

# Radioactivity in the Marine Environment 1999



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## Preface

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A new, comprehensive national programme for monitoring of Radioactivity in the Marine Environment (RAME) was established in 1999. This program is based on a proposal developed by the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR), the Directorate for Nature Conservation (DN) and the State pollution authorities (SFT) on behalf of the Ministry of Environment. NRPA, as the responsible authority on radiation protection, co-ordinates the programme whilst sampling at sea is conducted in close co-operation with IMR as part of the regular monitoring of the marine environment and its living resources. The principal objective of the programme is to document levels, distributions and trends of anthropogenic and naturally occurring radionuclides in the North Sea, the Norwegian Sea, the Barents Sea and along the Norwegian coast. The programme also collects updated information on both Norwegian and other sources of radioactive contamination, and carries out assessments of radiation exposures of humans and biota.

This new national monitoring programme has been co-ordinated with existing programmes funded by the Ministry of Fisheries. The monitoring programme for Marine Fish and Seafood was established in 1994. In previous reports from the programme established in 1994, (Sickel et al, 1995; Brungot et al, 1997, 1999) information regarding radioactivity in sea water, sediments and seaweed was included. However, the main purpose of this program is to document levels of anthropogenic radionuclides in fish and other seafood caught in Norwegian waters. This information is then made available to the relevant authorities, fishing industries and the general public as documentation regarding the quality of the marine products. The work in this programme is performed as a co-operation between the NRPA and the Directorate of Fisheries. In addition, results from the monitoring program conducted by the National Food Control Authority are also included in the present report.

The authors that have prepared the report are indicated at the start of each chapter.

Several institutions have collected samples and analysed results for the coordinated monitoring programme. They are, the Norwegian Radiation Protection Authority (including the environmental unit in Tromsø), the Institute of Marine Research, Institute for Energy Technology, the Directorate of Fisheries (including the local office in Tromsø) and the Norwegian Food Control Authority (local office in Salten).

Appendix 1 gives general information about radionuclide analysis and provides details of analytical methods employed by each institution.

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# 1. Introduction

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The issue of actual and potential radioactive contamination in the marine environment has received substantial attention in Norway in recent years. In the late nineteen-eighties several accidents and incidents involving nuclear powered submarines, demonstrated that the risk of releases of radionuclides into the Barents Sea should be considered more carefully. In particular, it was soon evident that better documentation concerning the radioactivity levels in fish and other seafood was important for the seafood export industries. Furthermore, in the early nineteen-nineties, information concerning the dumping of nuclear waste emerged through the bilateral environmental co-operation 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 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 discharges from European reprocessing facilities have been detected in the Norwegian marine environment. In 1994 and 1995, discharges of  $^{99}\text{Tc}$  from the reprocessing facilities at Sellafield in the United Kingdom increased sharply. There were public concerns about the consequences of such releases, because the radionuclides discharged to the Irish Sea have been transported by ocean currents via the North Sea into the Norwegian coastal current and the Barents Sea. In response to such concerns, programmes for monitoring of radioactivity in the marine environment were established. Due to the economic importance of the fishing industry and its vulnerability to any rumours of radioactive contamination, one of the main objectives is to document levels and trends of radionuclides in the Norwegian marine environment.

Human activities, such as mining and oil production may also affect the discharges of natural radionuclides (UNSCEAR, 1993, 1996, 2000) to sea.

In Norway there are currently two monitoring programmes concerned with radioactivity in the

marine environment, both co-ordinated by the Norwegian Radiation Protection Authority (NRPA). One is funded by the Ministry of Environment and the other by the Ministry of Fisheries. Results of both these programmes are presented in this report. In addition, data from the Food Control Authorities monitoring programme that is concerned with radioactivity in marine fish have been included.

The marine monitoring programmes include the collection of discharge data from Norwegian sources in addition to the collection of data that are concerned with the long-range transport of radionuclides from various sources. Liquid discharge data for 1999 from nuclear installations and recent trends in such discharges have been summarised, together with the available information concerning nuclear weapons fallout and outflow of  $^{137}\text{Cs}$  of Chernobyl origin from the Baltic Sea.

In addition, data from 1996 regarding the discharges of technologically enhanced naturally occurring radionuclides (TENORM) in production water from the North Sea oil and gas industry have been included. The natural and anthropogenic radionuclides carried by rivers, sediment and water into the sea have not been studied in 1999, but may be included in the monitoring programme in the future.

In 1999 samples were collected for the monitoring programme in the Barents Sea, the North Sea, 12 coastal stations including Svalbard and 11 fjords. In addition, control samples were collected in Nitelva near the outlet of discharges from the nuclear facilities of the Institute for Energy Technology (IFE) at Kjeller. The results of the analysis of the samples are presented and discussed in chapters 4, 5 and 6.

Work is also continuing to improve consequence assessment models for calculation of radiation doses to humans and marine biota, and to obtain better empirical values for the model parameters. The results obtained are described in chapter 7.

In chapter 8, a summary of the findings in 1999 and conclusions are given. In the appendix 1, technical information regarding sample preparation techniques and analysis methods employed in the laboratories are given.

## 2. Sources of radionuclides in the marine environment

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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 integrated part of the marine monitoring program RAME, and the main purpose 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 sources is included.

The largest Norwegian sources of anthropogenic radionuclide discharges in terms of radiation doses to the public are the nuclear facilities of the Institute for Energy Technology (IFE). These facilities have discharge authorisations issued by the Norwegian Radiation Protection Authority (NRPA). Unsealed radioactive materials used in medicine and science will primarily be released to the marine environment via the sewage treatment plants. The utilisation of such open sources is regulated through guidelines issued by the NRPA.

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

The discharges of radionuclides from Norwegian sources are only detectable in the local environment, near the discharge points. The long-range transport of radionuclides originating from nuclear weapons fallout, reprocessing of nuclear fuel and from the Chernobyl accident are still the main contributors to the general levels of anthropogenic radionuclides found in Norwegian waters. In Nitelva, the main source of radionuclide contamination is early discharges from the Institute for Energy Technology at Kjeller.

### 2.1 Discharges of anthropogenic radionuclides from Norwegian Sources

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#### 2.1.1 Institute for Energy Technology (IFE) and Isopharma, Kjeller

This nuclear site is located about 20 km north of Oslo. The facilities include a heavy water cooled and moderated research reactor with thermal effect 2 MW, JEEP II, a metallurgical laboratory, production facilities of 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. It is then transported through special pipelines to the Radwaste Plant. The plant has a system of large tanks for storage of liquid waste, as well as facilities for filtration and purification of the wastewater by ion-exchange and evaporation. The short-lived radionuclides are normally allowed to decay significantly prior to discharge.

Liquid effluent is discharged through a designated pipeline to the river Nitelva about 100 km from the sea. The river, which has an annual mean flow rate of 5 m<sup>3</sup>/second, empties into Lake Øyeren where the water is mixed with the water from the Glomma river which has an annual mean flow rate of 400 m<sup>3</sup>/second. The river Glomma empties into the Oslo Fjord at Fredrikstad City.

A plant manufacturing radiopharmaceutical products, managed by the private company, Isopharma AS, is operating in close collaboration with IFE. The release authorisation for IFE also includes the releases from this production facility.

The discharge limit authorised by the Norwegian Radiation Protection Authority (NRPA) is based on the annual dose to any member of a critical group of the population along the Nitelva River, and shall not exceed 1 µSv.

Each year, IFE reports discharge data, results of their environmental monitoring programme and calculations of effective doses resulting from discharges, to the Norwegian Radiation Protection Authority (IFE, 2000). The reported discharges for the period 1994-1999 are summarised in Table 2.1.

Table 2.1: Liquid discharges from IFE, Kjeller 1994-1999

SITE	Radionuclides	MBq 1994	MBq 1995	MBq 1996	MBq 1997	MBq 1998	MBq 1999
<b>Institute for Energy Technology</b>	Cs-137	99.6	112	80	21	110	30
	Cs-134	6.4	2.5	1.7	2.7	5.0	1.9
	I-131	7.1	209	1.41E+3	74	350	1.35E+3
	I-125	46.3	249	760	117	190	182
<b>Kjeller</b>	Gd-153	0.3	-	-	-	-	-
	Zn-65	3.9	4.2	273	15	0.87	0.75
	Co-60	27.6	40.9	106	16	11	6.9
	S-35	369	690	695	820	280	18.5
	Sr-90	5.9	12.1	12.2	4.6	81	161
	Pu-239,40	0.102	1.0	0.782	0.15	0.11	0.016
	H-3	3.27E+5	3.06E+5	7.23E+5	0.11E+5	1.52E+5	0.85E+5

Table 2.2 Effective doses to the critical group from river. water exposure pathways as reported by IFE Kjeller

	1994	1995	1996	1997	1998	1999
<b>Effective dose (µSv)</b>	0.12	0.23	0.71	0.06	0.24	0.53

Effective doses have been calculated by IFE for a hypothetical critical group exposed through annual consumption of 20 kg of fish from the river and 100 hours/year occupancy on the riverbanks. In 1999, the calculated effective dose to this critical group was 0.53 µSv, corresponding to 53 % of the annual dose constraint. The total annual discharge varies, as shown in Table 2.2, corresponding to between 10 and 70 % of the dose constraint, with an average of about 30 % for the years 1994-99.

### 2.1.2 Contamination of Nitelva sediments due to early discharges from IFE Kjeller

Nuclear activities began at IFE Kjeller in 1951 with the start-up of the first research reactor, "JEEP 1". Since 1952, radionuclides have been produced and this has been the main source of discharges of the short-lived radionuclide <sup>131</sup>I. A new reactor ("NORA") was in operation between 1961-1968. At the same time, an experimental plant for purification of uranium was in operation. During the decommissioning of the purification plant, discharges of plutonium and americium increased considerably. However, the discharges were still within the limits that existed at that time.

The available information regarding early discharges has been collected in a report at NRPA.

(NRPA, 1999:11). Discharge data exists in reports from 1964 onwards, when the first discharge authorisation was issued. Before 1967, liquid waste was released through the sewage pipeline to Sogna, a branch of the Nitelva river with low circulation of river water. In 1967, a new pipeline (the NALFA-pipeline) was constructed, releasing waste water near Nybrua in Lillestrøm (Figure 2.1). The discharge history of the IFE Kjeller facilities is shown in Figure 2.2 and Figure 2.3.

Discharges of plutonium peaked in 1969, due to the decommissioning of the uranium purification plant. In 1971, the IFE conducted an investigation into the contamination of Nitelva sediments. Radioactive material was found to be concentrated in an area of about 200 m<sup>2</sup> close to the outlet of the NALFA pipeline. Plutonium concentrations up to 2.3 MBq kg<sup>-1</sup> were found. In 1974, the IFE removed 1800 kg of contaminated sediment. It was assumed that the plutonium concentration was now below about 7.4 kBq kg<sup>-1</sup> and in the following years, the internal monitoring program of IFE showed only relatively low levels of contamination in surface sediments. However, in 1996, when a survey of deep sediments was performed, concentrations of plutonium of up to 2,0 MBq kg<sup>-1</sup> were found. These high levels of plutonium contamination were of great concern for the NRPA. The water level of Nitelva is regulated, but varies substantially through the year, particularly during the spring, when the water level may be lowered to avoid flo-

oding as a result of snow melting. In such situations, the contaminated sediments may be exposed and available to the public. As a result, the Ministry of Health and Social affairs requested a report from the NRPA regarding the radioactive discharges from IFE Kjeller from 1948 to 1999 (NRPA, 1999:11). In 1999, the NRPA also decided to collect additional samples in Nitelva as part of the national monitoring program. The results of the measurements of surface sediment samples are given in chapter 4.



Fig. 2.1 Map showing discharge pipelines from IFE Kjeller facilities. The old pipeline was used until 1967. The NALFA pipeline was used until 1999. In the year 2000, the outlet for discharges was moved and part of the NALFA pipeline is no longer used.

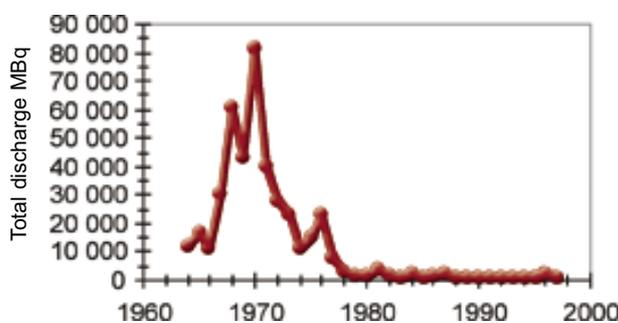


Fig. 2.2 Total discharges from IFE Kjeller 1962- 1997 estimated on the basis of gross alpha and gross beta measurements. After 1983, the discharges have been calculated as the sum of the activities of individual radionuclides.

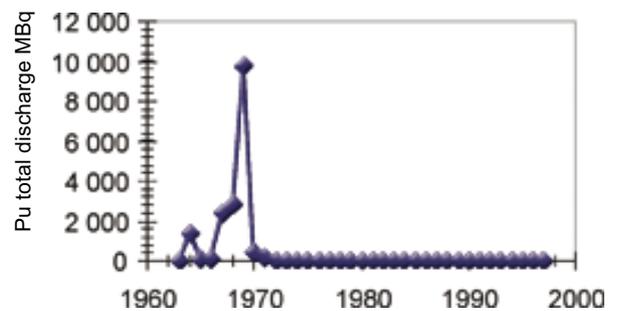


Fig. 2.3 Discharges of plutonium from IFE Kjeller 1963-1997. The levels of plutonium discharges in the period 1963-78 have been estimated on the basis of gross alpha measurements.

In February 2000, the NRPA decided that all sediments with summed concentrations of plutonium and americium in excess of  $10 \text{ Bq g}^{-1}$  had to be removed. In a clean-up operation in April 2000, a total of  $181 \text{ m}^3$  of contaminated sediments were removed by IFE. Later the NALFA pipeline was replaced with a new pipeline, where the wastewater was released to the free water masses midstream.

### 2.1.3 Institute for Energy Technology OECD Halden Reactor Project

Halden Boiling Water Reactor (HBWR) is located in the town of Halden, in the south-east of Norway, close to the Swedish border. The HBWR is heavy water cooled and moderated and has three main systems: the primary system (heavy water) and two light water heat removal systems where the secondary system is a closed loop system. The reactor was commissioned in 1959.

Liquid waste destined for disposal is retained and cleaned, with the help of ion-exchange columns, to a level considered acceptable for release, relative to the applicable release limits. The water flows to a  $10 \text{ m}^3$  delay tank that has a constant water level of  $5 \text{ m}^3$ . Activity control is performed continuously on the water leaving the delay tank to the sewage system.

In the case of abnormally high water activity, a main outlet valve will close automatically and the water is collected in the three delay tanks (total volume  $30 \text{ m}^3$ ). If necessary, an additional  $80 \text{ m}^3$  of water can be pumped in to a pit in the reactor hall, thus providing a storage capacity of  $110 \text{ m}^3$ . This water can then be cycled through a clean-up system with a particle filter and anion-exchange column, before release.

Liquid effluent was supposed to be released to the river Tista, which discharges into Iddefjord, leading to Skagerrak. However in May 2000, it was discovered that the discharges were actually directed to the local sewage system plant from 1992 to 2000. This plant in turn discharges treated water to Iddefjorden. The consequence for the marine environment was in fact a slightly reduced discharge because some radionuclides were retained in the sewage sludge. This issue will be discussed in more detail in the monitoring report for the year 2000.

The discharge authorisation limits the exposure of critical groups from liquid discharges to 1 micro Sievert per year. In Table 2.3 the liquid discharges from the period 1994-1999 reported by IFE Halden are listed.

The critical group is hypothetical and only defined by their food consumption and living habits. The calculation of effective dose to the critical group is based on:

- Annual consumption of 30kg fish from the fjord
- 200 hours/year occupancy on the fjord beaches
- 50 hours/year bathing in the fjord
- 1000 hours/year of boating on the fjord

In Table 2.4 the doses to the critical group as calculated by IFE are listed.

## 2.2 Unsealed radioactive substances in medicine, 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 Norwegian Radiation Protection Authority. Given this authorisation, laboratories and hospitals are allowed to discharge activity into the sewage system according to predefined limits. The present regulations are now under revision.

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 excretion of urine and faeces.

For some tracer experiments in offshore oil industry, tritium (<sup>3</sup>H) is injected into formations or drilled wells, and a fraction of this will be recovered. Water-based material is discharged to sea whereas mud-containing oil must be stored.

Table 2.3 Annual liquid discharges 1994-1999, IFE Halden

SITE	Radionuclides	MBq 1994	MBq 1995	MBq 1996	MBq 1997	MBq 1998	MBq 1999
<b>Institute for Energy Technology</b>	Cs-137	130	850	530	250	410	530
	Co-60	960	540	140	260	220	380
	Cs-134	10	93	51	29	28	29
	H-3	330.000	430.000	371.000	336.000	894.000	671.000
	Co-58	200	24	18	37	22	19
<b>Halden Reactor Project</b>	Nb-95	200	32	9	53	20	60
	Cr-51	660	86	240	540	330	240
	Mn-54	2	2	1	5.4	6	5.6
	Zr-95	20	3		24	7.6	14
	Sb-124	7	4		4.9		
	Sb-125				110	4.4	39
	Ag-110m		7	1		1.4	
	Ru-106			7.5			
	I-131			1.8	31		6.5
	Ce-144			4.8		18	8.3
	Mn-56					2.7	

Table 2.4 Average annual effective dose from marine exposure pathways as reported by IFE Halden

	1994	1995	1996	1997	1998	1999
<b>Effective dose (µSv)</b>	0.19	0.11	0.04	0.06	0.05	0.08

However, for the consequence assessments required by NRPA, it is conservatively assumed that all the  $^3\text{H}$  is discharged to the open sea.

Releases of radionuclides used in research laboratories are in general considered less important, partly because only small activity quantities are used and parts of the waste is collected and sent to the Institute for Energy technology (IFE) for storage as radioactive waste. Furthermore, most of these radionuclides have short half-lives and the activity is rapidly reduced by storage.

Exact data on the discharges from the above sources are not available, however, the purchasers of open radioactive sources are required to report their sales to NRPA monthly, listing customers, types of radionuclides and quantity (activity) as an inventory.

In Table 2.5 the purchase of open radioactive sources in Norway in 1999 is listed. As for the sales of  $^{99\text{m}}\text{Tc}$ , it is important to consider the short half-life of this radionuclide, and a large fraction will decay before it is even used.

*Table 2.5 Unsealed radioactive sources purchased in Norway 1999. Only radionuclides where the total activity exceeds 1 GBq have been included in the table.*

Radionuclide	Half-life	Activity purchased in Norway 1999 (GBq)
Mo-99/Tc-99m	66h/6h	24,376
I-131	8d	1,446
In-111	2.8d	16.5
Cr-51	27.7d	1.9
Ga-67	3.3d	7.6
C-14	5.73E+3y	50.8
Xe-133	5.3d	187.8
I-125	60d	6.1
P-32	14.3d	13.3
H-3	12.3y	8.22
S-35	87d	7.0

In a survey of man-made radionuclides in sewage sludge and wastewater in 1995-1996 at a sewage treatment plant near Oslo, iodine ( $^{131}\text{I}$ ) was detected in most samples, whereas chrome ( $^{51}\text{Cr}$ ) was only detected in low quantities in sewage sludge. On the basis of the radioactivity measurements it was estimated that the annual releases of  $^{131}\text{I}$  to the Oslo fjord from this plant was about 780 GBq.

## 2.3 Technological enhanced naturally occurring radioactive material (TENORM)

All minerals and rocks in the earth's crust contain small, but measurable amounts of naturally occurring radioactive materials - often referred to as NORM. NORM includes long-lived isotope of potassium ( $^{40}\text{K}$ ), isotopes of uranium (mainly  $^{238}\text{U}$ ) and thorium (mainly  $^{232}\text{Th}$ ).  $^{238}\text{U}$  and  $^{232}\text{Th}$  are parent radionuclides of two natural radioactive decay series. Naturally occurring radionuclides are released to the marine environment by natural processes like erosion for instance carried by river sediments, other particles, or dissolved in river water.

In certain industrial or other technological processes naturally occurring radionuclides can be concentrated to levels orders of magnitude higher than 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. Some data exists regarding the discharges of production water from the North Sea oil and gas production, but data on runoff from mine tailings are lacking. However, there are plans to extend the marine monitoring programme to include monitoring data for mine tailings from 2002.

### 2.3.1 TENORM in oil and gas production in Norway

Two isotopes in the uranium and thorium series are important in relation to water discharges and radioactive deposits in oil and gas production - namely radium ( $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ ).

The occurrence of natural radionuclides in North Sea oil and gas production were first discovered in 1981, and enhanced levels of radioactivity are now found in the production system of several North Sea oil fields (Strand et al, 1997). The activity concentrations range from background level to several hundred Bq  $\text{g}^{-1}$  of  $^{226}\text{Ra}$  (Smith, 1987). Doses to workers involved in handling contaminated equipment or waste are usually low, but the main problems related to radioactive deposits are waste disposal and discharges of produced water.

When oil and natural gas are extracted from the ground they are accompanied by solids and formation water. Under certain circumstances, naturally radioactive salts that are dissolved in the formation water will precipitate deposit onto the inner walls of production equipment as sulphate and carbonate scales. Particles of clay and sand co-produced from the reservoir may also act as surfaces and initiate scale deposition.

Sea water is injected into the reservoir to maintain the pressure as the oil is removed. Mixing of sea water and formation water creates incompatible solutions, and sulphates (mainly BaSO<sub>4</sub> and SrSO<sub>4</sub>) are precipitated. The dominating radioactive elements in geologic formations in the sea floor are potassium and the elements in the uranium- and thorium series. Radium exists in both series as <sup>226</sup>Ra and <sup>228</sup>Ra. Radium, under certain circumstances will leak from the formation and be dissolved in the formation water. Barium, strontium and radium are members of group IIA in the periodic Table and have similar chemical properties. Consequently radium is co-precipitated with barium and strontium as radium sulphate (RaSO<sub>4</sub>), even though its solubility product is not exceeded. Turbulent flow in the production system will then cause the precipitated sulphate salts to attach to the walls and form deposits with enhanced levels of radioactivity. These deposits are often referred to as LSA (Low Specific Activity) scale.

Generally, the activity concentrations of <sup>226</sup>Ra, <sup>228</sup>Ra and decay products in such deposits and sludge may vary from normal levels in soils and rocks (less than 0.1 Bq g<sup>-1</sup>) to more than 1000 Bq g<sup>-1</sup> (Strand et al, 1997; Lysebo et al, 1997).

Radioactive deposits represent a significant waste problem for the oil industry. Since the problem was discovered in 1981, waste from the cleaning of contaminated equipment has been sent to a temporary storage facility at Kjeller, north of Oslo. The total amount of LSA scale waste stored at Kjeller is at present about 80 tonnes, with a mean activity of 25 Bq g<sup>-1</sup> of <sup>226</sup>Ra. The expected amount in the future is 20 tonnes per year.

In 1995, a temporary exemption level of 10 Bq g<sup>-1</sup> of <sup>226</sup>Ra was introduced by the NRPA. All waste from the oil industry with activity above this level is defined as “radioactive waste”. Originally, this waste was to be sent to the national repository for final storage. It has now become clear that the

amounts of waste will exceed the amount expected when planning the repository and the authorities have decided that an alternative solution should be found.

### 2.3.2 Discharges of production water

Most of the radioactivity from the reservoir is dissolved in produced water and discharged into the sea. In 1996, extensive measurements of levels of radioactivity in produced water and deposits were performed by NRPA for several North Sea installations (Strand et al, 1997).

The samples were taken from 11 production units during normal operation and analysed at the NRPA. The results of the measurements are shown in Table 2.6.

Table 2.6 Activity concentration of <sup>226</sup>Ra and <sup>228</sup>Ra in production water from 11 installations on the Norwegian continental shelf.

Installation	Sampling Point	<sup>226</sup> Ra (Bq l <sup>-1</sup> )	<sup>228</sup> Ra (Bq l <sup>-1</sup> )
Statfjord A	Flotation cell	n.d.	0.3 - 0.6
Statfjord A	Degassing tank	n.d.	0.5 - 0.7
Statfjord B	Flotation cell	3.5	1.5 - 4.2
Statfjord B	Degassing tank	0.7 - 7.6	0.5 - 1.4
Statfjord C	Flotation cell	1.9 - 2.5	1.7 - 2.1
Statfjord C	Degassing tank	2.2	2.4 - 2.7
Gullfaks A	Flotation cell	n.d.	1.3 - 2.0
Gullfaks B	Flotation cell	n.d.	1.7 - 4.3
Gullfaks C	Flotation cell	1.3	0.5 - 1.8
Veslefrikk	Degassing tank	n.d.	n.d.
Sleipner A	Degassing tank	2.5	1.1 - 3.3
Oseberg	Degassing tank	10.4	10.0
Brage	Degassing tank	7.7	7.8
Valhall	Valve	3.0 - 5.6	0.8 - 1.1

With reference to Table 2.6, <sup>226</sup>Ra and <sup>228</sup>Ra activity concentrations appear to be in a narrow range: 0.7 - 10.4 Bq l<sup>-1</sup> and 0.3 - 10.0 Bq l<sup>-1</sup>, respectively. The mean concentration was 4.1 Bq l<sup>-1</sup> of <sup>226</sup>Ra and 2.1 Bq l<sup>-1</sup> of <sup>228</sup>Ra. This is approximately three orders of magnitude higher than the mean concentration in sea water (IAEA, 1990). The highest single measurement of <sup>226</sup>Ra in our study was 10.4 Bq l<sup>-1</sup>. These results are comparable to activity concentrations data for produced water in USA, Russia and Germany (Snaveley 1989 and API, 1991).

Large amounts of production water are released into the North Sea each year. A total volume of 160 x 10<sup>6</sup> m<sup>3</sup> was released in 1991. Assuming that the mean radium concentrations of production water in our study are representative values for the oil production of the North Sea this would corres-

pond to a total release of  $6.6 \times 10^{11}$  Bq and  $3.4 \times 10^{11}$  Bq for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  respectively. It has been estimated that the release of production water will increase significantly in the years to come owing to the fact that some of the larger production fields will reach a later stage in the production phase. It has been estimated that Norway contributes 6 % of the total release, while Great Britain, the Netherlands and Denmark contribute 88 %, 5 % and 1 % respectively (E & P Forum, 1993).

Sometimes, deposition of scales may interfere with the production process by blocking transport through the production zone, flow lines and produced water lines. Consequently, the oil companies try to prevent deposition of scales through the application of inhibitors. The scale inhibitors will prevent the deposition of radium salts in the system but will increase the concentration and the release of radioactivity in the produced water. These inhibitors are organic compounds and they may also increase the biological uptake of radioactivity in marine ecosystems. There has been an increase in the use of inhibitors during the last decade, and owing to present and future demands to reduce the production costs, the increase in the use of inhibitors in the years to come may be significant.

## 2.4 Long-range transport of radionuclides

There are several sources of radioactive contamination to the North East Atlantic and the Arctic marine environment. The primary sources are fallout from atmospheric nuclear weapons testing, transport of discharged radionuclides from reprocessing plants and fallout from the Chernobyl accident.

In addition, there are several sources of interest, which in the future may release radioactive contamination into the marine environment. In Figure 2.4 the locality of actual and potential sources are shown.

Figure 2.5 shows the main features of the water circulation of the North Atlantic. Discharges of radionuclides from European reprocessing plants are transported from the Irish Sea and the English Channel via the North Sea and into the Norwegian Coastal Current, to the Barents Sea and beyond. Baltic water contaminated with fallout from the

Chernobyl accident flows through Kattegat and enters the Norwegian Coastal Current in the Skagerrak area.

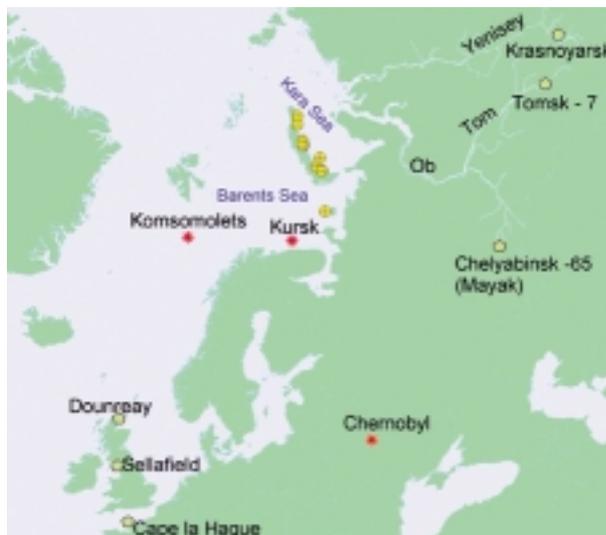


Fig.2.4 Sources of radionuclides in to the northern marine environment: Chernobyl in the Ukraine, the reprocessing plants at Sellafield, La Hague and Dounreay, the dumping sites of nuclear waste in the Kara Sea, the sunken submarines Komsomolets and Kursk and Russian nuclear installations (Mayak, Tomsk-7 and Krasjonoyarsk-26) releasing radionuclides to the Russian rivers Ob and Yenisey.

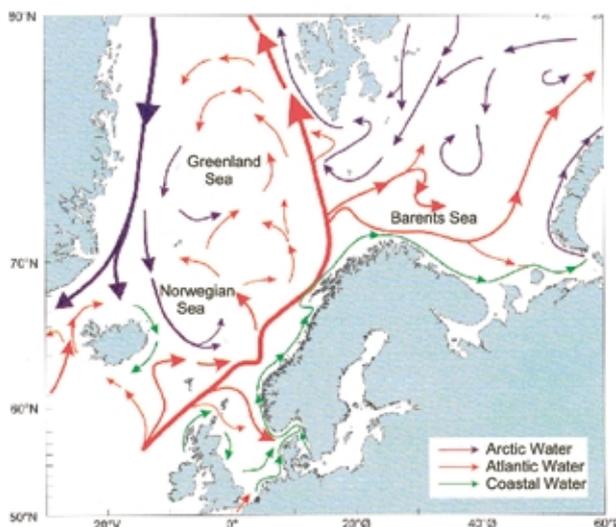


Fig. 2.5 The surface circulation pattern of the Northern Seas (adapted from Aure et al. (1999)).

### 2.4.1 Discharges of radionuclides from European nuclear installations

For decades, authorised discharges from nuclear reprocessing facilities in Europe have been an important source of anthropogenic radionuclides in Norwegian coastal areas. These facilities include Sellafield in England, Dounreay in Scotland, and Cap De La Hague in France. Releases of radionuclides to the marine environment also occur from nuclear fuel fabrication plants and nuclear power stations.

A nuclear fuel reprocessing plant recovers spent nuclear fuel for reuse in fission reactors. The remaining radionuclides are either stored pending a final management option or released to the environment (UNSCEAR, 1993). Liquid radioactive wastes from the operation of these plants are discharged via pipelines directly into the Irish Sea, Scottish coastal waters and into the English Channel respectively. Soluble radionuclides from these sources are subsequently transported further northwards with regional oceanic currents.

Sellafield has been the main contributor to radioactivity releases among the three Western European reprocessing plants. Maximum discharges of cesium ( $^{137}\text{Cs}$ ) and the actinides plutonium ( $^{239+240}\text{Pu}$ ) and americium ( $^{241}\text{Am}$ ) from Sellafield occurred during the mid-late 1970s (Gray et al., 1995). The introduction of the Site Ion-Exchange Effluent Plant (SIXEP) in 1985 subsequently led to a substantial reduction in discharges of strontium ( $^{90}\text{Sr}$ ),  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (Gray et al., 1995). Increased throughputs and processing of residues, led to increased discharges of plutonium and americium during the early to mid 1970s.

Throughout the 1980s and early 1990s (1981-1993), technetium ( $^{99}\text{Tc}$ ) was discharged from Sellafield at a rate of 1.9-6.6 TBq per year. In 1994, the Enhanced Actinide Removal Plant (EARP) at British Nuclear Fuel's (BNFL) Sellafield plant in north west England began operations to treat a backlog of stored wastes (Gray et al., 1995). This resulted in a step increase of  $^{99}\text{Tc}$  from a level of approximately 5 TBq  $\text{a}^{-1}$  to a level of 72-190 TBq  $\text{a}^{-1}$  in the period 1994-1996. In Figure 2.6 the discharges of  $^{99}\text{Tc}$  from Sellafield from the fifties to the present day are shown.

The initial impact, in terms of increased activity levels in biota and sea water, of the higher  $^{99}\text{Tc}$  discharges from Sellafield was observable in Norwegian coastal environments by 1997 (Brown et al., 1999).

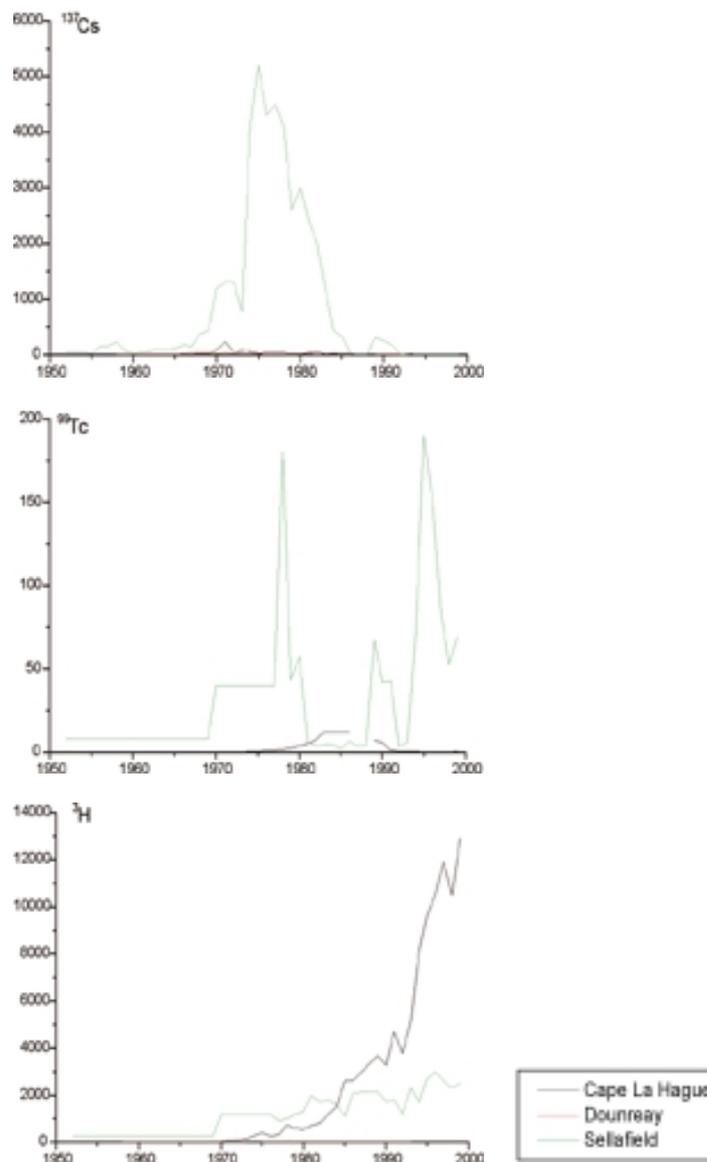


Fig. 2.6 Annual discharges of  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$  and  $^3\text{H}$  to the marine environment from Sellafield, Cape la Hague and Dounreay from the fifties and up to today, given in TBq.

In Table 2.7 the discharges of some radionuclides in 1999 from Sellafield, Cap la Hague, Springfields and Dounreay are shown.

In Figure 2.6 the discharges to the marine environment of  $^{99}\text{Tc}$ ,  $^{137}\text{Cs}$  and  $^3\text{H}$  from Cap La Hague, Sellafield and Dounreay from the fifties to the present day are shown. The Figure demonstrate that, except for tritium, discharges from Cape la Hague and Dounreay have been considerably lower than Sellafield discharges most of the time. The radiotoxicity of  $^3\text{H}$  is very low and the radiological importance of that nuclide is much less than for other radionuclides. To some extent the tritium discharges can be regarded as an indicator of the amount of reprocessed spent nuclear fuel.

Table 2.7 Discharges of liquid radioactive effluent from Sellafield, La Hague, Springfields and Dounreay to the marine environment in 1999 (OSPAR 2001).

Liquid discharges of radionuclides 1999 (TBq)				
Location	La Hague -English Channel	Sellafield -Irish Sea	Springfields -Irish Sea	Dounreay -Scottish coastal waters
<b>Discharges to Radionuclides</b>				
Tritium	1.29E+4	2.52E+3	-	1.37E-1
Total-a	3.95E-2	1.33E-1	2.38E-1	1.73E-3
Total-b	1.70E+3	1.10E+2	1.28E+2	2.79E-1
Co-60	3.21E-1	8.90E-1	-	3.61E-3
Sr-90	8.49E-1	3.12E+1	-	1.63E-1
(Sr-90+Cs-137)	2.14E+0	-	-	-
Tc-99	4.27E-1	6.88E+1	3.87E-2	-
I-129	1.83E+0	4.85E-1	-	-
Cs-134	5.79E-2	3.39E-1	-	-
Cs-137	1.29E+0	9.11E+0	-	1.57E-1
Plutonium- $\alpha$	1.61E-2	1.15E-1	-	-
Pu-241	2.21E-1	2.87E+0	-	8.67E-3
Am-241	3.49E-3	3.49E-2	-	-
Np-237	2.09E-4	-	3.00E-4	-

In Tables 2.8 and 2.9 the total alpha and total beta discharges to the North East Atlantic Ocean in the period 1994-1999 from different types of nuclear installations are shown (OSPAR, 2001). The discharges of alpha emitting radionuclides in 1994 were higher than in the following years. This corresponds roughly to the reduction of discharges from Sellafield in 1994-1995 due to the operation of the Enhanced Actinide Removal

The Dounreay facilities on the northern coast of Scotland were established in 1955, and have mainly been used in the development of technology of fast breeder reactors. Compared with the discharges from the other two reprocessing plants, the discharges to the marine environment are small. The amount of reprocessed nuclear fuel is also considerably lower.

Springfields is located in the west coast of United Kingdom, and liquid discharges are made to the Ribble estuary. This facility is mainly concerned with the manufacture of fuel elements for nuclear reactors and the production of uranium hexafluoride. The discharge of liquid radioactive waste from Springfields consists mainly of thorium and uranium and their daughter products.

Plant (EARP). This reduction in reprocessing discharges combined with a slight increase in discharges from nuclear fuel fabrication (mainly Springfields) resulted in a relative contribution of total alpha discharges of only 41.6% in 1999 compared to 80.9 % in 1994. The same trend can be seen for total beta discharges: The relative contribution of reprocessing discharges were reduced from 60.8 % in 1994 to 49.1% in 1999.

Discharges from Research and Development facilities have been reduced by roughly an order of magnitude in the six-year period. Nuclear power stations also discharge small amounts of radionuclides, but contributed at most a few percent of the total beta discharges.

Table 2.8 Assessment of liquid radioactive discharges of total alpha from nuclear installations in 1999 (OSPAR, 2001)

Total Alpha	1994	1995	1996	1997	1998	1999
All Nuclear Installations (TBq)	1.36	0.68	0.57	0.38	0.43	0.42
Reprocessing Plants (TBq)	1.1	0.47	0.32	0.23	0.22	0.17
% of all installations	80.9	69.1	56.1	60.5	51.2	41.6
Nuclear Power Plants (TBq)	-	-	-	-	-	-
% of all installations	-	-	-	-	-	-
Nuclear Fuel Fabrication (TBq)	0.16	0.12	0.12	0.12	0.20	0.24
% of all installations	11.8	17.6	21.1	31.6	46.5	57.7
Research & Development Facilities (TBq)	0.1	0.09	0.13	0.03	0.01	0.003
% of all installation	7.3	13.3	22.8	7.9	2.3	0.7

Table 2.9 Assessment of liquid radioactive discharges of total beta from nuclear installations in 1999 (OSPAR, 2001)

Total Beta (excluding Tritium for NPP)	1994	1995	1996	1997	1998	1999
All Nuclear Installations (TBq)	321	365	332	315	265	256
Reprocessing Plants (TBq)	195	243	169	167	112	126
% of all installations	60.8	66.5	50.9	53.0	42.4	49.1
Nuclear Power Plants (TBq)	2.8	3.4	5.2	7.4	2.0	2.0
% of all installations	0.9	0.9	1.6	2.3	0.8	0.7
Nuclear Fuel Fabrication (TBq)	114	112	150	140	150	128
% of all installations	35.5	30.7	45.1	44.4	56.6	50.0
Research & Development Facilities (TBq)	9.1	7.0	8.1	1	0.66	0.36
% of all installations	2.8	1.9	2.4	0.3	0.2	0.1

In addition to the direct discharges from reprocessing, remobilisation of cesium and plutonium from contaminated sediments in the Irish Sea acts as a secondary source of radionuclides to 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 seventies and early eighties 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 1.19 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 0.59 TBq. The half-lives of plutonium and americium in Irish Sea sediments were estimated to be 58 years and ~1000 years respectively. It has therefore been concluded that remobilisation of radionuclides from Irish Sea sediments is a major source of anthropogenic radionuclides to the North Sea and Norwegian coastal areas, and will remain so for a long time.

## 2.5 Fallout from atmospheric nuclear weapons testing and the Chernobyl accident

### 2.5.1 Global fallout from nuclear weapons testing

From a global perspective, the dominating radioactive contamination source is global fallout of debris from the atmospheric nuclear bomb tests conducted in different parts of the world between 1945 and 1980. In total, 520 atmospheric nuclear explosions were carried out; the periods of the most intensive

testing being in the years 1952-1954, 1957-1958 and 1961-1962 (UNSCEAR, 1993). The United States, the Soviet Union, the United Kingdom, France and the People's Republic of China mainly conducted atmospheric nuclear weapons tests. The three major tests sites for atmospheric testing were Novaya Zemlya (Russia) in the Arctic Ocean, the Bikini and Eniwetok Islands (USA) in the Pacific Ocean and the Nevada test site (USA) (Strand et al., 1998). It is assumed that 12% of the fallout activity has been deposited close to the test site, 10% has been deposited in a band around the earth at the latitude of the test site (tropospheric fallout) and 78% of the fallout activity has been spread over larger areas, mainly in the same hemisphere as the test site (UNSCEAR, 1993). Since most of the weapons tests were carried out in the northern hemisphere, the largest radioactive contamination is found there. These weapons tests have contributed to an overall background contamination level of long-lived fission products and transuranics in the northern marine environment. In terms of specific radionuclide releases, some 600 PBq  $^{90}\text{Sr}$  were released in the period 1952-1962 compared to a 910 PBq release of  $^{137}\text{Cs}$  (Balonov, 1997).

### 2.5.2 Outflow of Chernobyl fallout radionuclides from the Baltic Sea to the Norwegian Coastal Current

The accident at the Chernobyl nuclear power plant April 26, 1986, resulted in large-scale fallout in Europe. The accident was a consequence of uncontrolled fission in the reactor, followed by a powerful explosion and fire. The radioactive materials released were transported by air currents in the form of gases and dust particles. The prevailing

meteorological conditions at the time of the accident resulted in considerable radioactive fallout in Norway and Sweden.

Outflow of water from the Baltic Sea through Skagerrak is one of the major sources of radionuclides to the Norwegian coastal current and further into Arctic waters. According to Josefsson (1998), more than 90% of the net outflow of  $^{137}\text{Cs}$  from the Baltic Sea originates from the Chernobyl accident. The relatively high concentration of  $^{137}\text{Cs}$  in the water masses of the Baltic Sea is partly due to runoff from the Baltic drainage area. The general circulation pattern in the Skagerrak/Kattegat area is anticlockwise (Figure 2.5). The Baltic current carries the brackish surface waters out of the Baltic Sea and into the Skagerrak where it enters the Norwegian Coastal Current.

Higher salinity water enter the Baltic through a subsurface current from Skagerrak. Radionuclides released from La Hague and Sellafield are transported by this current into the Kattegat, where mixing with Baltic water occurs. The rate of water exchange between Skagerrak and the Baltic depends on weather conditions, and is subject to strong seasonal variations. Events of major inflows of saline waters to the Baltic can take place under specific weather conditions. Such an event occurred most recently in January 1993, leading to a correspondingly high outflow to Skagerrak.

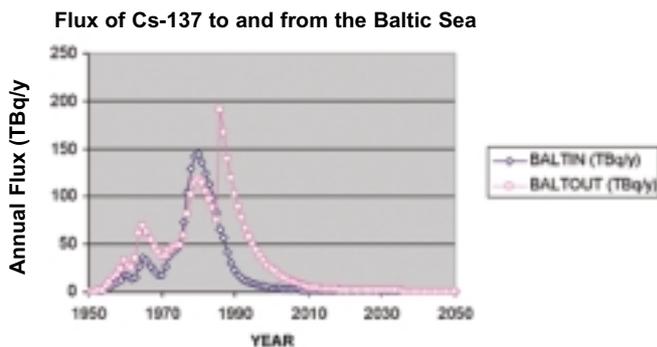


Fig. 2.7 Model calculations of  $^{137}\text{Cs}$  fluxes into and out of the Baltic Sea (Nielsen et al, 2000).

The net outflow of cesium from the Baltic Sea was estimated based on the observed concentrations of  $^{137}\text{Cs}$  and estimates of the average water exchange between Kattegat and Skagerrak. Based on the parameter values given in a box model (Helcom, 1995), and the average values of  $^{137}\text{Cs}$  in the North Sea and in the Kattegat area, an outflow of  $^{137}\text{Cs}$  of

100 TBq/year has been estimated for the year 1997 (Rudjord et al, 1999). This is an order of magnitude higher than reprocessing discharges, and comparable to the estimates for remobilised  $^{137}\text{Cs}$  for the Irish Sea. It should be emphasised however, that the calculation is based on activity concentrations in surface water from a single sampling campaign only therefore the values may not be representative for average concentrations of these water masses over a one-year period. In Figure 2.7, model calculations of the of  $^{137}\text{Cs}$  fluxes as functions of time are shown (Nielsen et al, 2000). For 1999, the model calculations gave an influx to the Baltic Sea of 5 TBq and outflow of 28 TBq  $^{137}\text{Cs}$ .

## 2.6 Other actual and potential sources of radioactivity to the northern marine environment

In addition to discharges from the western European reprocessing plants, northern marine waters have also received an input of different radionuclides from Russian reprocessing plants situated in the tributaries of the Russian rivers Ob and Yenisey (see Figure 2.4). These are the Mayak Production Association in the southern Urals, the Siberian Chemical Combine (Tomsk-7) north of the town of Tomsk and the Krasnoyarsk Mining and Chemical Combine (KMCC). Uncertainty exists in relation to the magnitude of radionuclide inputs 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 of 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 and Norwegian seas have been relatively low. Nonetheless, a potential for significant radionuclide contamination might exist following large accidental releases of radioactivity from these nuclear complexes.

Several other sources exist which also represent potential sources of radionuclides to 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 submarine Komsomolets in the Norwegian Sea. The dumping areas and the location of the sunken submarine Komsomolets are given in Figure 2.4. Several investigations have been conducted to detect any leakage from these sources. The Joint Russian Norwegian expert group has been investigating the condition of the contained dumped objects and analysed sediment and water samples at these sites. According to Strand et al. (1997), elevated levels of radionuclides in sediments collected in the vicinity of the dumped objects demonstrated that leakage had occurred, but that no inputs of radionuclides to the open sea from this source have been detected. The Russian nuclear submarine Komsomolets, following an accident on April 7th, 1989, 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.

### 3. Collection of samples

*Anne Lene Brungot, Norwegian Radiation Protection Authority (NRPA), Hilde Elise Helldal, Lars Føyn, The Institute of Marine Research (IMR).*

In 1999, samples for the monitoring programs for radioactivity in the marine environment were collected in the Barents Sea, the Norwegian Sea, the North Sea and Skagerrak, in selected fjords and at coastal stations including Svalbard. The participating institutions in the monitoring programme were, the Norwegian Radiation Protection Authority (NRPA), the Institute of Marine Research (IMR), the Norwegian Directorate of Fisheries, the Institute for Energy Technology (IFE) and the Norwegian Food Control Authority.

Various institutions collected the samples, but not all the samples were analysed for radionuclides. The rationale for this was that the cost of radionuclide analysis is a limiting factor. The extra samples are stored in case further analysis is needed for verification purposes or for more detailed investigations. Table 3.1 gives an overview of the sampling areas and number of analysed samples.

*Table 3.1 Overview of sampling areas and number of analysed samples 1999.*

	Sea water	Sediments	Fish	Sea weed	Other biota
Barents Sea	47	3 x 2	25 pooled 358 single		26
Norwegian Sea	1		5 pooled 18 single		11
North Sea/ Skagerrak	8	6	-		13
Fjords	10	11	22 pooled		1
Coastal stations Including Svalbard	11	-	-	72	2
Nitelva	-	4 x 9	-		

#### 3.1 Sea water

Sea water samples were collected at several locations along the Norwegian coast and in adjacent ocean waters at different depths. The water was collected during expeditions conducted by the Institute of Marine Research. In addition, monthly water samples for <sup>99</sup>Tc analysis were collected from Hillesøy in Troms and yearly samples were collected from locations in the southern part of Norway.



*Photo 3.1, B. Lind: Water samples collected on an expedition to the Barents Sea, (Jan.- Feb.1999) on the research vessel Johan Hjort (IMR).*

### 3.2 Sediment

In October 1999, NRPA participated in a sampling campaign close to the discharge point of the NALFA pipeline in the Nitelva river. In this location there is an area contaminated with plutonium and americium, mainly due to high discharges in the period 1968-1970. The contaminated area and discharge history has been described earlier (NRPA 1999:11) and also briefly in this report under chapter 2.1.2. Parallel samples were collected by the IFE as part of their regular monitoring programme.

Sediment samples from the North Sea areas and the fjords from Nordfjord in the south to Tanafjorden in the north were collected by the IMR during 1999. Sediments were retrieved from the seabed using a Smøgen box corer. The sediment samples were collected from the upper 2 cm layer. In addition, sediment samples were collected during the Barents Sea expedition in January and February 1999 on the research vessel Johan Hjort. Sediments were originally collected using a box-corer and a surface grab. Two profiles were collected from each box by slowly inserting PVC tubes of 100 mm diameter to a depth of 15-25 cm. The sediment samples were frozen shortly after sampling onboard the research vessel, and kept frozen until they were analysed.



Photo 3.2, B. K. Føyn: Collection of sediment samples in Iddefjord, onboard G.M. Dannevig (IMR) in 2001.

### 3.3 Fish and shrimps

The Norwegian Directorate of Fisheries collected fish and shrimp samples from commercial fishing locations. Individual fish were collected from five different locations around the Norwegian coast, but mainly from the Barents Sea. Four times per year, the Directorate of Fisheries collects fish and prepares pooled samples by combining muscle samples of 50 grams from 100 individual fish. Additionally, the Food Control Authority collected fish and shrimps and prepared pooled samples, each containing the meat from 25 fish.

Fish samples of cod, polar cod, capelin, flatfish, haddock and shrimps were collected during the expedition with R/V "Johan Hjort" to the Barents Sea in January and February in 1999.

In 1999, a total of approximately 2500 fish were analysed either individually or as pooled samples of no less than 25 fish; corresponding to more than 400 radionuclide analyses.

### 3.4 Lobsters, crabs and mussels

Lobsters and crabs were collected by local fishermen in different areas along the western and southern parts of the Norwegian coastline. Additionally King crab and starfish were collected during the Barents Sea expedition on the R/V "Johan Hjort" in January and February 1999.

At Hillesøy in northern Norway, mussels are collected four times each year. In southern Norway, mussels are collected once each year. The NRPA have analysed samples of lobster, crab and mussel for  $^{99}\text{Tc}$  and gamma emitting nuclides.

### 3.5 Sea Mammals

The Institute of Marine Research collected and analysed samples of porpoise from five different locations in Norway from Agder in the south to the coast of Finnmark in the north. Samples were collected opportunistically from porpoises caught accidentally by commercial bottom-set gillnet fisheries between April and June 1999, and April 2000. Thirty-six porpoises were collected (13 females, 23 males) from 5 different counties in Norway.

### 3.6 Seaweed and algae

The NRPA collected seaweed (*Fucus vesiculosus*) samples from two shoreline stations along the Norwegian coast. At Hillesøy in northern Norway, seaweed is collected every month to document seasonal fluctuations in radioactivity levels. In southern Norway, seaweed is collected once per year. From mid-1998, algae (*Laminaria hyperborea*) samples were collected from various locations ranging from the northern Norwegian Sea to the southern areas of the North Sea. IFE maintains a time series of seaweed (*Fucus vesiculosus*) sampling at ten locations along the Norwegian coastline from the Russian border in the north to the Swedish border in the south. Analysis results for 1999 sampling have been included in RAME. The sampling locations of all coastal stations are shown in Figures 6.3 and 6.4.

In September 1999 the NRPA local unit in Tromsø collected algae (*Laminaria hyperborea*) in Isfjorden at Svalbard.

All samples of seaweed and algae were collected and stored for later laboratory analyses for  $^{99}\text{Tc}$  and gamma emitting nuclides. Laboratory analyses were conducted at NRPA and IFE laboratories.



Photo 3.3, T. Bjerk: Collection of seaweed

## 4. Environmental radioactivity from Norwegian sources

*Anne Liv Rudjord, Norwegian Radiation Protection Authority (NRPA).*

### 4.1 Environmental contamination in Nitelva sediments

As the responsible operator of nuclear facilities requiring authorisation for radionuclide discharges, the Institute for Energy Technology (IFE) is responsible for carrying out monitoring of discharges and the resulting levels of radionuclides in the environment. The results of their monitoring programme are reported each year to the NRPA (NRPA 1999:11). NRPA, as the responsible authority on radiation protection, may carry out independent investigations, or collect separate control samples.

Based on the available information of the early discharges of radionuclides (mainly plutonium) to Nitelva, the NRPA decided to take part in the sampling of sediments near the discharge point in 1999, and to perform independent analysis of parallel samples.

The results of the analysis of the surface sediments are given in table 5.1. One of the samples has been analysed twice for americium and plutonium isotopes, and the result indicates some inconsistencies. This is not unexpected, as these radionuclides are highly particle reactive, and there may be some hot particles present.

It should be noted that the highest concentration was seen 2 meters west of the expected maximum point.

For comparison, the average concentrations as obtained by IFE are also shown. It should be emphasised that although the samples of IFE and NRPA were collected simultaneously, they were not identical. Some discrepancy is therefore expected in the results. However levels of contamination found here are consistent with the results of the internal IFE monitoring program.

Table 5.1 Concentration of radionuclides in surface sediments, Nitelva 1999.

Sample	Cs-137 Bq kg-1 d.w.	Co-60 Bq kg-1 d.w.	Pu-241 Bq kg-1 d.w.	Am-241 Bq kg-1 d.w.	Pu-238 Bq kg-1 d.w.	Pu-239,240 Bq kg-1 d.w.
North 3m	50 ± 2.5	n.d.	< 100	3.7 ± 0.9 5.7 ± 1.1	1.4 ± 0.4 1.3 ± 0.4	159 ± 10 126 ± 7
East 3m	138 ± 7	1.3 ± 0,13.	170 ± 70	20 ± 2.2	7.1 ± 0.95	737 ± 38
South 3m	109 ± 6	n.d.	< 120	7.2 ± 1.20	1.9 ± 0.5	185 ± 12
West 3m	52.6 ± 2.6	n.d.	< 100	13 ± 1.3	0.65 ± 0.2	54.0 ± 3
North 2m	77 ± 4	2.9 ± 0,23	1500 ± 180	15 ± 2.0	1.0 ± 0.3	141 ± 7
East 2m	117 ± 6	3.7 ± 0,26	460 ± 80	37 ± 3.1	10 ± 1.2	1150 ± 65
South 2m	104 ± 5	1.8 ± 0,16	< 110	6.6 ± 1.1	1.8 ± 0.4	182 ± 9
West 2m	401 ± 20	4.8 ± 0,29	1010 ± 120	139 ± 9	40 ± 3.3	4560 ± 260
Center	150 ± 0.7	1.9 ± 0,17	< 60	11 ± 1.3	2.8 ± 0.4	273 ± 12
Average	133	1.8	349 - 403	28.7	67	841
IFE results (average)	143	-	-	-	-	702

Internal investigations performed earlier by IFE showed that the radioactivity concentration was even higher in deeper sediments. This is due to a high sedimentation rate in the area, and contamination that is due mainly to discharges in the sixties and early seventies.



Photo 4.1, S. Borghuis: The contaminated area in the river Nitelva.

The general public and the NRPA were concerned by the high levels of plutonium contamination. The water levels of the Nitelva are regulated, and may vary strongly, especially during the spring season, when the water level may be lowered in order to avoid flooding as a result of snow-melt. In such situations, the contaminated sediments may be exposed possibly resulting in radiation doses to the public exceeding present dose limits. Taking the above into account, the NRPA ordered a clean-up operation. All sediments with concentrations of transuranics (plutonium + americium) higher than 10 Bq g<sup>-1</sup> were removed in early spring 2000.

## 5. Radioactivity in sea water and sediments

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Ocean currents transport water masses containing anthropogenic nuclides to the Norwegian coast as mentioned in Chapter 2.4. In 1999, surveys were carried out in the Barents Sea and in ten different Norwegian Fjords. A few samples were also collected in the Norwegian Sea, the North Sea and Kattegat. The observed activity concentrations of <sup>137</sup>Cs, <sup>99</sup>Tc, <sup>238</sup>Pu and <sup>239+240</sup>Pu in sea water and sediments are reported below.

### 5.1 Caesium-137 in sea water and sediments

Sea water concentrations of <sup>137</sup>Cs in the eastern Barents Sea, ten different Norwegian Fjords and in one sample from Kattegat are presented in Figure 5.1.

The activity concentrations of <sup>137</sup>Cs in the eastern Barents Sea vary in the range from 2.8 ± 0.1 Bq m<sup>-3</sup> to 4.0 ± 0.2 Bq m<sup>-3</sup> with a mean value of 3.4 Bq m<sup>-3</sup>. This is similar to the results from the “Gauss” cruise in 1995, where the mean concentration of <sup>137</sup>Cs at three stations in the Barents Sea was 3.6 Bq m<sup>-3</sup> (range 3.3-4.2 Bq m<sup>-3</sup>) (Grøttheim, 2000).



Concentrations of  $^{137}\text{Cs}$  in surface sediments collected in the North Sea and in Kattegat are shown in Figure 5.2. The levels are lower compared to the levels in the Norwegian fjords.

Two near-bottom water samples, from Station 3 and Station 6 in the Barents Sea were also analysed for  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$ . These data are compared with surface water results in Table 5.1.

These data are interesting because the  $^{99}\text{Tc}$  activity concentrations in the upper water column are distinctly higher than those in the lower part of the water column. This is despite the fact that, at both locations, there was little difference in the salinity recorded in the top and bottom waters. The water columns at Stations 3 and 6 were not vertically stratified and apparently not formed from more than one water mass. The activity concentrations of  $^{137}\text{Cs}$ , in contrast, appear to be more uniform with depth, although at station 6 the concentration in the near bottom water seems to be lower than in the surface water. In Atlantic water unaffected by reprocessing discharges, the concentrations of  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$  from nuclear weapons fallout are expected to be approximately  $2.3 \text{ Bq m}^{-3}$  (decay corrected) and  $0.005 \text{ Bq m}^{-3}$  respectively (Dahlgard et al, 1995). Results from work conducted in the summer of 1995 (Grøttheim, 2000) show a fairly uniform  $^{137}\text{Cs}$  activity concentration with depth for sampling locations in the Barents Sea.

Table 5.1  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  activity concentrations ( $\text{Bq m}^{-3}$ ) in surface and bottom sea water samples collected in the Barents Sea.

Station	Depth (m)	Salinity ‰	$^{99}\text{Tc}$ ( $\text{Bq m}^{-3}$ )	$^{137}\text{Cs}$ ( $\text{Bq m}^{-3}$ )
3	Surface	0	0.97 ± 0.08	3.65 ± 0.17
	Bottom	132	0.54 ± 0.04	3.41 ± 0.17
6	Surface	0	0.52 ± 0.04	3.35 ± 0.13
	Bottom	230	< 0.10 ± 0.08	2.5 ± 0.13

## 5.2 Technetium-99 in sea water

Activity concentrations of  $^{99}\text{Tc}$  in sea water from the Norwegian Sea and the Barents Sea in 1999 ranged from  $0.09\text{--}1.14 \text{ Bq m}^{-3}$ , Figure 5.4. The highest levels of  $^{99}\text{Tc}$  were observed in the eastern part of the Seas. The results reflect the general surface circulation pattern of the North Atlantic and adjacent seas.



Fig. 5.4 Activity concentrations of  $^{99}\text{Tc}$  ( $\text{Bq m}^{-3}$ ) in sea water samples collected in 1999.

Towards the central parts of the Barents Sea, Atlantic water carrying relatively low levels of  $^{99}\text{Tc}$  predominates, while sea water from the Norwegian Coastal Current with enhanced levels of  $^{99}\text{Tc}$  flows into the southern part of the Sea.

In September 1999, the concentration of  $^{99}\text{Tc}$  in one sea water sample collected from Isfjorden at Svalbard was  $0.13 \text{ Bq m}^{-3}$ .

Since July 1997, samples of sea water have been collected regularly at Hillesøy in northern Norway. The  $^{99}\text{Tc}$  activity concentrations as function of time are shown in Figure 5.5.

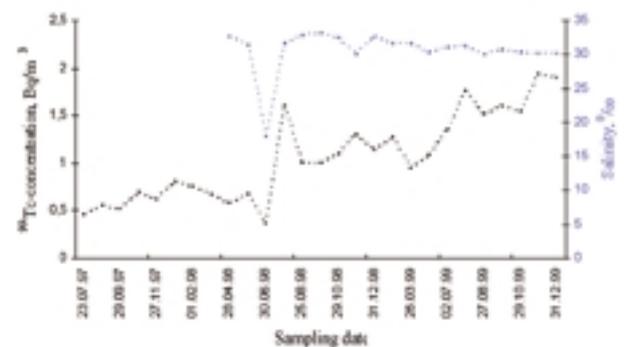


Fig. 5.5 Activity concentrations of  $^{99}\text{Tc}$  in sea water samples collected at Hillesøy in Troms.

The  $^{99}\text{Tc}$  level in sea water increased by a factor of four between July 1997 and December 1999. The mean value for the years 1997 (six samples), 1998 and 1999 were  $0.61 \text{ Bq m}^{-3}$ ,  $0.91 \text{ Bq m}^{-3}$  and  $1.5 \text{ Bq m}^{-3}$  respectively.

The levels of  $^{99}\text{Tc}$  in the North Sea ranged from  $0.46\text{--}7.2 \text{ Bq m}^{-3}$  and are consistent with data from H. Nies et. al., 2000. Both surveys have found concentrations of around  $2 \text{ Bq m}^{-3}$  close to Aberdeen and a maximum concentration of  $^{99}\text{Tc}$  at around  $7 \text{ Bq m}^{-3}$  at the west coast of Denmark.

### 5.3 Plutonium-238 and plutonium (239+240) in sea water

Activity concentrations of  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$  in sea water samples collected in the Barents Sea and the Norwegian Sea are shown in Figure 5.6. The levels of  $^{239+240}\text{Pu}$  in surface waters vary from  $6.6 \pm 0.5 \text{ mBq m}^{-3}$  to  $9.9 \pm 1.0 \text{ mBq m}^{-3}$ . Activity concentrations were fairly uniform over a large part of the sampling transect but fell slightly at locations very

Station	Depth (m)	Salinity	$^{239+240}\text{Pu}$ ( $\text{mBq m}^{-3}$ )	$^{238}\text{Pu}$ ( $\text{mBq m}^{-3}$ )	$^{238}\text{Pu}/^{239+240}\text{Pu}$
3 Surface	0	34.63 ‰	$9.1 \pm 0.8$	$0.73 \pm 0.24$	$0.08 \pm 0.03$
3 Bottom	132	34.64 ‰	$12.8 \pm 1.1$	$3.0 \pm 0.4$	$0.23 \pm 0.04$
6 Surface	0	34.80 ‰	$9.9 \pm 1.0$	$0.49 \pm 0.18$	$0.05 \pm 0.02$
6 Bottom	230	34.87 ‰	$7.3 \pm 1.0$	$0.56 \pm 0.25$	$0.07 \pm 0.02$

close to the Kola Bay. In comparison based on 15 measurements a mean activity concentration of  $7.1 \pm 1.7 \text{ mBq m}^{-3}$  was found in 1995 by Grøttheim, (Grøttheim, 2000). The pattern observed in the case of  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$ , where elevated activity concentrations appear to be associated with the North Cape Current (NCC), was absent for  $^{239+240}\text{Pu}$ .



Fig. 5.6 Activity concentrations of  $^{239+240}\text{Pu}$  ( $\text{mBq m}^{-3}$ ) in surface waters in 1999.

In general, activity ratios can be an important tool in identifying the sources of radioactive contamination, and in Table 5.2 the  $^{238}\text{Pu} / ^{239,240}\text{Pu}$  activity ratios commonly associated with different sources are listed.

In surface waters collected in the Barents Sea the  $^{238}\text{Pu}$  to  $^{239+240}\text{Pu}$  activity ratios were in the range of 0.025 and 0.14. The ratios indicate that global fallout is the main contributor to the plutonium inventory in the Barents Sea waters, but other sources may be contributing in a few samples.

Table 5.2  $^{238}\text{Pu} / ^{239+240}\text{Pu}$  activity ratio associated with radionuclide sources.

Source	$^{238}\text{Pu} / ^{239+240}\text{Pu}$ activity ratio
Global fallout	$\sim 0,04$ (0,03)
Sellafield, historical	$\sim 0,20$
Sellafield, recent	$\sim 0,30$
Chernobyl fallout	$\sim 0,50$

Table 5.3  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  activity concentrations ( $\text{mBq m}^{-3}$ ) in surface and bottom sea water samples from Stations 3 and 6 in the Barents Sea.

Two bottom water samples from Station 3 and Station 6 in the Barents Sea were also analysed for  $^{239+240}\text{Pu}$  and  $^{238}\text{Pu}$ . These data are presented in Table 5.3 along with data for surface waters for comparison purposes. The results show that at Station 3 bottom water activity concentrations were higher than surface concentrations, whereas at Station 6 bottom water concentrations were lower than surface concentrations. At station 3 (bottom water) the activity ratio of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  of 0.23 may indicate that there is a contribution from earlier discharges from Sellafield, but a single result is not sufficient to provide conclusive evidence.

## 6. Radioactivity in fish and other biota

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The Norwegian Radiation Protection Authority (NRPA) in co-operation with the Institute of Marine Research (IMR) and Directorate of Fisheries collected and measured samples of fish, lobster, mussels, starfish, harbour porpoises and shrimps in 1999. In addition, results from the Food Control Authorities monitoring program of fish are presented in this chapter. The NRPA and the Institute of Energy Technology (IFE) have also collected and measured samples of seaweed along the Norwegian coast and at Svalbard in 1999.

### 6.1 Fish

Since 1994 the NRPA and the Norwegian Directorate of Fisheries have conducted a monitoring programme to determine the levels of  $^{137}\text{Cs}$  in fish. This involved the analysis of approximately 300 fish samples per annum using NaI(Tl) detectors. The  $^{137}\text{Cs}$  results for all these samples were below the limit of detection for this equipment (d.l. =  $5.5 \text{ Bq kg}^{-1}$ ). In the same screening program the NRPA also analysed pooled fish samples using equipment with a limit of detection in the area of  $0.1$  to  $0.2 \text{ Bq kg}^{-1}$  wet weight. These results are given in Table 6.1 on page 24.

Samples of fish were also collected during an expedition to the Barents Sea on board the research vessel R/V Johan Hjort in January and February 1999. The samples have been analysed for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  and the results are shown in Table 6.1.

In the Barents Sea the observed level of  $^{137}\text{Cs}$  in 1999 was  $0.5 \text{ Bq kg}^{-1}$  or less for cod. Concentrations of  $^{90}\text{Sr}$  in cod and plaice ranged from  $0.0094 \text{ Bq kg}^{-1}$  to  $0.025 \text{ Bq kg}^{-1}$  in muscle samples.  $^{90}\text{Sr}$  behaves like an analogue for calcium, and higher concentrations are therefore found in fish bones.



Photo 6.1 E. Svensen: Cod (*Gadus morhua* L.).

In the Norwegian Sea the concentrations of  $^{137}\text{Cs}$  ranged from  $0.4$  to  $0.8 \text{ Bq kg}^{-1}$  (wet weight). The concentration of  $^{137}\text{Cs}$  in cod was  $0.5 \text{ Bq kg}^{-1}$  (wet weight).

In Figure 6.1 the typical  $^{137}\text{Cs}$  concentrations observed in fish caught in different areas can be compared. The levels in the seas around Norway are very low. In the Baltic Sea, the uptake of cesium in fish is higher due to low salinity waters. In the Irish Sea, levels are influenced by past and present discharges from Sellafield.

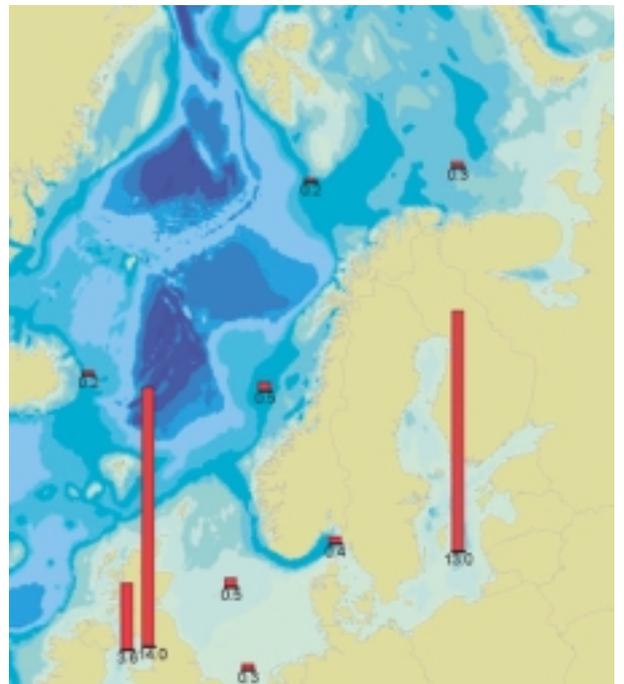


Fig.6.1 Levels of  $^{137}\text{Cs}$  in cod from different areas data from MAFF&SEPA, 2000 and this report.

Table 6.1 <sup>137</sup>Cs and <sup>90</sup>Sr activity concentrations (Bq kg<sup>-1</sup> wet weight) for various fish species collected from the Norwegian and Barents Sea in 1999. Data obtained by the NRPA, IMR, the Local office of the Directorate of Fisheries in Troms, IFE or the Norwegian Food Control Authority

Species	Location	<sup>137</sup> Cs (Bq kg <sup>-1</sup> ) Meat	<sup>90</sup> Sr* (Bq kg <sup>-1</sup> ) Meat/bone	Number of samples	Number of fish
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	0.2-0.4	-	12 pooled samples	1200 muscle
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	0.2-0.5	-	13 pooled samples	325 muscle
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	0,2 – 0,3	0.0094 / 0.089	20 samples	20 muscle
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	-	0.025	1 sample	1 meat
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	< 5.5	-	300 samples	300 muscle
Polar cod ( <i>Boreogadus saida</i> )	Barents Sea	n.d.- 0,2	-	5 samples	5 muscle
Haddock ( <i>Mellangorammus aeglefinus</i> )	Barents Sea	n.d.- 0,3	< 0.007 / -	7 samples	7 muscle
Capelin ( <i>Mallotus villosus</i> )	Barents Sea	n.d.- 0,2	0.0055 / -	10 samples	10 muscle
Norway redfish ( <i>Sebastes viviparus</i> )	Barents Sea	0,2 – 0,3	-	2 samples	2 muscle
Flatfishes ( <i>Pleuronectiformes</i> )	Barents Sea	n.d.- 0,4	0.015 / -	6 samples	6 muscle
European Plaice ( <i>Pleuronectes platessa</i> )	Barents Sea	0,3 – 0,4	0.023 / -	2 samples	2 muscle
Raudåte ( <i>Calanus finmarchius</i> )	Barents Sea	n.d.-0,2	-	3 samples	3 muscle
Fry of Saithe ( <i>Pollachius virens</i> )	Barents Sea	0,1	-	1 sample	1 pooled
Cod ( <i>Gadus morhua</i> L.)	Norwegian Sea	0,5	-	1 muscle	1 muscle
Atlantic Herring ( <i>Clupea harengus</i> )	Norwegian Sea	n.d. -0,1	-	2 sample	2 muscle
Mackerel ( <i>Scomber scombrus</i> )	Norwegian Sea	0,1	-	1 samples	1 muscle
Cusk ( <i>Brosme</i> )	Norwegian Sea	0.4-0.8	-	2 pooled samples	50 muscle
Haddock ( <i>Mellangrammus aeglefinus</i> )	Norwegian Sea	0.3-0.6	-	3 pooled samples	75 muscle
Fry of Atlantic herring ( <i>Clupea harengus</i> )	Norwegian Sea	n.d. -0,1	-	2 samples	2 pooled
Fry of Saithe ( <i>Pollachius virens</i> )	Norwegian Sea	n.d. - 0,1	-	11 samples	11 pooled

In 1999 the IMR and the Norwegian Food Control Authority sampled and analysed fish caught in nine different Norwegian fjords. The results are given in Table 6.2.

The levels of <sup>137</sup>Cs in fish caught in the fjords may be slightly higher than in the open sea, but all the observed activity concentrations are still less than 1 Bq kg<sup>-1</sup> wet weight. Slightly higher concentrations of <sup>137</sup>Cs in fish from fjords can be expected, partly because uptake in fish may be enhanced by lower salinity in the fjord waters. Also, the <sup>137</sup>Cs concentrations in fjord water seem to be slightly higher than the Barents Sea concentrations (see Chapter 5.1).

## 6.2 Crustacea and molluscs

In 1999 samples of lobster, crabs, mussel, starfish and shrimps were collected and analysed for <sup>99</sup>Tc and <sup>137</sup>Cs.

Following the increase of <sup>99</sup>Tc discharges from Sellafield in 1994, high concentrations of this radionuclide were detected in crustacea and molluscs in the Irish Sea. Highest levels were observed in lobster. Lower concentrations were observed in shrimps and in mussels. In the Irish Sea, excluding the areas close to Sellafield, <sup>99</sup>Tc levels in lobsters of several thousands Bq kg<sup>-1</sup> have been measured.

Table 6.2 <sup>137</sup>Cs activity concentrations (Bq kg<sup>-1</sup> wet weight) for fish collected from Norwegian fjords in 1999, analysed by the Norwegian Food Control Authority in Salten and the Institute of Marine Research.

Species	Location	<sup>137</sup> Cs (Bq kg <sup>-1</sup> )	No & type of sample	No of fish & type of sample
Atlantic Salmon ( <i>Salmo salar</i> )	Varangerfjorden	0.8	1 pooled	25 muscle
Haddock ( <i>Melanogrammus aeglefinus</i> )	Porsangerfjorden	0,13	1 pooled	5 muscle
Long rough dab ( <i>Hippoglossoides platessoides</i> )	Porsangerfjorden	0,20	1 pooled	Many fish
Atlantic Salmon ( <i>Salmo salar</i> )	Vestfjorden	0.3 – 0.4	5 pooled	125 muscle
Atlantic Salmon ( <i>Salmo salar</i> )	Lyngen	0.44	1 pooled	25 muscle
Saithe ( <i>Pollachius virens</i> )	Skreigrunnen v/ Freia	0.3	1 pooled	25 muscle
Atlantic Salmon ( <i>Salmo salar</i> )	Gammelvær på Seiland	0.8	1 pooled	25 muscle
Sprat ( <i>Sprattus sprattus</i> )	Namsfjorden	0.8	2 pooled	Many fish
Haddock (small) ( <i>Melanogrammus aeglefinus</i> )	Romsdalsfjorden	0.8	1 pooled	Many fish
Haddock ( <i>Melanogrammus aeglefinus</i> )	Romsdalsfjorden	0.6	1 pooled	3 muscle
Whiting ( <i>Merlangius merlangus</i> )	Romsdalsfjorden	0.7	1 pooled	5 muscle
Sprat ( <i>Sprattus sprattus</i> )	Romsdalsfjorden	0.5	2 pooled	Many fish
Three-spined stic ( <i>Gasterosteus aculeatus</i> )	Nordfjord	0.4	1 pooled	Many fish
Krill ( <i>Euphausiidae</i> )	Nordfjord	0.4	3 pooled	Three bags with krill

The levels of <sup>99</sup>Tc are considerably lower along the Norwegian coast. The results from 1998 and 1999 are given in Table 6.3. Activity concentrations of <sup>137</sup>Cs were at or below detection limits in many cases for crustacean and mollusc samples, especially in the Barents Sea areas. The concentration of <sup>239,240</sup>Pu in starfish (Asteroidea) collected in the Barents Sea was 62.0 mBq kg<sup>-1</sup>. <sup>99</sup>Tc and <sup>137</sup>Cs in starfish from the Barents Sea were not detectable.

### 6.3 Seaweed

In 1999 the NRPA and the Institute of Energy Technology collected samples of seaweed at coastal stations for analysis of <sup>137</sup>Cs and <sup>99</sup>Tc.

Seaweed (*Fucus vesiculosus*) has previously been used as an indicator species for radionuclides in sea

water. Long-term monitoring of the Norwegian coast shows that <sup>137</sup>Cs levels in seaweed species have responded to changes associated with the main sources of radioactivity in the marine environment.

Levels of <sup>99</sup>Tc were relatively high (in the range of 50 – 70 Bq kg<sup>-1</sup> dry weight) in Norwegian coastal areas during the 1980s (Dahlgard et al, 1995) reflecting the elevated discharges from Sellafield in 1978 and La Hague in the preceding years.

In the late 1980 and the early 1990s, a significant decrease in levels was observed corresponding to the throughput of this radionuclide at Sellafield (before operation at EARP began) and the declining discharges from La Hague. The levels fell to approximately 30 Bq kg<sup>-1</sup>.



Photo 6.2, E. Svensen. Lobster (*Homarus vulgaris*)

From 1997 to 1999 a significant increase of  $^{99}\text{Tc}$  in seaweed (*Fucus vesiculosus*) was observed at Hillesøy (Figure 6.2) in the north of Norway. Seaweed from Hillesøy was collected monthly from July 1997 to the end of 1999 and analysed for trends. The maximum concentration ( $335 \text{ Bq kg}^{-1}$  dry weight) was found in December 1999.

Monthly sampling of seaweed (*Fucus vesiculosus*) was also conducted at Utsira. For 1999, the  $^{99}\text{Tc}$  concentrations varied in the range from  $244 \text{ Bq kg}^{-1}$  to  $510 \text{ Bq kg}^{-1}$  (dry weight), with an average of  $401 \text{ Bq kg}^{-1}$ .



Photo 6.3, E. Svensen. Seaweed.

The results for  $^{99}\text{Tc}$  in seaweed (*Fucus vesiculosus*) from 10 coastal stations in 1999 are shown in Figure 6.3. The numbers given for Hillesøy and Utsira represent average concentrations of monthly samples with the minimum and maximum values given in brackets.

Table 6.3 Concentrations of  $^{40}\text{K}$ ,  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$  in crustacea and molluscs collected in 1998 and 1999.

Species	Sampling place	$^{40}\text{K}$ -activity concentration $\text{Bq kg}^{-1}$ w.w.	$^{137}\text{Cs}$ -activity concentration $\text{Bq kg}^{-1}$ w.w.	$^{99}\text{Tc}$ -activity concentration $\text{Bq kg}^{-1}$ w.w.
Shrimps, ( <i>Pandalus borealis</i> )	Barents Sea	77 - 94	0,1 – 0,2	n.d*
Kamchatca crab	Barents Sea	77 - 82	n.d – 0,2	n.d*
Starfishes ( <i>Asteroidea</i> )	Barents Sea	11	< 0,02	n.d*
Shrimps, ( <i>Pandalus borealis</i> )	Porsangerfjord	-	0,18 (+/- 0,05)	n.d*
Mussels incl. exoskeleton ( <i>Mytilidae</i> )	Hillesøy	66- 200	*Bdl – 0,2	0,40 ±0,05 0,50 ±0,06
Crabs, ( <i>Brachyura</i> )	Norwegian Sea (Gulen)	46 - 66	*Bdl	-
Norway lobster ( <i>Nephrops norvegicus</i> )	Coast of Rogaland	-	*Bdl	12 ±1,2
Shrimps, ( <i>Caridea</i> )	Skagerrak (Larvik)	25	0,05	-
Edible Crabs, ( <i>Cancer pagurus</i> )	Skagerrak (Larvik)	50 - 100	n.d.	-
Edible Crab exoskeleton ( <i>Cancer pagurus</i> )	Skagerrak (Larvik)	150	*Bdl – 0,3	-
Lobster ( <i>Homarus gammarus</i> )	Outer Oslofjord	-	0.13-0.16	14,4 ±1,5 26,2 ±2,6
Lobster exoskeleton ( <i>Homarus gammarus</i> )	Outer Oslofjord	70 - 150	0,2 – 0,4	-

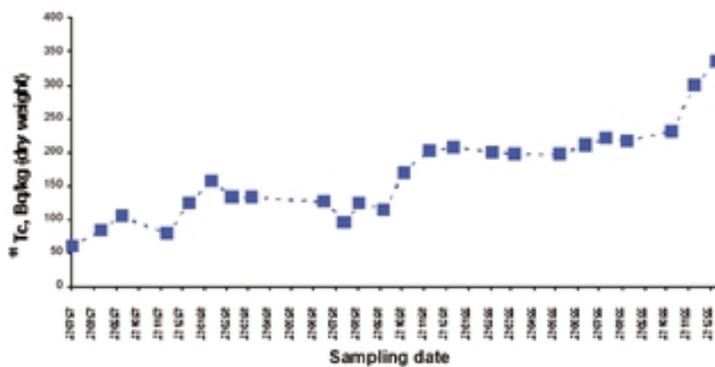


Fig. 6.2. Concentrations of  $^{99}\text{Tc}$  in seaweed (*Fucus vesiculosus*) collected at Hillesøy in the period from 1997 to 1999.



Fig. 6.3 Concentrations of  $^{99}\text{Tc}$  in seaweed (*Fucus vesiculosus*) collected along the Norwegian coastline in 1999. Data obtained by IFE and NRPA. Measurement uncertainties are in the range 3-22%.

The highest concentrations were found at Utsira and Tromsøy in southern Norway and at Indre Kiberg in the north. On the west coast, from Sognesjøen to Vikna, concentrations were fairly constant, in the range of 254 to 289 Bq kg<sup>-1</sup>. The largest variations are seen in northern parts of Norway, where concentrations range from 107-347 Bq kg<sup>-1</sup>. Samples are collected in August or September, therefore seasonal variations cannot explain these observations. Also, it seems unlikely that salinity or temperature effects should produce such a large variation. A possible explanation may be fluctuations in the sea water concentrations due to pulsed discharges from Sellafield and/or mixing

with Atlantic waters with low concentrations of  $^{99}\text{Tc}$ . The sea water data at Hillesøy, shows that concentrations vary through the year by approximately a factor of two in 1999, although the salinity is reasonably constant (about 30 ‰).

In Table 6.4 the levels of  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$  in kelp (*Laminaria hyperborea*) stems and leaves from Svalbard are reported. The  $^{137}\text{Cs}$  concentrations range from below detection limit up to 1.1 Bq kg<sup>-1</sup> (dry weight). Concentrations of  $^{99}\text{Tc}$  are in the range of 2.5-3.25 Bq kg<sup>-1</sup> in the leaves and 8.0-10.3 Bq kg<sup>-1</sup> in the stems. This corresponds to average concentration factors of 23.000 for the leaves and 69.000 for the stems. This can be compared with data from the west coast of Norway in 1998, where concentration factors for *Laminaria Hyperborea* varied from 7.300–8.600 for the leaves and 11.000-17.000 for the stems (Kolstad A.K. and Rudjord A.L, 2000). It should be noted, however, that this calculation is based on a single measurement of  $^{99}\text{Tc}$  sea water from Isfjorden, Svalbard. This may indicate a substantially higher concentration factor for  $^{99}\text{Tc}$  in *Laminaria hyperborea* at Svalbard compared to the west coast of Norway, but more data are required to confirm this.

Table 6.4 Levels of  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$  in kelp (*Laminaria hyperborea*) collected at Isfjorden, Svalbard in September 1999.

Kelp ( <i>Laminaria hyperborea</i> )	$^{137}\text{Cs}$ -activity concentration Bq kg <sup>-1</sup> d.w.	$^{99}\text{Tc}$ -activity concentration Bq kg <sup>-1</sup> d.w.
Leaves	n.d.	3.3 ±0,4
Stem	n.d.	10.3 ±1,0
Leaves	0.35	3.1 ±0,4
Stem	n.d.	8.0 ±0,8
Leaves	0.42	2.5 ±0,3
Stem	1.10	8.5 ±0,9

(n.d. = not detected)

In 1999, six seaweed (*Fucus vesiculosus*) samples collected at Hillesøy were analysed for  $^{137}\text{Cs}$ . The average concentration was 0.55 Bq kg<sup>-1</sup> (dry-weight). This is similar to the average concentrations from 1997 (three samples) and 1998 (nine samples) of 0.49 Bq kg<sup>-1</sup> and 0.54 Bq kg<sup>-1</sup> respectively. In samples analysed from 1997 to 1999 the results were in the range of 0.2 to 0.8 Bq kg<sup>-1</sup>. In Figure 6.4 the results of  $^{137}\text{Cs}$  from the Norwegian coast are shown.



Fig. 6.4 Concentrations of  $^{137}\text{Cs}$  in seaweed (*Fucus vesiculosus*) collected along the Norwegian coastline in 1999. Data obtained by IFE and NRPA. Measurement uncertainties are in the range 6-10 %, except for Gresse Jakobselv (uncertainty 26 %)

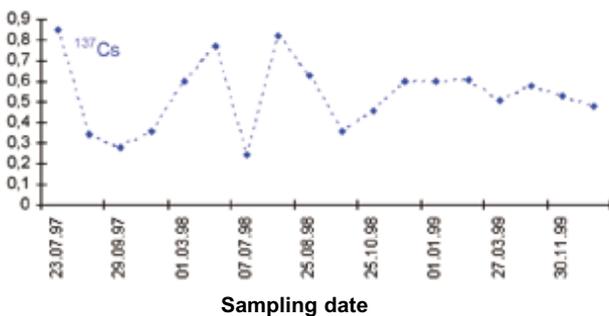


Fig. 6.5 Concentrations of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  dry weight) in seaweed (*Fucus vesiculosus*) collected at Hillesøy in the period from 1997 to 1999. Measurement uncertainties are in the range 15-23 %, if  $^{137}\text{Cs}$  were detected.

Along most of the western coast of Norway, the concentration of  $^{137}\text{Cs}$  in seaweed is similar to levels found in 1993 and 1995. At the Tromsø station, a trend of increasing concentrations is indicated ( $2.7 \pm 0.2 \text{ Bq kg}^{-1}$ ,  $3.7 \pm 0.2 \text{ Bq kg}^{-1}$  and  $4.7 \pm 0.3 \text{ Bq kg}^{-1}$  in 1993, 1995 and 1999, respectively, Sickel et al 1995, Brungot et al 1997 and 1999). This is probably due to fluctuations in salinity and/or variations in the outflow from the Baltic Sea.

The highest values are found at Hvaler ( $9.4 \pm 0.3 \text{ Bq kg}^{-1}$ ,  $6.6 \pm 0.6 \text{ Bq kg}^{-1}$ , and  $9.1 \pm 0.6 \text{ Bq kg}^{-1}$  for 1993, 1995 and 1999, respectively). The relatively high concentrations observed at Hvaler are due to low salinity waters (average approximately 17 ‰) combined with the influence of Chernobyl contaminated Baltic waters.

## 6.4 Harbour porpoise

Geographic variations in the levels of radioactive  $^{137}\text{Cs}$  have been examined from the axial musculature of harbour porpoises (*Phocoena phocoena*) from five locations along the coast of Norway (Tolley KA, Heldal HE, 2001).

The levels of  $^{137}\text{Cs}$  in harbour porpoises ranged from  $0.3 - 3.8 \text{ Bq kg}^{-1}$  wet weight. Porpoises caught in Vest-Agder, the location situated closest to the main sources, contained the highest levels of  $^{137}\text{Cs}$ . There were no significant differences in the  $^{137}\text{Cs}$  levels among the groups from Rogaland, Hordaland and Nordland. The levels of  $^{137}\text{Cs}$  in porpoises caught in Rogaland are lower than expected based on ambient levels of  $^{137}\text{Cs}$  in sea water. In southern Norway, the Coastal Current (NWCC), which transports  $^{137}\text{Cs}$  northwards, is relatively narrow. If porpoises in southwest Norway (i.e. Rogaland) are undergoing movements out of the narrow NWCC and into less contaminated Atlantic water, they may show levels of  $^{137}\text{Cs}$  in their tissue. At higher latitudes, the NWCC mixes with Atlantic water, and the zone of  $^{137}\text{Cs}$  contamination tends to extend westwards. Thus, variability associated with the NWCC is less of a factor in northern regions.

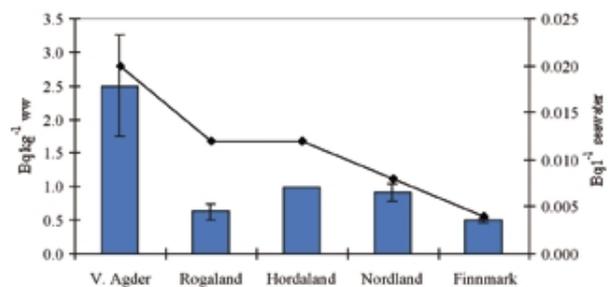


Fig. 6.6 Mean levels and standard errors of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$  wet weight) in harbour porpoises (*Phocoena phocoena*) from five locations in Norway (bars). The solid line shows median concentrations of  $^{137}\text{Cs}$  in sea water within each area.

## 7. Development of assessment tools

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### 7.1 Transfer factors and transit times for Sellafield <sup>99</sup>Tc discharges

Time series data concerning <sup>99</sup>Tc activity concentrations in sea water at Hillesøy have been used to derive transfer times and transfer factors from the Sellafield reprocessing plant in the UK to northern Norwegian coastal areas (Brown et al, 2001 in press). This has been achieved through the comparison of monthly <sup>99</sup>Tc discharge data (smoothed to allow for the dispersion of contamination downstream) with activity concentrations in sea water (normalised to a salinity of 33 ‰) sampled from

BCF (l/kgw.w.)	Cs	Tc	Sr (muscle)	Sr
Shrimp	24-54	< 444	11-19	26 (shell)
Crustaceans - IAEA*	30 (10-50)	1000 (500-50000)	2 (0.1-5)	
Fish	-	-	6-15	52 (bone)
Fish - IAEA*			2 (0.3-10)	

Hillesøy at monthly intervals using a technique known as cross-correlation analysis.

The results from this analysis produced a transit time of 42 months and a transfer factor of 6 Bq m<sup>-3</sup> per PBq a<sup>-1</sup>. The transit time falls within the range of those reported in earlier studies (e.g. Dahlgard, 1995) whereas the transfer factor is slightly lower than earlier results.

The reason for the discrepancy in the transfer factors is unclear but may reflect differences in the locations for which the time series environmental activities were derived. Clearly an open sea area receiving inputs of marine-derived contamination might be expected to produce a different “signal” to a coastal area where any marine-derived signal may be diluted by freshwater inputs. Predictions based on the analyses of the discharge pattern from Sellafield suggested that activity concentrations in northern Norwegian coastal waters were unlikely to increase dramatically above the levels observed in 1998. This appears to have been confirmed by recent data collated following this research work.

### 7.2 Uptake and transfer of radionuclides in marine food-webs

The expedition to the Barents Sea conducted in February 1999 has been the basis for research considering the uptake and transfer of radionuclides in marine food-webs. The research work was conducted under the Norwegian “Transport and Effects” programme.

Although concentration factors are based on an assumption of equilibrium conditions they do provide an indication, at least, of the degree of accumulation for defined radionuclides within organisms. From this, a basic assessment can be made as to whether certain trophic levels are more prone to accumulation than others. The degree of uptake will depend to some extent on the chemical environment and the physicochemical form of the radionuclide.

*Table 7.1 Calculated concentration factors compared with IAEA (1985) recommended BCF values.*

Preliminary analysis of the data shows that <sup>137</sup>Cs concentration factors fall within the generic values reported by the IAEA. The activity concentrations of <sup>137</sup>Cs have been falling slowly over the last decade or so which means that the sediment-water-biota system might be considered to be as close to an equilibrium situation as one might observe in the marine environment. The situation for <sup>99</sup>Tc is somewhat different because the northern marine system may still be responding to the step increase in discharges of this radionuclide which occurred following the commissioning of the Enhanced Actinide Removal Plant (EARP) at Sellafield in 1994. It is also known that different species of crustacean exhibit quite different levels of <sup>99</sup>Tc uptake (Busby et al., 1997; Brown et al., 1999). Shrimp are a crustacean species that are remarkable for their relatively low uptake of this radionuclide (Brown et al, 1999).

### 7.3 Doses to biota

Data from the Norwegian marine monitoring programme have been used to conduct basic environmental impact assessments for radionuclides present in Norwegian coastal environments (Strand et al., 2000). Dose rates to biota are derived using an equilibrium absorbed dose constant, modified in the case of g-radiation for the absorbed fraction of energy. Doses have been calculated for a suite of radionuclides including  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ ,  $^{238}\text{Pu}$ ,  $^{239+240}\text{Pu}$ ,  $^{241}\text{Am}$  and the natural radionuclides  $^{210}\text{Po}$  and  $^{40}\text{K}$ . For the suite of radionuclides considered, total doses varied between  $1.35 \mu\text{Gy a}^{-1}$  and  $2.5 \mu\text{Gy a}^{-1}$ . The doses were essentially attributable to the natural radionuclides  $^{40}\text{K}$  and  $^{210}\text{Po}$ . For mussels and seaweed, the internal component of the dose was predominant, forming 73 % and 62 % of the total dose respectively, whereas for lobster, the external component and internal component were of equal magnitude. In terms of the anthropogenic radionuclides,  $^{99}\text{Tc}$  was the most important dose-forming radionuclide for lobsters and seaweed. Doses arising from  $^{99}\text{Tc}$  were calculated as  $20 \mu\text{Gy a}^{-1}$  for lobster and  $23 \mu\text{Gy a}^{-1}$  for seaweed almost entirely attributable to the internal component of dose. The doses calculated using this methodology are far below the  $1 \mu\text{Gy h}^{-1}$  chronic dose rate below which populations of animals and plant are unlikely to be harmed (IAEA, 1992).

### 7.4 Model validation

Experimental data for discharges of  $^{99}\text{Tc}$  from Sellafield (Brown et al., 1999; Leonard et al., 1998) were used for validation of the NRPA box model for evaluation of consequences from releases of radionuclides into marine environment, which is described by Iosjpe et al. (1997 and 2001).

Figure 7.1 shows the correlation between experimental and predicted data, where solid lines correspond to total agreement (1:1 relationship) between experimental data and calculations. Correlation analysis for experimental and predicted data shows that statistical estimation for correlation coefficient  $r$  is 0.82 with the confidence interval [0.62, 1.03] for the confidence coefficient 0.95 (significance level 0.05). A regression analysis shows that regression  $Y=K \cdot X$  is best at least in class of linear functions (here,  $Y$  and  $X$  correspon-

ding to experimental and predicted data,  $K$  is a regression coefficient). Statistical estimation for  $K$  is 0.98 and the confidence interval [0.88, 1.08] which corresponding to the confidence coefficient 0.95 for parameter  $K$ .

This analysis shows that the comparison between calculations and experiment has a satisfactory agreement and is consistent with a hypothesis concerning the relationship  $Y=X$  for predicted and experimental data.

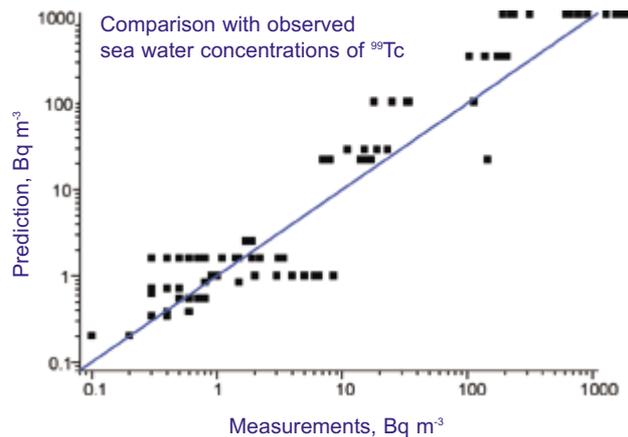


Fig. 7.1. Correlations of calculation and experimental.

## 8. Summary and conclusions

In Norway there are currently two monitoring programmes concerned with radioactivity in the marine environment, both co-ordinated by the Norwegian Radiation Protection Authority (NRPA) and funded by the Ministry of Environment and the Ministry of Fisheries, respectively. Results of both these programmes are presented in this report. In addition, data co-ordinated by the Food Control Authorities from a monitoring programme concerned with radioactivity in marine fish is 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 both included in the marine monitoring programme. Liquid discharge data for 1999 from nuclear installations and recent trends in such discharges are summarised, together with the

available information concerning nuclear weapons fallout and outflow of  $^{137}\text{Cs}$  of Chernobyl origin from the Baltic Sea. In addition, data from 1996 regarding the discharges of technologically enhanced naturally occurring radionuclides (TENORM) in production water from the North Sea oil and gas industry are included.

In 1999 samples of water, sediment, fish and/or other biota were collected for the monitoring programme in the Barents Sea, the North Sea, 12 coastal stations including Svalbard and 11 fjords. In addition, control samples were collected in Nitelva near the outlet from the nuclear facilities of the Institute for Energy Technology (IFE) at Kjeller.

## 8.1 Sources

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1. The liquid discharges from the nuclear facilities of IFE at Kjeller and in Halden have been estimated to be within authorised limits. The effective doses (as estimated by IFE) to the critical groups from discharges at Kjeller were  $0.53 \mu\text{Sv}$  in 1999, which corresponds to 53 % of the dose constraint of  $1 \mu\text{Sv year}^{-1}$  limit. About 75% of the dose was due to discharges of  $^{131}\text{I}$ . The effective doses to the critical group from IFE Halden liquid discharges has been estimated by IFE to  $0.08 \mu\text{Sv}$ .

2. Regarding the use of unsealed radioactive substances at hospitals, research and industrial applications, and the most significant radionuclides in relation to discharges are  $^{131}\text{I}$ . This radionuclide was detected in sewage system wastewater in a survey in 1995-1996, and the annual discharge was estimated at 780 GBq.

3. Production water from offshore oil production contains elevated levels of naturally occurring radionuclides. Estimates based on a survey from 1996 indicate a total discharge in the North Sea of about  $0.66 \text{ TBq } ^{226}\text{Ra}$  and  $0.34 \text{ TBq } ^{224}\text{Ra}$ . It has been estimated that Norwegian installations contribute approximately 6 % of the total release and it is likely that these releases were higher in 1999.

4. The discharges of radionuclides from Norwegian sources are only detectable in the environment close to the discharge points and have no significant impact on the large-scale distribution of anthropogenic radionuclides in the marine environment.

5. 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 of anthropogenic radionuclides found in Norwegian waters. In addition to direct discharges from reprocessing, remobilisation of  $^{137}\text{Cs}$  and plutonium from contaminated Irish Sea sediments acts as a secondary source of radionuclides to the marine environment.

## 8.2 Environmental contamination in Nitelva sediments

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1. Discharges of radionuclides from IFE Kjeller in the 1960s and early 1970s have caused radioactive contamination of Nitelva sediments. IFE, as the responsible operator, is responsible for carrying out monitoring of the area. In 1999, NRPA collected parallel samples of surface sediments. Concentrations of plutonium in surface sediment samples were in the range  $54 \text{ Bq kg}^{-1}$  (dry weight) to  $4560 \text{ Bq kg}^{-1}$  (dry weight), and concentrations of  $^{137}\text{Cs}$  were in the range of  $52.6 \text{ Bq kg}^{-1}$  to  $401 \text{ Bq kg}^{-1}$ . The average concentrations were similar to the results reported by IFE.

2. Earlier investigations by IFE showed that concentrations were considerably higher in deeper sediments. Based on this, the NRPA ordered a clean-up operation, and sediments with concentrations of transuranics (plutonium and americium) higher than  $10 \text{ Bq g}^{-1}$  were removed early spring 2000.

## 8.3 Radioactivity in sea water and sediments

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1. Concentrations of  $^{137}\text{Cs}$  in the eastern Barents Sea surface water were in the range of  $2.8 \text{ Bq m}^{-3}$  to  $4.0 + 0.2 \text{ Bq m}^{-3}$  with a mean value of  $3.4 \text{ Bq m}^{-3}$ . This is similar to levels obtained from the "Gauss" cruise in 1995. In ten Norwegian fjords, the  $^{137}\text{Cs}$  concentrations of sea water varied between  $4.5 \text{ Bq m}^{-3}$  and  $8.4 \text{ Bq m}^{-3}$ , with an average concentration of  $6 \text{ Bq m}^{-3}$ .

2. Concentrations of  $^{137}\text{Cs}$  in surface sediment samples collected in the eastern Barents Sea, western North Sea and in Kattegat varied between

1.9 Bq kg<sup>-1</sup> and 17.4 Bq kg<sup>-1</sup>. In the fjords, a larger variation was seen. In Nordfjord, Beiarfjorden and Namsfjorden, concentrations exceeded 100 Bq kg<sup>-1</sup>. This is probably due to the generally high levels of Chernobyl fallout in Trøndelag and central parts of Norway.

3. The concentration of <sup>99</sup>Tc in Barents Sea surface water varied between 0.10 Bq m<sup>-3</sup> and 1.14 Bq m<sup>-3</sup>. In the North Sea, concentrations ranged from 0.46 Bq m<sup>-3</sup> to 7.2 Bq m<sup>-3</sup>. At the regular sampling station at Hillesøy, average concentrations of <sup>99</sup>Tc increased by a factor of four from July 1997 to December 1999. Annual averages increased from 0.61 Bq m<sup>-3</sup> in 1997 to 0.91 Bq m<sup>-3</sup> and 1.5 Bq m<sup>-3</sup> in 1998 and 1999 respectively. The elevated levels are due to recent Sellafield discharges of this radionuclide.

4. Concentrations of plutonium in the eastern Barents Sea and three stations in the Norwegian Sea were in the range of 6.6 mBq m<sup>-3</sup> and 9.9 mBq m<sup>-3</sup>. The <sup>238</sup>Pu/<sup>239,240</sup>Pu activity ratios indicated that global nuclear weapons fallout is the main contributor, but in a few samples there were clear indications of an additional source, however, there are not enough data to confirm this.

#### 8.4 Radioactivity in fish and other biota

1. Concentrations of <sup>137</sup>Cs in various species of fish caught in the Barents Sea, the Norwegian Sea and nine fjords were low. The maximum concentration observed was 0.8 Bq kg<sup>-1</sup> (wet weight). Concentrations of <sup>90</sup>Sr in fish meat were in the range of 9.4 mBq kg<sup>-1</sup> to 23 mBq kg<sup>-1</sup> (wet weight).

2. Concentrations of <sup>137</sup>Cs in shrimps, crabs, lobsters and mussels were very low, the highest concentration observed was 0.4 Bq kg<sup>-1</sup> (wet weight). <sup>99</sup>Tc was detected in mussels, norway lobster and lobster. The highest concentrations of <sup>99</sup>Tc were observed in lobsters 14-26 Bq kg<sup>-1</sup> (wet weight). For shrimps, starfish and crabs, the concentrations were below the detection limit (< 0.25 Bq kg<sup>-1</sup>).

3. Seaweed is used as an indicator for <sup>99</sup>Tc and other radionuclides in the marine environment. Concentrations of <sup>99</sup>Tc in *Fucus vesiculosus* at

Hillesøy in Troms increased by a factor 5.5 from July 1997 to December 1999 due to elevated Sellafield discharges from mid-1994. In 1999, levels of <sup>99</sup>Tc in *Fucus vesiculosus* at ten regular sampling stations varied from 107 Bq kg<sup>-1</sup> (dry weight) at Vestvågøy up to an average of 401 Bq kg<sup>-1</sup> (dry weight) at Utsira.

4. Levels of <sup>137</sup>Cs in the seaweed are higher in the Oslofjord and at the Skagerrak coast than in northern parts of Norway. This is attributable to higher concentrations of <sup>137</sup>Cs in sea water due to Baltic outflow of Chernobyl contaminated water masses in combination with enhanced uptake by seaweed due to low salinity waters.

5. Concentrations of <sup>137</sup>Cs in Harbour Porpoises ranged from 0.3- 3.8 Bq kg<sup>-1</sup> (wet weight). Porpoises caught near the coast of Vest-Agder contained the highest levels.

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## Appendix 1

### Detection and measurement of radionuclides in environmental samples

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The technique used to measure the activity concentration of a particular radionuclide in an environmental material is dependent on the nature of radiation that is emitted by that radionuclide.

Radionuclides have unstable nuclei and spontaneously undergo radioactive decay in order to produce more stable nuclides. The product nuclide, also referred to as the daughter nuclide, of the radioactive decay process may be stable or may be radioactive and also undergo radioactive decay.

Several types of radioactive decay are known to occur, but only three types are described here:

- Alpha decay, which involves the emission of a helium-4 nucleus;
- Beta decay, which involves the emission of an electron (negatron) or a positively charged electron (positron);
- Gamma decay, which involves the emission of very short wavelength electromagnetic radiation. Many alpha and beta decays are accompanied by gamma decay.

Figure 1 illustrates the different ways in which these types of emitted radiation interact with matter. Alpha-radiation is the least penetrating and is easily absorbed by a thin sheet of paper or a few centimetres of air whereas gamma-radiation has much greater penetrating power. These properties are taken into account when selecting a suitable detection method for the radionuclides of interest in the environment.

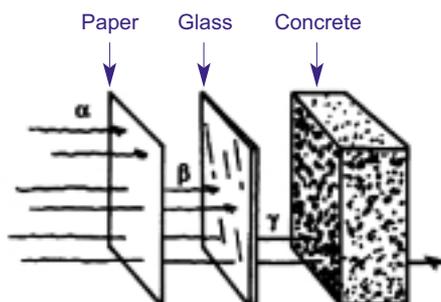


Fig. 1 Penetration of alpha-, beta- and gamma radiations in various materials.

In addition to detecting the radioactive emissions, the radionuclide from which the emissions originated needs to be identified and quantified. Identification can be achieved using detector systems that can determine the energy of the emitted radiation. The measured energy can then be compared to the known emission energies of radionuclides of interest. This approach may not always be possible and a chemical separation process may be required to prevent radioactive emissions from other radionuclides interfering with the measurement of the radionuclide of interest.

Table 1 lists some of the most important anthropogenic radionuclides present in the marine environment along with the type of radiation and their half-life.

### Gamma emitting radionuclides

<sup>137</sup>Cs, <sup>134</sup>Cs and <sup>60</sup>Co

The detection and measurement of gamma-emitting radionuclides in environmental materials is usually conducted by gamma spectrometry using either a thallium activated sodium iodide detectors (NaI(Tl)), which is a solid scintillator detector, or the so-called high purity germanium detector, which is a semi-conductor detector. Sediment and biota samples are dried or ashed and homogenised before they are transferred to a suitable beaker for measurement. Sea water samples normally contain very low concentrations of <sup>137</sup>Cs, and therefore the radionuclide is concentrated by pumping up to several hundred litres through a filter system with caesium-binding sorbents. The filters are ashed before counting.

The semiconductor detector system detects gamma radiation as a function of energy, and the radionuclides are identified by the energy of the peaks in the gamma spectrum. The intensity of the peaks is proportional to the concentration of the radionuclide in the sample. The counting time needed depends on the amount of material and the radioactivity level in the sample. In order to obtain sufficiently well-defined peaks with a germanium detector system, typical counting times for marine samples are 1-4 days in a shielded low-background counting room. The samples that are measured with sodium iodide detectors, have substantially shorter counting times, but the detection limits are higher for this measurement technique.

Table 1. Radionuclides with half-lives and radiation characteristics (emission probability in brackets).

Isotope	Half-life	a-energy keV	b-energy (max) keV	g-energy keV
Americium-241	432.7 years	5485 (85%)		59,5(35,9%)
Plutonium-238	87.7 years	5499,3 (71%)		
Plutonium-239	24,113 years	5156,2 (73,3%)		
Plutonium-240	6,563 years	5168,3 (72,9%)		
Plutonium-241	14.4 years		20,81 (99,99%)	
Technetium-99	2,113•10 <sup>5</sup> years		293,6 (99,99%)	
Strontium-90	28.64 years		546,2 (100%)	
Cesium-134	2.065 years			604,71(97,6%)*)
Cesium-137	30.17 years			661,66 (90%)
Potassium-40	1,28•10 <sup>9</sup> years			1460,8 (10,7%)
Iodine-131	8.04 days			364,48 (81,2%)

\*) Cesium-134 has several other strong  $\gamma$ -peaks

### Alpha emitters: <sup>238</sup>Pu, <sup>239,240</sup>Pu and <sup>241</sup>Am

Determination of alpha-emitters is more time consuming and labour intensive than gamma spectrometric measurements. The analytical procedure starts with 200 litres of sea water or 10-20 grams of dried sediment or biotic material. Prior to counting many days of laboratory work is needed to separate and purify the element in question from other elements present in the sample. In order to monitor the recovery of the radionuclide, a controlled quantity of yield tracers, usually <sup>242</sup>Pu and <sup>243</sup>Am, are added to the sample before the radiochemical separation starts. In the final step, plutonium or americium is electrodeposited on stainless steel discs. Impurities must be avoided in order to get a reliable measurement result, since any material can easily absorb alpha radiation. Since alpha radiation can also be stopped by a few centimetres of air the samples must be counted in a vacuum chamber. Counting times of one or two weeks are commonly required for alpha-emitters in environmental samples.

### Beta emitters: <sup>90</sup>Sr, <sup>99</sup>Tc and <sup>241</sup>Pu

<sup>99</sup>Tc and <sup>90</sup>Sr, are pure beta emitters. Like the alpha emitters, the laboratory analysis includes a chemical separation procedure prior to counting. The gamma emitting radionuclide <sup>99m</sup>Tc is used as a yield monitor for <sup>99</sup>Tc analysis.

The procedure for <sup>99</sup>Tc starts with 50-100 litres of sea water (Photo 1) or 10-20 grams of dried sediment or biotic material. In the final step, technetium is electrodeposited on stainless steel discs and counted with a beta counter.

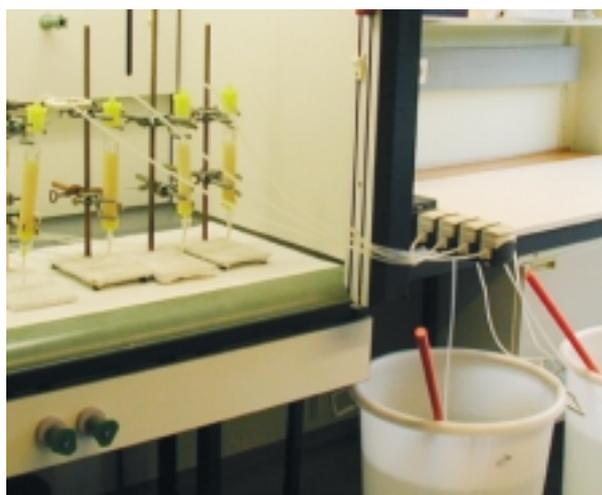


Photo 1: <sup>99</sup>Tc analysis. At first 50 litres of sea-water is passing true an ion exchange column.



Photo 2: Dried material of sample in different containers ready for gamma-measurements. To the right, <sup>99</sup>Tc preparations ready for counting.

The beta emitter <sup>241</sup>Pu can be analysed by dissolving the plutonium from the alpha analysis discs, and then mixing it with a fluid that emits light when exposed to the beta radiation from <sup>241</sup>Pu. The light is then measured in a liquid scintillation counter.

## Analytical methods

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During the last few years, laboratories at the Norwegian Radiation Protection Authority, the Local office of the Directorate of Fisheries in Tromsø, the Norwegian Food Control Authority in Salten and the Institute of Marine Research have all been accredited according to the requirements of NS-EN 45001 and ISO/IEC Guide 25. The scope and conditions of the accreditation for the institutes mentioned above are quite different, but they have all been accredited for gamma spectrometric measurements. The analytical techniques employed at each institution are described below. The results in this report are not given up as accredited results because not all results that are accredited.

### The Norwegian Radiation Protection Authority

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NRPA have been analysing alpha, beta and gamma emitters, according to procedures described in this appendix.

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

The concentrations of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$  were measured in samples of 200 litres of sea water.  $^{242}\text{Pu}$  was added for chemical yield determination. The precipitation from the water samples was made according to the analytical procedure described by Chen et al.(1991). Different radiochemical separation techniques were applied to separate plutonium from other nuclides using solvent extraction with 10% TIOA/Xylene solution and ion exchange chromatography with BIO-RAD AG1-X4 (100-200 mesh). Purified samples were electrodeposited on stainless steel discs and counted in semiconductor silicon detectors. As alpha spectrometry is not able to distinguish between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  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. Chemical yields obtained from the  $^{242}\text{Pu}$  spike were about 40% - 75 %.

The starfish samples was treated differently, four pieces of starfish were dried and homogenised and

then pooled together for gamma emitting analysis,  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$ .

25g dried starfish sample was analysed for alpha emitters (Chen, 1994 pers.comm.). Chemical yield obtained for the  $^{242}\text{Pu}$  spike was 28 %.

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

To determine the concentrations of  $^{99}\text{Tc}$  in sea water, samples of 50 litres were filtered through a 1 $\mu\text{m}$  polypropylene cartridge to remove suspended matter. Seaweed and other biota samples were dried, milled and homogenised. A 10 - 20 g dried sample was transferred to a specially designed bottle and carbonised and 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.

The analytical procedure used is a modified version of that developed by Risø National Laboratory, Denmark. The procedure is outlined briefly below. Technetium was separated from the matrix by ion exchange chromatography using BIO-RAD AG1-X4 (100-200 mesh) then other separation techniques such as precipitation and solvent extraction were used before technetium was electro-deposited onto stainless steel discs. The chemical yields were determined by gamma counting from the  $^{99\text{m}}\text{Tc}$  tracer on a NaI-well detector. Typically yields varied between 70 % and 85 %. After one week,  $^{99}\text{Tc}$  activity was measured using a low background anti-coincidence beta counter.

Total uncertainties for  $^{99}\text{Tc}$  analysis is around 10%. The limit of detection may vary slightly owing to variations in chemical yield, counting efficiency and mass of sample.

#### **Detection of gamma emitters**

At NRPA analyses of gamma emitting nuclides are performed on low level, high-purity, germanium (HPGe) detectors. All the detectors are situated in a low-background laboratory which gives 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 covered the energy interval of 50-2000 keV, and two covered the interval 20-2000 keV.

The three filters from the filtering system for sea water samples were separately dried at 105°C and ashed at 450°C before measuring took place. Samples were then counted from 1 to 4 days.

Samples of fish and seaweed were dried at 105°C and homogenised, and placed in containers prior to gamma counting. Fish samples were each counted for a minimum of 3 days.

In the fish and shrimp samples the detection limit of <sup>137</sup>Cs at the NRPA ranges from 0.2 to 0.4 Bq kg<sup>-1</sup>.

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 <sup>137</sup>Cs and <sup>134</sup>Cs in the sediment samples ranged from approximately 0.3 - 2 Bq kg<sup>-1</sup> (dry weight).

## **The Institute of Marine Research, IMR**

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### ***Detection of gamma emitters***

Biota samples were carried deep-frozen to IMR, where the samples were subsequently ground, freeze-dried, homogenised and filled into polyethylene counting boxes of appropriate size prior to analysis. The gamma counting for <sup>137</sup>Cs was performed in IMR's low-background laboratory using an HPGe detector with 30 % efficiency and an extra low-background HPGe detector with 60 % efficiency. Both detectors have electric cryostat cooling systems, and 10 cm lead shielding.

## **Institute for Energy Technology, IFE**

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### ***Detection of beta emitters***

Samples of dried and homogenised *Fucus vesiculosus* were analysed for <sup>99</sup>Tc. The pretreatment was performed at Kjeller and the analysis at Lund University. The following analytical procedure was used at Lund: Technetium was extracted by TBP from sulphuric acid-hydrogen fluoride media. Back extraction is done from sodium hydroxide solution from which media technetium is electro-deposited

onto stainless steel discs and <sup>99</sup>Tc is used as a radiochemical yield determinant. After decay of the yield determinant technetium is measured by an anti coincidence shielded GM counter (Holm et al., 1984).

### ***Detection of gamma emitters***

At IFE analyses of gamma emitting nuclides are performed on low level, high-purity, germanium (HPGe) detectors. Samples of seaweed were dried at 105°C and homogenised, and placed in containers prior to gamma counting.

## **The Norwegian Food Control Authority (SNT) in Salten**

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### ***Detection of gamma emitters***

The Food Control Authority in Salten uses both NaI and HPGe detectors for gamma measurements. The NaI detector is a Canberra series 10 portable detector. The NaI detectors have higher efficiency than the HPGe detectors, but the resolution is poor and it is hard to separate the different gamma emitters. The detector is measuring the total amount of radio caesium, both <sup>134+137</sup>Cs. The detection limit is 20Bq kg<sup>-1</sup>.

The Laboratory is using a HPGe detector when measuring samples of fish and shrimp with low activity. It's a EG&G Ortec GEM (p-type) detector with 45% relative efficiency. 500g dried muscles meat (pooled sample of 25 fishes) was put in a Marinelli beaker and counted for 2 to 3 days. The detection limit is estimated to be 0.1 Bq kg<sup>-1</sup> (wet weight).

## **The laboratory of the Local office of the Directorate of Fisheries in Tromsø**

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### ***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 detectors. The detection limit for fish, as reported by the Directorate of Fisheries in Tromsø, is approximately 11 Bq kg<sup>-1</sup>. Samples of 200 g fresh fish containing both meat and bone were analysed.

## **Harwell Scientifics Ltd**

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### ***Detection of alpha emitters***

Radiochemical analysis of <sup>241</sup>Am, <sup>238</sup>Pu, <sup>239+240</sup>Pu and <sup>241</sup>Pu of sediment samples collected at IFE were carried out by Harwell Scientifics Ltd on a contract with NRPA. Samples were dried and ground prior to analysis. An aliquot of the sample was removed and spiked with a known activity of <sup>242</sup>Pu and <sup>243</sup>Am tracers. The sample was then digested with aqua regia, filtered and the filtrate evaporated to dryness. The residue was dissolved in 2M HNO<sub>3</sub> and the Fe reduced to Fe(II) with ascorbic acid. Pu and Am were then isolated and separated from each other using a TRU resin column. Pu was subsequently purified using anion exchange chromatography before being electrodeposited and counted by alpha spectrometry. Any americium was separated from rare earth elements using anion exchange chromatography. The purified americium was electro-deposited and counted using alpha spectrometry. Activities of plutonium alpha isotopes and <sup>241</sup>Am were determined relative to the tracer spikes. A method blank and reference material were also analysed as part of the data quality control.

<sup>241</sup>Pu fraction was leached from the electro-deposited disc with plutonium and extracted into trioctyl phosphine oxide in toluene. This organic extract was mixed directly with a liquid scintillation cocktail and counted on a Wallac 1220 Quantulus low-level liquid scintillation counter. The <sup>241</sup>Pu activity was determined relative to the Plutonium (alpha) activity that was simultaneously measured on the liquid scintillation counter.

