

# Radioactive contamination in the marine environment

Report no 3 from national surveillance programme

*Anne Lene Brungot, Lars Føyn, JoLynn Carroll, Anne Kathrine Kolstad, Justin Brown, Anne Liv Rudjord, Bjarne Bøe, Turid Hellstrøm*

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## SUMMARY

During 1996-1998 a number of samples of marine biota as well as sediment and seawater were collected from various fishing grounds of interest for the Norwegian commercial fishing industry. The main emphasis of the sampling programme was in areas of the North Sea, the Barents Sea, the Norwegian Sea and Skagerrak. A total of 3540 fish were analysed either individually or as pooled samples of no less than 25 fish for a total of 726 analyses. Radioactivity measurements of gamma-emitting radionuclides were performed on these samples. A variety of seafood products in addition to fish, namely lobster, mussels, and shrimps were also analysed for gamma-emitting radionuclides and for  $^{99}\text{Tc}$ . In addition, seaweed, sediment and seawater radionuclide levels were documented for these same areas.

In general, levels of radioactive contamination in the ocean and seas surrounding Norway remain low. As in previous years of this surveillance programme, the level of  $^{137}\text{Cs}$  in fish and shrimps is in the order of 1 Bq/kg. This may be compared with the action limit for radiocaesium in most food products which is 600 Bq/kg in Norway. Other radionuclides,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{60}\text{Co}$  and  $^{241}\text{Am}$ , are found in extremely low concentrations and in many cases, are below detection limits. Measurements further indicate that low levels of these radionuclides currently exist in sediments and seawater from the North Sea, the Norwegian Sea and the Barents Sea. Measurements of biota indicate that radioactivity levels decrease towards the north of Norway. This is because relative to the north of Norway, the south of Norway is in closer proximity to two major sources of artificial radionuclides. Water from the Baltic Sea contains radioactivity from Chernobyl and the Irish Sea contains discharges from the main nuclear waste reprocessing facility, Sellafield in UK.

However, an increasing of the level of  $^{99}\text{Tc}$  in samples of seaweed, lobsters and seawater is observed. This is due to increased discharges from the new waste treatment plant EARP at Sellafield in UK after 1994.

## 1. INTRODUCTION

In 1994 a national monitoring programme was initiated by the Norwegian authorities in response to information about radioactive sources and potential contamination of northern marine areas. The purpose of the programme is to continuously monitor the trends of radioactive contamination in the marine environment and to document levels in fish and other commercially important marine species. Results of the monitoring programme for the years 1994 and 1995/96 have been published earlier Sickel et al. (1995) and Brungot et al. (1997). These summary reports can be obtained from the NRPA.

This report presents surveillance results for the period December 1996 to early in 1998. Samples of fish, seaweed, and other biota as well as sea water and sediment were collected for analysis of gamma-emitting nuclides and radionuclides of plutonium, americium, and technetium.

The analytical results and sampling for this report were provided by the following institutions:

- Norwegian Radiation Protection Authority
- The Institute of Marine Research
- The Directorate of Fisheries
- The Local office of the Directorate of Fisheries in Troms
- Norwegian Food Control Authority

The program is coo-funded by the Ministry of Fisheries.

## 2. SOURCES OF RADIOACTIVE CONTAMINATION

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 radionuclides discharged from reprocessing plants.
- Fallout from the Chernobyl accident in 1986.

In addition, there are several sources of interest which in the future may release radioactive contaminants into the marine environment. Major potential sources that are currently known to exist in the Barents Sea and in the North Atlantic Ocean are:

- Land-based nuclear installations and accidental releases of radioactive contamination subsequently transported into northern seas.
- Dumped solid radioactive waste and spent nuclear fuel in the Kara Sea.
- Civilian and military nuclear installations on the Kola Peninsula, including submarines and icebreakers with nuclear power reactors.

### 2.1 Fallout from nuclear weapons testing

Global fallout from atmospheric nuclear weapons testing is one of the main sources of radioactive contamination in the Barents region. Since 1945, the declared nuclear powers (United States, Russia, Britain, France, and China) acknowledged conducting a total of 2.036 nuclear tests (BAS, United Kingdom, 1995).

The island of Novaya Zemlya was the site of 132 of Russia's 718 nuclear tests (BAS, 1995). In the early sixties underwater testing was carried out in the Chernaya Fjord on the south west coast of Novaya Zemlya. The detonations resulted in high levels of radioactive contamination that persist today in the sediments of the fjord (Smith et al., 1995). In addition, underground detonations were carried out in the period 1963-89 at Novaya Zemlya. While fission products have been identified in air after underground nuclear detonations, (Bjurman et al., 1990), such detonations are assumed to have no significant impact on the level of radioactive contamination in the marine environment.

### 2.2 Discharges from European reprocessing plants

Another main source of anthropogenic radionuclides to the North Atlantic and the North Sea has been major nuclear reprocessing facilities in Europe. These facilities include Sellafield in the United Kingdom, Dounreay in Scotland, and Cap de La Hague in France. The largest discharges of nuclear waste have occurred from the reprocessing facilities at Sellafield, UK (*Table 1*) In the late sixties until the mid-eighties, releases of radiocaesium from Sellafield were a factor of 100 higher than the releases from Dounreay and Cap de La Hague. Due to stronger regulatory controls and plant improvements which have been implemented since the mid-seventies, releases of several of the main radionuclides, including radiocaesium, have declined markedly over the ensuing years. In 1994 the Enhanced Actinide Removal Plant (EARP) and the Thermal Oxide Reprocessing Plant (THORP) began operations. While the

operation of these plants resulted in reduced discharges of some radionuclides such as plutonium and americium, plant operations increased the discharges of others, especially  $^{99}\text{Tc}$  but also  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{90}\text{Sr}$ . As a result, in 1997  $^{99}\text{Tc}$  levels in the marine waters surrounding Norway were higher than in previous years (Brown et al.1998).

The Dounreay facilities on the northern coast of Scotland, were established in 1955, and have mainly been used for development of technology of fast breeder reactors. Beginning in the 1990's, there were efforts to attract commercial contracts to Dounreay primarily for the reprocessing of spent nuclear fuel and other nuclear wastes. However in June 1998, Dounreay was ordered by the Government to take on no further commercial reprocessing contracts. Reprocessing at Dounreay will end once the plant completes the reprocessing of current inventories and satisfies all of its outstanding contracts (MAFF&SEPA, 1998).

The Cap La Hague reprocessing facility in France is also a source of radioactive discharges to the North Sea. Discharges from this facility have also been reduced over the last ten years (Betis, et al., 1994).

*Table 1 Discharges of liquid radioactive waste from Sellafield, La Hague and Dounreay during 1994 and 1997. (MAFF-ISSN 0142-2499)(MAFF&SEPA, 1998)*

Radionuclide		Discharge limit,		Discharges	
		TBq		TBq	
		1994	1997	1994	1997
<b>Sellafield</b>	$^{137}\text{Cs}$	75	75	6.75	7.94
	Pu-alpha	7	0.7	1.08	0.147
	$^{241}\text{Am}$		3 0.3	0.74	0.0505
	$^{90}\text{Sr}$	30	48	28.9	37.3
	$^{99}\text{Tc}$		200	80	84.2
<b>Dounreay</b>	$^{137}\text{Cs}$	50	50	4.70	0.326
	Total-alpha	0.75	0.75	0.03	0.0255
	$^{241}\text{Pu}$	15	15	0.945	0.252
	$^{90}\text{Sr}$	12	12	1.60	0.223
<b>La Hague</b>	$^{137}\text{Cs}$			10	2.46
	Pu-alpha			0.11	0.0153
	$^{90}\text{Sr}$			10.5	3.73
	$^{99}\text{Tc}$			7	0.13
	$^{125}\text{Sb}$			30	1.34

\* In November 1997, a significant increase of  $^{106}\text{Ru}$  discharges occurred at one of the Sellafield plants resulting in an incident of regulatory non-compliance at the plant (MAFF&SEPA, 1998).

## 2.3 Chernobyl accident

The most serious accident of nuclear reactor operation occurred at the Chernobyl plant in Ukraine in April 1986. Radionuclides were released to the atmosphere contaminating both the

local environment and areas across the Scandinavian countries of Finland, Sweden and Norway. Fallout levels of radiocaesium of up to 100-200 kBq/m<sup>2</sup> were found in central Norway (Backe et al, 1986). It is estimated that during the Chernobyl accident approximately 140 PBq of radiocaesium (<sup>134</sup>Cs and <sup>137</sup>Cs) and 10 PBq of <sup>90</sup>Sr and 0.1 PBq of plutonium isotopes were released. Direct fallout from the atmosphere and run-off from contaminated land areas, especially in the Baltic Sea, have influenced the marine areas surrounding Norway.

## 2.4 Other Sources

### 2.4.1 River transport

Several nuclear facilities are located in the drainage basins of the large Russian rivers, Ob and Yenisey. Discharges or accidental releases from these facilities combined with surface run-off of radioactive fallout from atmospheric nuclear weapons testing may enter the river drainage basins and be transported downstream to the Kara Sea. It is difficult to estimate the impact on the Barents Sea from these sources both because the transport routes and the magnitude of transport are not sufficiently documented and because the data of radioactivity in the open literature contain inconsistencies. Observations conducted by Roshydromet (Vakulovsky et al., 1993) indicate that about 1PBq <sup>90</sup>Sr and 0.1 PBq <sup>137</sup>Cs was transported by the Ob and Yenisey rivers during 1961-1989. In addition, approximately 200 TBq of <sup>137</sup>Cs have been transported to the Barents Sea by the rivers Pechora, Onega and Severnaya Dvina (Vakulovsky et al., 1993).

### 2.4.2 Dumping around Novaya Zemlya

From 1961 to 1991 nuclear waste was dumped in the Barents Sea and the Kara Sea, the dumped waste ranged from low level liquid waste to highly active spent nuclear fuel. Studies have been carried out to assess the actual and potential risks from the dumped material and to measure the extent of actual contamination of the marine environment. The joint Russian-Norwegian expert group has been investigating current contamination levels and the potential for future releases at the dumpsites (JRNEG 1996). The expert group has concluded that enhanced levels of anthropogenic radionuclides in sediments collected in close proximity to dumped objects indicates leakages from these objects. However no contribution from dumped radioactive waste can be observed in the open sea. Preliminary radiological dose assessments indicate that regional and global populations will receive a very small dose from this source of radioactive contamination in the environment even if the dumped activity should be released instantaneously (JRNEG, 1996).

### 2.4.3 Nuclear Submarine - Komsomolets

In 1989 the Soviet nuclear submarine Komsomolets caught fire and sunk south east of Bear Island. The submarine contained a nuclear reactor and two nuclear warheads. The reactor radionuclide inventory includes 2.7 PBq <sup>90</sup>Sr and 3.0 PBq <sup>137</sup>Cs, while the warheads contain approximately 16 TBq of <sup>239</sup>Pu (CCMS 1995). Underwater monitoring has shown elevated levels of <sup>137</sup>Cs and also sign after <sup>134</sup>Cs near the reactor section, indicating some leakage of radionuclides has occurred (Kolstad, 1995). However, conservative modelling of the possible releases of <sup>137</sup>Cs indicates that the radionuclide concentration in sea water and fish caused by past, present and future releases from Komsomolets are at least a factor 100 lower than current concentrations in these media (CCMS 1995).

#### *2.4.4 Russian Civilian and Military Nuclear Fleet*

On the Kola Peninsula, the Russian Northern Fleet and the civilian nuclear ice breaker fleet have their main bases. Approximately 100 nuclear vessels operate from these bases, containing together about 200 nuclear reactors. Besides the operating vessels, about 80 decommissioned nuclear submarines awaiting dismantling are stationed at the military bases. Most of these submarines still contain their nuclear reactors and fuel. Plans exist to decommission 125 submarines during the next several years. However, many challenges exist in performing the dismantling work minimising the risks of accidents and doses to the workers. Disposing of the nuclear materials poses a further challenge as storage facilities for spent nuclear fuel in the Kola Peninsula are already filled to capacity (NEFCO, 1996).

### **3. COLLECTION AND TREATMENT OF SAMPLES**

#### **3.1 Fish and shrimps**

Fish and shrimp samples were collected at commercial fishing locations by the Norwegian Directorate of Fisheries. Individual fish were collected from five different areas around the Norwegian coast, but mainly from the Barents Sea. Four times each year, the Directorate of Fisheries collect fish to make pooled samples: pooled samples (50 g each) were made after combining the meat of one hundred fish. Additionally, in 1997 fish samples of haddock, mackerel, and saithe were collected by the Institute of Marine Research (IMR). The shrimp samples were collected by the Directorate of Fisheries mainly from the Barents Sea and areas surrounding Spitsbergen.

The Local office of the Directorate of Fisheries in Troms routinely analyse 25 samples of cod each month for  $^{137}\text{Cs}$ . Additionally the Food Control Authority (SNT) also collect fish and shrimp to make pooled samples each containing meat from 25 fish each. The samples were counted for  $\gamma$ -emitting nuclides by HPGc detector and the results are reported and included in the surveillance program.

The Norwegian Radiation Protection Authority (NRPA) analysed shrimps and the pooled samples of cod for  $\alpha$ ,  $\beta$ - and  $\gamma$ -emitting nuclides.

Fish collected by the IMR were also analysed for  $\gamma$ -emitters by the NRPA.

#### **3.2 Lobsters and mussels**

Lobsters were collected by local fishermen in different areas along the western and southern parts of the Norwegian coastline. Mussels were collected four times yearly from Tromsø and in the south part once in the year. The Norwegian Radiation Protection Authority have analysed the samples of lobsters and mussels for  $\beta$ - and  $\gamma$ -emitting nuclides.

#### **3.3 Seaweed and algae**

Seaweed samples were collected from two shoreline stations along the Norwegian coast. In northern Norway, seaweed is collected every month to document seasonal fluctuations in radioactivity levels. In southern Norway, seaweed is collected once each year. From mid-1998, algae samples were collected from various locations ranging from the northern Norwegian Sea to the southern areas of the North Sea. All samples of seaweed and algae were collected and stored for later laboratory analyses for  $\beta$ - and  $\gamma$ -emitting nuclides. Laboratory analyses were conducted at the NRPA.

### 3.4 Sea water

Sea water samples were collected from different locations in the North Atlantic Ocean. In addition, beginning in 1996, monthly water samples for  $\beta$ -analyses were collected from Hillesøy in Troms and yearly water samples for  $\alpha$ ,  $\beta$  and  $\gamma$ -analyses were collected from locations in the southern part of Norway.

Sea water for determination of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  was pumped directly to a filtering system consisting of three consecutive cartridge filters, the first one to remove suspended matter  $>1\mu\text{m}$  and the following two coated with a Cs-sorbent,  $\text{Cu}_2\text{Fe}(\text{CN})_6$ . A flowmeter mounted at the outlet of the third filter registered the amount of filtered water. Surface water was collected while the ship was moving from one station to the next.

Sea water for determination of Pu and Am analysis were collected from different depths in large precipitation tanks. The water was adjusted to pH 2 by adding conc. HCl, and then  $\text{FeSO}_4\cdot 7\text{H}_2\text{O}$ ,  $\text{Na}_2\text{S}_2\text{O}_5$  and  $^{242}\text{Pu}/^{243}\text{Am}$ -tracer for chemical recovery. After again adjusting the pH to 9.5 by NaOH the resulting precipitate became transferred to a 25 l polyethylene container for further radiochemical separation in the laboratory prior to alpha counting.

Laboratory analyses were conducted at the NRPA.

### 3.5 Sediment

Sediment samples were collected in Skagerrak and the North Sea areas during 1996 by the Institute of Marine Research. Sediments were retrieved from the seabed using a Smøgen box corer. The sediments samples are from the upper 2 cm layer. The samples were frozen and stored until further preparation and analyses for  $\gamma$ - and  $\alpha$ - emitters by the Norwegian Radiation Protection Authority.

## 4. ANALYTICAL PROCEDURES

### 4.1 Analysis of gamma emitting nuclides

#### 4.1.1 Analysis of radionuclides using HPGe detectors

Analyses of gamma emitting nuclides were performed on low level, high-purity, germanium (HPGe) detectors. 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. Sediment samples were freeze-dried, weighed, and placed in containers prior to gamma counting. Samples were counted for a minimum of 2 days. Cesium sorbents were dried at 105°C and ashed at 450°C prior to measurements. Samples have been counted from 1 to 4 days and the water volume who passed the filter system varied from approximately 500 litres up to 1300 litres.

The HPGe detectors have relative efficiencies in the range of 23% to 40%. The full width at half maximum (FWHM) at 1332 keV is less than 1.9 keV for all detectors.

In addition the Food Control Authority (SNT) also measured their fish and shrimp samples using a HPGe detector. The samples were counted for minimum 3 days. Generally the detection limit is different from one sample to an other depending on the measuring time, efficiencies of the detector, sample volume, the background spectrum and the influence of naturally occurring radionuclides in each sample. At NRPA the samples are measured in a low background room to make the detection limit low. In the fish and scrimp samples the detection limit of  $^{137}\text{Cs}$  at the NRPA ranges from 0.2 to 0.4 Bq/kg.

#### 4.1.2 Analysis of radiocaesium using NaI detectors

The laboratory of The Local office of the Directorate of Fisheries in Tromsø is equipped with a Canberra series 10 portable NaI detector. The efficiency of the NaI detector is approximately 5%, and the resolution is approximately 7% for  $^{137}\text{Cs}$ . The detection limit for fish, as reported by the Directorate of Fisheries in Tromsø, is 11 Bq/kg. Samples of fresh fish containing both meat and bone and weighing about 200 g each, were analysed.

### 4.2 Analysis of beta emitters

Technetium is separated from samples by ion exchange chromatography. In preparation for radiochemical separation procedures, 50-liter water samples are filtered through a 1µm polypropylene cartridge to remove suspended matter and then stored in 70-liter tanks. Fresh biota samples are dried, milled and homogenised. A 10-20 gram dried sample is transferred to a specially designed bottle for carbonising and dissolving.

$^{99\text{m}}\text{Tc}$  is added to the samples for the later determination of chemical recoveries. Uranium and Thorium were removed using solvent extraction with 5% TIOA/Xylene solution. After back-extraction from the organic phase, technetium is electro-deposited on stainless steel discs.

The chemical yields, determined by gamma counting of the  $^{99m}\text{Tc}$ -tracer on a NaI well detector, are usually above 70%. Sample activities of  $^{99}\text{Tc}$  were determined on a low background anti-coincidence beta counter. Detection limits are estimated to be in the range 0.2-0.5 Bq/kg for biota samples and 0.03-0.06 Bq/m<sup>3</sup> for samples of sea water.

### 4.3 Analyses of alpha emitters

Radiochemical separation techniques were applied to precipitate from each 200 l sea water sample. Following acid leaching of seaweed and sediments samples, these samples were treated identically to water samples as described below, only the fish samples were treated differently because of the material in the samples. The fish samples were not analysed for  $^{241}\text{Am}$ . After gamma analysis, samples of fish from the same area were pooled together for analyses of  $\alpha$ -emitters  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ . The total wet weight of the pooled samples was about 5 kg (Chen, 1994 pers.comm.).

After the addition of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  as a chemical yield tracer, plutonium and americium was separated by extraction with a 10% TIOA/Xylene solution. Next Pu was back-extracted from the organic phase into 8M HCl, and separated by ion exchange chromatography. Purified samples were electro-deposited on stainless steel discs and counted in semiconductor silicon detectors (Aarkrog et al, 1991). Chemical yields obtained from the  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  spike were about 40 - 65 %.

## 5. RESULTS AND DISCUSSION

A variety of samples have been collected in the marine environment including samples of fish, shrimps and other seafood products, seaweed, water and sediments. These samples have been analysed for several anthropogenic radionuclides:  $^{134,137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{241}\text{Am}$ ,  $^{210}\text{Po}$ ,  $^{99}\text{Tc}$ ,  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$ . The samples, collected over a period of two years beginning in November 1996 form the basis of the conclusions drawn in this updated assessment of the levels of radioactive contamination in areas of importance to the Norwegian fishing industry and the Norwegian authorities .

### 5.1 Fish

As part of the Norwegian Directorate of Fisheries screening program, levels of  $^{137}\text{Cs}$  were determined for approximately 700 fish samples using NaI detectors. For all of the samples analysed in the screening program,  $^{137}\text{Cs}$  levels were below the limit of detection (d.l.) for this equipment. The detection limit for NaI detectors (d.l.=11 Bq/kg w.w.) is much higher than for HPGe detectors (d.l.=0.05 Bq/kg w.w.).

Pooled samples of fish, analysed using HPGe detectors, indicate that the levels of  $^{137}\text{Cs}$  in fish are typically less than 1 Bq/kg (w.w). This finding is corroborated by the analyses performed on individual fish. Comparing data from all areas, maximum  $^{137}\text{Cs}$  levels were observed in samples of Whiting (*Merlangius merlangus*) and Cod (*Gadus morhua* L) from the Skagerrak region. Yet even in the Skagerrak region,  $^{137}\text{Cs}$  levels in fish were only 1.1-1.2 Bq/kg (w.w). For comparison, cod fish collected from the Baltic Sea, an area more contaminated with  $^{137}\text{Cs}$  from the Chernobyl accident, were reported at 21 Bq/kg (w.w) in 1993 (MAFF, 1993), and 12 Bq/kg (w.w) in 1997 (MAFF&SEPA, 1998). This is partly due to a higher uptake of  $^{137}\text{Cs}$  in fish in the low salinity waters of the Baltic Sea. In the Irish Sea, near Sellafield, a  $^{137}\text{Cs}$  activity concentration of 19 Bq/kg (w.w.) was reported by MAFF (1993), and in 1997 reported to be from 16 Bq/kg (w.w) and less (MAFF&SEPA, 1998).

The data reported in the National Surveillance Programme suggest that the concentrations of anthropogenic radionuclides in fish from important Norwegian fishing areas remain low. A similar conclusion was reached in previous years (Sickel et al. 1995; Brungot et al. 1997).

### 5.2 Other Seafood

A variety of other marine products important in the seafood industry have been analysed for  $^{137}\text{Cs}$  and  $^{99}\text{Tc}$ . Samples of shrimp (*Pandalus borealis*), lobster (*Homarus vulgaris*), mussel (*Mytilus edulis*), and crab (*Cancer pagurus*) were collected from coastal areas of the Norwegian Sea, the Outer Oslo Fjord and the North Sea.

Shrimps and mussels contained very low levels of  $^{137}\text{Cs}$ . The highest activity observed in these marine biota was 0.2 Bq/kg (w.w.). While the crustaceans contained higher activity concentrations of  $^{137}\text{Cs}$ , the levels were still equal to or below 1 Bq/kg (w.w.).

The highest levels of  $^{99}\text{Tc}$  in these marine biota samples were recorded in lobster. Recent studies in the Irish Sea have also shown higher uptake by these biota. Shrimps and mussels have concentrated  $^{99}\text{Tc}$  to a much lower degree. The content of  $^{99}\text{Tc}$  in these samples were less than 1 Bq/kg (w.w.).

Table 2:  $^{137}\text{Cs}$  activities (Bq/kg wet weight) for fish collected from marine waters in 1997 and 1998.

Species	Location	$^{137}\text{Cs}$ (Bq/kg)	No and type of sample	No of fish per sample
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	0.2-0.5	7 pooled	25 each
			17 pooled	100 each
Cod ( <i>Gadus morhua</i> L.)	Barents Sea	< 11	700	1
Saithe ( <i>Pollachius virens</i> )	Barents Sea	0.3-0.4	3 pooled	25 each
Cod ( <i>Gadus morhua</i> L.)	Norwegian Sea	0.2-0.7	21 pooled	25 each
Saithe ( <i>Pollachius virens</i> )	Norwegian Sea	0.4-0.6	6 pooled	25 each
Haddock ( <i>Melanogrammus aeglefinus</i> )	Norwegian Sea	0.3-0.6	3 pooled	25 each
Cusk ( <i>Brosme</i> )	Norwegian Sea	0.4-0.8	2 pooled	25 each
Atlantic Salmon ( <i>Salmo salar</i> )	Norwegian Sea	0.8	1 pooled	25 each
Saithe ( <i>Pollachius virens</i> )	North Sea	0.1-0.4	10	1
Haddock ( <i>Melanogrammus aeglefinus</i> )	North Sea	0.1-0.2	10	1
Mackerel ( <i>Scomber scombrus</i> )	North Sea	0.1-0.4	10	1
Cod ( <i>Gadus morhua</i> L.)	Skagerrak	1.0-1.1	3	1
Whiting ( <i>Merlangius merlangus</i> )	Skagerrak	1.2	1	1
Plaice ( <i>Pleuronectes platessa</i> )	Skagerrak	0.5	1	1

### 5.3 Seaweed

The seaweed (*Fucus vesiculosus*), has previously been used as an indicator species for radionuclides in sea water. A large data set exists containing data on radionuclide levels in algae for areas along northern European coastlines. Long-term monitoring of the Norwegian coastline shows that  $^{137}\text{Cs}$  levels in seaweed species have responded to changes associated with the main sources of radioactivity to the marine environment. In northern Norway,  $^{137}\text{Cs}$  levels in seaweed samples are lower than in the southern part of Norway. For samples collected in 1997 and 1998 from Hillesøy in Troms the levels are less than 1 Bq/kg (d.w.) of  $^{137}\text{Cs}$ . Compared to the southern part of Norway samples collected in Kristiansand and Outer Oslofjord show levels from 1.7 to 3.9 Bq/kg (d.w.) of  $^{137}\text{Cs}$ , in 1997 and 1998. For the  $^{60}\text{Co}$  in seaweed no significant changes are found compared to data from 1993 and 1995.

Table 3: Radionuclide levels (Bq/kg wet weight) in seafood products collected in 1997 and 1998.

Species	No of samples	Location	Year	$^{137}\text{Cs}$ (Bq/kg)	$^{99}\text{Tc}$ (Bq/kg)
Lobster ( <i>Homarus vulgaris</i> )	4	Bergen	December 1997	0.8-1.0	42-35
Lobster exoskeleton ( <i>Homarus vulgaris</i> )	2	Bergen	December 1997	n.d.	
Lobster ( <i>Homarus vulgaris</i> )	2	Outer Oslofjord	November 1997		11.1 -14.6
Lobster ( <i>Homarus vulgaris</i> )	2	Outer Oslofjord	1998	0.13-0.16	
Crab ( <i>Cancer pagurus</i> )	2	Sunnhordaland	1998	0.14-0.8	
Crab exoskeleton ( <i>Cancer pagurus</i> )	1	Sunnhordaland	1998	0.7	
Mussels ( <i>Mytilus edulis</i> )	2	Hillesøy, Troms	1997	< 0.08	0.5
Mussels ( <i>Mytilus edulis</i> )	2	Hillesøy, Troms	1998	< 0.08	
Mussels ( <i>Mytilus edulis</i> )	1	Outer Oslofjord	November 1997		0.7
Mussels ( <i>Mytilus edulis</i> )	1	Outer Oslofjord	December 1998	0.2	
Shrimps, fresh ( <i>Pandalus borealis</i> )	1	Norwegian Sea, North Sea	June 1998	0.12	
Shrimps, fresh ( <i>Pandalus borealis</i> )	1	North Sea	February 1998	0.14	
Shrimps, fresh ( <i>Pandalus borealis</i> )	2	Outer Oslofjord	November 1997		0.3 - 0.8
Shrimps, cooked ( <i>Pandalus borealis</i> )	1	Outer Oslofjord	December 1998	0.18	

Levels of  $^{99}\text{Tc}$  were relatively high (in the range of 50-70 Bq/kg, (d.w)) during the early and mid 1980s (Dahlgard et al., 1997) reflecting the elevated discharges from Sellafield and La Hague in 1978 the preceding years. In the late 1980s and early 1990s, a significant decrease in levels was observed corresponding to the low throughput of this radionuclide at Sellafield (before operations at EARP began) and the declining discharges from La Hague. Levels were generally below 30 Bq/kg (d.w) after 1990. The  $^{99}\text{Tc}$  activity concentration in seaweed samples from outer Oslofjord has increased significantly from 1996 to 1997 (Brown et al., 1998). The effect of the increased discharges of  $^{99}\text{Tc}$  from Sellafield is also seen at locations as far north as Troms.

Table 4: Ranges of concentration for various radionuclides in seaweed from 1996 to 1998, dry weight.

Species	No of samples	Location	Year	$^{137}\text{Cs}$ (Bq/kg)	$^{99}\text{Tc}$ (Bq/kg)	$^{60}\text{Co}$ (Bq/kg)
Seaweed ( <i>Laminaria hyerborea</i> )	8	West coast of Norway	1998	< 0,2 to 0,5*		< d.l
Seaweed ( <i>Fucus serratus</i> )	1	Outer Oslofjord	November 1996	$3.6 \pm 0.2$	$36 \pm 4$	$0.7 \pm 0.08$
Seaweed ( <i>Fucus serratus</i> )	1	Outer Oslofjord	November 1997	$2.7 \pm 0.14$	$170 \pm 20$	$1 \pm 0.1$
Seaweed ( <i>Fucus serratus</i> )	1	Outer Oslofjord	December 1998	$3 \pm 0.24$	$285 \pm 34$	< d.l
Seaweed ( <i>Fucus vesiculosus</i> )	1	Outer Oslofjord	December 1998	$3.9 \pm 0.3$	$255 \pm 31$	< d.l
Seaweed ( <i>Fucus serratus</i> )	1	Kristiansand	January 1998	$1.7 \pm 0.15$		$0.5 \pm 0.1$
Seaweed ( <i>Fucus vesiculosus</i> )	4	Hillesøy	1997	< 0.3 - $0.8 \pm 0.1$	$79 \pm 8$	< d.l
Seaweed ( <i>Fucus vesiculosus</i> )	10	Hillesøy	1998	$0.4 \pm 0.1$ - $0.8 \pm 0.1$	$60 \pm 8$ - $203 \pm 25$	< d.l

\*The activity concentration in 50% of the samples were below the limit of detection.

#### 5.4 Sea water

Samples collected each year of surface sea water in the Outer Oslofjord have been analysed for a variety of anthropogenic radionuclides ( $\pi$ ). These samples were collected over the period November 1996-1998. In addition,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  concentrations were determined in samples of sea water collected from the North Sea and surrounding waters in both 1996 and 1997 (Figure 1 and 2). The ratio between  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$  show that the radioactive contamination in the Southern coastline is influenced by the Chernobyl accident. In November 1996 the activity ratios  $^{134}\text{Cs} / ^{137}\text{Cs}$  from the Chernobyl fallout is calculated to be approximately 0.023, in November 1997 approximately 0.017 and in 1998 approximately 0.013. The results from 1996 and 1997 the ratios  $^{134}\text{Cs} / ^{137}\text{Cs}$  are between 0.0094 to 0.010 in sea water samples collected near the southern coastline of Norway. In the outer Oslofjord the ratio is calculated

to be approximately 0.015 in 1996 and in Kattegat in 1997 between 0.014 to 0.015. These results indicate that outflow of Baltic water to the Norwegian current contributes substantially to the radiocesium contamination along the Norwegian coast. The highest  $^{134}\text{Cs} / ^{137}\text{Cs}$  ratios were however found near the Danish and Belgian coastline in 1997 ( in the range 0.014 - 0.017 ) The reason for this is not clear. In general, radiocesium concentrations have been relatively constant in the marine area surrounding Norway since 1990.

Transuranic elements like  $^{239,240}\text{Pu}$  reached a maximum concentration in the North Sea surface water in 1980 of  $20 \text{ mBq/m}^3$  (Kautsky, H. 1986). In 1995 sampling and analyses of  $^{239,240}\text{Pu}$  depth profiles water show higher concentration of  $^{239,240}\text{Pu}$  at a few hundred to thousands of meter depth, than Kautsky reported in 1986 for the surface water. Surface water in the North Atlantic surrounding the Norwegian coast was in 1995 and 1997 reported to be less than  $10 \text{ mBq/m}^3$  of  $^{239,240}\text{Pu}$  (Grøttheim, 1999).

*Table 5: Concentration of various radionuclides in sea water from the Outer Oslofjord (1996 to 1998).*

Year	$^{137}\text{Cs}$ (Bq/m <sup>3</sup> )	$^{134}\text{Cs}$ (Bq/m <sup>3</sup> )	$^{238}\text{Pu}$ (mBq/m <sup>3</sup> )	$^{239,240}\text{Pu}$ (mBq/m <sup>3</sup> )	$^{241}\text{Am}$ (mBq/m <sup>3</sup> )
1996	$16 \pm 0.8$	$0.24 \pm 0.02$			
1997	$11.4 \pm 0.6$	$0.12 \pm 0.012$	< 1.6	$2.2 \pm 0.5$	< 1.0
1998	$12 \pm 0.6$	$0.08 \pm 0.013$			

The radionuclides,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were also analysed in sea water samples from different water depths and locations in Norwegian coastal waters in 1997 (Table 6). The concentrations are almost constant at all depths.

The ratio  $^{238}\text{Pu}/^{239,240}\text{Pu}$  measured in sea water from the North Sea and Skagerrak show that the results can be explained by a combination of radionuclides derived from Western European reprocessing plants in addition to fallout from atmospheric nuclear weapons testing during the fifties and the sixties.

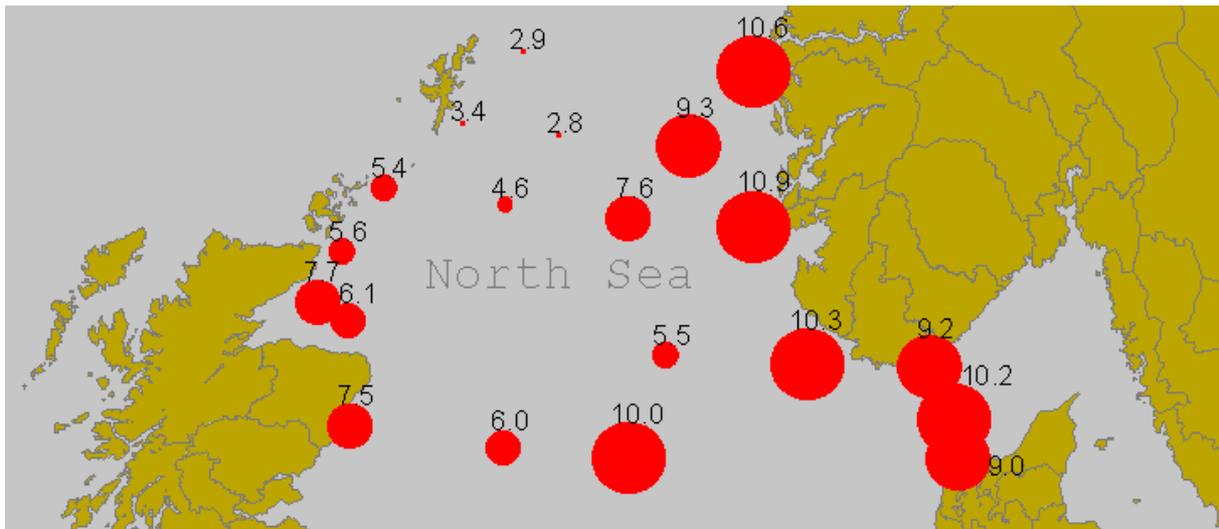


Figure 1:  $^{137}\text{Cs}$  concentrations in sea water in 1996, in Bq/m<sup>3</sup>.

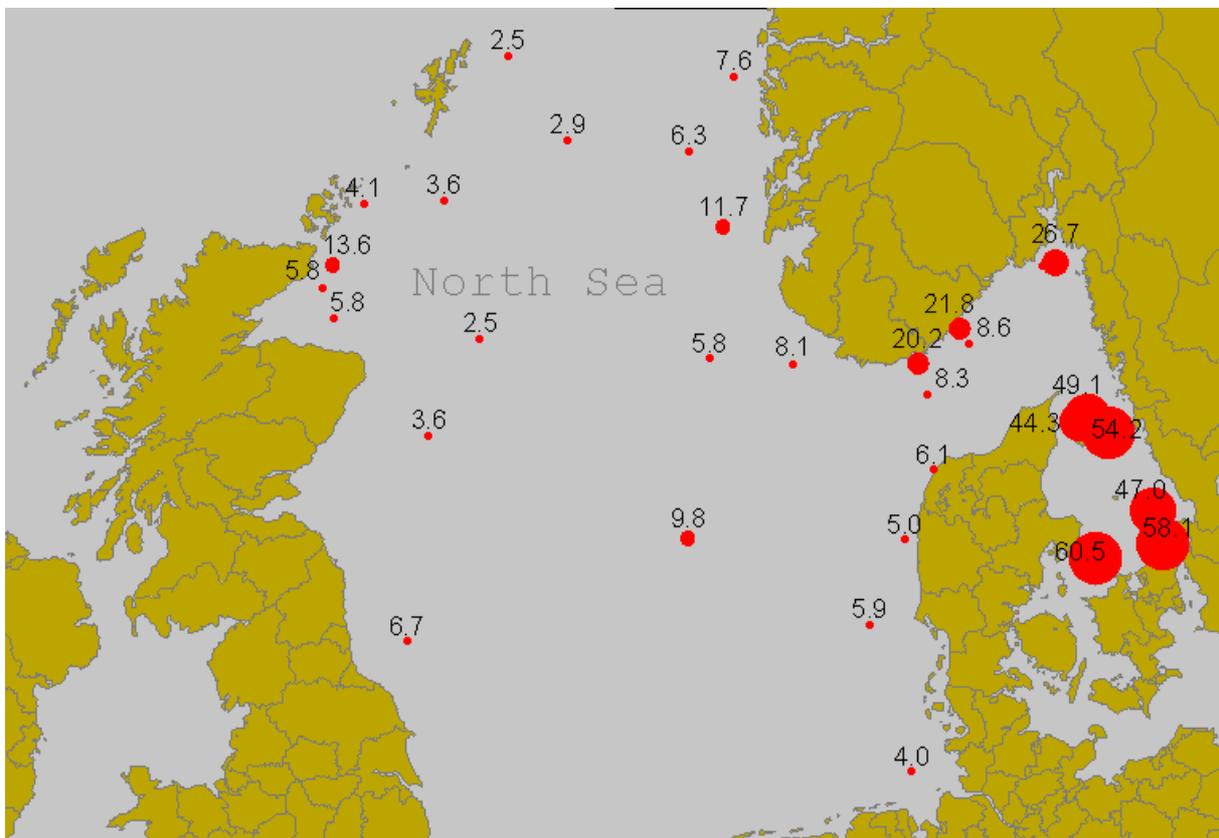


Figure 2:  $^{137}\text{Cs}$  concentrations in sea water in 1997, in Bq/m<sup>3</sup>.

As previously mentioned, the levels of  $^{99}\text{Tc}$  have been increasing in response to the elevated discharges from Sellafield from 1994. Levels of  $^{99}\text{Tc}$  in Norwegian coastal waters are generally in the range of 0.9-6.5 Bq/m<sup>3</sup> (Figure 3 and Figure 4). Levels of  $^{99}\text{Tc}$  in Norwegian coastal waters have increased by almost an order of magnitude between 1991 and the end of 1996 (Brown et al. 1998). The highest level of  $^{99}\text{Tc}$  recorded in this surveillance programme

was  $8.5 \text{ Bq/m}^3$ , recorded north east of the Shetland Islands. The plume of  $^{99}\text{Tc}$  from Sellafield has reached the Norwegian coast before November 1996 and the results indicate a transport time for the arrival of the first pulse of contamination of less than 2.5 years (Brown et al., 1998).

## 5.5 Sediments

Sediment samples were collected from a limited number of stations in the Skagerrak and the North Sea. Samples were collected from the upper 2 cm of the sediment deposit. The samples were analysed for  $\gamma$ -emitting nuclides  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ . Samples from all stations generally contained low levels of these radionuclides. At all stations except one,  $^{137}\text{Cs}$  levels are below  $4 \text{ Bq/kg}$  (d.w.). Only near Skagerrak was the  $^{137}\text{Cs}$  level higher,  $11 \text{ Bq/kg}$  (d.w.) (Figure 5). The  $^{137}\text{Cs}$  level at this station could be explained by the combination of radionuclides derived from weapons fallout, Western European reprocessing plants, and outflow of Baltic water contaminated by the Chernobyl fallout.

Table 6: Concentration of transuranics in sea water (1997).

St.nr.	Depth, m	Location	$^{238}\text{Pu}$ , $\text{mBq/m}^3$	$^{239,240}\text{Pu}$ , $\text{mBq/m}^3$	$^{241}\text{Am}$ , $\text{mBq/m}^3$
St. 5	0	Norwegian coast	$< 3$	$9.5 \pm 1.4$	$3.8 \pm 0.6$
	150	Slotterøy	$< 1$	$12.5 \pm 1.2$	$2.6 \pm 0.5$
	260	$60^\circ 00' \text{N} - 03^\circ 30' \text{E}$	*	*	$5.9 \pm 0.8$
St. 32	0	Skagerrak	$1.6 \pm 0.3$	$9.9 \pm 0.8$	$5.2 \pm 0.6$
	50		$2.0 \pm 0.5$	$13.1 \pm 1.5$	$3.0 \pm 0.5$
	385	$57^\circ 44' \text{N} - 08^\circ 17' \text{E}$	$1.5 \pm 0.3$	$14.9 \pm 1.1$	$18.9 \pm 1.4$
St. 45	0	Danish coastline $56^\circ 15' \text{N} - 08^\circ 00' \text{E}$	$5.5 \pm 0.6$	$9.1 \pm 0.8$	$3.6 \pm 0.7$
St.47	0	Outer Oslofjord $59^\circ 11' \text{N} - 10^\circ 38' \text{E}$	*	*	$0.5 \pm 0.1$

\*Not measured

The levels of  $^{239,240}\text{Pu}$  varied from  $0.07$ - $2.8 \text{ mBq/kg}$  and for  $^{238}\text{Pu}$  from below  $0.01$  up to  $0.27$  (Figure 6). As was previously observed for  $^{137}\text{Cs}$ , the highest levels occur near Skagerrak. While  $^{241}\text{Am}$  was detected in sediments from all stations, the levels are very low;  $\leq 0.8 \text{ mBq/kg}$ .



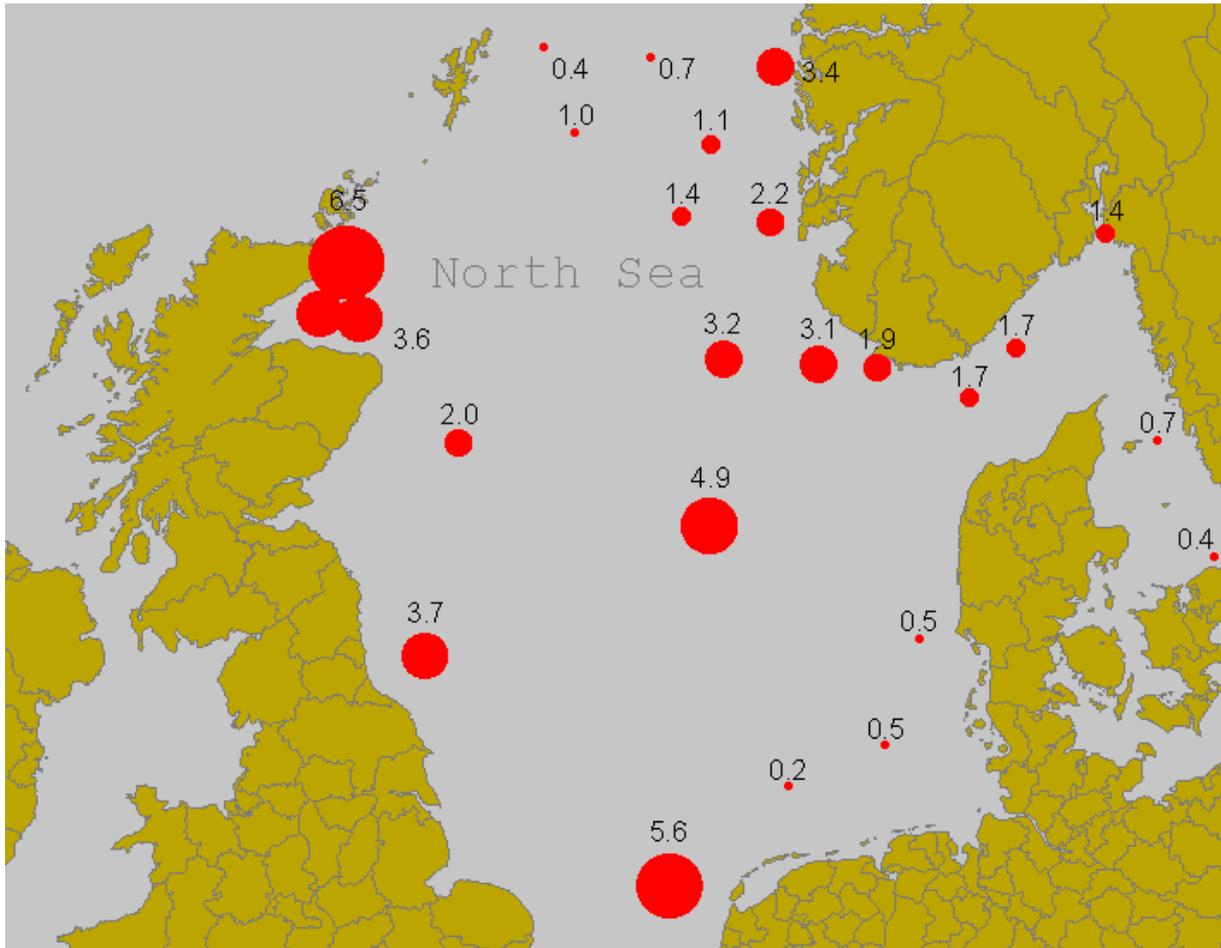


Figure 4: <sup>99</sup>Tc concentrations in sea water in 1997, expressed in Bq/m<sup>3</sup>.

### 5.6 Comparison with other areas

Two of the most important areas in northern Europe with respect to radioactive contamination are the Irish Sea which is the immediate receiver of radioactive discharges from Sellafield and the Baltic Sea which has acted as a large catchment basin for Chernobyl-related radionuclides. The Baltic and the Irish seas contain higher levels of anthropogenic radionuclides than in Norwegian waters in both biological and non-biological components of the marine environment. For the commercial fisheries industry, the most important anthropogenic radionuclide is <sup>137</sup>Cs. As previously noted, <sup>137</sup>Cs concentrations in fish caught from areas around Norway currently are below 1.2 Bq/kg (w.w), whereas in the Irish and Baltic Sea, the levels can be a factor of 10 higher (MAFF & SEPA, 1998). In more open sea areas, the levels in fish are considerably reduced due to the dilution of sea water concentrations with increasing distance from the sources.

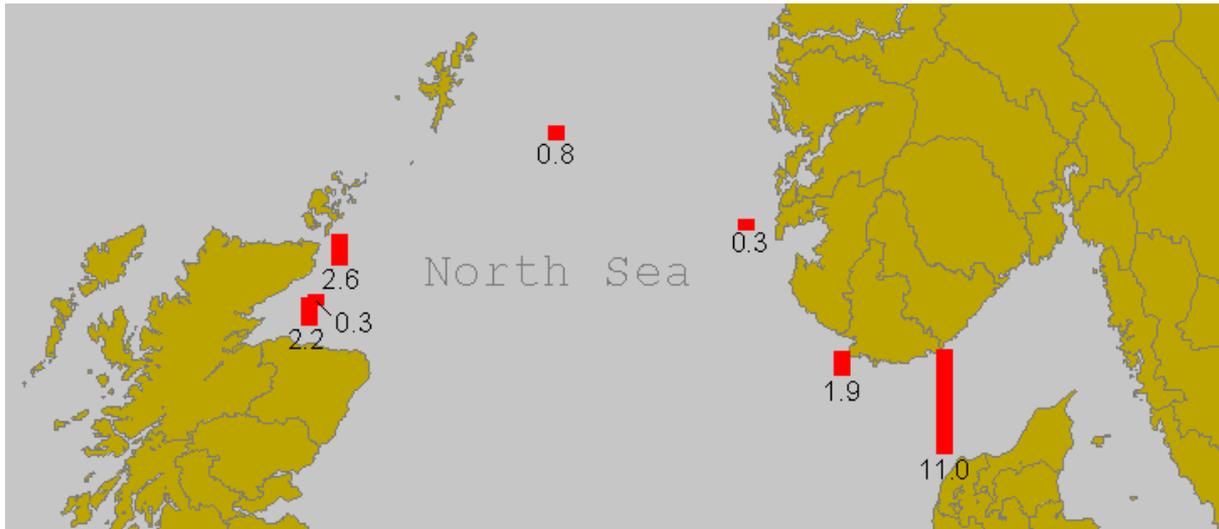


Figure 5: Levels of  $^{137}\text{Cs}$  in sediments from the Skagerrak and the North Sea in 1996, in Bq/kg (d.w).

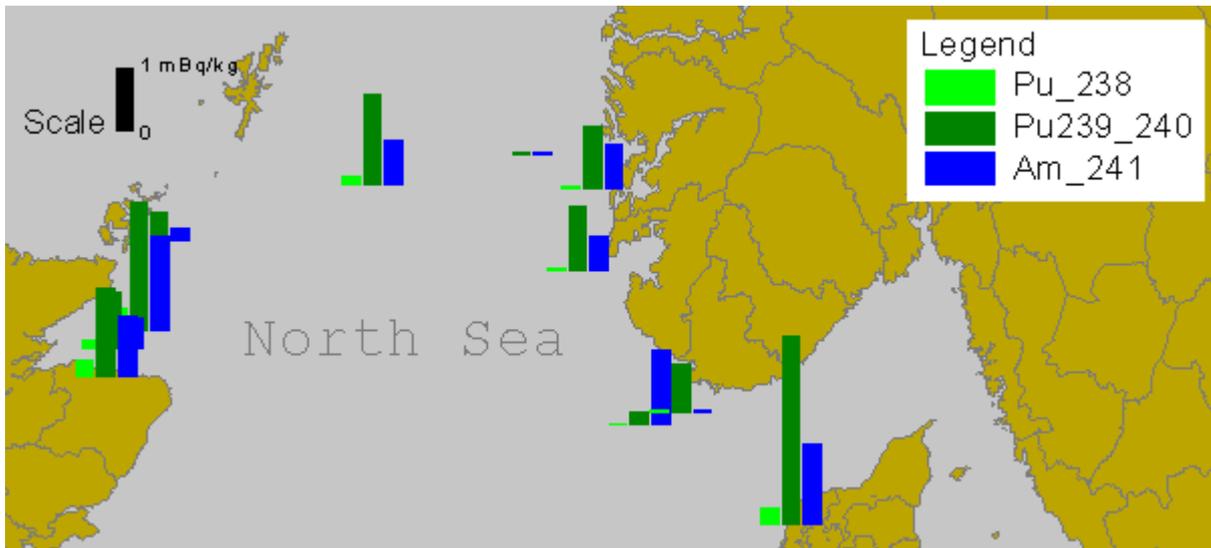


Figure 6: Levels of  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  expressed in mBq/kg in sediments from the Skagerrak and the North Sea in 1996.

The levels of many of the primary anthropogenic radionuclides supplied by the nuclear reprocessing facilities in Europe have been decreasing since the end of the seventies and the beginning of the eighties. However the increase in discharge of  $^{99}\text{Tc}$  in mid 1994 indicate that the  $^{99}\text{Tc}$  plume reached the Norwegian south-west coast before November 1996, a travel time of less than 2.5 years. Compared to  $^{99}\text{Tc}$  levels in the Irish Sea, the levels in waters adjacent to Norway are still quite low (Brown et al., 1998)(Figure 3 and 4). Further monitoring may provide additional information regarding transport and behaviour of  $^{99}\text{Tc}$  in the marine environment.

## 6. CONCLUSIONS

Presently the main source of radioactive contamination of Norway's marine waters are releases from the reprocessing plants in Sellafield as well as from fallout from the Chernobyl accident and from nuclear weapons testing. Other sources, such as the sunken submarine Komsomolets or the dumped radioactive waste in the Kara Sea have had no significant impact on the large-scale distribution of radioactivity in the marine environment.

The data collected as part of the National Surveillance Programme indicate that radioactivity in the waters surrounding Norway remains at low levels. In fish and shrimps,  $^{137}\text{Cs}$  activity concentrations are approximately 1.2 Bq/kg or less.  $^{137}\text{Cs}$  levels in the waters surrounding Norway have decreased significantly since their peak concentrations detected around 1980. However, in recent years the variations in radiocesium concentration in the sea water can largely be explained by variations in the water exchange with the Baltic Sea. The influence of Chernobyl fallout on the concentrations of these radionuclides is clearly seen. The levels decrease with increasing distance away from the Baltic Sea. Other radionuclides, i.e.  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ ,  $^{60}\text{Co}$  and  $^{241}\text{Am}$  were found in low concentrations only.

The reprocessing plant at Sellafield in United Kingdom began operating a new waste treatment plant in 1994. These has resulted in changes in the composition of radionuclides being discharged into the sea as waste. As a result, the concentrations of  $^{99}\text{Tc}$  in the waters surrounding Norway have increased in recent years and the highest levels of radioactivity detected in marine biota during the surveillance program were for  $^{99}\text{Tc}$  in lobster. The increase in  $^{99}\text{Tc}$  is also clearly observed in seaweed.

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