopes. Sediment was also sampled and later analysed for  $^{137}\text{Cs}$ . A few samples were also collected in the Kategatt in April by IMR.



Fig. 3.2. R/V G O Sars in Bergen (Photo: NRPA).

### 3.1 Seawater

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

For the analysis of  $^{137}\text{Cs}$ ,  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters are used as sorbents (Roos *et al.*, 1994). The system consists of a prefilter (1 micron) and two  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters connected in series (Fig 3.3). Assuming identical collection efficiency for the  $\text{Cu}_2[\text{Fe}(\text{CN})_6]$ -impregnated cotton filters, the amount of caesium passing through the prefilter can be calculated by considering the two filters as the first two terms in a geometrical series, which has the sum:

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

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



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

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



*Fig. 3.4. CTD/Rosette multi-bottle sampler.*

### **3.2 Sediment**

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

### **3.3 Seaweed**

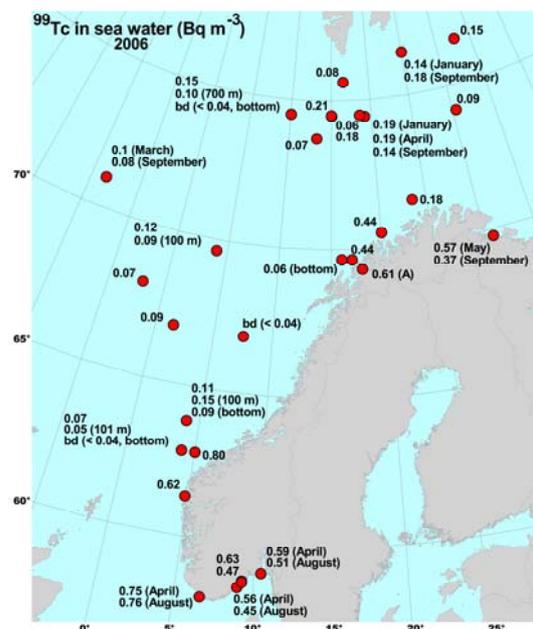
The NRPA collected seaweed (*Fucus vesiculosus*) samples from eight stations along the Norwegian coastline. At Hillesøy in northern Norway, seaweed is collected every month and at the other locations once per year. In addition, IFE performs monthly or annual seaweed sampling at eleven locations along the coastline, from the Russian border in the north to the Swedish border in the south.

## 4 Radioactivity in seawater and sediments

### 4.1 Technetium-99 in seawater

Tc-99 has a physical half-life of  $2.13 \cdot 10^5$  years, and originates from the decay of  $^{99}\text{Mo}$ , which is formed either as a fission product from  $^{235}\text{U}$  or  $^{239}\text{Pu}$  or by neutron activation of  $^{98}\text{Mo}$ . The main source of  $^{99}\text{Tc}$  in Norwegian waters is liquid discharge from the reprocessing plant at Sellafield. In oxygenated seawater  $^{99}\text{Tc}$  is present as the highly soluble pertechnetate ion ( $\text{TcO}_4^-$ ). Due to its conservative behaviour in seawater,  $\text{TcO}_4^-$  has the ability to be transported by ocean currents without being significantly affected by sedimentation processes. From the Irish Sea,  $^{99}\text{Tc}$  is transported by ocean currents to the North Sea and via the Norwegian Coastal Current up to the Barents Sea. The transit time (the time between a specific discharge and the maximum activity concentration from that discharge reaching the sampling location) for  $^{99}\text{Tc}$  to reach this area (Hillesøy) from the Irish Sea has been estimated to 3-4 years (Brown *et al.*, 2002; Dahlgaard, 1995).

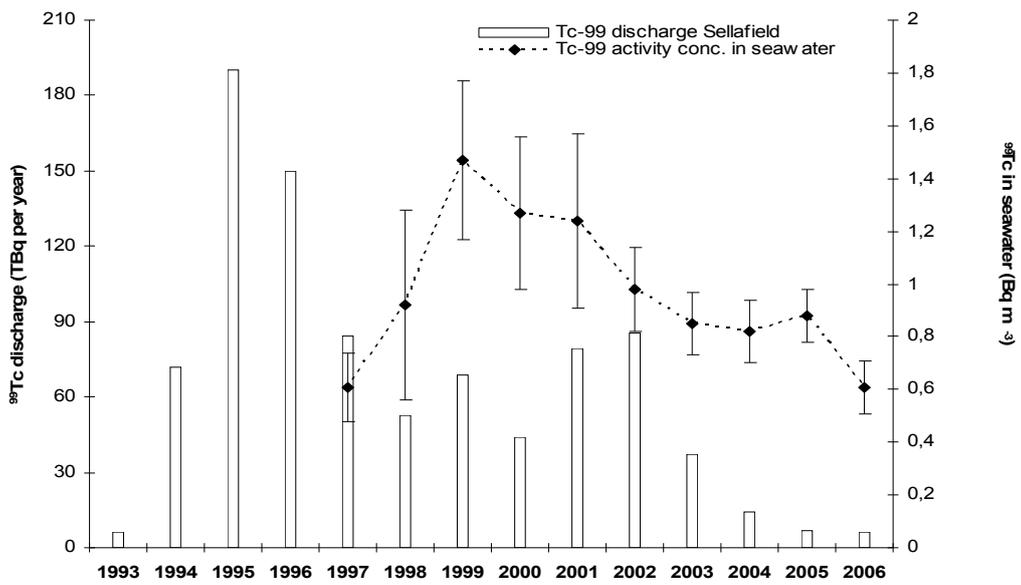
During 2006, seawater samples were collected in the Norwegian Sea, the Barents Sea and along the coast and analysed for  $^{99}\text{Tc}$ . The results are presented in Figure 4.1. In the Norwegian Sea the activity concentration of  $^{99}\text{Tc}$  ranged from  $< 0.04$  to  $0.8 \text{ Bq m}^{-3}$ . The activity concentration of  $^{99}\text{Tc}$  in the Skagerrak ranged from  $0.45$  to  $0.76 \text{ Bq m}^{-3}$ . Highest activity concentration was observed in samples collected in the Norwegian coastal current.



**Figure 4.1.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{99}\text{Tc}$  in seawater samples collected in the Norwegian Sea and along the coast in 2006.

The activity concentrations found in the Norwegian Sea and the Skagerrak are generally lower than those observed in 2003 (NRPA, 2005). Monthly samples have also been collected at Hillesøy (Fig. 4.2). Average activity concentration in seawater at Hillesøy in 2006 was  $0.61 \pm 0.10 \text{ Bq m}^{-3}$ , which is slightly lower than reported in 2005.

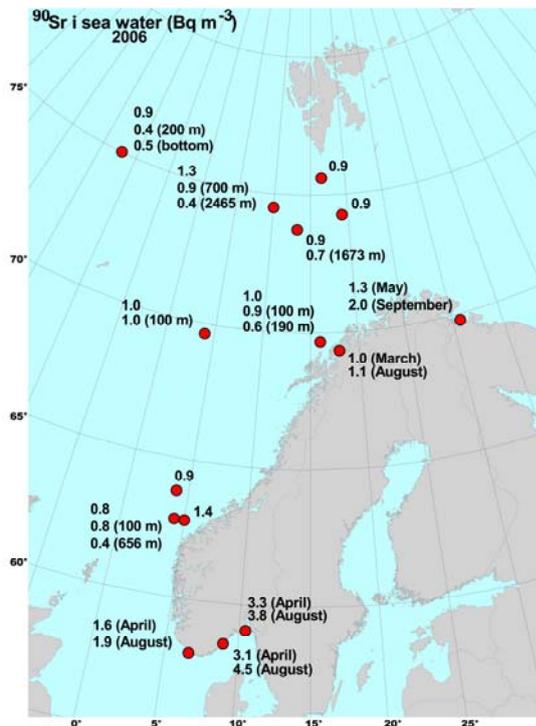
The activity concentrations of  $^{99}\text{Tc}$  found at the coastal stations at Tjøme, Narestø and Lista in the southern part of Norway are slightly lower than those found in 2005.



**Figure 4.2.** Annual average activity concentration ( $Bq\ m^{-3}$ ) of  $^{99}Tc$  in seawater at Hillesøy from monthly sampling and annual discharge of  $^{99}Tc$  from Sellafield.

## 4.2 Strontium-90 in seawater

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



**Figure 4.3.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{90}\text{Sr}$  in surface water samples collected in the Norwegian Sea and the Skagerrak in 2006.

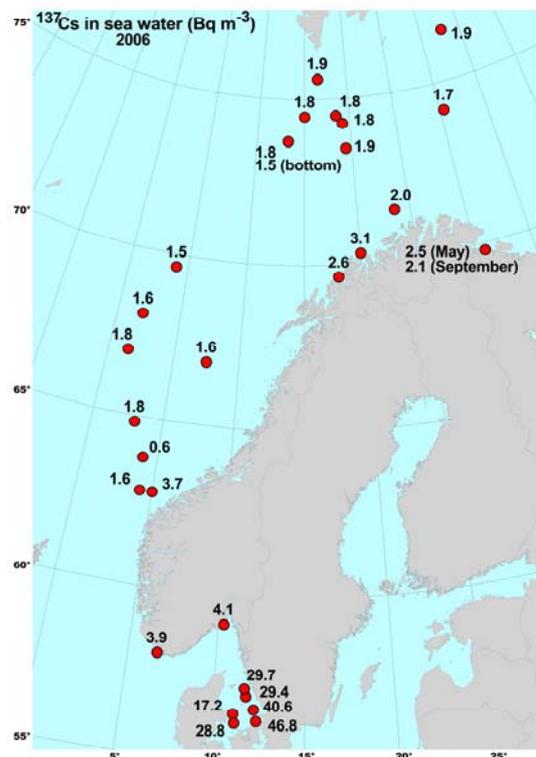
Seawater has been sampled in the Norwegian Sea and the Skagerrak and analysed for  $^{90}\text{Sr}$ . The main sources of  $^{90}\text{Sr}$  in these waters are discharge of liquid waste from reprocessing plants (mainly Sellafield), fallout from atmospheric nuclear weapons tests conducted mainly in the 1950s and 1960s and outflow of water from the Baltic Sea. The annual discharge of  $^{90}\text{Sr}$  from Sellafield between 1998 and 2006 has been in the range of 5 to 31 TBq. The results for 2006 are presented in Figure 4.3. Typical activity concentration in Baltic Sea seawater has been reported to be in the range from 8 to 12  $\text{Bq m}^{-3}$  (Vartti *et al.*, 2006).

In the Norwegian Sea and in the Skagerrak, the activity concentration ranged from 0.4  $\text{Bq m}^{-3}$  to 4.5  $\text{Bq m}^{-3}$ . The highest concentrations were found off the southern coast of Norway in the Skagerrak. The activity concentrations are

similar to the levels observed in 2003 in the same area (NRPA, 2005).

## 4.3 Caesium-137 in seawater and sediment

Caesium-137 is a fission product with a half-life of 30 years. The main sources of  $^{137}\text{Cs}$  in the North Sea and the Skagerrak are fallout from atmospheric nuclear weapons tests in the 1950s and 60s, outflowing water from the Baltic Sea and  $^{137}\text{Cs}$  remobilised from Irish Sea sediments. Runoff from land, from the areas with the highest Chernobyl fallout can also contribute locally in coastal water. Like  $^{99}\text{Tc}$  and  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  is also a conservatively behaving radionuclide in seawater.



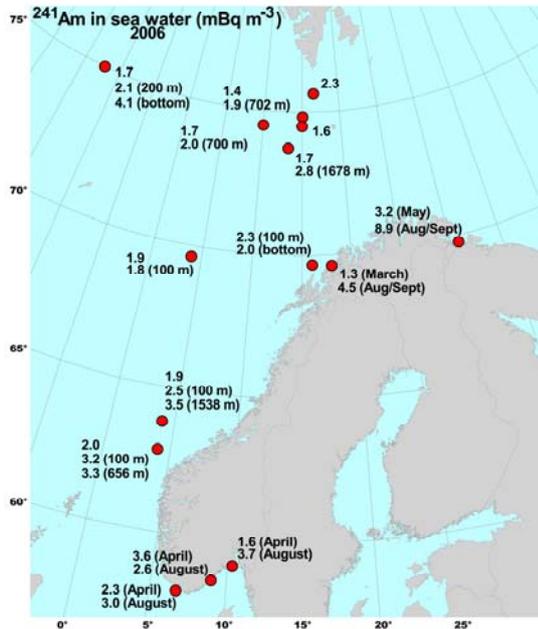
**Figure 4.4.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{137}\text{Cs}$  in seawater samples (surface water) collected in the Norwegian Sea, the Skagerrak and the Barents Sea in 2006.

Observed levels of  $^{137}\text{Cs}$  in the Norwegian Sea, the Barents Sea, the Skagerrak and the Kattegat in 2006 are shown in Figure 4.4.

The activity concentration in surface water ranged from 0.6 to 3.7  $\text{Bq m}^{-3}$ , in the Norwegian Sea/Barents Sea area. This is similar to the levels observed in 2003 in the same area.



years. It is produced, for example, in nuclear reactors by successive neutron capture by  $^{239}\text{Pu}$ . Am-241 is finally formed by beta-decay of  $^{241}\text{Pu}$  ( $T_{1/2} = 14.35$  y). Main sources of  $^{241}\text{Am}$  in the environment are fallout of  $^{241}\text{Pu}$  from nuclear weapon tests in the 1950s and '60s and the discharge of  $^{241}\text{Am}$  and  $^{241}\text{Pu}$  from reprocessing plants.



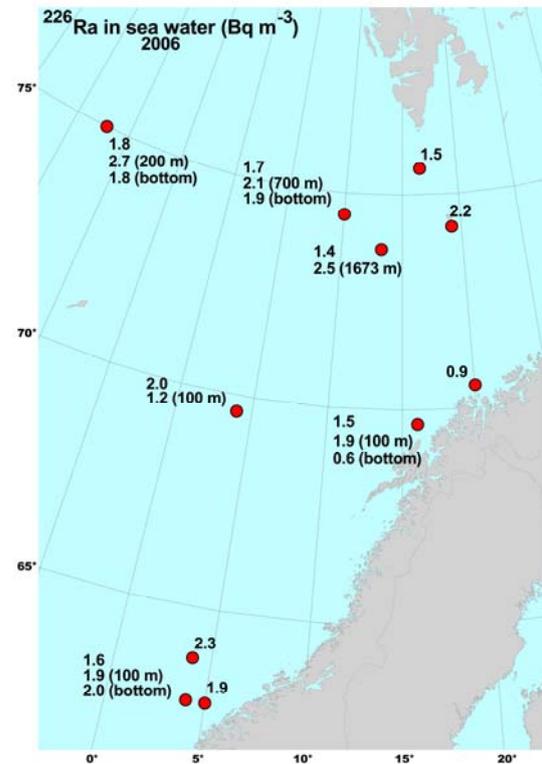
**Figure 4.8.** Activity concentration ( $\text{mBq m}^{-3}$ ) of  $^{241}\text{Am}$  in seawater from the Norwegian Sea and along the coast in 2006.

Measured activity concentrations of  $^{241}\text{Am}$  in the Norwegian Sea in 2006 are presented in Figure 4.8 and range from 1.3 to  $8.9 \text{ mBq m}^{-3}$ . The levels of  $^{241}\text{Am}$  are similar to what was found in the Norwegian Sea in 2003.

#### 4.6 Radium-226 in seawater

Radium-226 is a naturally occurring radionuclide with a physical half-life of 1 600 years. As a member of the natural decay chain starting with  $^{238}\text{U}$  ( $t_{1/2} = 4.47 \cdot 10^9$  years), it is continuously produced by the decay of  $^{230}\text{Th}$ , and can be found in different concentrations in the environment. In the marine environment  $^{226}\text{Ra}$  is naturally supplied from both the sediments (the mother nuclide  $^{230}\text{Th}$  is particle reactive and is mainly found attached to sediment particles) and by river water to the oceans. Typical activity concentration of  $^{226}\text{Ra}$  in Atlantic surface water has been reported to be about  $1.3 \text{ Bq m}^{-3}$  (IAEA, 1990). An anthropogenic source of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in the

North Sea is produced water from oil reservoirs. Due to the relatively high solubility of radium, produced water can contain more than a thousand times the activity concentration normally found in seawater.



**Figure 4.9.** Activity concentration ( $\text{Bq m}^{-3}$ ) of  $^{226}\text{Ra}$  seawater from the Norwegian Sea in 2006.

The activity concentration of  $^{226}\text{Ra}$  observed in the Norwegian Sea in 2006 were in the range of  $0.6 - 2.7 \text{ Bq m}^{-3}$  (Fig. 4.9).

## 5 Radioactivity in biota

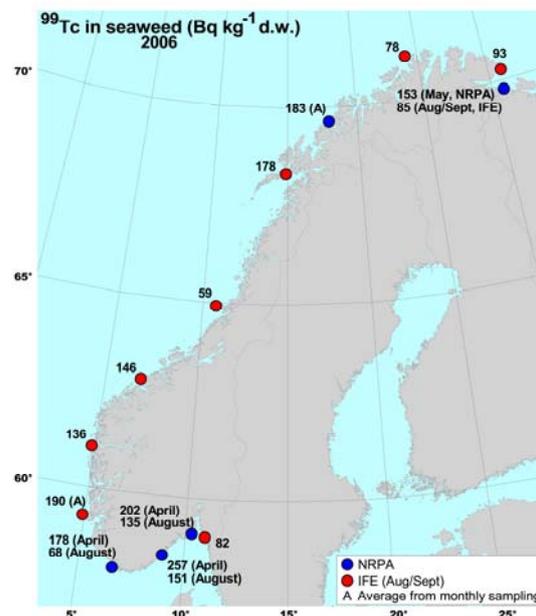
### 5.1 Technetium-99 in seaweed



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

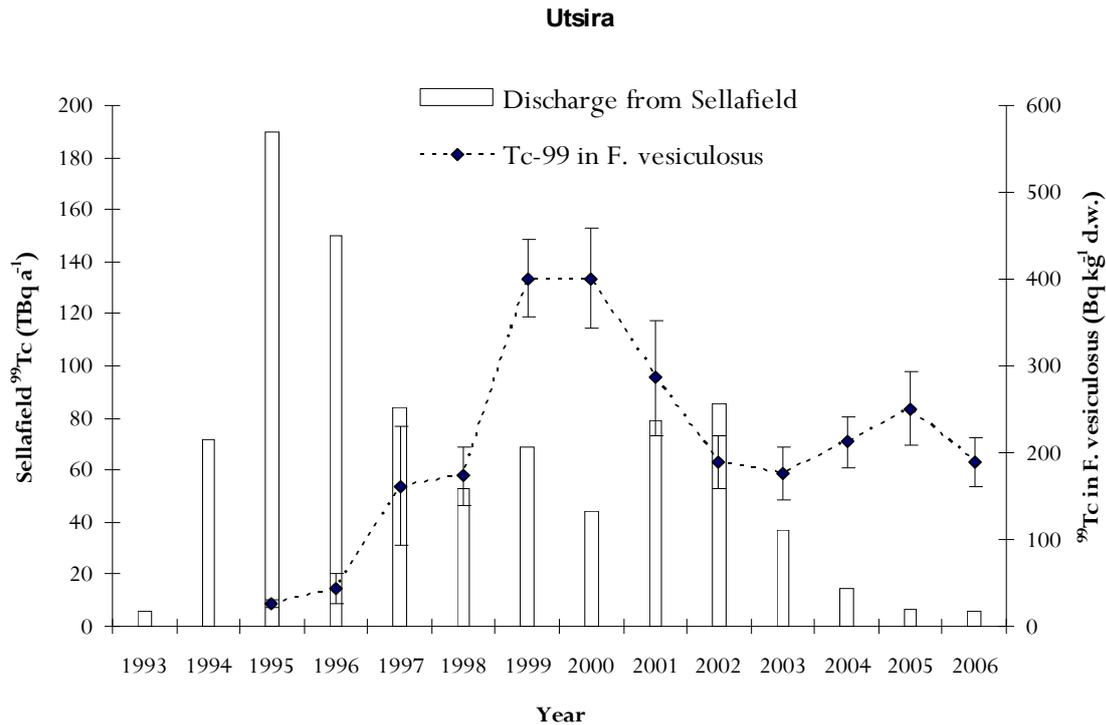
Seaweed is a useful bioindicator for  $^{99}\text{Tc}$  in the marine environment. It has a high ability to concentrate  $^{99}\text{Tc}$  from seawater and is easy accessible in most coastal areas. During 2005, seaweed (*Fucus vesiculosus*, Fig 5.1) was collected at the permanent coastal sampling stations along the Norwegian coastline and analysed for  $^{99}\text{Tc}$ . At two locations, Hillesøy and Utsira, sampling was performed monthly. At the remaining sites sampling was conducted in August or September. The results of the analyses are presented in Figure 5.2 and range from 59 to 257  $\text{Bq kg}^{-1}$  (d.w.), where the highest activity concentration (294  $\text{Bq kg}^{-1}$  d.w.) was found in one of the monthly samples collected at Utsira. Compared with the results from 1999-2001 (Rudjord *et al.*, 2001; Gäfvert *et al.*, 2003), the levels of  $^{99}\text{Tc}$  have decreased at most sampling sites. The trend can also be seen in Figures 5.3 and 5.4 which show the annual average activity concentration of  $^{99}\text{Tc}$  in *Fucus vesiculosus* at Utsira and Hillesøy, together with the annual discharge of  $^{99}\text{Tc}$  from Sellafield. The maximum levels in Figures 5.3 and 5.4 are the response to the increased discharge from Sellafield in the mid 1990s. From Figures 5.3 and 5.4, one can see that the peak in  $^{99}\text{Tc}$  levels in *F. vesiculosus* occurred after about 3-4 years at Utsira and about 4-5 years at Hillesøy further down-stream. The levels of  $^{99}\text{Tc}$  in *Fucus vesiculosus* from Hillesøy and Utsira are slightly lower in 2006 compared to 2005. The levels observed in 2006

are however significantly lower than the peak levels observed in 2000-2001 (Mann-Whitney U-test:  $z = 4.45$ ,  $n = 33$ ,  $p < 0.0001$ ).

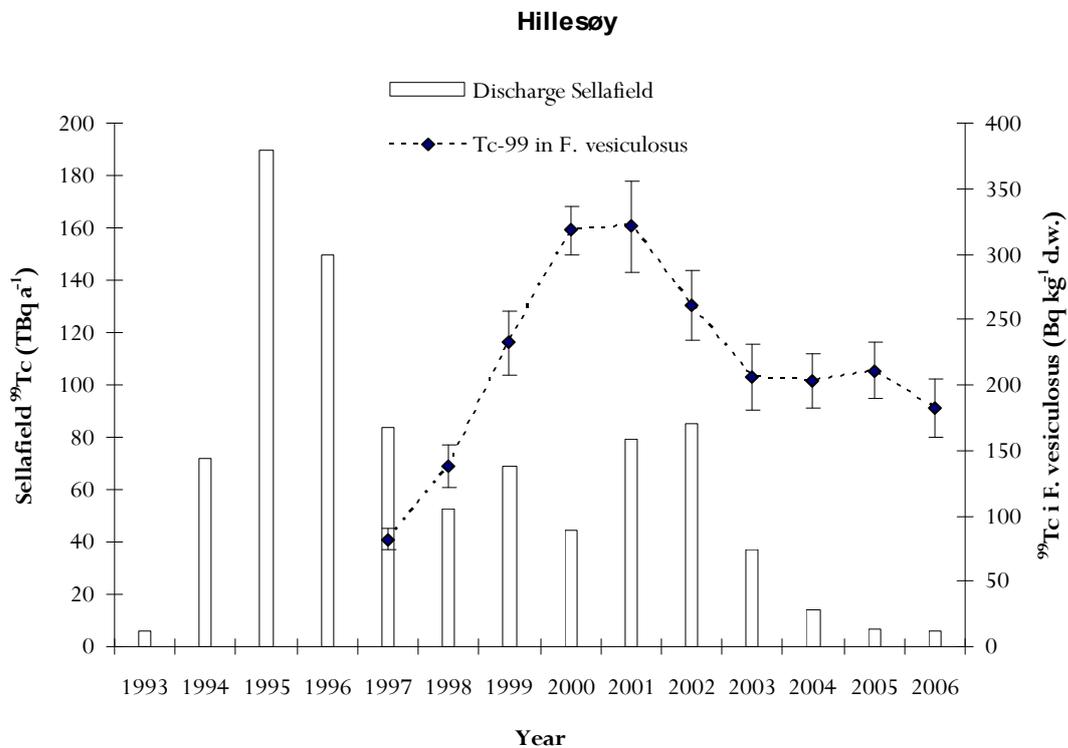


**Figure 5.2.** Levels of  $^{99}\text{Tc}$  in *Fucus vesiculosus* sampled along the Norwegian coastline in 2006.

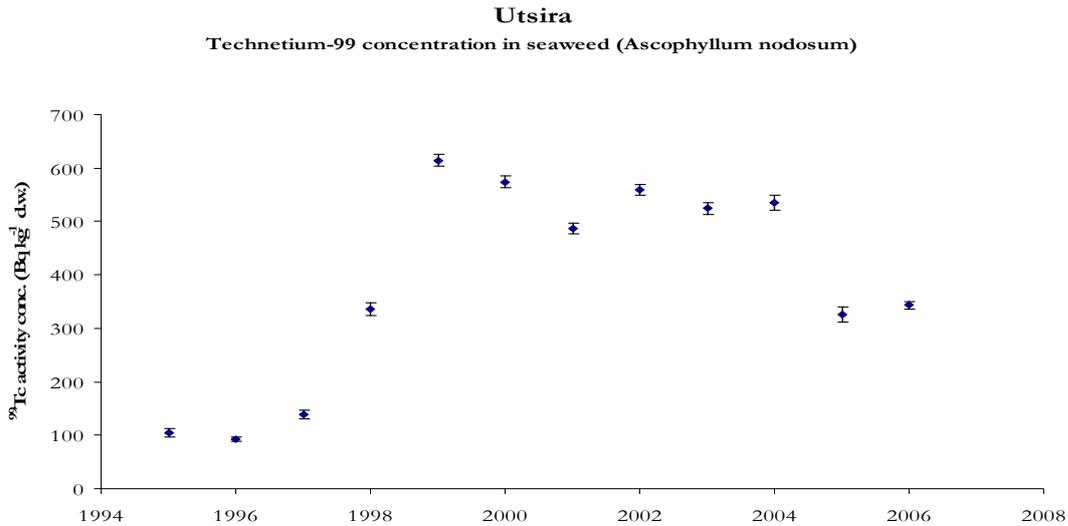
$\text{Tc-99}$  was also analysed in samples of *Ascophyllum nodosum* collected at Utsira in the period 1997 - 2006 (Fig. 5.5). *Ascophyllum nodosum* is known to have a relatively high concentration factor for  $^{99}\text{Tc}$ . Holm and Rioseco (1986) have shown that the concentration factor for  $^{99}\text{Tc}$  in *Ascophyllum nodosum* are about twice as high as for *Fucus vesiculosus*. The highest level found at Utsira, 614  $\text{Bq kg}^{-1}$  (d.w.), was observed in 1999. Since then the levels have decreased and in 2006 the activity concentration was found to be 343  $\text{Bq kg}^{-1}$  (d.w.).



**Figure 5.3.** Annual liquid discharge of <sup>99</sup>Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) <sup>99</sup>Tc activity concentration in brown algae (*Fucus vesiculosus*) sampled at Utsira (data provided by IFE) in the period 1995-2006 (secondary axis).



**Figure 5.4.** Annual liquid discharge of <sup>99</sup>Tc from Sellafield (primary axis) and annual average (with 95 % confidence limits) <sup>99</sup>Tc activity concentration in brown algae (*Fucus vesiculosus*) sampled at Hillesøy in the period 1997-2005 (secondary axis).

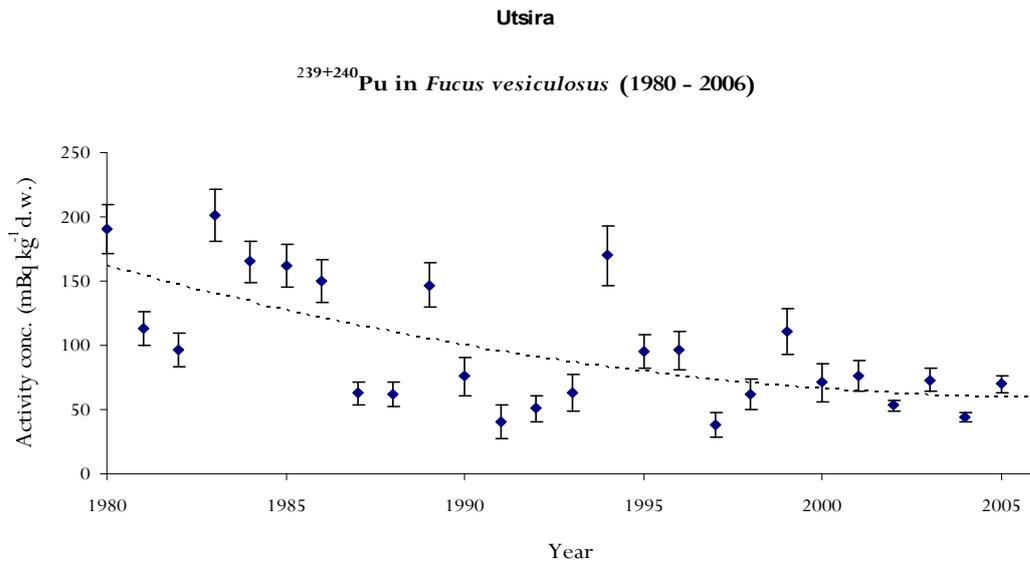


**Figure 5.5.** Activity concentration ( $Bq\ kg^{-1}\ d.w.$ ) of  $^{99}Tc$  in seaweed (*Ascophyllum nodosum*) sampled at Utsira in the period 1995 to 2006 (data provided by IFE).

## 5.2 Plutonium-239+240 in seaweed

*Fucus vesiculosus* has been collected and analysed for  $^{239+240}Pu$  at Utsira since 1980. The results from the period 1980 to 2006 are presented in Figure 5.6. The activity concentrations in these samples were in the

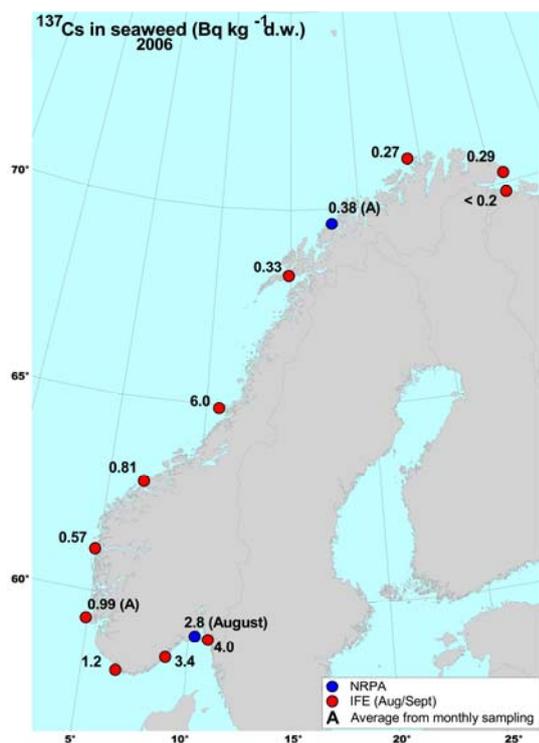
range 36 to 201  $mBq\ kg^{-1}$ , with relatively large fluctuations from year to year. One can also see that the level of  $^{239+240}Pu$  has remained at the same level in recent years. The levels in recent years are however lower than those observed in the early 1980s



**Figure 5.6.**  $^{239+240}Pu$  levels ( $mBq\ kg^{-1}\ d.w.$ ) in *Fucus vesiculosus* at Utsira in the period 1980 to 2006 (data provided by IFE).

### 5.3 Caesium-137 in seaweed

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



**Figure 5.7.** Levels of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1} \text{ d.w.}$ ) in *Fucus vesiculosus* sampled along the Norwegian coastline in 2006.

In 2006, samples of *Fucus vesiculosus* from the permanent coastal stations (see Figure 3.1) were analysed with respect to  $^{137}\text{Cs}$ . The results are presented in Figure 5.7, and range from  $< 0.2 \text{ Bq kg}^{-1} \text{ (d.w.)}$  in the north to  $6.0 \text{ Bq kg}^{-1} \text{ (d.w.)}$  at Vikna. The peak in the  $^{137}\text{Cs}$  levels in seaweed collected at Vikna can be explained by run-off of  $^{137}\text{Cs}$  from land, since this area was affected by fallout from the Chernobyl accident in 1986.

The reason for the higher activity concentration in *Fucus vesiculosus* in the southern part of Norway is a combination of two effects. First, the salinity in the water is

lower and secondly, that the activity concentration of  $^{137}\text{Cs}$  in the seawater is higher due to outflowing Baltic Sea water contaminated by the Chernobyl accident.

Comparing those results with the levels found at the coastal sampling stations in the period 2001-2005 (Gäfvert *et al.*, 2003; NRPA, 2004, NRPA 2005, NRPA 2006 and NRPA 2007), one can see that the activity concentration of  $^{137}\text{Cs}$  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  $6.0 \text{ Bq kg}^{-1} \text{ (d.w.)}$ , while in the northern part all results have been below  $1 \text{ Bq kg}^{-1} \text{ (d.w.)}$ .

## 5.4 Technetium-99 in lobster

In 2006, samples of lobster (*Homarus gammarus*) were collected at Sandøya and at Værlandet in Sogn og Fjordane (see Figure 3.1) and analysed for  $^{99}\text{Tc}$ . Lobsters have previously been identified as a marine organism with a high ability to accumulate  $^{99}\text{Tc}$  from seawater (Swift, 1985; Smith *et al.*, 1998; Smith *et al.*, 2001). The observed levels are presented in Table 5.1 and range from 1.2 to 12.7 Bq kg<sup>-1</sup> w.w. The levels in Sandøya and Værlandet are lower than those found in recent years at the same sites. The results show, however, a relatively large variation, even for samples collected in the same area. It is also observed a clear difference between female and male lobsters where female lobsters have a significantly higher uptake of  $^{99}\text{Tc}$ .

**Table 5.1.** Average activity concentration and range of  $^{99}\text{Tc}$  in lobster (*Homarus gammarus*) tail muscle in 2006.

| Location  | Gender (F/M) | n | $^{99}\text{Tc}$<br>Average<br>(range)<br>(Bq kg <sup>-1</sup> w.w.) |
|-----------|--------------|---|--|
| Sandøya   | F            | 7 | 4.0<br>(1.2 - 8.3)   |
|           | M            | 5 | 2.0<br>(1.8 - 2.3)   |
| Værlandet | F            | 7 | 8.9<br>(5.3 - 12.7)  |
|           | M            | 1 | 1.4  |

## 6 Summary and conclusions

In 2006 samples of seawater, sediment, and seaweed were collected in the Norwegian Sea, the Skagerrak and at a number of coastal stations and fjords. Data on radioactivity levels and trends in these areas together with data on liquid discharges from nuclear installations and recent trends in such discharges are summarised in this report.

### 6.1 Sources

The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden in 2006 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 0.48  $\mu\text{Sv}$  in 2006, which correspond to 48 % of the dose limit. The effective dose to the critical group from IFE Halden liquid radioactive discharge was estimated to be 0.02  $\mu\text{Sv}$  in 2006, corresponding to 2 % 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.

Produced water from offshore oil production may contain enhanced levels of naturally occurring radium isotopes. The reported total discharge of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  in produced water from Norwegian platforms in 2006 was 460 GBq and 390 GBq, respectively, where about 40 % was discharged from 2 platforms, Troll B and Troll C.

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

sediments act as secondary sources of radionuclides to the Norwegian marine environment.

### 6.2 Radioactivity in seawater and sediment

In 2006, samples of seawater and sediment were collected in the North Sea and the Skagerrak and analysed with respect to  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{226}\text{Ra}$ . 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 seawater and sediment

Observed levels of  $^{137}\text{Cs}$  in surface water in the Norwegian Sea and Barents Sea in 2006 ranged from 0.6 to 3.7  $\text{Bq m}^{-3}$ . This is similar to the levels observed in 2003 in the same area. The highest activity concentrations were found in the Kattegat. The relatively high activity concentrations in the Kattegat can be explained by outflowing seawater from the Baltic Sea, which was significantly affected by the Chernobyl accident in 1986.

#### 6.2.2 Technetium-99 in seawater

Samples of seawater were collected mainly in the Norwegian Sea and at a number of coastal stations. Levels of  $^{99}\text{Tc}$  ranged from below detection limit ( $< 0.04 \text{ Bq m}^{-3}$ ) to 0.8  $\text{Bq m}^{-3}$ . Highest activity concentrations were observed in coastal water. Average activity concentration from monthly sampling at Hillesøy was  $0.61 \pm 0.10$ , which is significantly lower than the peak values observed in 1999/2000, and slightly lower than the average activity concentration found in 2005. Due to the reduced discharge from Sellafield in 2003, the levels are expected to continue to decrease in 2007.

#### 6.2.3 Strontium-90 in seawater

The activity concentration in seawater sampled in the Norwegian Sea and in the Skagerrak ranged from 0.4  $\text{Bq m}^{-3}$  to 4.5  $\text{Bq m}^{-3}$  in 2006. The highest concentrations were found off the southern coast of Norway in the Skagerrak. The levels of  $^{90}\text{Sr}$  in both the Norwegian Sea

and the Skagerrak are similar to those observed in 2003.

#### **6.2.4 Plutonium-239+240 and americium-241 in seawater**

Observed levels of  $^{239+240}\text{Pu}$  in the Norwegian Sea and the Skagerrak in 2006 ranged from 2.8 to 18 mBq m<sup>-3</sup>, where the highest level was found off the coast of Scotland. The observed levels of  $^{239+240}\text{Pu}$  in 2006 are similar to those found in the same area in 2000 and 2003.

The activity concentration of  $^{241}\text{Am}$  in seawater from the Barents Sea ranged from 1.3 to 8.9 mBq m<sup>-3</sup>. Except for two samples with slightly elevated levels of  $^{241}\text{Am}$ , the activity concentration of  $^{241}\text{Am}$  in the Barents Sea are similar to what was observed in 2002.

#### **6.2.5 Radium-226 in seawater**

Radium-226 was analysed in seawater samples collected in the Norwegian Sea. The activity concentration of  $^{226}\text{Ra}$  in the samples ranged from 0.6 to 2.7 Bq m<sup>-3</sup>. Typical activity concentration of  $^{226}\text{Ra}$  in Atlantic surface water has been reported to be about 1.3 Bq m<sup>-3</sup> (IAEA, 1990).

### **6.3 Radioactivity in biota**

#### **6.3.1 Technetium-99 in seaweed**

Samples of *Fucus vesiculosus* sampled at the permanent coastal stations showed activity concentrations in the range 59 to 257 Bq kg<sup>-1</sup> (d.w.). For most stations the levels were similar or slightly lower in 2006 compared to observed levels in the period 2002 - 2005. The levels in 2006 are however significantly lower than the peak values observed in the period 1999-2001. Monthly sampling at Hillesøy and Utsira all indicate that the peak of  $^{99}\text{Tc}$  from the Sellafield discharge in 1995 and 1996 has passed these sites.

#### **6.3.2 Caesium-137 in seaweed**

The activity concentration of  $^{137}\text{Cs}$  in *F. vesiculosus* sampled at the coastal stations was in the range < 0.2 to 6.0 Bq kg<sup>-1</sup> (d.w.), where the highest levels were found at Vikna. The peak in the  $^{137}\text{Cs}$  levels in seaweed collected at Vikna can be explained by run-off of  $^{137}\text{Cs}$  from land, since this area was affected by

fallout from the Chernobyl accident in 1986. A comparison with results from 2000 and onwards shows that the levels have been relatively stable in recent years.

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## Appendix

### Analytical methods

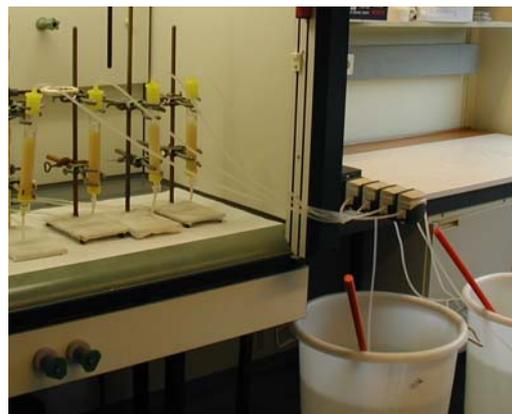
During recent years, laboratories at the Norwegian Radiation Protection Authority, and the Institute of Marine Research have both been accredited according to the requirements of NS-EN ISO/IEC 17025. Laboratories and organisations that are not named here have not been accredited according to the requirements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

### The Norwegian Radiation Protection Authority (NRPA)

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

#### *Determination of $^{99}\text{Tc}$ activity*

To determine the activity concentration of  $^{99}\text{Tc}$  in seawater, 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  $\text{H}_2\text{SO}_4$  followed by  $\text{HNO}_3$ .  $^{99\text{m}}\text{Tc}$  was added to all samples for chemical recovery determination.



**Figure A.1.** Fifty litres of seawater 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  $^{99\text{m}}\text{Tc}$  tracer in a NaI well-type detector. Typically, the radio-chemical yields varied between 70 % and 85 %. After one week, the  $^{99}\text{Tc}$  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,  $^{99}\text{Tc}$  preparations ready for counting.

The limits of detection for 10 g seaweed and 50 l seawater have been calculated to be approximately  $0.5 \text{ Bq kg}^{-1}$  (d.w.) and  $0.10 \text{ Bq m}^{-3}$ , 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  $^{99}\text{Tc}$  analysis is normally around 10 %.

### *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 seawater samples were dried separately at 105°C and ashed at 450°C before the activity was determined with an HPGe detector. The counting time varied from 1 to 4 days.

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

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

### *Detection of gamma emitters*

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

cryostat cooling systems, and 10 cm lead shielding.

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

Before the activity concentration of  $^{99}\text{Tc}$  can be determined, a radiochemical separation is necessary. Briefly, rhenium is added as a yield monitor in the form of  $\text{KReO}_4$ , and a preliminary extraction of  $^{99}\text{Tc}$  (and Re) based on anion-exchange separation is performed. After iron hydroxide scavenging,  $^{99}\text{Tc}$  and Re are further extracted by a second anion-exchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts are isolated.  $^{99}\text{Tc}$  is beta-counted using a low-background anti-coincidence beta counter (Model Risø GM-25-5), and the yield of the rhenium tetraphenyl arsonium salt is determined gravimetrically. See Harvey *et al.* (1992) for a detailed description of the method.

## **Institute for Energy Technology (IFE)**

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

Samples of dried and homogenized *Fucus vesiculosus* were analysed with regard to  $^{99}\text{Tc}$  content. The pretreatment was performed at 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,  $^{99}\text{Tc}$  was measured with an anti-coincidence-shielded GM counter (Holm *et al.*, 1984).

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

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

### *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.  $^{242}\text{Pu}$  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 was then electrodeposited on a stainless steel disc and counted using alpha spectrometry.

## **Risø National Laboratory**

### *Determination of $^{226}\text{Ra}$ in seawater*

Radium was coprecipitated with  $\text{MnO}_2$  from 10 l of seawater, after  $^{133}\text{Ba}$  had been added as a yield determinant. The  $\text{MnO}_2$  precipitate was then dissolved in hydrochloric acid and hydrogen peroxide, and after the addition of sulphuric acid,  $\text{K}_2\text{SO}_4$  and  $\text{Pb}(\text{NO}_3)_2$ , radium was coprecipitated with  $\text{PbSO}_4$ . The precipitate was dissolved in EDTA at  $\text{pH} = 10$ , and transferred to a liquid scintillation vial. After measuring the radiochemical yield with gamma spectrometry, a liquid scintillation cocktail was added (OptiFluor O). The sample was then stored for about four weeks after which the activity of  $^{226}\text{Ra}$  was measured through  $^{214}\text{Po}$  with a low-background, liquid scintillation counter (Quantulus). The laboratory work was carried out at Risø National Laboratory.

## **Department of Radiation Physics, Lund University**

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

The concentrations of  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$  and  $^{241}\text{Am}$  were measured in samples of 200 litres of seawater.  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  were added for chemical yield determination. Precipitation

from the water samples was achieved using the analytical procedure described by Chen *et al.*, (1991). 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.



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Virksomhetsplan 2008

**StrålevernRapport 2008:2**

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**StrålevernRapport 2008:4**

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**StrålevernRapport 2008:5**

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Strålevernet si overvåking av radioaktivitet i luft – resultatrapport for luftfilterstasjonar 2005–2006

**StrålevernRapport 2008:7**

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**StrålevernRapport 2008:8**

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**StrålevernRapport 2008:14**

Radioactivity in the Marine Environment 2006