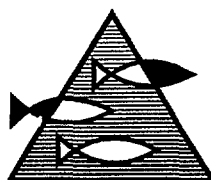


DISSERTATION FOR THE DEGREE OF DOCTOR SCIENTIARUM  
2001

# RADIOACTIVITY IN NORWEGIAN WATERS: DISTRIBUTION IN SEAWATER AND SEDIMENTS, AND UPTAKE IN MARINE ORGANISMS

HILDE ELISE HELDAL



INSTITUTE OF MARINE RESEARCH, BERGEN, NORWAY



DEPARTMENT OF CHEMISTRY,  
UNIVERSITY OF BERGEN, NORWAY

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## LIST OF PAPERS

- I. Heldal, H.E., Varskog, P. and Føyn, L. Distribution of selected anthropogenic radionuclides ( $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ ) in marine sediments with emphasis on the Spitsbergen-Bear Island area. *The Science of the Total Environment*. Accepted for publication.
- II. Heldal, H.E., Stupakoff, I. and Fisher, N.S., 2001. Bioaccumulation of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  by five marine phytoplankton species. *Journal of Environmental Radioactivity* 57 (3), 231-236.
- III. Heldal, H.E., Føyn, L. and Varskog, P. Bioaccumulation of  $^{137}\text{Cs}$  in pelagic food webs in the Norwegian and Barents Seas. Submitted to *Journal of Environmental Radioactivity*.
- IV. Tolley, K.A. and Heldal, H.E., 2001. Inferring ecological separation from regional differences in radioactive caesium in harbour porpoises (*Phocoena phocoena*). *Marine Ecology Progress Series*. In press.
- V. Heldal, H.E., Kershaw, P.J. and Mork, K.A. Technetium-99: a renewed transient tracer of Nordic Seas and Arctic circulation. Submitted to *Deep-Sea Research*.

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## CONTENTS

<b>1. INTRODUCTION .....</b>	<b>1</b>
1.1 ANTHROPOGENIC RADIOACTIVITY IN NORWEGIAN AND ARCTIC SEAS - MAIN SOURCES ..	1
1.1.1 Releases from nuclear weapons testing .....	2
1.1.2 The Chernobyl accident .....	3
1.1.3 Discharges from European nuclear fuel reprocessing plants.....	4
1.1.4 Discharges from Russian nuclear fuel reprocessing plants.....	6
1.1.5 The sunken nuclear submarines Komsomolets and Kursk.....	6
1.1.6 Dumped radioactive wastes in the Barents and Kara Seas .....	7
1.1.7 The Thule nuclear weapons accident.....	7
1.2 RADIOACTIVE CONTAMINATION OF FISH IN THE BARENTS SEA DURING THE 1960'S AND TODAY .....	8
1.3 NATURAL VS. ANTHROPOGENIC RADIOACTIVITY IN THE MARINE ENVIRONMENT .....	9
1.4 ANTHROPOGENIC RADIOACTIVITY IN TERRESTRIAL VS. MARINE FOOD WEBS .....	10
1.5 WHY DO WE MEASURE RADIOACTIVE CONTAMINATION IN THE MARINE ENVIRONMENT? ..	11
1.6 BACKGROUND AND TOPICS OF THE THESIS .....	11
<b>2. SAMPLE COLLECTION .....</b>	<b>13</b>
2.1 SEDIMENTS.....	13
2.2 BIOTA .....	14
2.3 SEAWATER .....	15
<b>3. ANALYSES.....</b>	<b>16</b>
3.1 GAMMA RAY SPECTROSCOPY USING AN HPGe DETECTOR.....	17
3.2 <sup>137</sup> Cs IN SEDIMENTS (IMR AND IFE) .....	18
3.3 <sup>137</sup> Cs IN BIOTA (IMR AND IFE) .....	18
3.4 <sup>137</sup> Cs IN SEAWATER (CEFAS) .....	19
3.5 <sup>99</sup> Tc IN SEAWATER (CEFAS).....	19
3.6 <sup>238</sup> Pu, <sup>239,240</sup> Pu AND <sup>241</sup> Am IN SEDIMENTS (CEFAS) .....	20
3.7 DATING OF SEDIMENTS BY THE <sup>210</sup> Pb/ <sup>210</sup> Po METHOD (RISØ NATIONAL LABORATORY)...	20
3.8 UPTAKE STUDIES OF <sup>137</sup> Cs AND <sup>57</sup> Co IN MARINE PHYTOPLANKTON (MSRC) .....	21
<b>4. SUMMARY OF MAIN RESULTS.....</b>	<b>22</b>
4.1 DISTRIBUTION OF <sup>137</sup> Cs, <sup>238</sup> Pu, <sup>239,240</sup> Pu AND <sup>241</sup> Am IN MARINE SEDIMENTS WITH EMPHASIS ON THE SPITSBERGEN-BEAR ISLAND AREA ( <i>PAPER I</i> ).....	22
4.2 UPTAKE AND ACCUMULATION OF <sup>137</sup> Cs IN MARINE FOOD WEBS ( <i>PAPER II, III AND IV</i> ) ...	23
4.3 TRANSPORT OF <sup>99</sup> Tc FROM SELLAFIELD TO ARCTIC ENVIRONMENTS ( <i>PAPER V</i> ).....	26
<b>5. SUGGESTIONS FOR FURTHER WORK .....</b>	<b>29</b>
<b>6. REFERENCES .....</b>	<b>30</b>
<b>PAPERS I-V</b>	

## **1. INTRODUCTION**

Norway controls some of the richest fishing grounds in the world. The North Sea, Norwegian coast, the Barents Sea and the polar front in the Norwegian Sea are all highly productive areas. Yearly catches amount to 2.5-3 million tons fish of which about 90% are exported (Norwegian Seafood Export Council). Seafood is, after oil, Norway's second largest export commodity, bringing in earnings of NOK 31.5 billion in 2000 (Norwegian Seafood Export Council). Anthropogenic (man-made) radioactivity has represented a potential threat to Norwegian fisheries since it was introduced to the marine environment with atmospheric testing of thermonuclear bombs, reaching a peak in the late 1950's and early 1960's (UNSCEAR 1993). To place the current levels of radioactive contamination of the marine environment into perspective, it is useful to relate them to earlier levels of radioactive contamination and levels of natural radioactivity in the marine environment, and to contrast these to the levels of radioactive contamination in terrestrial food webs. A brief introduction to these topics is given below together with an overview of the most important sources for radioactive contamination in the Northern Hemisphere. The introduction is ended with a description of the topics and background of this thesis.

### **1.1 Anthropogenic radioactivity in Norwegian and Arctic Seas - Main Sources**

Nuclear weapons explosions have provided the largest inventory of both fission and neutron activation products in the global environment. In addition to local contamination, small contaminated particles produced in high-yield explosions were transported into different atmospheric layers. The radionuclides have been transferred back to aquatic and terrestrial ecosystems, mainly by wet deposition, in rain or snow (AMAP 1998).

As a result of the use of fission reactors for electrical power generation, there are large quantities of fission and activation products in spent nuclear fuel and reactor assemblies. The spent nuclear fuel can either be stored or reprocessed for recovery of plutonium for further use in the electrical generating industry. The latter has involved releases of substantial quantities of radioactive wastes to the environment. Various nuclear accidents have further contributed to the inventory of anthropogenic radionuclides in the environment.

### ***1.1.1 Releases from nuclear weapons testing***

Releases of anthropogenic radionuclides to the environment became a reality on 16 July 1945 when the first fission weapon test was performed in New Mexico, USA. Fission weapons tests detonated in the years 1945-1952 resulted in depositions locally and at latitude bands where the sites were situated, and did not have any influence on the radioactive contamination of the polar regions (Aarkrog 1994). The major test sites were the Bikini Islands (USA) in the Pacific Ocean, Nevada (USA) and Semipalatinsk (Kazakhstan). The global fallout, which contaminated all parts of the Earth, started when USA tested the first thermonuclear bomb (H-bomb) at the Eniwetok Island in the Pacific Ocean on 1 November 1952. The thermonuclear bombs yielded higher explosive power than the first fission weapons did. The major sites for atmospheric testing of thermonuclear weapons were Novaya Zemlya in the arctic region of the former USSR, Bikini and the Eniwetok Islands in the Pacific Ocean and the Nevada test site.

Most of the atmospheric release occurred in the two periods 1952-1958 and 1961-1962 (UNSCEAR 1993), separated by a temporary test ban treaty in 1959-1960. In 1963, Great Britain, USA and the USSR signed a treaty banning all atmospheric tests, which resulted in a large decrease in the total global fallout. In total, 520 atmospheric nuclear tests have taken place, performed mostly by the USA and the former USSR (UNSCEAR 1993). Since 1980, there has been no atmospheric testing carried out by any country. The total production of selected radionuclides by atmospheric nuclear tests is shown in Table 1.1. Atmospheric releases have led to 'background' levels of  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  near the Azores in the northeastern Atlantic, in surface seawater, of 0.005, 1.6 and 2.5 Bq m<sup>-3</sup>, respectively (Dahlggaard et al. 1995a).

*Table 1.1. Total releases of selected radionuclides by past atmospheric nuclear tests (UNSCEAR, 1993)*

	Total production (PBq) (1 PBq = 1 · 10 <sup>15</sup> Bq)
Strontium-90 ( $^{90}\text{Sr}$ )	604
Caesium-137 ( $^{137}\text{Cs}$ )	912
Plutonium-239 ( $^{239}\text{Pu}$ )	6.5
Plutonium-240 ( $^{240}\text{Pu}$ )	4.3
Plutonium-241 ( $^{241}\text{Pu}$ )	142

In 1955, 1957 and 1961, three underwater nuclear weapons tests took place at Novaya Zemlya (JNRE 1996; AMAP 1998). Two tests were conducted within the Chernaya Bay (located on the southwestern coast of Novaya Zemlya), while one was conducted in the vicinity. Elevated levels of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ , and  $^{239,240}\text{Pu}$  (levels exceeding 100, 300, and 15,000  $\text{Bq kg}^{-1}$ , respectively) have been measured in sediments in the deep (60-80 m) central part of the Chernaya Bay (Smith et al. 2000). These levels are orders of magnitude higher than fallout levels (e.g.  $< 1 \text{ Bq kg}^{-1}$  for  $^{239,240}\text{Pu}$  in sediments). Levels of  $^{239,240}\text{Pu}$  in Chernaya Bay bottom water is also relatively high ( $\sim 4 \text{ Bq m}^{-3}$ ), but restricted exchange over the fjord sill limits the rate of  $^{239,240}\text{Pu}$  transport from the Chernaya Bay into the eastern Barents Sea (Smith et al. 2000). In contrast, the concentration of  $^{239,240}\text{Pu}$  in surface seawater south of Spitsbergen was  $\sim 7 \text{ mBq m}^{-3}$  in 1999 (Heldal, unpubl. data). Low  $^{240}\text{Pu}:^{239}\text{Pu}$  atom ratios in sediment cores collected in the eastern Barents Sea indicate, however, that significant offshore transport of plutonium has occurred in the past, probably at the time of the nuclear tests (Smith et al. 2000).

A total of 42 underground weapons tests were carried out at Novaya Zemlya from 1961 to 1990 (JNRE 1996). The underground tests are assumed to have no significant impact on the level of sea contamination, but local contamination due to leakages cannot be excluded (JNRE 1996).

### ***1.1.2 The Chernobyl accident***

The Chernobyl nuclear power plant accident took place in Ukraine on April 26, 1986. In the order of 100 PBq  $^{137}\text{Cs}$  ( $1 \text{ PBq} = 10^{15} \text{ Bq}$ ) were released to the atmosphere and deposited in the Northern Hemisphere, mainly in Europe and the USSR (Aarkrog 1994). Due to runoff from land, the Baltic Sea still receives significant amounts of  $^{137}\text{Cs}$  originating from the Chernobyl accident. This is transported to Norwegian coastal areas as described in section 1.1.3.

As a result of the test ban treaty signed in 1963 (see above), and the following decrease in global fallout, the monitoring of anthropogenic radioactivity in the marine environment was gradually reduced to a minimum. Following the Chernobyl accident, however, funds were made available for purchase of new instrumentation, implementation of new radio-analytical methods and training of workers. It may well be said that research in the field of radioecology was resurrected at that time.



### 1.1.3 Discharges from European nuclear fuel reprocessing plants

The reprocessing plants of main importance in Northwestern Europe are Sellafield (formerly Windscale) in Cumbria, UK, and Cap de La Hague near Cherbourg, France (Fig. 1.1). Sellafield is operated by British Nuclear Fuels plc (BNFL), and has discharged radioactive wastes into the Irish Sea since 1951 (Grey et al. 1995). The nuclides representing the principle beta/gamma emitters released from Sellafield (excluding  $^3\text{H}$ ) are  $^{137}\text{Cs}$ ,  $^{241}\text{Pu}$ ,  $^{90}\text{Sr}$  and  $^{106}\text{Ru}$  (Fig. 1.2). Releases of alpha emitters are dominated by  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  (Fig. 1.3). The La Hague reprocessing plant, which discharges into the English Channel, was brought into operation in 1965 (AMAP 1998). The total discharges from this site are much

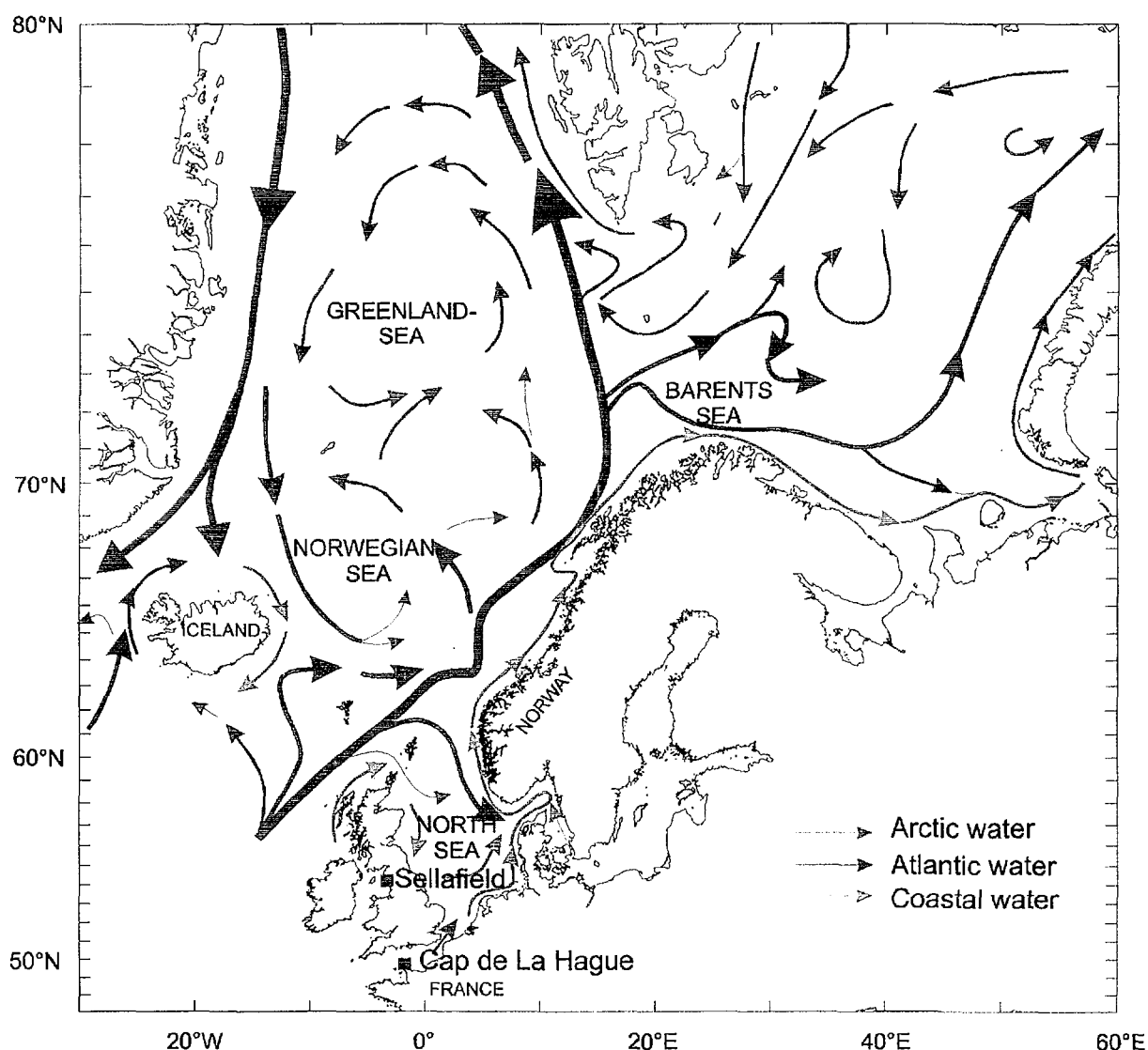


Figure 1.1. Circulation of surface waters of the North, Norwegian, Greenland and Barents Seas. The reprocessing plants Sellafield and Cap de La Hague are shown.

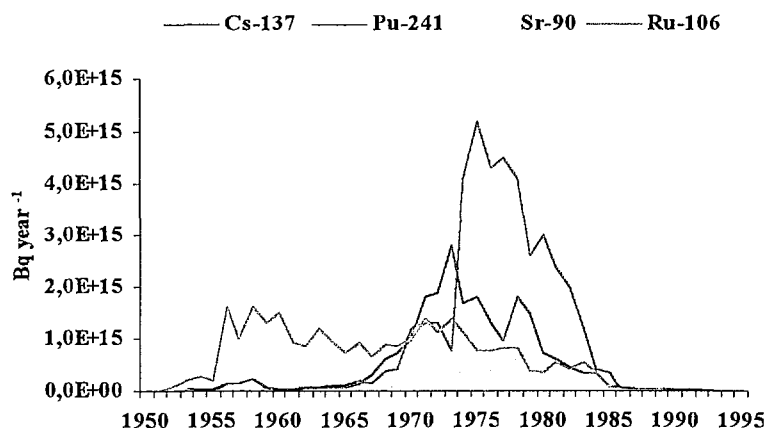


Figure 1.2. Discharges of principle beta/gamma emitters in liquid effluent from Sellafield 1952-1992 (after Gray et al. 1995)

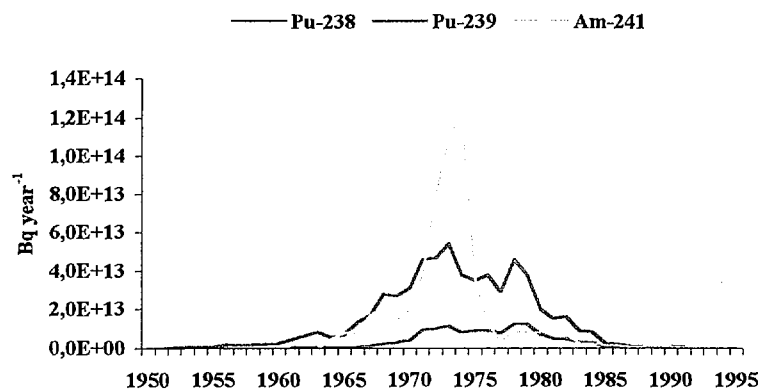


Figure 1.3. Discharges of principle alpha emitters in liquid effluent from Sellafield 1952-1992 (after Gray et al. 1995)

less than those from Sellafield. The dominant contributor to activity releases has been the beta emitter  $^{106}\text{Ru}$ , followed by  $^{90}\text{Sr}$  (AMAP 1998).

The radioactive wastes are transported with ocean currents from Sellafield and La Hague into the North Sea and further into the Skagerrak, where they merge with waters from the Baltic Sea containing significant amounts of  $^{137}\text{Cs}$  originating from fallout from the Chernobyl accident (Kershaw and Baxter 1995) (Fig. 1.1). They are transported further northwards in the Norwegian Coastal Current (NWCC), which gradually mixes with and contaminates the Norwegian Atlantic Current (NWAC). The NWAC splits into two branches off northern Norway: the North Cape Current (NCC) and the West Spitsbergen Current (WSC), which transport the radioactive wastes eastwards into the Barents Sea and northwards into the Arctic Ocean, respectively.

#### **1.1.4 Discharges from Russian nuclear fuel reprocessing plants**

There are three reprocessing plants for spent nuclear fuel in Russia: Mayak (Chelyabinsk), Krasnoyarsk and Tomsk. Mayak was the first plant established in the USSR for production of nuclear weapons material and began operation in 1948. Radioactive wastes from Mayak and Tomsk are discharged into the drainage area of the Ob River, while those from Krasnoyarsk are discharged directly into the Yenisey River. The radioactive wastes discharged into these rivers can be transported downstream (northwards) and enter the Kara Sea (JNRE 1997; AMAP 1998; Balonov et al. 1999). These wastes may spread further into the Barents Sea. Large quantities of liquid radioactive waste have been discharged from Mayak. Operational releases from Mayak were, however, considerably lowered following the shutdown of the last of the five uranium-graphite reactors in 1990.

#### **1.1.5 The sunken nuclear submarines *Komsomolets* and *Kursk***

On 7 April 1989, the Soviet nuclear submarine *Komsomolets* sank 180 km southwest of Bear Island in the Norwegian Sea. The wreck presently rests at a depth of 1658 m (Føyn 1994). The sunken submarine contains one nuclear reactor with an inventory of long-lived radionuclides comprising  $2.8 \cdot 10^{15}$  Bq  $^{90}\text{Sr}$  and  $3.1 \cdot 10^{15}$  Bq  $^{137}\text{Cs}$  along with other fission and activation products, and two mixed uranium/plutonium nuclear warheads containing about  $1.6 \cdot 10^{13}$  Bq weapons-grade plutonium (AMAP 1998; Balonov et al. 1999). Once a year, water and sediments are collected in the vicinity of the wreck and analysed for  $^{137}\text{Cs}$ , as part of the Norwegian Institute of Marine Research's (IMR) routine monitoring program. Results from this program show levels of  $^{137}\text{Cs}$  in the range of 1-10 Bq kg<sup>-1</sup> and 1-30 Bq m<sup>-3</sup> in sediments and seawater collected close to *Komsomolets*, respectively (Føyn 2001). Detectable amounts of  $^{134}\text{Cs}$  in these sediments indicate some leaching from the reactor (Føyn 2001). Due to the deep position of the wreck, radioactive releases are considered to be of low importance to the biological active upper layer of the ocean.

On 12 August 2000, the Russian nuclear submarine *Kursk* sank to a depth of 116 m in the Barents Sea, north-east of Murmansk, about 250 km from Norway and 80 km from the coast of Kola. *Kursk* is equipped with two pressurized water reactors, which, according to official Russian sources, were shut down during the accident. The submarine was not carrying nuclear weapons (Amundsen et al. 2001). The submarine was recovered 8 October 2001.

Continuous measurements of radioactivity in seawater, sediment and fish indicate no leakages from *Kursk* so far (October 2001), but the most critical phase, the decommissioning of the wreck, is still left. *Kursk* adds up to about 100 Russian nuclear submarines waiting for decommissioning along the Kola Peninsula, which all represent a danger for future releases.

#### ***1.1.6 Dumped radioactive wastes in the Barents and Kara Seas***

For over three decades (1959-1991), the USSR dumped radioactive waste in shallow waters of the Arctic Seas. In 1993, the Russian Federation published the so-called 'White Book', which reported the total amount of radioactive wastes dumped in Arctic Seas to be approximately 90 PBq at the time of dumping (Office of the President of the Russian Federation (OPRF) 1993). Of the total inventory, 89 PBq was high-level radioactive waste. The dumped items include six nuclear submarine reactors and a shielding assembly from an icebreaker reactor each containing spent fuel, ten nuclear reactors without fuel, liquid low-level waste and solid intermediate and low-level waste. The solid wastes were dumped in the Kara Sea, mainly in the shallow fjords at the eastcoast of Novaya Zemlya at depths of between 12 and 135 m and in the Novaya Zemlya Trough at depths up to 380 m. Liquid low-level wastes were discharged in the open Barents and Kara Seas. A joint Norwegian-Russian Expert Group has investigated the radioactive contamination of northern areas as a result of the dumping of nuclear waste in the Barents and Kara Seas. Elevated levels of radionuclides were found in the close vicinity of almost all dumped objects. However, the contamination were at background levels only a few meters from the objects (Føyn and Nikitin 1994). The levels in seawater, sediments and biota in the open Kara Sea were found to be generally low, and lower than the existing levels in the Irish, Baltic and North Seas (JNRE 1994; Strand et al. 1994; JNRE 1996).

#### ***1.1.7 The Thule nuclear weapons accident***

In January 1968, an American B-52 aircraft carrying four nuclear weapons crashed on the sea-ice of the Bylot Sund 11 km west of the Thule Air Base, Greenland. This resulted in dispersion of plutonium onto the ice. A clean-up operation involving recovering of weapons and aircraft debris and removing of the upper layer of contaminated snow and ice, was very

efficient. Approximately 1 TBq (1 TBq= $10^{12}$  Bq) ( $\pm 50\%$ ) of plutonium remained on the ice (Aarkrog 1994; Aarkrog et al. 1984). This was deposited on the seabed after the ice melted, and is assumed to have entered marine food webs in this area.

## 1.2 Radioactive contamination of fish in the Barents Sea during the 1960's and today

As mentioned above, the majority of the total atmospheric releases occurred in the two periods 1952-1958 and 1961-1962. Investigations performed by IMR showed that the highest levels of anthropogenic radioactivity (at that time measured as the total mean beta activity minus potassium-40 ( $^{40}\text{K}$ )) in fish during the 1960's were found in 1962-1963 (Fig. 1.4), clearly as a response to the previous atmospheric releases. The contamination levels in fish decreased substantially during the late 1960's as a result of the decrease in global fallout, biological elimination of the radioactive elements and possibly short physical half-lives of some beta emitters.

Today, the levels of radioactive contamination in fish in areas important to Norwegian fisheries are low. Although not directly comparable to the levels of 'total mean beta activity minus  $^{40}\text{K}$ ', the levels of one of the most abundant anthropogenic gamma emitting radionuclides,  $^{137}\text{Cs}$ , are generally below 1 Bq  $\text{kg}^{-1}$  ww (wet weight) in fish today (Rissanen et al. 1997; Brungot et al. 1999; the present study).

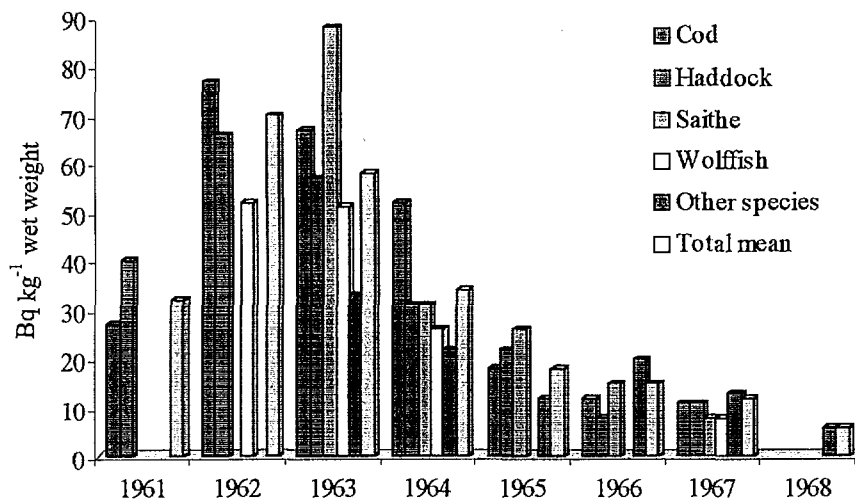


Figure 1.4. Radioactivity in fish in the Barents Sea 1961-1968. The activities are expressed as 'total mean beta-activity' minus potassium-40 ( $^{40}\text{K}$ ) (after Føyn et al. 1999).

### 1.3 Natural vs. anthropogenic radioactivity in the marine environment

More than 60 radionuclides occur naturally in the earth's environment (Joseph et al. 1971). Natural radionuclides present in rocks and minerals are carried to the oceans by runoff from land or as gases or particulates by the wind. If the total amount of anthropogenic radionuclides were diluted in the oceans, the total activity would be dominated by the naturally occurring radioactivity (Aarkrog 1997). Activities of selected natural radionuclides in seawater are shown in Table 1.2.  $^3\text{H}$  and  $^{14}\text{C}$  are also produced by nuclear detonations. Activities of selected anthropogenic radionuclides in seawater are shown in Table 1.3 for comparison.

Table 1.2. Activities of selected natural radionuclides in seawater (Joseph et al. 1971)

Radionuclide	Half-life	Mode of decay	Activity (Bq m <sup>-3</sup> seawater)
Uranium-238 ( $^{238}\text{U}$ )	4.5·10 <sup>9</sup> y	$\alpha$	33-42
Thorium-234 ( $^{234}\text{Th}$ )	24.1 d	$\beta$	37
Polonium-210 ( $^{210}\text{Po}$ )	138.4 d	$\alpha$	3
Lead-210 ( $^{210}\text{Pb}$ )	22.3 y	$\beta$	3
Potassium-40 ( $^{40}\text{K}$ )	1.3·10 <sup>9</sup> y	$\beta$	11000
Tritium ( $^3\text{H}$ )	12.3 y	$\beta$	0.6
Carbon-14 ( $^{14}\text{C}$ )	5730 y	$\beta$	3-5
Rubidium-87 ( $^{87}\text{Rb}$ )	4.8·10 <sup>10</sup> y	$\beta$	1100

Table 1.3. Activities of selected anthropogenic radionuclides in seawater

Radionuclide	Half-life	Mode of decay	Activity (Bq m <sup>-3</sup> seawater)	Sampling location	Source
$^{137}\text{Cs}$	30 y	$\beta$ ( $\gamma$ )	3.3-8.1	Barents and Kara Seas	Strand et al., 1994
$^{137}\text{Cs}$	30 y	$\beta$ ( $\gamma$ )	2.0-4.4	Greenland and Norwegian Seas	The present study
$^{90}\text{Sr}$	29.1 y	$\beta$	3.1-11.4	Barents and Kara Seas	Strand et al., 1994
$^{99}\text{Tc}$	2.1·10 <sup>5</sup> y	$\beta$	(63-197)·10 <sup>-3</sup>	Barents and Kara Seas	Strand et al., 1994
$^{99}\text{Tc}$	2.1·10 <sup>5</sup> y	$\beta$	Bd-1.61	Greenland, Norwegian and Barents Seas	The present study
$^{239,240}\text{Pu}$	*	$\alpha$	(1.8-7.7)·10 <sup>-3</sup>	Barents and Kara Seas	Strand et al., 1994
$^{239,240}\text{Pu}$	*	$\alpha$	(4.0-14.4)·10 <sup>-3</sup>	Greenland and Norwegian Seas	Heldal, unpubl. data

\*Half life: 24119 y ( $^{239}\text{Pu}$ ) and 6570 y ( $^{240}\text{Pu}$ )

Bd=Below detection limit

As shown in Tables 1.2 and 1.3, seawater concentrations of the natural radionuclide  $^{40}\text{K}$  are several orders of magnitude higher than seawater concentrations of the anthropogenic radionuclides  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in the Barents and Kara Seas. The activity of  $^{137}\text{Cs}$  in fish collected from 10 main trawling sites in the Barents Sea varied from below detection limit to  $0.8 \text{ Bq kg}^{-1} \text{ ww}$  (Rissanen et al. 1997). In contrast, the activities of  $^{40}\text{K}$  in the same samples varied from 70 to  $130 \text{ Bq kg}^{-1} \text{ ww}$ . Concentrations of  $^{40}\text{K}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in regional seafood diet in Wales in 1998 are reported to be 78, 0.54 and  $0.05 \text{ Bq kg}^{-1} \text{ ww}$ , respectively (MAFF and SEPA 1999). The present dose to humans from naturally occurring  $^{210}\text{Po}$  in fish is about 100-fold larger than the dose from all anthropogenic radionuclides in the marine environment, apart from in the immediate vicinity of major localised sources (Aarkrog 1997).

#### 1.4 Anthropogenic radioactivity in terrestrial vs. marine food webs

For representative foodstuffs, the degree of contamination generally decreases as follows (AMAP 1998):

*reindeer, mushrooms, freshwater fish > lamb meat, goat cheese > potatoes, vegetables >>  
marine fish, whale and seal meat*

The Chernobyl accident initiated many studies of uptake and transport of anthropogenic radioactivity in terrestrial food webs, for example in *lichen-reindeer-man*. Levels of  $^{137}\text{Cs}$  as high as  $85,000$  and  $50,000 \text{ Bq kg}^{-1}$  were found in lichen and reindeer meat, respectively, following the Chernobyl accident (Bretten et al. 1993; Gunnerød et al. 1989). High contamination levels were also found in mushrooms, over  $30,000 \text{ Bq kg}^{-1}$  were found in bracelet cort (*Cortinarius armillatus*) and gypsy (*Rozites caperata*) in mid-Norway in 1993 and 1992, respectively (Liland et al. 2001). These high contamination levels were transferred up the food chain and reflected in animals such as moose, sheep, goats, cattle, roe deer and reindeer. Freshwater fish also contained high  $^{137}\text{Cs}$  levels after the Chernobyl accident. An estimate of 14,000 Swedish lakes contained fish with  $^{137}\text{Cs}$  levels above  $1,500 \text{ Bq kg}^{-1}$  during the autumn of 1987 (Håkanson et al. 1992). The levels of  $^{137}\text{Cs}$  in terrestrial food webs have decreased during the 1990's, but levels as high as  $700 \text{ Bq kg}^{-1}$  were found in lamb in mid-Norway in 1998 (Liland et al. 2001).

In contrast, the concentrations of  $^{137}\text{Cs}$  in seawater along the Norwegian southern coast were  $\sim 110 \text{ Bq m}^{-3}$  in May/June 1986, about a month after the Chernobyl accident (Nies

and Wedekind 1988) (1985 background levels: 30-59 Bq m<sup>-3</sup> (Kershaw and Baxter 1995)). The seawater concentrations of <sup>137</sup>Cs decreased rapidly due to dilution, and were ~ 15-29 Bq m<sup>-3</sup> along the Norwegian coast in 1989, lower than the 1985 pre-Chernobyl concentrations. The marked decrease of <sup>137</sup>Cs levels in seawater was due to lower discharges of <sup>137</sup>Cs from Sellafield and La Hague. The concentrations of <sup>137</sup>Cs in seawater along the Norwegian coast have continued to decrease, although, as mentioned above, runoff from land and outflow from the Baltic Sea still contributes significantly with Chernobyl related <sup>137</sup>Cs.

### 1.5 Why do we measure radioactive contamination in the marine environment?

Prior to the detonation of the first thermonuclear bomb, small amounts of radioactivity, for example in mineral water, were considered to be health enriching. Negative experiences related to thermonuclear bombs and several nuclear accidents have, however, changed people's attitude towards radioactivity during the past 40-50 years. Today, there is a common concern for regular and potential accidental releases of radioactivity from sources such as Sellafield. Although this is important, incorrect assessments of the effects of these releases (e.g. created by uncritical journalism) have the potential to harm the country's fisheries and economy. Therefore, it is of major importance to document up-to-date levels of radioactive contamination of the marine environment, and be able to place these into the proper perspectives.

### 1.6 Background and topics of the thesis

The main topics of the thesis may be summarised as follows:

- 1) *Distribution of <sup>137</sup>Cs, <sup>238</sup>Pu, <sup>239,240</sup>Pu and <sup>241</sup>Am in sediments with emphasis on the Spitsbergen-Bear Island area (Paper I)*
- 2) *Uptake of <sup>137</sup>Cs in phytoplankton representative for the Barents and Norwegian Seas phytoplankton communities (laboratory experiments) (Paper II)*
- 3) *Bioaccumulation of <sup>137</sup>Cs in food webs in the Barents and Norwegian Seas (Paper III)*
- 4) *Geographical variations of <sup>137</sup>Cs in harbour porpoises (Phocoena phocoena) along the Norwegian coast (Paper IV)*
- 5) *Transport times for <sup>99</sup>Tc from Sellafield to various locations along the Norwegian coast and the Arctic Ocean (Paper V)*



A background for each study is presented below:

- 1) The levels of  $^{137}\text{Cs}$  were measured in marine sediments at 102 locations in the Barents Sea and Spitsbergen area during 1991-1993, as a part of IMR's monitoring program established after the Chernobyl accident. Results from this program indicated that sediments in the Spitsbergen area contain elevated levels of  $^{137}\text{Cs}$  (Føyn and Sværen 1997). As part of the present study, a more extensive sampling of sediments in the Spitsbergen-Bear Island area was conducted in order to test their results. Furthermore, concentrations of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were measured in these sediments, as different actinide activity ratios may provide information about the contamination source. Five samples collected in the deep Norwegian Sea were included for comparison.
- 2) A substantial ice melting takes place in the Spitsbergen area during spring and summer, giving rise to a stratified and nutrient rich euphotic zone, which supports a high primary production (Sakshaug and Skjoldal 1989). Føyn and Sværen (1997) suggested that phytoplankton in the water column in the Spitsbergen area might actively act as scavenging elements by absorption of  $^{137}\text{Cs}$  and subsequent deposition onto the seabed. Initially, a field study of the uptake of  $^{137}\text{Cs}$  in phytoplankton in the Spitsbergen area was conducted in order to test this suggestion. However, attempts to collect large enough phytoplankton samples in the Spitsbergen area for quantification of  $^{137}\text{Cs}$  were not successful for reasons given in section 4.2. As an alternative approach, uptake studies of  $^{137}\text{Cs}$  (and for comparison: cobalt-57 ( $^{57}\text{Co}$ )) in five marine phytoplankton species representative of the Barents and Norwegian Seas phytoplankton communities were performed in the laboratory.
- 3) Føyn and Sværen (1997) further hypothesised that if phytoplankton in the Spitsbergen area absorbs  $^{137}\text{Cs}$ , this might be an important mechanism for introduction of  $^{137}\text{Cs}$  to marine food webs, as these phytoplankters are heavily grazed by zooplankton (Hassel et al., 1991). In the present study, the accumulation of  $^{137}\text{Cs}$  in zooplankton and higher trophic levels of food webs in the Barents and Norwegian Seas was studied.
- 4) This study was initiated as a cooperation with Krystal Tolley from the marine mammal division at IMR about midway during the work with this thesis. Although the main purpose of her study (inferring ecological separation of harbour porpoises) is beyond the

scope of the present work, it is important for this thesis, as it is the most extensive study on  $^{137}\text{Cs}$  accumulation in marine mammals in Norwegian waters so far. The  $^{137}\text{Cs}$  measurements in harbour porpoises collected outside Finnmark were also used in *Paper III*.

- 5) Both knowledge of mechanisms for uptake and accumulation of radionuclides in marine food webs and knowledge of transport times from important sources to the fishing grounds are valuable to Norwegian fisheries, and necessary when predicting consequences of potential future releases. As mentioned above, Sellafield is an important source for radioactive contamination in the North Atlantic and the Barents Sea. The increased releases of technetium-99 ( $^{99}\text{Tc}$ ) from Sellafield, starting in 1994, provided an excellent opportunity to study the transport time for this conservative radionuclide to ocean areas important to Norwegian fisheries. The results are also placed in an appropriate oceanographic setting.

## 2. SAMPLE COLLECTION

The collection of most samples took place during three cruises conducted by IMR: in July 1998 aboard R/V 'Michael Sars' and in June/July 1999 and May/June 2000 aboard R/V 'Johan Hjort' (Rey 1999; Rey 2001). Colleagues participating on other cruises have collected a few additional samples. Sediments, seawater, phytoplankton and juvenile fish were collected in the Spitsbergen area during the cruise in 1998, which was dedicated to the work with this thesis. In 1999, sediments, biota and seawater were collected, while only biota and seawater were collected during the cruise in 2000. Researchers with various interests participated at the cruises in 1999 and 2000. As a result, the cruise tracks were compromises of the various interests. The cruise tracks for all three years are shown in Fig. 2.1 a-c.

### 2.1 Sediments

Surface sediments and sediment cores were collected using a 'Smøgen' boxcorer (30 cm x 30 cm inner dimensions) (Fig. 2.2 a). Approximately 100 cm<sup>2</sup> of the upper 1 cm was collected for each surface sample. Cores were collected in open PVC tubes of 50 cm length and 10 cm inner diameter. All samples were kept frozen at - 20°C and transported to IMR.

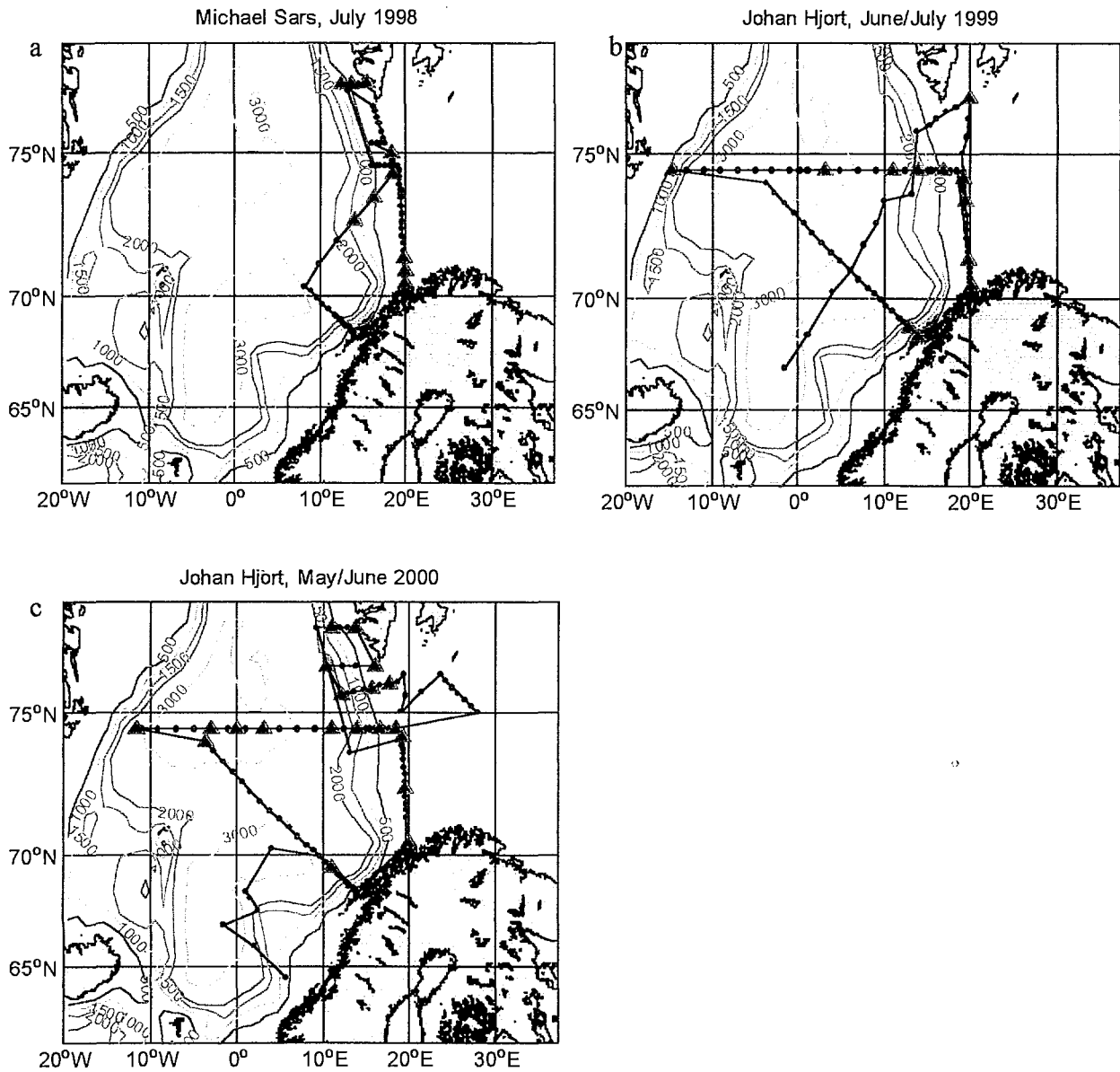


Figure 2.1. Cruise tracks. (a) 1998; (b) 1999; (c) 2000. The blue triangles show sampling stations for seawater, which was analysed for  $^{99}\text{Tc}$ .

## 2.2 Biota

In order to collect phytoplankton, 1000 L seawater was pumped from  $\sim 30$  m depth (where the highest phytoplankton concentrations were expected to be found), using a 'FLYGT' pump attached to the end of a reinforced hose (largest capacity:  $350 \text{ L min}^{-1}$ ) (Fig. 2.2 b). The seawater was thereafter filtered onboard the ship using a 'MILLIPORE' tangential flow filter system with a 'Pellicon'  $0.45 \mu\text{m}$  filter and a Millipore peristaltic pump. The fraction containing particles  $> 0.45 \mu\text{m}$ , which contains phytoplankton, was concentrated down to a few litres and transported to IMR.

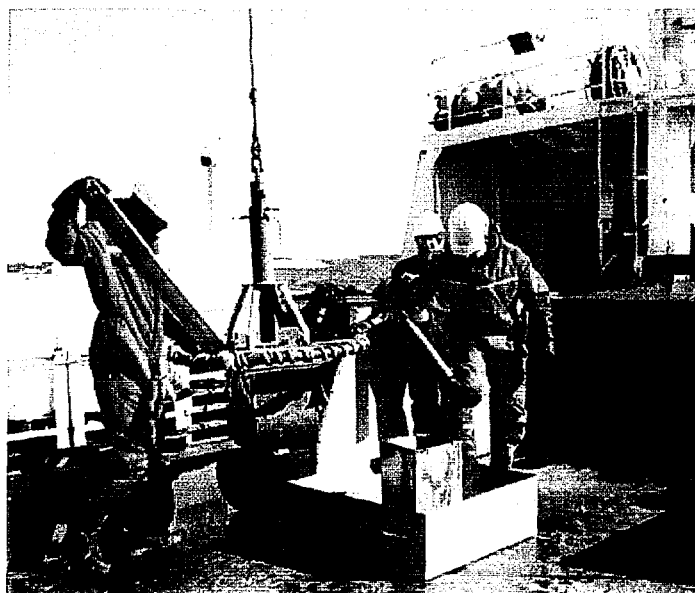
Zooplankton was collected using vertical tows at selected depth intervals by means of a WP-2 plankton net (56 cm opening, 180  $\mu$ m mesh size). The samples were kept frozen at - 20°C and transported to IMR.

Fish and benthic organisms were collected by means of pelagic and bottom trawls, kept frozen at - 20°C and transported to IMR.

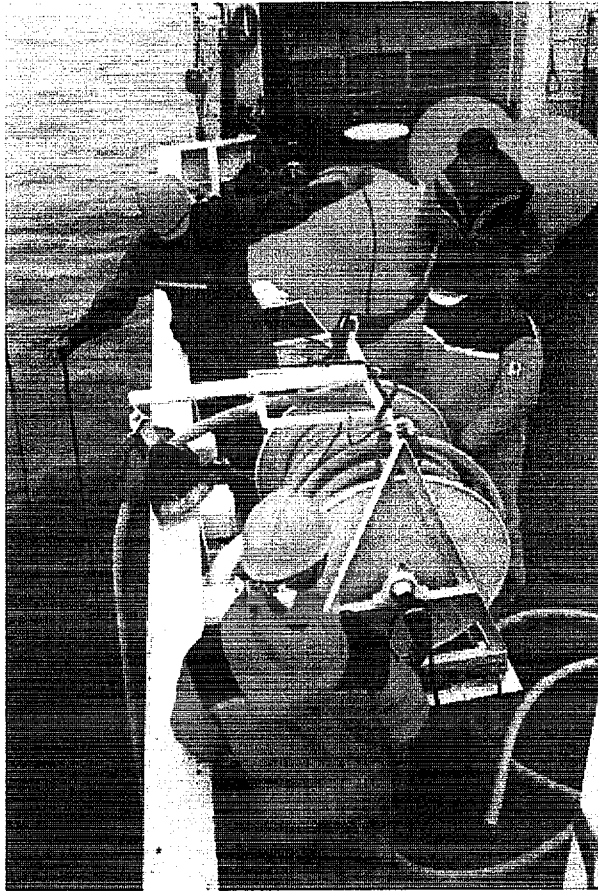
Harbour porpoises were collected opportunistically by commercial bottom-set gillnet fisheries along the coast of Norway between April - June 1999, and April 2000. All samples were stored deep-frozen and transported to IMR.

### 2.3 Seawater

Surface seawater (5 m) was collected with a shipboard pump. For sampling of seawater deeper than 5 m, we made use of a 'FLYGT' pump attached to the end of a reinforced hose, as described under '2.2 Biota'. 100-150 L seawater was collected for both  $^{137}\text{Cs}$ - and  $^{99}\text{Tc}$ -analyses.



*Fig. 2.2 (a) Collection of sediment samples using a 'Smøgen' boxcorer*



*Fig. 2.2 (b) Collection of large volumes of seawater using a 'FLYGT' pump attached to the end of a reinforced hose*

### 3. ANALYSES

Radioactivity is the property of spontaneous disintegration (decay) of atomic nuclei accompanied by the emission of ionizing radiation. The three major forms of ionizing radiation emitted during radioactive decay are *alpha particles* (charged helium nuclei), *beta particles* (electrons) and *gamma rays* (photons or electromagnetic radiation). At IMR, the analyses of anthropogenic radionuclides are mainly restricted to the gamma emitters  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ . A brief introduction to the analytical method used at IMR, gamma ray spectroscopy using an HPGe detector, is given below. Analyses of alpha/beta emitting radionuclides have been performed during stays at other laboratories or purchased, and the analytical methods will not be described in detail.

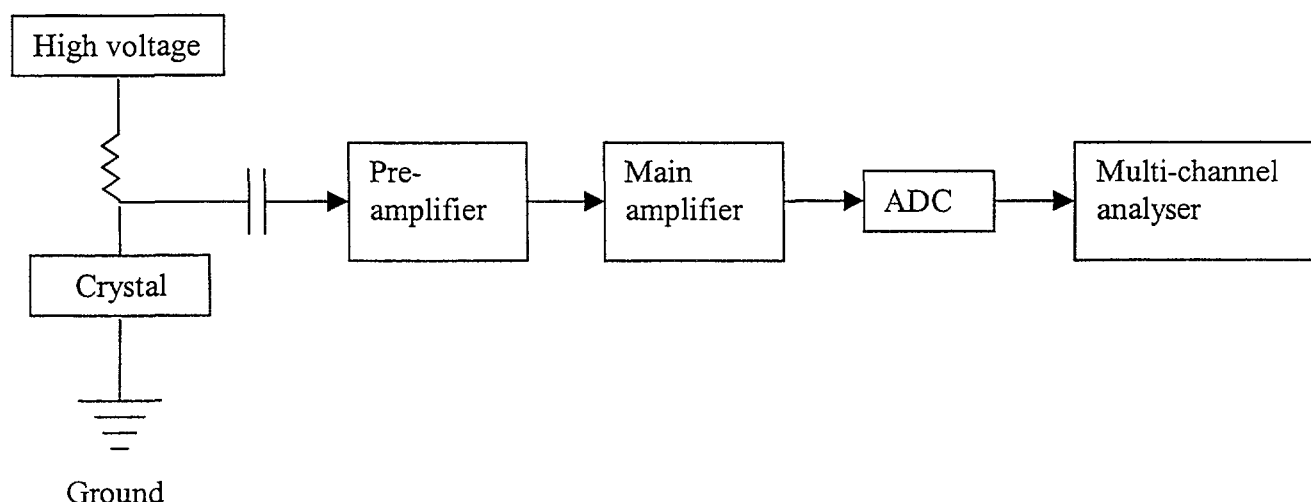
### 3.1 Gamma ray spectroscopy using an HPGe detector

Gamma rays are chargeless particles and do not cause ionisation directly. There are, however, three processes in which gamma rays interact with electrons, and when these occur inside the detector material, they allow us to infer the energy of the gamma rays. These three processes are *photoelectric absorption*, *Compton scattering* and *pair production*. In each of these processes an ionizing particle, usually an electron, is produced which can subsequently be detected. HPGe (High Purity Germanium) detectors are semiconductors. When an ionizing particle enters the detector material, it loses its energy by exciting electrons in the atoms of the germanium crystal from the valence band across the band gap to the conduction band. These excited electrons soon drop back to the valence band and emit a photon in the process. Since a typical ionizing particle causes many such excitations before it has lost its energy, many photons are emitted. Thus the energy of the particle has been transformed to a small pulse of light, which is subsequently transformed to a small charge pulse that is collected and integrated in a pre-amplifier (Fig. 3.1). With the aid of an applied bias voltage, the pre-amplifier produces an output voltage pulse that is proportional to the input charge (and hence the energy of the photon). A main amplifier shapes and amplifies the pulse arising from the pre-amplifier for optimum analogue to digital conversion. An analogue to digital converter (ADC) converts the analogue output signal from the main amplifier into a digitised form suitable for routing into the spectral analyser (MCA (Multi-channel Analyser)). The output from the MCA is an energy spectrum, where the different gamma emitting radionuclides can be identified by the energy of their peaks. The energy of gamma rays varies from 0.1-1.5 MeV. For example,  $^{137}\text{Cs}$  emits gamma radiation with an energy of 0.662 MeV<sup>1</sup>. The intensity of the peak is proportional to the activity of the radionuclide.

Both the detector and sample being gamma counted are shielded by pre-nuclear age lead in order to prevent cosmic and/or terrestrial background radiation to reach the detector. The detector is cooled in order to reduce the number of electrons in the conduction band, thereby reducing the background current and make it much easier to detect the extra excitations due to the gamma ray excitations.

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<sup>1</sup> The gamma radiation released is actually a property of  $^{137}\text{Ba}$ .  $^{137}\text{Cs}$  is, strictly speaking, a beta emitter. 93.5 % of the beta decays from  $^{137}\text{Cs}$  go to an excited nuclear state of  $^{137}\text{Ba}$  while 6.5 % go directly to the ground state of  $^{137}\text{Ba}$ . The excited state of  $^{137}\text{Ba}$  emits gamma radiation with energy of 0.662 MeV while it de-excites to the ground state of  $^{137}\text{Ba}$ . This energy is conventionally regarded as the ' $^{137}\text{Cs}$  gamma' energy.



*Figure 3.1 Schematic diagram of a typical gamma spectrometer*

### 3.2 $^{137}\text{Cs}$ in sediments (IMR and IFE)

After the sediment samples were brought back to IMR, the sediment cores were cut into slices of 1 cm thickness. The 7 upper and 3 lowest slices were subject of further sample preparation. The core slices and surface samples were thereafter freeze-dried, homogenized and packed in polyethylene counting beakers of known geometry prior to gamma counting.

At IMR, the gamma counting for  $^{137}\text{Cs}$  was performed in a low-background laboratory using a Canberra HPGe detector with 30 % efficiency and an extra low-background Ortec HPGe detector with 60 % efficiency. Both detectors are cooled to  $< -170^{\circ}\text{C}$  by electric cryostat cooling systems (helium filled refrigeration systems) and have 12 and 15 cm lead shielding, respectively.

At the Institute for Energy Technology (IFE), the gamma counting for  $^{137}\text{Cs}$  was performed using Canberra low-level detectors with relative efficiencies of 30-50 % and LN-cryotank cooling systems.

The counting times varied from 10 to 72 hours and the detection limits were  $\sim 0.5 \text{ Bq kg}^{-1}$  dry weight.

### 3.3 $^{137}\text{Cs}$ in biota (IMR and IFE)

The phytoplankton sample was evaporated to 0.5 L at IMR, and transferred to a Marinelli polyethylene counting beaker prior to gamma counting. Other biota samples were subsequently ground, freeze-dried, homogenised and filled into counting beakers of

appropriate size prior to gamma counting. The dry weights of zooplankton- and fish-samples varied from 12-60 g and 60-220 g, respectively. Approximately 500-750 g of muscle were obtained from each harbour porpoise and the dry weights of the samples varied from 60-230 g. Skin, bone and entrails were removed from large fish prior to grinding, while fry and juvenile fish were ground whole. The gamma counting was performed as described above for  $^{137}\text{Cs}$  in sediments. The counting times varied from 24 to 72 hours and the detection limits were generally in the range of 0.3-0.5 Bq kg<sup>-1</sup> wet weight.

### **3.4 $^{137}\text{Cs}$ in seawater (CEFAS)**

Caesium-137 was extracted from seawater by pumping the samples, already acidified to pH ~ 2 with nitric acid, through cartridges (24 ml plastic syringes) containing the Cs-sorbent ASG (ammonium-dodeca-molybdophosphate) on silica gel. The cartridges were sent to the Centre for Environment, Fisheries & Aquaculture Sciences (CEFAS), Lowestoft, UK, where they were directly gamma counted on NaI well detectors by CEFAS personnel. Details of the method and its efficiency are described by Baker (1975) with Steele (1989) reporting quality assurance procedures.

### **3.5 $^{99}\text{Tc}$ in seawater (CEFAS)**

$^{99}\text{Tc}$  is a pure beta emitter ( $E_{\text{max}} = 292 \text{ keV}$ ) decaying from  $^{99}\text{Mo}$ , and a radiochemical separation of this radionuclide is required for quantitative analysis. The detailed analytical method adopted is described by Harvey et al. (1992). Briefly, after adding rhenium in the form of  $\text{KReO}_4^-$  as a yield monitor, a preliminary extraction of  $^{99}\text{Tc}$  (and Re) based on anion-exchange separation was performed. After iron hydroxide scavenging,  $^{99}\text{Tc}$  and Re were further extracted by a second anion-exchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts were isolated.  $^{99}\text{Tc}$  was beta counted, and the yield of the rhenium tetraphenyl arsonium salt was determined gravimetrically. The samples were not filtered prior to analysis, as they were collected in waters with low concentrations of suspended materials. All the chemical treatment and beta counting were carried out at CEFAS by CEFAS personnel, with the exception that the first radiochemical separation step (addition of yield monitor and preliminary anion-exchange) in June/July 2000 was performed aboard R/V 'Johan Hjort' during the cruise. The analytical method has been quality assured through



internal accreditation (UKAS-NAMAS) and international intercomparison exercises (Dahlgard et al., 1995b).

### 3.6 $^{238}\text{Pu}$ , $^{239,240}\text{Pu}$ and $^{241}\text{Am}$ in sediments (CEFAS)

$^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  are often produced when heavy atomic nuclei (e.g. uranium) are bombarded with light atomic nuclei or neutrons. For example,  $^{239}\text{Pu}$  is produced when  $^{238}\text{U}$  captures a neutron and emits two beta particles. The analytical method used in the present study does not distinguish between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ; they are therefore determined together and quoted as  $^{239,240}\text{Pu}$ . A separation of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  can only be achieved using mass-spectrometric techniques.

$^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  are alpha emitters, and as for beta emitters, a radiochemical separation is required. During a stay at CEFAS in November-December 1999, eleven surface samples were analysed for  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  by alpha spectrometry. A detailed description of the method, which is somewhat complex, is given by Lovett et al. (1990). Briefly, the radionuclides were coprecipitated with iron hydroxide, extracted radiochemically using ion-exchange techniques, electro-deposited onto polished stainless steel discs and alpha counted on silicon surface barrier detectors.

### 3.7 Dating of sediments by the $^{210}\text{Pb}/^{210}\text{Po}$ method (Risø National Laboratory)

$^{210}\text{Pb}$  enters seawater by removal from the atmosphere by rain and by decay of  $^{226}\text{Ra}$  within the water. It is rapidly scavenged by particles in seawater, and is therefore useful in the determination of scavenging rates. The activity of  $^{210}\text{Pb}$  at some depth  $h$  below the water-sediment interface is related to the time elapsed since deposition, provided the initial activity has remained constant (Faure 1986). If the rate of sedimentation is also constant, then the activity of  $^{210}\text{Pb}$  is linearly related to  $h$ :

$$\ln ^{210}\text{Pb}_A = \ln ^{210}\text{Pb}_{A0} - ((\lambda h) / a)$$

where  $\lambda$  is the decay constant of  $^{210}\text{Pb}$  and  $a$  is the annual rate of accumulation. A complication arises because of the presence of a small amount of  $^{210}\text{Pb}$  that is supported by decay of  $^{226}\text{Ra}$ . This component can be identified by analysing sediment in which the unsupported  $^{210}\text{Pb}$  has already decayed (e.g. deep cores).

$^{210}\text{Po}$  ( $t_{1/2} = 138$  d) is a daughter of  $^{210}\text{Pb}$  ( $t_{1/2} = 22.4$  y), and they are both daughters of the primordial radionuclide  $^{238}\text{U}$ . In the context of sediment dating,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are assumed to be in secular equilibrium. The activity of  $^{210}\text{Pb}$  in sediments may therefore indirectly be determined by analysis of  $^{210}\text{Po}$ . In this thesis,  $^{210}\text{Po}$  was analysed in three sediment cores, eight slices from each, by alpha spectrometry at Risø National Laboratory (DK) by Risø personnel. Unsupported  $^{210}\text{Pb}$  was determined by subtracting the background level (an average of the  $^{210}\text{Po}$  concentration in the two deepest layers) from the total level of  $^{210}\text{Po}$ . Sedimentation rates were estimated using an exponentially curve fit based on the determinations of unsupported  $^{210}\text{Pb}$  in the core slices.

### 3.8 Uptake studies of $^{137}\text{Cs}$ and $^{57}\text{Co}$ in marine phytoplankton (MSRC)

Pilot experiments of the uptake of  $^{134}\text{Cs}$  in *Isochrysis galbana* and *Phaeocystis pouchetii* took place during 1998. Phytoplankton cultures were grown at the Department of Microbiology and Department of Fisheries and Marine Biology at the University of Bergen. The cultures were brought to IMR where they were exposed to  $^{134}\text{Cs}$ , filtered and gamma counted after certain time intervals. The method used was, however, incomplete and the results were not consistent.

Uptake experiments were resumed during a stay at Marine Sciences Research Center (MSRC), State University of New York, Stony Brook, USA, in March-May 1999. The uptake of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  were studied in *Emiliania huxleyi*, *Isochrysis galbana*, *Phaeocystis globosa*, *Skeletonema costatum* and *Thalassiosira pseudonana*. To determine the bioaccumulation of the radionuclides, experimental protocols described by Fisher et al. (1983) were generally followed. Two experiments were performed under identical experimental conditions, one on non-growing cells and one on growing cells. Growing cells were exposed to cyclic light (L:D 14:10) provided by cool-white fluorescent lamps ( $170 \mu\text{E m}^{-2}\text{s}^{-1}$ ). The non-growing cells were kept in darkness to prevent growth. Both non-growing and growing cells were maintained without shaking at  $12^\circ \pm 1^\circ\text{C}$ . In both experiments, growth was checked by counting cells microscopically with a hemacytometer. Radioisotope additions were added, via Eppendorf micropipette, as micro litre quantities of 0.1 N HCl ( $^{57}\text{Co}$ ) and 1.0 N HCl ( $^{137}\text{Cs}$ ). The pH in the seawater solutions was unaffected by the additions. The concentrations of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  in the experimental solution were about 300 and 40 Bq  $\text{ml}^{-1}$ , respectively. Over a six-day period (non-growing cells) and ten-day period (growing cells), cells were periodically filtered onto Nuclepore polycarbonate membranes and the fraction of radioactivity associated with

cells was determined for each isotope at each sample time (Fisher et al. 1983). Blanks that contained only media and isotopes were treated in the same manner and corrections were made for isotope sorption to the filters. Gamma activities of the two radionuclides was measured using a large-well NaI(Tl) detector with automatic sample changer (LKB Compugamma). The counting time was 5 minutes each sample. Propagated counting errors for all samples were < 5 %.

#### 4. SUMMARY OF MAIN RESULTS

##### 4.1 Distribution of $^{137}\text{Cs}$ , $^{238}\text{Pu}$ , $^{239,240}\text{Pu}$ and $^{241}\text{Am}$ in marine sediments with emphasis on the Spitsbergen-Bear Island area (*Paper I*)

The concentrations of  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in sediments near Spitsbergen reached over 50 % higher levels than in adjacent areas. The results of Føyen and Sværen (1997), indicating elevated levels of  $^{137}\text{Cs}$  in this area, are thereby confirmed. Their suggestion of phytoplankton as a source of  $^{137}\text{Cs}$  to marine sediments is, however, questionable, as it is concluded in *Paper II* that the uptake of  $^{137}\text{Cs}$  in phytoplankton is low. In the present paper, other sources for  $^{137}\text{Cs}$  in sediments near Spitsbergen, and the geographical origin of the sediments, are briefly discussed.

The plutonium activity ratios ( $^{238}\text{Pu}$ :  $^{239,240}\text{Pu}$ ) ( $\sim 0.03$ ) found in sediments south of Spitsbergen were consistent with that of global fallout. Based on this, we could exclude the European reprocessing industry as a contamination source. However, we could not distinguish between the following origins of sediments: 1) particles washed out from Spitsbergen, or 2) particles originating from Russian rivers draining into the Barents and Kara Seas and transported to the Spitsbergen area via sea ice; as both these areas are contaminated by global fallout. The plutonium activity ratios found in two samples collected in the deep Norwegian Sea were above 0.1, indicating that these locations are contaminated with plutonium from the reprocessing industry. However, these samples contained low plutonium concentrations and large errors, which might have biased the results.

Using an exponential curve fit based on determinations of unsupported  $^{210}\text{Pb}$  in sediment cores, sedimentation rates of  $0.28 \text{ cm y}^{-1}$ ,  $0.05 \text{ cm y}^{-1}$  and  $0.26 \text{ cm y}^{-1}$  were calculated at locations south of Spitsbergen, near the Greenland ice-edge and in the deep Norwegian Sea, respectively. Sedimentation rates are expected to be lower in the deep Norwegian Sea than in the shallow, highly productive area close to Spitsbergen. The results

showing similar sedimentation rates at these sampling locations were therefore unexpected. The results are, however, supported by the vertical distributions of  $^{137}\text{Cs}$ . The  $^{137}\text{Cs}$ -maximum at 2-4 cm depth at both locations corresponds with deposition from the Chernobyl accident. The lower sedimentation rate near the Greenland ice-edge is probably due to ice cover and resulting low biological production large portions of the year.

#### 4.2 Uptake and accumulation of $^{137}\text{Cs}$ in marine food webs (*Paper II, III and IV*)

Quantification of  $^{137}\text{Cs}$  in phytoplankton collected in the field was not successful, probably due to a combination of low phytoplankton concentrations in the collected amount of seawater (1000 L) and low  $^{137}\text{Cs}$  concentrations in phytoplankton. A larger and more powerful filtration system than the one used in this work is needed in order to collect enough sample material (preferably 100-200 g dry weight or 500-1000 g wet weight).

Bioaccumulation of  $^{137}\text{Cs}$ , and for comparison: cobalt-57 ( $^{57}\text{Co}$ ), was, as an alternative, studied in phytoplankton representative for the Barents and Norwegian Seas phytoplankton communities in the laboratory (*Paper II*). The results showed that  $^{57}\text{Co}$  was systematically taken up by all the species investigated (Fig. 4.1 a and b). The uptake of  $^{137}\text{Cs}$  was, however, generally much lower, and the uptake pattern seemed unsystematic (Fig. 4.1 c and d).

The bioaccumulation of  $^{137}\text{Cs}$  in species belonging to higher trophic levels of the Barents and Norwegian Seas food webs was studied in *Paper III* (Figure 4.2). The concentrations of  $^{137}\text{Cs}$  were low in all the investigated species. The mean concentration of  $^{137}\text{Cs}$  found in seawater  $3.0 \text{ Bq m}^{-3}$  ( $0.003 \text{ Bq L}^{-1}$ ) was used to calculate Concentration Factors ( $\text{CF} = \text{Bq kg}^{-1} \text{ wet tissue} / \text{Bq L}^{-1} \text{ seawater}$ ). The CFs increased markedly from low to high trophic levels (Figure 4.3). The CFs varied within each trophic level, probably as a result of different feeding habits within each trophic level.

It was concluded in *Paper III* that the concentrations of  $^{137}\text{Cs}$  in marine organisms in the Barents and Norwegian Seas were generally low. In *Paper IV*, variations in the  $^{137}\text{Cs}$  concentrations in harbour porpoises (representing the upper level of the food web in *Paper III*) along the Norwegian coast were studied.

The concentrations of  $^{137}\text{Cs}$  in seawater are known to differ along the coast of Norway due to distance from point sources, such as Sellafield and the outflow from the Baltic Sea (Herrmann et al. 1995; Kershaw et al. 1997). We hypothesised that if harbour porpoises are philopatric (resident in an area), their muscle tissue would reflect the concentrations of  $^{137}\text{Cs}$

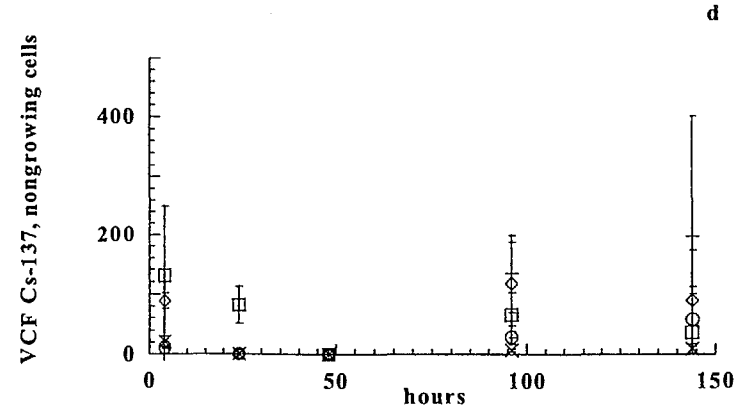
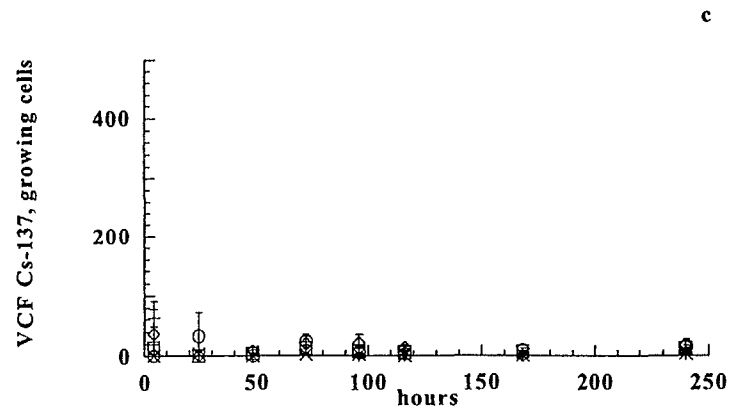
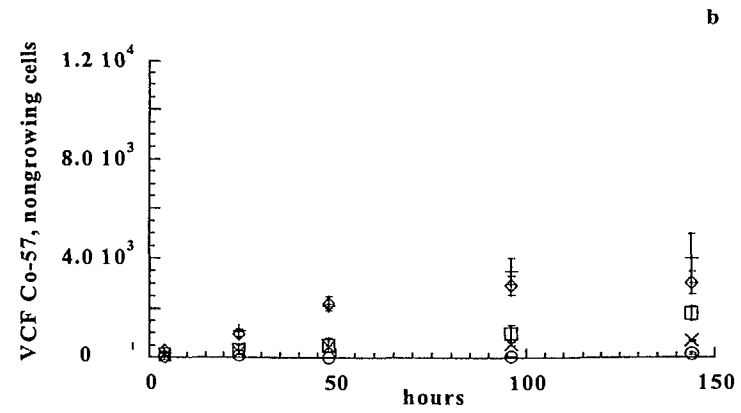
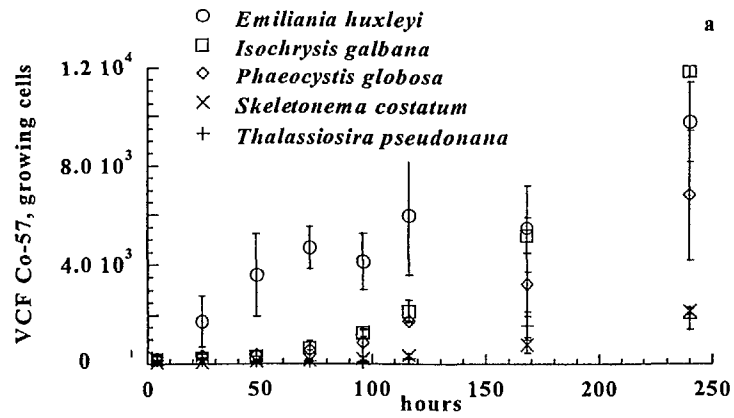


Figure 4.1. (a) VCF  $^{57}\text{Co}$  (Volume Concentration Factors, radioactivity  $\mu\text{m}^{-3}$  cell divided by radioactivity dissolved  $\mu\text{m}^{-3}$  seawater), growing cells; (b) VCF  $^{57}\text{Co}$  non-growing cells; (c) VCF  $^{137}\text{Cs}$ , growing cells; (d) VCF  $^{137}\text{Cs}$ , non-growing cells.

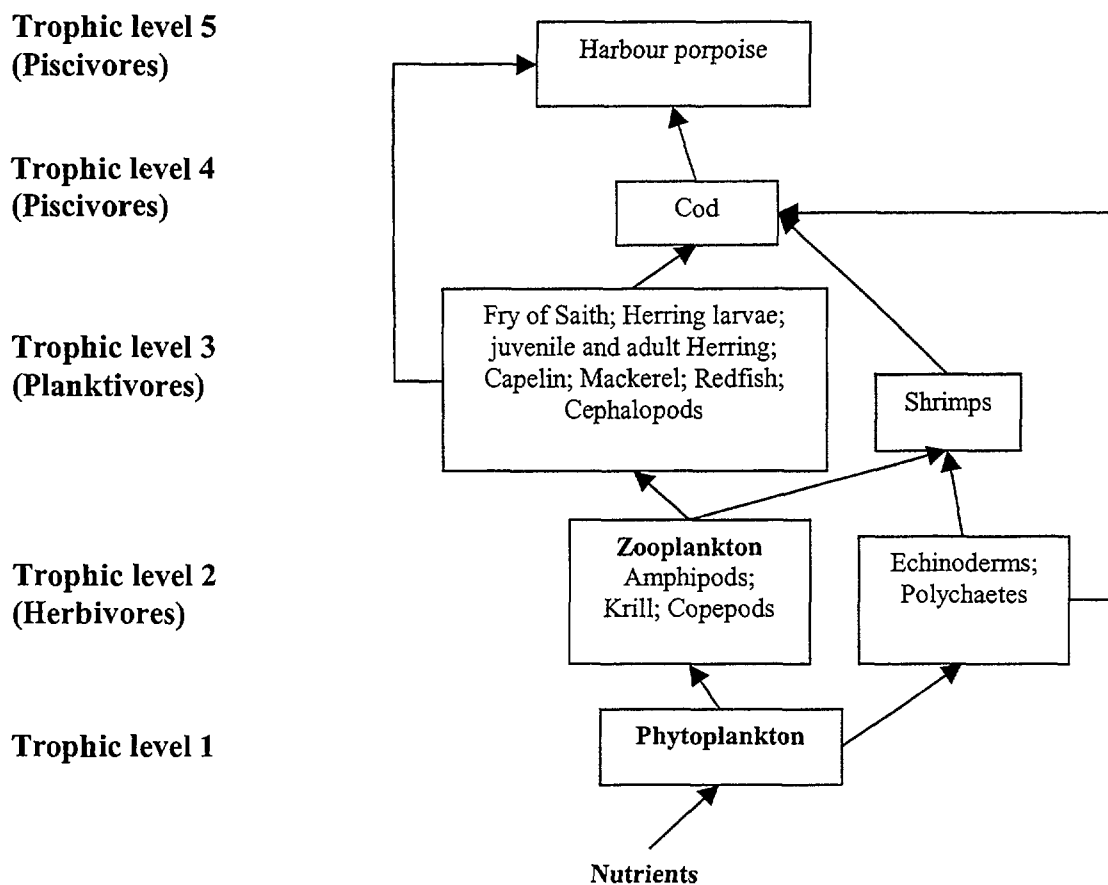


Figure 4.2. Investigated species assigned to different trophic levels.

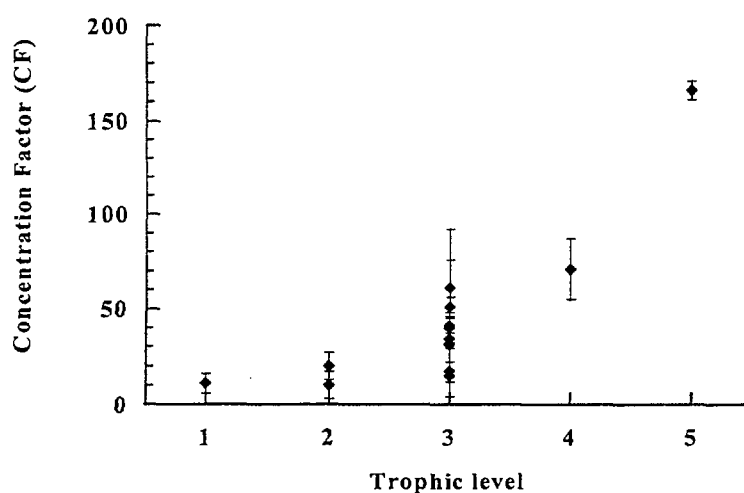


Figure 4.3. Concentration Factors ( $CF = Bq\ kg^{-1}\ wet\ tissue / Bq\ L^{-1}\ seawater$ ) for various trophic levels of the Barents and Norwegian Seas food webs. The CFs were calculated individually for each biotic sample. The CFs shown are mean values.

in their area of residence. Alternatively, if harbour porpoise movements are not restricted along the coast of Norway, levels of  $^{137}\text{Cs}$  in muscle tissue should not show a high correspondence to geographic location.

The  $^{137}\text{Cs}$  levels found in harbour porpoise muscle were consistent with that found in the area from which they were caught. Porpoises from the Barents Sea had lower levels than porpoises from the west and south coasts of Norway, reflecting concentration patterns of  $^{137}\text{Cs}$  in the waters of those areas (Fig. 4.4). This suggests ecological separation of harbour porpoises in these regions, and that porpoises are limited in their movements along the coast of Norway.

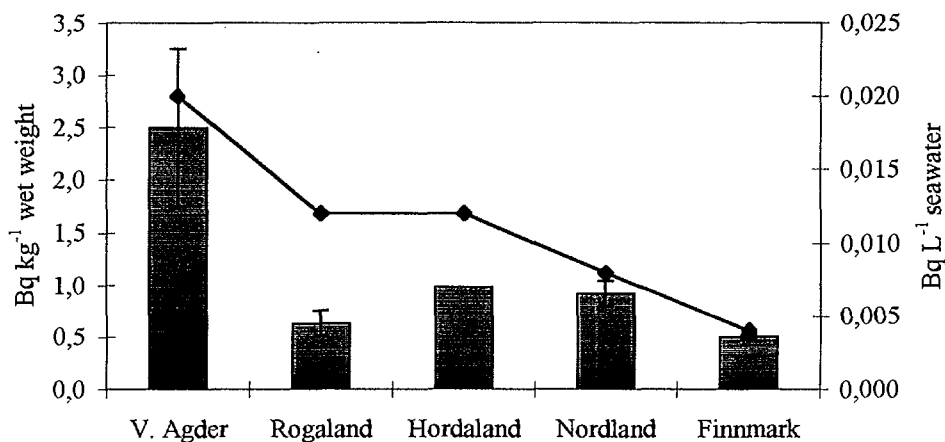
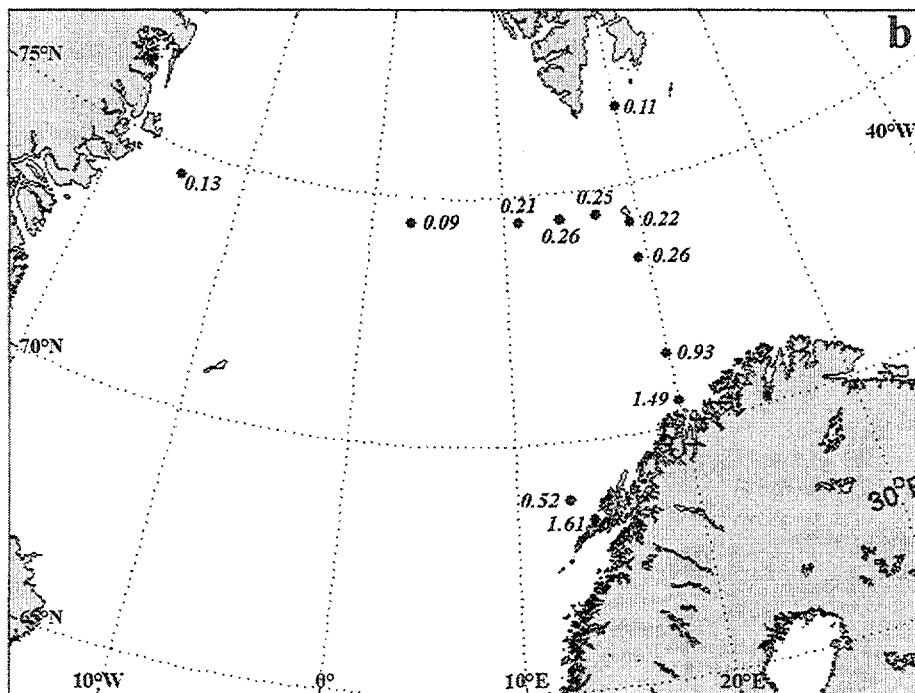
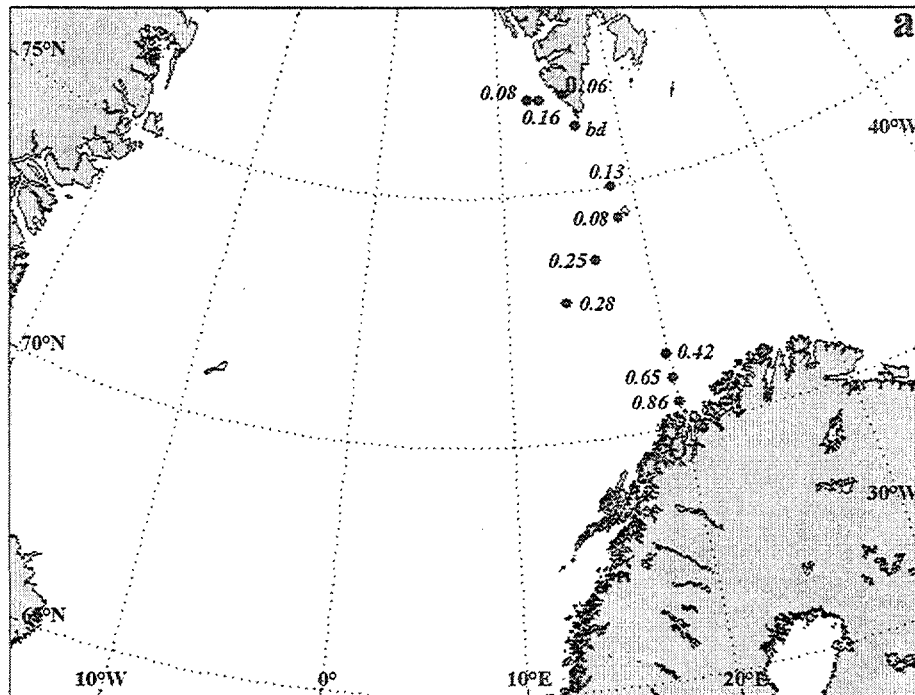


Figure 4.4. Mean levels and standard errors of  $^{137}\text{Cs}$  (Bq kg<sup>-1</sup> ww) in harbour porpoises from five locations in Norway (bars). Median concentrations of  $^{137}\text{Cs}$  in seawater within each area are shown by the solid line.

#### 4.3 Transport of $^{99}\text{Tc}$ from Sellafield to Arctic environments (*Paper V*)

Radionuclides are excellent tracers for e.g. transport of ocean currents. The advantage with radionuclides as tracers is that we are able to measure extremely low concentrations. For example, the total time-integrated (2000) input from Sellafield of  $4.1 \cdot 10^{16}$  Bq of  $^{137}\text{Cs}$  over 47 years corresponds to a mass of 12.8 kg only (Woodhead 2000).  $^{137}\text{Cs}$  has been used as an environmental tracer for a range of environmental processes. This emphasises how sensitive (and, therefore, effective) radionuclides are as environmental tracers. In *Paper V*,  $^{99}\text{Tc}$  is used as a tracer for transport of ocean currents.





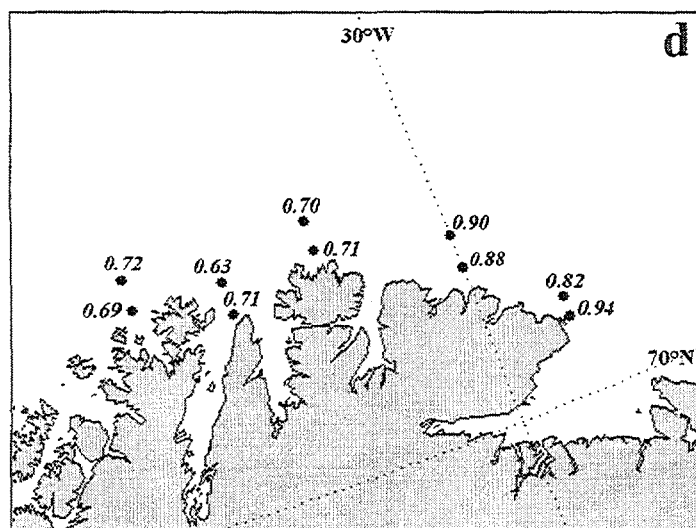
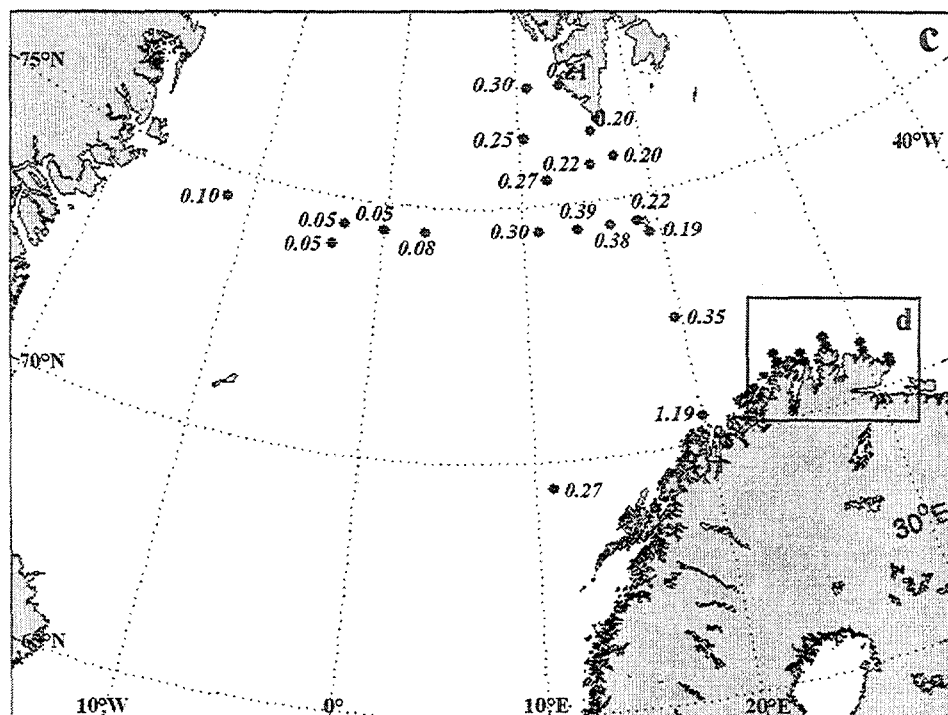


Figure 4.5. Spatial distribution of  $^{99}\text{Tc}$  in seawater ( $\text{Bq m}^{-3}$ ), in (a) 1998; (b) 1999; (c) 2000; (d) 2000

In April 1994, a new waste treatment plant, the Enhanced Actinide Removal Plant (EARP), began operation at Sellafield. EARP was introduced primarily to reduce the discharges of plutonium and americium, and does not remove  $^{99}\text{Tc}$  from the effluents. The discharges of  $^{99}\text{Tc}$  from Sellafield into the Irish Sea have therefore increased significantly since 1994. In *Paper V*, the concentrations of  $^{99}\text{Tc}$  have been measured in seawater in the Norwegian Coastal Current (NWCC) and the West Spitsbergen Current (WSC) in 1998, 1999 and 2000 (Figure 4.5 a-d). The study shows that the ' $^{99}\text{Tc}$ -front' had passed southwest of Bear Island in the WSC in July 1998,  $\sim 4$  years post-EARP (concentrations  $\sim 0.3 \text{ Bq m}^{-3}$ ,  $\sim 6$  times background levels), indicating a transport time to this location of 3-4 years. The ' $^{99}\text{Tc}$ -front' passed west of Spitsbergen in the WSC between July 1998 and May/June 2000 (concentrations  $\sim 0.3 \text{ Bq m}^{-3}$ ), demonstrating a transport time to this location of 4-6 years.

A Transfer Factor is the relationship between the environmental concentration at a given remote location and the average quantity discharged  $t$  years earlier, where  $t$  is the mean transport time to the location. The units for radionuclide TFs are usually given as  $\text{Bq m}^{-3}/\text{PBq a}^{-1}$ . A TF of 4 was calculated for a sample collected near Fugløy in 1998. The transport time from Sellafield to Fugløy is estimated to 3 years. This means, for example, that a  $^{99}\text{Tc}$  discharge from Sellafield of 200 TBq will give a seawater concentration near Fugløy of approximately  $0.8 \text{ Bq m}^{-3}$  three years later.

Based on data indicating that the ' $^{99}\text{Tc}$ -front' reached Utsira in January 1997 (Christensen et al. 2001) and Ingøy in August 1997 (G. Christensen, pers. comm.), we roughly calculated the mean propagation speed of  $^{99}\text{Tc}$ : Following the NWCC close to the coast from Utsira to Ingøy gives an average speed of  $7 \text{ cm s}^{-1}$  when a transport time of 7 months was used.

## 5. SUGGESTIONS FOR FURTHER WORK

- 1) A task for the future will be to develop a method (e.g. a large capacity filtering system) for collecting phytoplankton samples of such a size that proper measurements of various important radionuclides can be conducted. Such measurements are needed in order to determine the role of phytoplankton in radionuclide flux to deeper waters and accumulation in marine food webs.
- 2) It is shown in this thesis that  $^{137}\text{Cs}$  is accumulated in food webs in the Norwegian and Barents Seas. It is assumed that  $^{137}\text{Cs}$  is taken up both directly from seawater and via dietary sources, but the exact mechanisms for uptake and transport of  $^{137}\text{Cs}$  in marine food

webs are not well known. Increased knowledge of the mechanisms for uptake of  $^{137}\text{Cs}$  could be achieved through controlled laboratory experiments.

- 3) The increased releases of  $^{99}\text{Tc}$  from Sellafield have caused public concerns in Norway. Little is known about uptake of  $^{99}\text{Tc}$  in marine organisms. Very high uptakes have, however, been found in benthic organisms as lobsters and seaweed. A main task for the future will be to study the bioaccumulation of  $^{99}\text{Tc}$  in for example:

- Various benthic organisms along the Norwegian coast
- Species as wolf-fish (*Anarhichas* sp.), haddock (*Melanogrammus aeglefinus*), halibut (*Hippoglossus hippoglossus*) and plaice (*Pleuronectes platessa*), species often consumed by people, which have benthic organisms as their main diet
- Commercially important pelagic species

## 6. REFERENCES

- Aarkrog, A., 1994. Radioactivity in Polar Regions – Main Sources. *Journal of Environmental Radioactivity* 25, 21-35.
- Aarkrog, A., 1997. Radioactive contamination of the marine environment. *Ciba Foundation Symposia* 203, 68-70.
- Aarkrog, A., Dahlgard, H., Nilsson, K., Holm, E., 1984. Further studies of plutonium and americium at Thule, Greenland. *Health Physics* 46, 29-44.
- AMAP, 1998. AMAP Assessment Report: Arctic Pollution Issues. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway, pp. xii + 859.
- Amundsen, I., Lind, B., Reistad, O., Gussgaard, K., Iosjpe, M., Sickel, M., 2001. The Kursk Accident. NRPA report 2001:5. NRPA, Østerås, Norway.
- Baker, C.W., 1975. The determination of radiocaesium in sea and freshwater. Fisheries Research Technical Report, No. 16, MAFF Directorate of Fisheries Research, Lowestoft, UK, pp. 7.
- Balonov, M., Tsaturov, Y., Howard, B., Strand, P., 1999. Radioactive contamination of the Russian Arctic. Report of Russian Experts for AMAP. AMAP Data Centre for Radioactivity Report 1999:1. Østerås, Norway.

- Bretten, S., Gaare, E., Skogland, T., Steinnes, E., 1993. Investigations of radiocaesium in alpine ecosystems in Norway following the Chernobyl accident. In: Strand, P., Holm, E. (Eds.), *Environmental Radioactivity in the Arctic and Antarctic. Proceedings of the International Conference on Environmental Radioactivity in the Arctic and Antarctic*, Kirkenes, Norway, August 1993, pp. 432.
- Brungot, A.L., Føyn, L., Carroll, J.L., Kolstad, A.K., Brown, J., Rudjord, A.L., Bøe, B., Hellstrøm, T., 1999. Radioactive contamination in the marine environment. Report no. 3 from the National Surveillance Programme, NRPA Report 1999:6. NRPA, Østerås, Norway.
- Christensen, G.C., Strålberg, E., Holm, E., 2001. Time trends of anthropogenic radionuclides at Utsira. The Eight Nordic Seminar on Radioecology, 25.-28. February 2001, Rovaniemi, Finland.
- Dahlgaard, H., Chen, Q., Herrmann, J., Nies, H., Ibbett, R.D., Kershaw, P.J., 1995a. On the background level of  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the North Atlantic. *Journal of Marine Systems* 6, 571-578.
- Dahlgaard, H., Herrmann, J., van Weers, A.W., Masson, M., Ibbett, R.D., Chen, Q., 1995b. Quality assurance of the oceanographic tracers Technetium-99 and Antimony-125: Intercomparisons and recovery tests. *Journal of Marine Systems* 6, 391-396.
- Faure, G., 1986. *Principles of Isotope Geology*, 2nd edition. John Wiley & Sons, New York, pp. 589.
- Fisher, N.S., Bjerregaard, P., Fowler, S.W., 1983. Interactions of marine plankton with transuranic elements. I. Biokinetics of neptunium, plutonium, americium, and californium in phytoplankton. *Limnology and Oceanography* 28, 432-447.
- Føyn L., 1994. To move or not to move. In: Sætre, R. (Ed.), *The sunken nuclear submarine in the Norwegian Sea – a potential environmental problem? Fisker og havet No. 7*, Institute of Marine Research, Norway, pp. 5-12.
- Føyn, L., 2001. Radioactive wastes. In: Steele, J. H., Thorpe, S. A., Turekian, K. K. (Eds.), *Encyclopedia of Ocean Sciences*, Academic press.
- Føyn, L., Nikitin, A., 1994. The joint Norwegian/Russian Expedition to the dump sites for radioactive waste in the Abrosimov fjord and the Stepovogo fjord. August-September 1994. Report from the expedition on board R/V 'Viktor Buinitskiy', pp. 18.
- Føyn, L., Sværen I., 1997. Distribution and sedimentation of radionuclides in the Barents Sea. *ICES Journal of Marine Science* 54, 333-340.

- Føyn, L., Heldal, H.E., Sværen, I., 1999. The Barents Sea, Distribution and Fate of Radioactive Contaminants. Proceedings, International Symposium on Marine Pollution, Monaco, 5 - 9 October 1998. IAEA-TECDOC-1094, pp. 471 - 474. International Atomic Energy Agency, July 1999.
- Gray, J., Jones, S.R., Smith, A.D., 1995. Discharges to the environment from the Sellafield Site, 1951-1992. *Journal of Radiological Protection* 15 (2), 99-131.
- Gunnerød, T.B., Blakan, I., Ugedal, O., Skogland, K., 1989. Freshwater and alpine ecosystem response to Chernobyl fallout in Norway. In: *Environmental Contamination Following a Major Nuclear Accident*. IAEA-SM-306/41, International Atomic Energy Authority, Vienna, pp. 59-67.
- Harvey, B.R., Williams, K.J., Lovett, M.B., Ibbett, R.D., 1992. Determination of technetium-99 in environmental materials with rhenium as a yield monitor. *Journal of Radioanalytical and Nuclear Chemistry* 158, 417-436.
- Hassel, A., Skjoldal, H.R., Gjøsæter, H., Loeng, H., Omli, L., 1991. Impact of grazing from capelin (*Mallotus villosus*) on phytoplankton: a case study in the Northern Barents Sea in 1985. In: Sakshaug, E., Hopkins, C. C. E., Øritsland, N. A. (Eds.), *Proceedings of the Pro Mare Symposium on Polar Marine Ecology*, 12-16 May 1990. *Polar Research* 10, 371-388.
- Herrmann, J., Kershaw, P.J., Bailly de Bois, P., Guegueniat, P., 1995. The distribution of artificial radionuclides in the English Channel, southern North Sea, Skagerrak and Kattegat, 1990-1993. *Journal of Marine Systems* 6, 427-456.
- Håkanson, L., Andersson, T., 1992. Remedial measures against radioactive Caesium in Swedish lake fish after Chernobyl. *Aquatic Sciences* 54 (2), 141-164.
- Joseph, A.B, Gustavson, P.F., Russell, I.R., Schuert, E.A., Volchok, H.L., Tamplin, A., 1971. Sources of radioactivity and their characteristics. In: *Radioactivity in the Marine Environment*, pp. 6-41, National Academy of Sciences, Washington D. C.
- JNRE, 1994. Radioactive contamination at dumping sites for nuclear waste in the Kara Sea. Results from the Russian-Norwegian 1993 expedition to the Kara Sea. Joint Russian-Norwegian Expert Group for Investigations of Radioactive Contamination in the Northern Areas. NRPA, Østerås, Norway, pp. 82.
- JNRE, 1996. Dumping of radioactive waste and investigation of radioactive contamination of the Kara Sea. Results from 3 years of investigations (1992-1994) in the Kara Sea. Joint Norwegian-Russian Expert Group for Investigation of Radioactive Contamination in the Northern Areas. NRPA, Østerås, Norway, pp. 56.

- JNRE, 1997. Sources contributing to radioactive contamination of the Techa River and areas surrounding the “Mayak” production association, Urals, Russia. Programme of investigations of possible impacts of the “Mayak” PA activities on radioactive contamination of the Barents and Kara Seas. Joint Norwegian-Russian Expert Group for Investigation of Radioactive Contamination in the Northern Areas. NRPA, Østerås, Norway, pp.134.
- Kershaw, P.J., Baxter, A., 1995. The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep-Sea Research Part II* 42 (6), 1413-1448.
- Kershaw, P.J., Gurbutt, P., Woodhead, D., Leonard, K., Rees, J., 1997. Estimates of fluxes of <sup>137</sup>Cs in northern waters from recent measurements. *The Science of the Total Environment* 202, 211-223.
- Liland, A., Skuterud, L., Bergan, T., Forseth, T., Gaare, E., Hellstrøm, T., 2001. Overvåkning av radioaktiv forurensning i næringsmidler og det terrestre miljø 1986-1998 (in Norwegian). *StrålevernRapport 2001:1*. Statens Strålevern (NRPA), Østerås, Norway.
- Lovett, M.B., Boggis, S.J., Blowers, P., 1990. The determination of alpha-emitting nuclides of plutonium, americium and curium in environmental materials: Part 1. Sea water, *Aquatic Environment Protection: Analytical Methods No. 7*, MAFF, Directorate of Fisheries Research, Lowestoft, UK.
- MAFF & SEPA, 1999. Radioactivity in Food and the Environment, 1998 RIFE-4. Ministry of Agriculture, Fisheries and Food & Scottish Environment Protection Agency, London, pp. 176.
- Nies, H., Wedekind, Ch., 1988. The contamination of the North Sea and the Baltic Sea by the Chernobyl fallout. In: Guary, J.C, Guegueniat, P., Pentreath, R.J. (Eds.), *Radionuclides: A tool for oceanography*. Elsevier, London, pp. 227-239.
- OPRF, 1993. Facts and problems related to radioactive waste disposal in seas adjacent to the territory of the Russian Federation. In: Yablokov, A.V., Karasev, V.K., Rumyantsev, V.M., Kokeev, M.E., Petrov, O.J. (Eds.), *Materials for a report by the Government Commission on matters related to radioactive waste disposal at sea*. Created by Decree No. 613 of the Russian Federation President of October 24, 1994. White Book No. 3, Small World Publisher, Moscow.
- Rey, F., 1999. Havforskningsinstituttet, Interne Notat Nr. 04, 1999. Interne Toktrapporter Nr. 03, 1999 (IT 08/99) (Cruise Report). Institute of Marine Research, Bergen, Norway.
- Rey, F., 2001. Havforskningsinstituttet, Interne Notat Nr. 01, 2001. Interne Toktrapporter Nr. 01, 2001. (Cruise Report). Institute of Marine Research, Bergen, Norway.

- Rissanen, K., Ikäheimonen, T.K., Matishov, D., Matishov G.G., 1997. Radioactivity Levels in Fish, Benthic Fauna, Seals and Sea Birds Collected in the Northwest Arctic of Russia. Radioprotection – Colloques 32 (C2), 323-331.
- Sakshaug, E., Skjoldal, H. R., 1989. Life at the ice edge. *Ambio* 18, 60-67.
- Smith, J.N., Ellis, K.M., Polyak, L., Ivanov, G., Forman, S.L., Moran, S.B., 2000.  $^{239,240}\text{Pu}$  transport into the Arctic Ocean from underwater nuclear tests in Chernaya Bay, Novaya Zemlya. *Continental Shelf Research* 20, 255-279.
- Steele, A.K., 1989. Laboratory intercomparison of  $^{137}\text{Cs}$  in sea water. Fisheries Technical Note, No. RL1/89, MAFF, Directorate of Fisheries Research, Lowestoft, UK, pp. 25.
- Strand, P., Nikitin, A., Rudjord, A.L., Salbu, B., Christensen, G., Føyn, L., Kryshev, I.I., Chumichev, V.B., Dahlgard, H., Holm, E., 1994. Survey of Artificial Radionuclides in the Barents Sea and the Kara Sea. *Journal of Environmental Radioactivity* 25, 99-112.
- UNSCEAR, 1993. Sources and effects of ionizing radiation, United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR 1993 Report to the General Assembly, with Scientific Annexes, United Nations, New York.
- Woodhead, D., 2000. Whither marine radioactivity studies? *Journal of Environmental Radioactivity* 52, 1-3.

# **PAPER I**



Distribution of selected anthropogenic radionuclides ( $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$ ) in marine sediments with emphasis on the Spitsbergen-Bear Island area

Hilde Elise Heldal<sup>1\*</sup>, Per Varskog<sup>2</sup> and Lars Føyn<sup>1</sup>

<sup>1</sup>Marine Chemistry Division, Dept. of Marine Environment, Institute of Marine Research, P.O. Box 1870 Nordnes, N-5817 Bergen, Norway.

Telephone: +47 55 23 63 34, Fax: +47 55 23 85 84, Contact e-mail: [hilde.elise.heldal@imr.no](mailto:hilde.elise.heldal@imr.no)

<sup>2</sup>Institute for Energy Technology, P.O. Box 40, N-2007 Kjeller, Norway.

## Abstract

Measurements of anthropogenic radionuclides in marine sediment can provide good estimates of past and present radioactive contamination of the marine environment. The Barents Sea is one of the world's richest ocean areas, and it is therefore of major interest for Norwegian fisheries to document the levels of radioactive contamination of this and adjacent ocean areas. In this study, concentrations of  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  were measured in marine sediments collected mainly in the Spitsbergen-Bear Island region. Additional samples collected in the deep Norwegian Sea and near the Greenland ice-edge have been included for comparison. The highest radionuclide levels were found near Spitsbergen, reaching over 50 % higher than in adjacent areas. Determinations of the mineral content and particle size distribution indicate a relation between the content of clay minerals and radionuclide levels. The present study confirms earlier observations of elevated levels of  $^{137}\text{Cs}$  in sediments in the Spitsbergen-Bear Island area and documents elevated levels of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in these sediments. Using an exponential curve fit based on determinations of unsupported  $^{210}\text{Po}$  in sediment cores, sedimentation rates of  $0.28\text{ cm y}^{-1}$ ,  $0.05\text{ cm y}^{-1}$  and  $0.26\text{ cm y}^{-1}$  were calculated at locations south of Spitsbergen, near the Greenland ice-edge and in the deep Norwegian Sea, respectively.

**Keywords:** Caesium-137; Plutonium; Americium-241; Marine sediments; Spitsbergen

## 1. Introduction

There are several sources for radioactive contamination around Spitsbergen and in adjacent areas of the Barents Sea and Arctic Ocean. The most substantial include global fallout, fallout from the Chernobyl nuclear reactor accident and fallout from nuclear weapons testing near Novaya Zemlya by the former Soviet Union. In addition, there have been direct discharges from nuclear installations along the Russian rivers Ob and Yenisey, which drain into the Barents and Kara Seas, plus discharges from the nuclear reprocessing plants Sellafield (UK) and La Hague (France) (e.g. Aarkrog, 1994; Strand et al., 1994; Kershaw and Baxter, 1995; Salbu et al., 1997; Smith et al., 2000). Radioactive wastes dumped in the Barents and Kara Seas represent a potential contamination source. The Barents Sea, one of the world's richest ocean areas, is of major importance to Norwegian fisheries with yearly catches of 2-3.5 million tons. It is of great importance with respect to both local consumers and the export industry to document the levels of radioactive contamination in this and adjacent areas.

Certain areas of the Barents Sea may receive extra deposits of radioactive contamination, for example the seabed in ice-melting zones, where contaminated particles trapped in the ice may deposit during the ice melting. During spring and summer, a geographically fairly stable ice-melting zone arises along the polar front in the Spitsbergen area in the western Barents Sea (Loeng, 1991). Furthermore, the ice melting gives rise to a stratified and nutrient rich euphotic zone which supports high primary production (Sakshaug and Skjoldal, 1989). It has been suggested that living phytoplankton in the water column might actively act as scavenging elements and cause elevated levels of  $^{137}\text{Cs}$  in sediments in the Spitsbergen area (Føyn and Sværen, 1997).

The purpose of this study was to investigate the levels of  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in marine sediments with emphasis on the Spitsbergen-Bear Island area, and to characterise these sediments by means of their mineral compositions and particle size distributions. Five samples collected in the deeper Norwegian Sea and one sample collected near the Greenland ice-edge are included in the study for comparison. This study extends upon results from earlier surveys conducted by the Norwegian Institute of Marine Research (IMR) (1991-1993), where the levels of  $^{137}\text{Cs}$  in surface sediments in the Barents Sea and Spitsbergen area were mapped. Results from these surveys indicated elevated levels of  $^{137}\text{Cs}$  in surface sediments west of Spitsbergen and in the Spitsbergen-Bear Island (Føyn and Sværen, 1997).

## 2. Materials and methods

Sample collection took place during two cruises conducted by IMR: aboard RV ‘Michael Sars’ in July 1998 and aboard RV ‘Johan Hjørt’ in May/June 1999. Samples were collected in the Spitsbergen-Bear Island area ( $s = 17$ ,  $c = 3$ ), west of Spitsbergen ( $s = 4$ ,  $c = 0$ ), in the Norwegian Sea ( $s = 5$ ,  $c = 5$ ) and near the Greenland ice-edge ( $s = 1$ ,  $c = 1$ ) ( $s =$  surface sediments,  $c =$  sediment cores). Fig. 1 shows the study area and sampling locations. Table 1 shows sampling positions and water depths.

Both surface sediments and sediment cores were collected using a ‘Smøgen’ boxcorer (30 cm x 30 cm inner dimensions). Approximately 100 cm<sup>2</sup> of the upper 1 cm was collected for each surface sample. Cores were collected in open PVC tubes of 50 cm length and 10 cm inner diameter. All samples were kept frozen at -20°C until further preparation at a land-based laboratory took place.

Prior to analysis, the sediment cores were cut into slices of 1 cm thickness. The 7 upper and 3 lowest slices were subject of further sample preparation. Both core slices and surface samples were subsequently freeze-dried, homogenized and packed in containers of known geometry before the concentrations of <sup>137</sup>Cs were measured using high-resolution gamma spectrometry at IMR and Institute for Energy Technology (IFE). Typical counting times were 24 hours for the surface samples and 24 – 72 hours for the core layers. Eleven surface samples were analysed for <sup>238</sup>Pu, <sup>239,240</sup>Pu and <sup>241</sup>Am by  $\alpha$ -spectrometry at the Centre for Environment, Fisheries & Aquaculture Sciences (CEFAS), Lowestoft, UK. Briefly, the actinides (<sup>238</sup>Pu, <sup>239,240</sup>Pu and <sup>241</sup>Am) were extracted using ion-exchange techniques, electro-deposited onto polished stainless steel discs and alpha counted on silicon surface barrier detectors. A detailed description of the method is given by Lovett et al. (1990).

All surface samples were separated into fractions of silt/clay (particles < 63  $\mu$ m) and sand/gravel (particles > 63  $\mu$ m) by dry sieving in a Retsch analytical sieve shaker. This classification of sedimentary particles refers to the British Standards Institution (Brady, 1990). In addition, the particle size distribution in the silt/clay fraction was determined in 11 surface samples, using a Micromeritics SediGraph 5100 particle size analyser. The mineral content in these samples was determined qualitatively and semi quantitatively by X-ray diffraction (XRD) using a Philips PW 1700 X-ray diffractometer with a Cu X-ray tube. The particle size distribution in the silt/clay fraction and the sediment mineralogy were determined at the department of Geology, University of Bergen, Norway.

Analyses of  $^{210}\text{Po}$  in eight layers in three sediment cores were carried out by alpha spectrometry at Risø National Laboratory, Denmark. In the context of sediment dating,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  are assumed to be in secular equilibrium. Unsupported  $^{210}\text{Pb}$  was determined by subtracting the background level (an average of the  $^{210}\text{Po}$  concentration in the two deepest layers) from the total level of  $^{210}\text{Po}$ . Using each core layer's porosity (Equation 1), the thickness of layers below 1 cm was corrected for density compression (Equation 2). The corrected depth of each core layer was thereafter calculated using Equation 3. Sedimentation rates were finally estimated using an exponential curve fit based on the determinations of unsupported  $^{210}\text{Pb}$  in density corrected cores, according to Tadjiki and Erten (1994).

$$\text{Porosity (\%)} = \left[ \frac{\text{Mass (Sample (wet weight))} - \text{Mass (Sample (dry weight))}}{\text{Mass (Sample (wet weight))}} \right] \cdot 100 \quad (\text{Equation 1})$$

$$\text{CT}_i = \text{NT}_i + \text{NT}_i \left[ \frac{P_1 - P_i}{100 - P_1} \right] \quad (\text{Equation 2})$$

$$\text{CD}_i = \text{CD}_{i-1} + \left[ \frac{\text{CT}_{i-1}}{2} \right] + \left[ \frac{\text{CT}_i}{2} \right] \quad (\text{Equation 3})$$

$\text{CT}_i$  is corrected thickness of layer  $i$ ,  $\text{NT}_i$  is nominal thickness of layer  $i$ ,  $P_1$  is porosity of layer 1 (the upper layer),  $P_i$  is the porosity of layer  $i$ ,  $\text{CD}_i$  is corrected depth of layer  $i$  in the middle of the layer.  $\text{CT}_1$  and  $\text{CD}_1$  are estimated to be 1 cm and 0.5 cm, respectively.

### 3. Results and discussion

#### 3.1 Geographical distribution of radionuclides

The  $^{137}\text{Cs}$  concentrations in surface sediments varied from below the detection limit ( $\sim 0.5 \text{ Bq kg}^{-1} \text{ dw}$  (dry weight)) to  $9.3 \text{ Bq kg}^{-1} \text{ dw}$  (Table 2). The highest levels were found in the Spitsbergen area and the lowest levels in the deeper part of the Norwegian Sea, where the  $^{137}\text{Cs}$  concentrations were below the detection limit in four of five samples. One of these (NS-3) was collected in the vicinity of the wreck of the Russian nuclear submarine 'Komsomolets'. Thus, no significant leakage from the wreck has reached this particular sampling location. One sample collected in the Norwegian Sea (NS-1) and one sample collected near the Greenland ice-edge (GI-1) had  $^{137}\text{Cs}$  levels comparable to those found in the Spitsbergen area. The  $^{137}\text{Cs}$  concentrations in sediment cores decreases regularly with

collected at the same location south of Spitsbergen (SB-3), for which the highest concentrations were found at 2-4 cm depth (Fig. 2 a-i). The concentrations of caesium-134 ( $^{134}\text{Cs}$ ) were below the detection limit in all samples.

The concentrations of  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in surface sediments varied from  $1 \cdot 10^{-3}$  - 0.1, 0.01 - 2.4 Bq and  $7 \cdot 10^{-3}$  - 0.9 Bq  $\text{kg}^{-1}$  dw, respectively (Table 2). Again, the highest levels were found in the Spitsbergen area, and the lowest levels were found in the deeper part of the Norwegian Sea. The plutonium activity ratios ( $^{238}\text{Pu} : ^{239,240}\text{Pu}$ ) range from 0.03 to 0.12 (Table 2). Samples collected in the Spitsbergen area have plutonium activity ratios consistent with the ratio resulting from atmospheric weapons tests (0.03-0.04 (UNSCEAR, 1982)), whereas samples collected in the Norwegian Sea have higher ratios. Two samples collected in the Norwegian Sea (NS-2 and NS-4) have plutonium activity ratios above 0.1, indicating that these locations are contaminated with plutonium originating from the European reprocessing industry. However, these samples contain low plutonium concentrations and large errors, which might have biased the results.

The  $^{137}\text{Cs}$  levels found close to Spitsbergen in this study are comparable to those found by Føyn and Sværen (1997) ( $\sim 4.5$ -9 Bq  $\text{kg}^{-1}$  dw). The  $^{137}\text{Cs}$  levels found in the present study in the deeper parts of the Norwegian Sea are low compared to those found in a study by Grøttheim (1998). However, some of the samples in the latter study were collected further south in the Norwegian Sea, closer to point sources for  $^{137}\text{Cs}$  (such as the Baltic Sea outflow, which contains significant amounts of Chernobyl originated  $^{137}\text{Cs}$ , and the European reprocessing industry), while others were collected in shallower areas closer to the Norwegian coast, and may therefore be more influenced by terrestrial contributions of  $^{137}\text{Cs}$  than the deep Norwegian Sea samples collected in this study.

The present study indicates that surface sediments in the Spitsbergen area contain elevated levels of actinides. The concentration of  $^{239,240}\text{Pu}$  found at the sampling location closest to Spitsbergen (2.4 Bq  $\text{kg}^{-1}$  dw) is more than 50% higher than previously reported levels in the Norwegian, Greenland and Barents Seas. The few reported levels of  $^{239,240}\text{Pu}$  in the Norwegian and Greenland Seas are approximately 1 Bq  $\text{kg}^{-1}$  and below (Stepanov et al., 1999; Grøttheim, 1998). Levels of  $^{239,240}\text{Pu}$  in fine-grained depositional regimes in the Barents and Kara Seas are generally  $< 1$  Bq  $\text{kg}^{-1}$  (e.g. Baskaran et al., 1995; Smith et al., 1995).

### 3.2 Sedimentation rates

The distributions of  $^{210}\text{Po}$  were investigated in three vertical sediment cores collected in the Norwegian Sea (NS-1), near the Greenland ice-edge (GI-1) and south of Spitsbergen (SB-3) in order to calculate sedimentation rates (Fig. 3 a-c). The sedimentation rates at the three locations were calculated to be  $0.26 \text{ cm y}^{-1}$  ( $1300 \text{ g m}^{-2} \text{ y}^{-1}$ ),  $0.05 \text{ cm y}^{-1}$  ( $600 \text{ g m}^{-2} \text{ y}^{-1}$ ) and  $0.28 \text{ cm y}^{-1}$  ( $1000 \text{ g m}^{-2} \text{ y}^{-1}$ ), respectively. These sedimentation rates are similar to sedimentation rates found by Huh et al. (1997) in the Chukchi Slope and Laptev and Chukchi Shelves ( $0.089 \text{ cm y}^{-1}$  and  $0.2\text{-}0.7 \text{ cm y}^{-1}$ , respectively) but three orders of magnitude higher than those found in the deep western Arctic Ocean ( $1 \cdot 10^{-4} - 3 \cdot 10^{-4} \text{ cm y}^{-1}$ ) (Huh et al., 1997). Sedimentation rates are expected to be lower in the deep Norwegian Sea than in the shallow, highly productive area close to Spitsbergen. The results showing similar sedimentation rates at the locations NS-1 in the Norwegian Sea and SB-3 close to Spitsbergen were therefore unexpected. The results are, however, supported by the vertical distributions of  $^{137}\text{Cs}$ . The  $^{137}\text{Cs}$ -maximum at 2-4 cm depth at both locations (Figs. 2 b and d) corresponds with deposition from the Chernobyl accident. The cause of a high sedimentation rate at this location of 2922 meters depth is unknown, but may be related to seabed topography. The lower sedimentation rate near the Greenland ice-edge (GI-1) is probably due to ice cover and resulting low biological production large parts of the year.

### 3.3 Mineralogy and particle size distribution

It is well known that some clay minerals (i.e. three-layer clay minerals) have a high adsorption capacity for caesium. Coughtrey and Thorne (1983) stated that the degree to which  $^{137}\text{Cs}$  binds to sediments depends primarily on their content of the clay mineral illite, as  $^{137}\text{Cs}$  is expected to bind irreversibly to this mineral. X-ray diffraction (XRD) determination of the mineral content in 11 surface sediments showed that all contained the clay minerals illite and kaolinite, whereas smectite was apparent in all except one sample (SB-1) (Table 3). Three samples (SB-4, NS-3 and NS-4) contained chlorite, a mineral with layered structure similar to that of clays. XRD does not give a quantitative determination of the clay mineral content. However, as clay minerals are known to be found in the clay fraction (particles  $< 2 \mu\text{m}$ ), a determination of the particle size distribution in the silt/clay fraction in the 11 samples gave a rough quantitative estimate of the clay content in each sample (Fig. 4 a-k). The clay contents (%) are shown in Table 4. The  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  concentrations appear to increase with

increasing clay contents (%) (Fig. 5a and b). Exceptions to this pattern are samples collected at great depths in the deep Norwegian Sea (NS-2 (3479 m), NS-3 (1680 m), NS-4 (3190 m) and NS-5 (3414 m)), for which there seem to be no correlation between the content of clay and radionuclide levels. The silt/clay content (%) determined by dry sieving is shown in Table 4. The silt/clay content (%) plotted against  $^{137}\text{Cs}$  concentrations support the result showing increasing radionuclide concentration with increasing clay content (Fig. 5c). The results illustrate the importance of taking sediment mineralogy into account when assessments of radionuclides in sediments are made. The amount of organic materials in sediments may also influence on radionuclide levels, but are not discussed in the present study.

### *3.4 Transport of radionuclides via terrigenous particles*

When ice starts freezing in the Ob and Yenisey estuaries in the autumn, the coastal water masses contain a maximum of suspended material delivered by run-off from land via rivers. Both contaminated suspended material delivered by run-off from land and contaminated bottom sediments can be incorporated into ice in the estuaries in the Kara Sea (Vinje and Kvambekk, 1991; Landa et al., 1998; Dethleff et al., 2000; Smedsrud, 2000). Several investigations indicate that during winter, Kara Sea ice travels around the northern tip of Novaya Zemlya, into the Barents Sea south of Franz Josef Land and further southwest towards Svalbard and Spitsbergen (Vinje and Kvambekk, 1991; Nürnberg et al., 1994; Pfirman et al., 1995; Emery et al., 1997; Landa et al., 1998). During ice melting, contaminated material trapped in the ice can sink to the bottom in the melting zone, and thereby cause elevated levels of radioactive contamination in sediments in this area. Further, contaminated suspended material originating from Spitsbergen may also be delivered via rivers and/or glaciers. The latter contribution is dependent on the degree of contamination of global fallout and Chernobyl origin and the precipitation over Spitsbergen, but is not studied in detail here. The plutonium activity ratios found in sediments in the Spitsbergen area ( $\sim 0.03$ ) are consistent with that of global fallout. Both the areas draining into the Barents and Kara Seas and the Spitsbergen area were contaminated by global fallout. The results do therefore not allow distinguishing between these two possible sources. We can, however, exclude the European reprocessing industry as a source for contamination of this area.

### *3.5 Vertical transport of radionuclides in the water column by sorption to sinking phytoplankton*

As mentioned in the introduction, a geographically fairly stable ice-melting zone arises along the polar front in the Spitsbergen area in the western Barents Sea during spring and summer (Loeng, 1991). The ice-melting gives rise to a stratified and nutrient rich euphotic zone, which supports high primary production (Sakshaug and Skjoldal, 1989). A fraction of the phytoplankton stock will eventually sink out of the euphotic zone and reach the sediments. Several studies describe vertical transport of natural and anthropogenic radionuclides in the water column by adsorption onto sinking particulate organic matter (e.g. Fowler et al., 1987; Baumann and Wefer, 1991). Føyn and Sværen (1997) suggested that living phytoplankton in the water column might actively act as scavenging elements and cause elevated levels of  $^{137}\text{Cs}$  in sediments in the Spitsbergen area. During both sample collections, attempts were made to collect phytoplankton samples in the Spitsbergen area, large enough for direct measurements of gamma-emitting radionuclides (e.g.  $^{137}\text{Cs}$ ). The attempts failed, and we were not able to perform radionuclide analyses on environmental phytoplankton samples, analyses that were thought to confirm or invalidate the hypothesis of Føyn and Sværen (1997). As an alternative, uptake studies of  $^{137}\text{Cs}$  in five marine phytoplankton species representative for the Barents Sea phytoplankton community were performed in the laboratory (Heldal et al., 2001). The results showed that the uptake of  $^{137}\text{Cs}$  was low or even negligible in both growing and non-growing cells. Heldal et al. (2001) concluded that phytoplankton are unlikely to contribute as a source of  $^{137}\text{Cs}$  to marine sediments. This is in accordance with the findings of Fowler et al. (1987). Baumann and Wefer (1991) found, however, that sinking fecal pellets caused a strong vertical  $^{137}\text{Cs}$  flux in the Norwegian Sea following the Chernobyl accident. It is not possible to draw conclusions concerning the importance of uptake of transuranes in phytoplankton and subsequent deposition in the Spitsbergen area. It is, however, known that both Pu and Am are accumulated in phytoplankton to a much higher degree than Cs (e.g. Gromov, 1976). IAEA (1985) recommend Concentration Factors ( $\text{CF} = \text{Bq kg}^{-1} \text{ wet organism} / \text{Bq kg}^{-1} \text{ seawater}$ ) for Pu and Am in phytoplankton of  $1 \cdot 10^5$  and  $2 \cdot 10^5$ , respectively. In contrast, Heldal et al. (2001) found Volume Concentration Factors ( $\text{VCF} = \text{Bq } \mu\text{m}^{-3} \text{ cell} / \text{Bq } \mu\text{m}^{-3} \text{ seawater}$ ), generally close to 1 and not above 100 for  $^{137}\text{Cs}$  in growing phytoplankton cells. Low seawater concentrations of actinides in this area ( $\sim 7 \mu\text{Bq } ^{239,240}\text{Pu L}^{-1}$  and  $\sim 0.4 \mu\text{Bq } ^{241}\text{Am L}^{-1}$  in surface water; Heldal, unpubl. data) do, however, not support the above mentioned transport mechanism. More efficient equipment is needed to collect large enough samples in order to



determine the significance of transport of radionuclides via sinking particulate organic matter in the Spitsbergen area.

#### **4. Conclusions**

The concentrations of  $^{137}\text{Cs}$ ,  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  in marine sediments in the Spitsbergen area were found to reach 9.3, 0.1, 2.4 and 0.9 Bq kg<sup>-1</sup> dw, respectively. These concentrations are over 50% higher than in adjacent areas. The  $^{137}\text{Cs}$  concentration was below the detection limit in four of five samples collected in the deeper Norwegian Sea.

The plutonium activity ratios found in sediments south of Spitsbergen (~0.03) are consistent with that of global fallout. Based on this, we can exclude the European reprocessing industry as a contamination source for this area. We cannot, however, distinguish between the following origins of sediments: 1) sedimentary particles originating from Russian rivers draining into the Barents and Kara Seas and transported to the Spitsbergen area via sea ice, or 2) sedimentary particles washed out from Spitsbergen; as both these areas were contaminated by global fallout.

The plutonium activity ratios found in two sediment samples collected in the deep Norwegian Sea were above 0.1, indicating that these locations are contaminated with plutonium originating from the European reprocessing industry.

Sedimentation rates found in this study are comparable with those found by others in the Chucki Slope and Laptev and Chucki Shelves. The result showing similar sedimentation rates at a location in the deep Norwegian Sea and a location in the highly productive area close to Spitsbergen was, however, unexpected.

This study indicates a relation between the content of clay minerals and radionuclide levels for samples collected in the Spitsbergen-Bear Island area. With one exception, samples collected in the deeper Norwegian Sea deviated from this pattern.

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## References

- Aarkrog A. Radioactivity in Polar Regions - Main Sources. *J Environ Radioact* 1994; 25: 21-35.
- Baskaran M, Asbill S, Santschi P, Davis T, Brooks J, Champ M, Makeyev V, Khlebovich V. Distribution of  $^{239,240}\text{Pu}$  and  $^{238}\text{Pu}$  concentrations in sediments from the Ob and Yenisey rivers and the Kara Sea. *Appl Radiation Isot* 1995; 46: 1109-1119.
- Baumann M, Wefer G. Enhanced deposition of Chernobyl radiocesium by phytoplankton in the Norwegian Sea: Evidence from sediment trap deployments. In: Kershaw P, Woodhead D, editors. *Int. Symp. Radionuclides in the Study of Marine Processes*, 10-13 September 1991, Norwich, UK, 1991: 299-308.
- Brady NC. *The nature and properties of soils*: 10<sup>th</sup> edition. Macmillan Publishing Company, New York, USA, 1990.
- Coughtrey PJ, Thorne MC. Caesium in aquatic ecosystems. In: *Radionuclide distribution and transport in terrestrial and aquatic ecosystems*, Vol. 1, AA Balkema Publ., Rotterdam, 1983: pp 496.
- Dethleff D, Nies H, Harms IH, Karcher MJ. Transport of radionuclides by sea-ice and dense-water formed in western Kara Sea flaw leads. *J Mar Syst* 2000; 24: 233-248.
- Emery WJ, Fowler C, Maslanik J, Pfirman S. New Satellite Derived Sea Ice Motion Tracks Arctic Contamination. *Mar Pollut Bull* 1997; 35: 345-352.
- Fowler SW, Buat-Menard P, Yokoyama Y, Ballestra S, Holm E, Nguyen HV. Rapid removal of Chernobyl fallout from Mediterranean surface waters by biological activity. *Nature* 1987; 329: 56-58.
- Føyn L, Sværen I. Distribution and sedimentation of radionuclides in the Barents Sea. *ICES J Mar Sci* 1997; 54: 333-340.
- Gromov VV. Uptake of plutonium and other nuclear wastes by plankton. *Mar Sci Comm* 1976; 2 (3&4): 227-247.

- Grøttheim S. A preliminary report on radioactive contamination in the northern marine environment. Studies on distribution of radiocaesium, plutonium and americium in sea water and sediments, MSc thesis, Norwegian Radiation Protection Authority and Biological Institute, University of Oslo, Norway, 1998.
- Heldal HE, Stupakoff I, Fisher NS. Bioaccumulation of  $^{57}\text{Co}$  and  $^{137}\text{Cs}$  in five marine phytoplankton species. *J Environ Radioact* 2001; 57 (3): 231-236.
- Huh C-A, Pisias NG, Kelley JM, Maiti TC, Grantz A. Natural radionuclides and plutonium in sediments from the western Arctic Ocean: sedimentation rates and pathways of radionuclides. *Deep Sea Res II* 1997; 44 (8): 1725-1743.
- IAEA. Sediment  $K_d$ 's and Concentration Factors for Radionuclides in the Marine Environment. Tech. Rept. Ser. No. 247. International Atomic Energy Agency, Vienna, 1985.
- Kershaw P, Baxter A. The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep Sea Res II* 1995; 42 (6): 1413-1448.
- Landa ER, Reimnitz E, Beals DM, Pochkowski JM, Winn WG, Rigor I. Transport of  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  with Ice-rafted Debris in the Arctic Ocean. *Arctic* 1998; 51: 27-39.
- Loeng H. Features of the physical oceanographic conditions of the Barents Sea. *Polar Res* 1991; 10: 5-18.
- Lovett MB, Boggis SJ, Blowers P. The determination of alpha-emitting nuclides of plutonium, americium and curium in environmental materials: Part 1. Sea water, Aquatic Environment Protection: Analytical Methods No. 7, MAFF, Directorate of Fisheries Research, Lowestoft, UK, 1990.
- Nürnberg D, Wollenburg I, Dethleff D, Eicken H, Kassens H, Letzig T, Reimnitz E, Thiede J. Sediments in Arctic sea ice: Implications for entrainment, transport and release. *Mar Geol* 1994; 119: 185-214.
- Pfirman SL, Eicken H, Bauch D, Weeks WF. The potential transport of pollutants by Arctic sea ice. *Sci Tot Environ* 1995; 159: 129-146.
- Sakshaug E, Skjoldal HR. Life at the ice edge. *Ambio* 1989; 18: 60-67.
- Salbu B, Nikitin AI, Strand P, Christensen GC, Chumichev VB, Lind B, Fjellidal H, Bergan TDS, Rudjord AL, Sickel M, Valetova NK, Føyn L. Radioactive contamination from dumped nuclear waste in the Kara Sea-results from the joint Russian –Norwegian expeditions in 1992-1994. *Sci Tot Environ* 1997; 202: 185-198.
- Smedsrud LH. Frazil Ice Formation and Incorporation of Sediments into Sea Ice in the Kara Sea, PhD thesis, Geophysical Institute, University of Bergen, Norway, 2000.

- Smith JN, Ellis KM, Naes K, Dahle S, Matishov D. Sedimentation and mixing rates of radionuclides in Barents Sea sediments off Novaya Zemlya. *Deep Sea Res II* 1995; 42 (6): 1471-1493.
- Smith JN, Ellis KM, Polyak L, Ivanov G, Forman SL, Moran SB.  $^{239,240}\text{Pu}$  transport into the Arctic Ocean from underwater nuclear tests in Chernaya Bay, Novaya Zemlya. *Cont Shelf Res* 2000; 20: 255-279.
- Stepanov AV, Tsvetkov OS, Tishkov VP, Belyaev BN, Domkin VD, Ivanova LM, Osokina AA, Plekhov VS, Bobylev KL, Johannessen O. Isotopic composition of Plutonium in the bottom deposits of the Norwegian Sea and Greenland Sea and identification of the sources of contaminationheader. *Atomic Energy* 1999; 87: 745-752.
- Strand P, Nikitin A, Rudjord AL, Salbu B, Christensen G, Føyn L, Kryshev II, Chumichev VB, Dahlgaard H, Holm E. Survey of Artificial Radionuclides in the Barents Sea and the Kara Sea. *J Environ Radioact* 1994; 25: 99-112.
- Tadjiki S, Erten HN. Radiochronology of sediments from the Mediterranean Sea using natural  $^{210}\text{Pb}$  and fallout  $^{137}\text{Cs}$ . *J Radioanal Nucl Chem* 1994; 181: 447-459.
- UNSCEAR. Ionizing radiation: sources and biological effects. United Nations, New York, USA, 1982.
- Vinje T, Kvambekk AS. Barents Sea drift ice characteristics. *Polar Res* 1991; 10: 59-68.

Table 1. Sampling positions and water depths. Sample IDs refer to sampling areas (SB=Spitsbergen-Bear Island area, WS=west of Spitsbergen, NS=Norwegian Sea, GI=Greenland ice-edge)

Sample ID	Latitude	Longitude	Water depth (m)
<i>Spitsbergen – Bear Island area</i>			
SB-1	75° 30' N	19° 30' E	86
SB-2	76° 00' N	19° 40' E	121
SB-3	76° 35' N	20° 00' E	205
SB-4	76° 19' N	18° 10' E	246
SB-5	75° 50' N	15° 10' E	374
SB-6	74° 59' N	18° 18' E	94
SB-7	75° 15' N	16° 30' E	227
SB-8	75° 15' N	17° 30' E	149
SB-9	75° 25' N	17° 20' E	142
SB-10	75° 35' N	17° 07' E	175
SB-11	75° 50' N	16° 49' E	324
SB-12	76° 00' N	16° 36' E	333
SB-13	76° 10' N	16° 24' E	311
SB-14	74° 35' N	16° 30' E	248
SB-15	74° 35' N	18° 00' E	121
SB-16	74° 00' N	19° 13' E	135
SB-17	73° 50' N	19° 16' E	231
<i>West of Spitsbergen</i>			
WS-1	76° 55' N	13° 00' E	272
WS-2	76° 55' N	13° 30' E	101
WS-3	76° 55' N	14° 30' E	108
WS-4	76° 58' N	15° 30' E	150
<i>Norwegian Sea</i>			
NS-1	70° 24' N	08° 12' E	2922
NS-2	74° 05' N	03° 40' W	3479
NS-3	73° 44' N	13° 16' E	1680
NS-4	70° 15' N	04° 00' E	3190
NS-5	67° 00' N	01° 27' W	3414
<i>Greenland ice-edge</i>			
GI-1	74° 30' N	14° 37' W	267

Table 2. Concentrations of  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  and plutonium isotope ratios ( $^{238}\text{Pu}/^{239,240}\text{Pu}$ ) in surface sediments. Errors quoted for  $^{137}\text{Cs}$  are  $\pm 1$  standard deviation. Errors quoted for  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$  and  $^{241}\text{Am}$  are  $\pm 1$  sigma ( $\sigma$ ) percentage counting propagated errors only. bd=below detection limit; nd=not determined

Sample ID	$^{137}\text{Cs}$			$^{239,240}\text{Pu}$			$^{238}\text{Pu}$			$^{241}\text{Am}$			$^{238}\text{Pu}/^{239,240}\text{Pu}$	
	Bq	kg <sup>-1</sup>	dw	Bq	kg <sup>-1</sup>	dw	Bq	kg <sup>-1</sup>	dw	Bq	kg <sup>-1</sup>	dw		
SB-1	2.4	±	0.1	0.98	±	0.01	0.033	±	0.001	0.37	±	0.01	0.03	
SB-2	3.4	±	0.2	1.14	±	0.02	0.046	±	0.001	0.48	±	0.02	0.04	
SB-3	6.3	±	0.1	2.37	±	0.03	0.096	±	0.002	0.85	±	0.05	0.04	
SB-4	6.0	±	0.3	1.62	±	0.02	0.059	±	0.002	0.70	±	0.02	0.04	
SB-5	5.5	±	0.3	1.28	±	0.02	0.053	±	0.002	0.69	±	0.03	0.04	
SB-6	0.7	±	0.1	nd	-		nd	-		nd	-		-	
SB-7	4.1	±	0.1	nd	-		nd	-		nd	-		-	
SB-8	4.5	±	0.1	nd	-		nd	-		nd	-		-	
SB-9	3.2	±	0.1	nd	-		nd	-		nd	-		-	
SB-10	2.1	±	0.1	nd	-		nd	-		nd	-		-	
SB-11	3.8	±	0.1	nd	-		nd	-		nd	-		-	
SB-12	3.5	±	0.1	nd	-		nd	-		nd	-		-	
SB-13	5.0	±	0.1	nd	-		nd	-		nd	-		-	
SB-14	1.1	±	0.1	nd	-		nd	-		nd	-		-	
SB-15	3.4	±	0.1	nd	-		nd	-		nd	-		-	
SB-16	1.6	±	0.1	nd	-		nd	-		nd	-		-	
SB-17	3.0	±	0.1	nd	-		nd	-		nd	-		-	
WS-1	6.4	±	0.1	nd	-		nd	-		nd	-		-	
WS-2	3.9	±	0.1	nd	-		nd	-		nd	-		-	
WS-3	6.6	±	0.1	nd	-		nd	-		nd	-		-	
WS-4	9.3	±	0.1	nd	-		nd	-		nd	-		-	
NS-1	4.8	±	0.1	0.47	±	0.01	0.029	±	0.001	0.30	±	0.01	0.06	
NS-2	0.5	±	0.1	0.024	±	0.001	0.003	±	0.001	0.015	±	0.004	0.11	
NS-3	bd	-		0.100	±	0.003	0.008	±	0.001	0.09	±	0.01	0.08	
NS-4	bd	-		0.009	±	0.001	0.0011	±	0.0004	0.015	±	0.001	0.12	
NS-5	bd	-		0.037	±	0.001	0.0027	±	0.0003	0.007	±	0.001	0.07	
GI-1	5.4	±	0.1	0.37	±	0.01	0.025	±	0.001	0.25	±	0.02	0.07	

Table 3. The mineral content in surface samples as semi-quantitatively measured by X-ray diffraction. Abbreviations: Ill=Illite; Kaol=Kaolinite; Smec=Smectite; Chl=Chlorite; Cal=Calcite; Qu=Quartz; Fel=Feldspar

Sample ID	Clay minerals and Chlorite	Other minerals
SB-1	Ill > Kaol	Cal > Qu > Fel
SB-2	Ill > Kaol > Smec	Qu > Cal > Fel
SB-3	Kaol > Ill > Smec	Qu > Fel
SB-4	Kaol > Ill > Smec > Chl	Qu > Fel
SB-5	Kaol > Ill > Smec	Qu > Fel
NS-1	Kaol > Ill > Smec	Cal > Qu > Fel
NS-2	Kaol > Ill > Smec	Qu > Cal > Fel
NS-3	Ill > Kaol > Smec > Chl	Qu > Cal > Fel
NS-4	Ill > Kaol > Smec > Chl	Cal > Qu > Fel
NS-5	Ill > Kaol > Smec	Cal > Qu > Fel
GI-1	Ill > Kaol > Smec	Qu > Fel

Table 4. Percentage (%) silt/clay and clay. <sup>a</sup> Determined by dry sieving of undivided samples. <sup>b</sup> Determined by particle size analysis where particles > 250  $\mu$ m was removed from each sample prior to analysis. The percentages silt/clay and clay found by the different methods are not comparable.

Sample ID	Silt/Clay (%) <sup>a</sup>	Clay (%) <sup>b</sup>
SB-1	16	16.9
SB-2	31	22.9
SB-3	31	28.2
SB-4	13	24.4
SB-5	29	33.0
SB-6	nd	nd
SB-7	34	nd
SB-8	37	nd
SB-9	13	nd
SB-10	28	nd
SB-11	32	nd
SB-12	24	nd
SB-13	12	nd
SB-14	10	nd
SB-15	10	nd
SB-16	6	nd
SB-17	12	nd
WS-1	23	nd
WS-2	16	nd
WS-3	34	nd
WS-4	45	nd
NS-1	22	26.8
NS-2	21	31.0
NS-3	25	41.3
NS-4	17	43.4
NS-5	24	21.9
GI-1	20	16.1

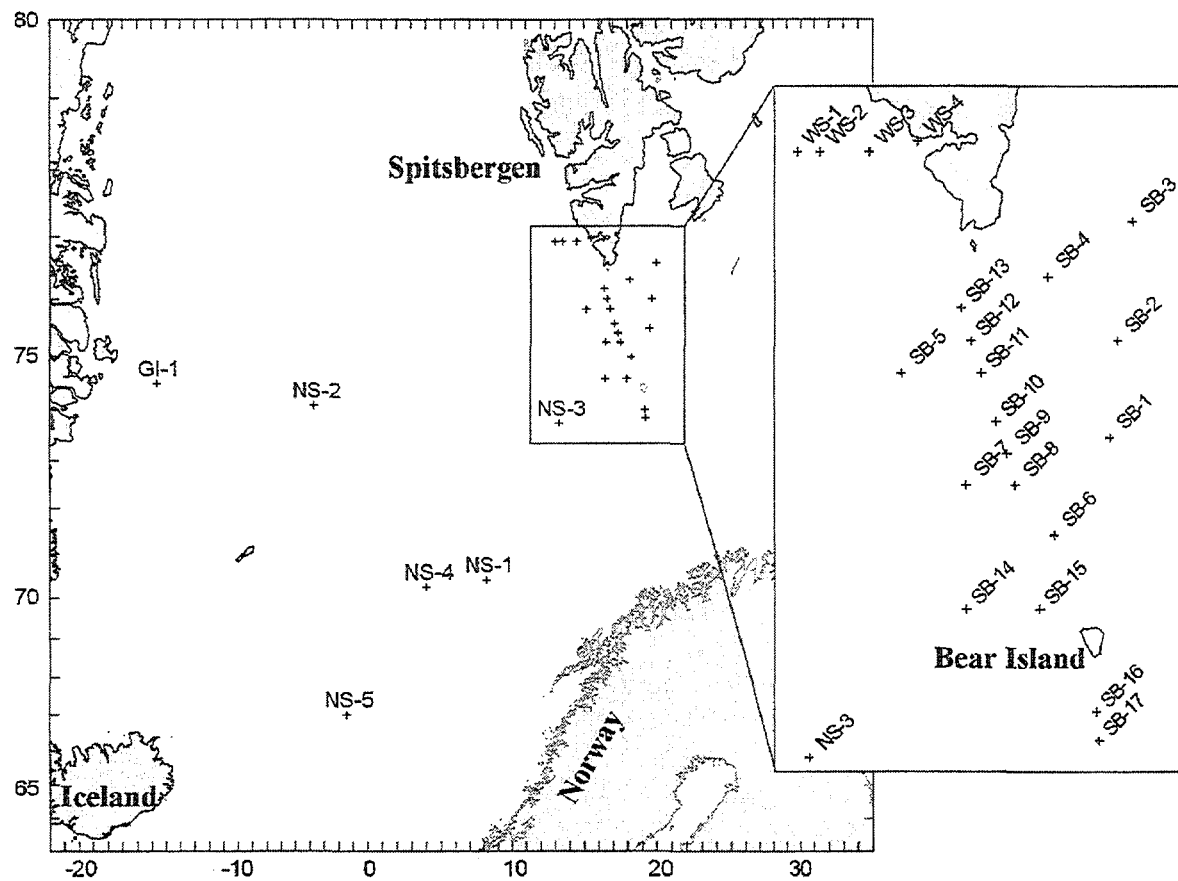


Figure 1. Map showing the study area and sampling locations



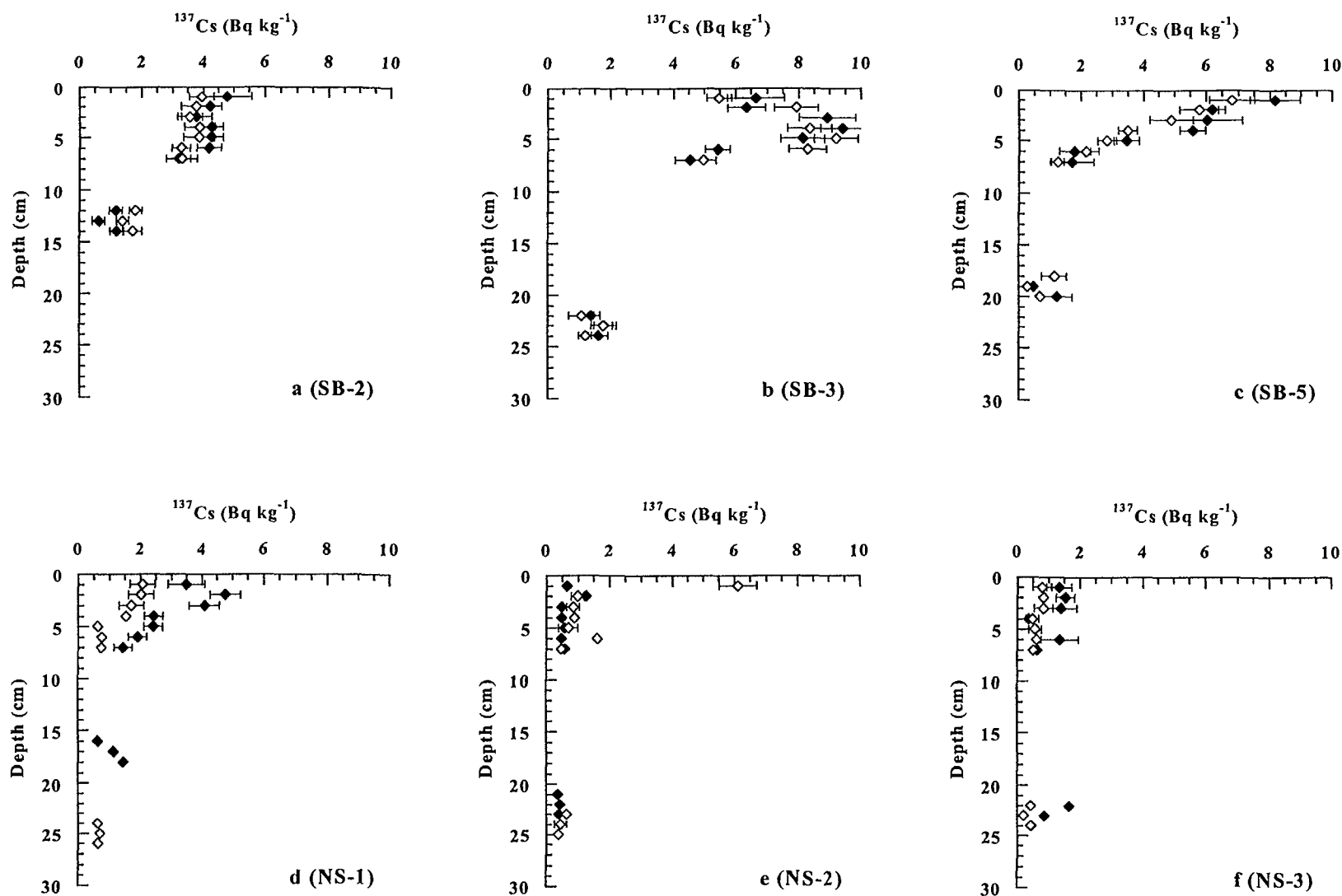


Figure 2. The distribution of  $^{137}\text{Cs}$  (Bq kg $^{-1}$  dry weight (dw)) in sediment depth profiles. The  $^{137}\text{Cs}$  activity is decay-corrected to collection date. Black and white diamonds are different cores collected from the same box corer. The detection limit (DL) activity is plotted for samples below the DL. The error (one sigma ( $\sigma$ ) total error) is shown with bars for each sample above the detection limit.

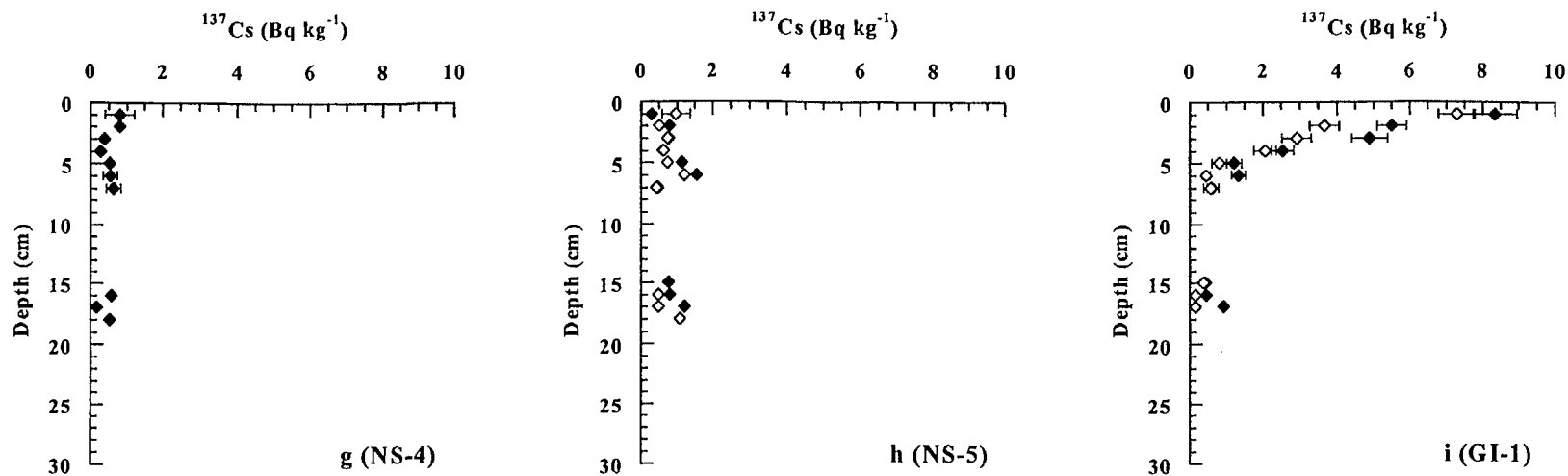


Figure 2 (cont.). The distribution of  $^{137}\text{Cs}$  (Bq kg $^{-1}$  dry weight (dw)) in sediment depth profiles. The  $^{137}\text{Cs}$  activity is decay-corrected to collection date. Black and white diamonds are different cores collected from the same box corer. The detection limit (DL) activity is plotted for samples below the DL. The error (one sigma ( $\sigma$ ) total error) is shown with bars for each sample above the detection limit.

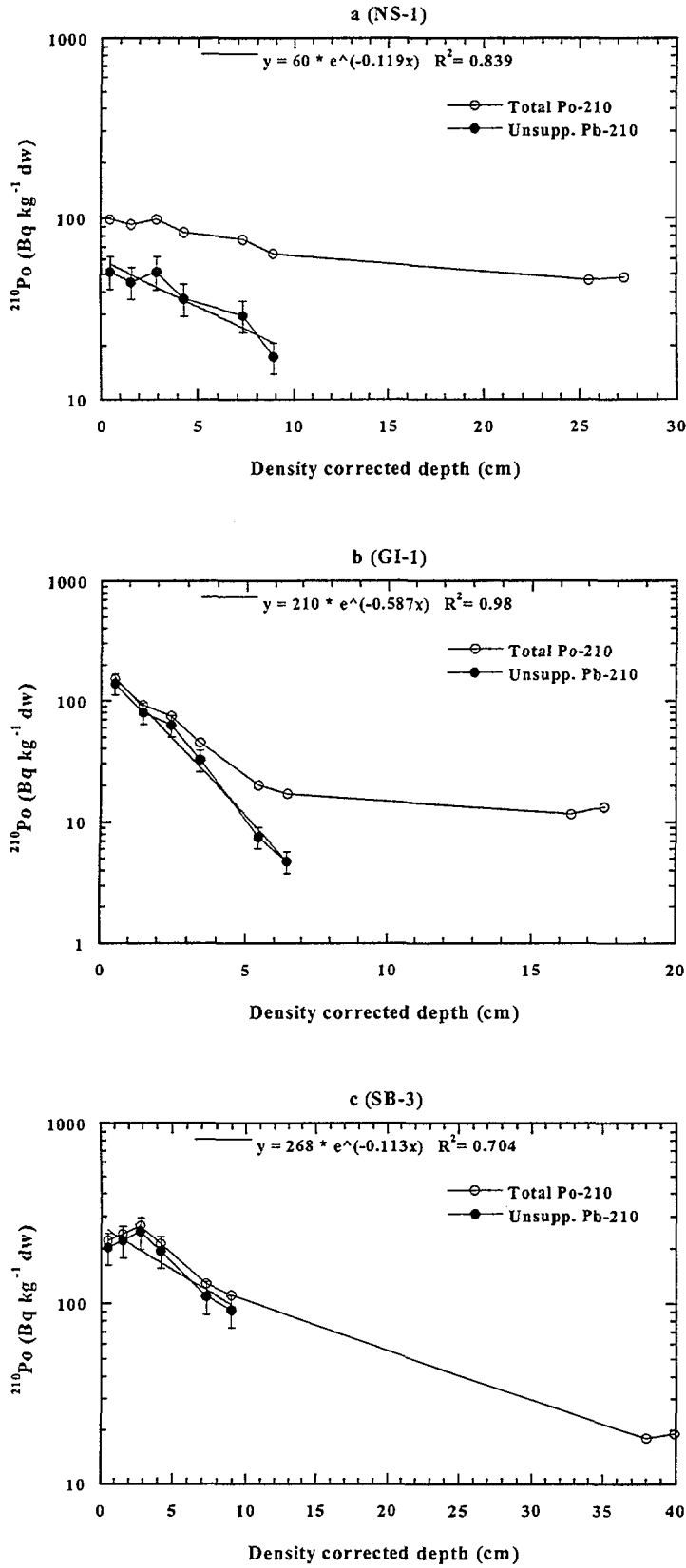


Figure 3.  $^{210}\text{Po}$  profiles in vertical sediment cores. Sedimentation rates were estimated using an exponential curve fit. Uncertainties in the unsupported  $^{210}\text{Pb}$  data are estimated to be 20 % and indicated with error bars.

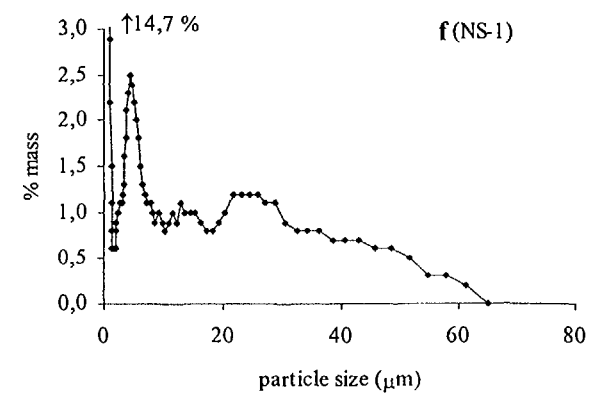
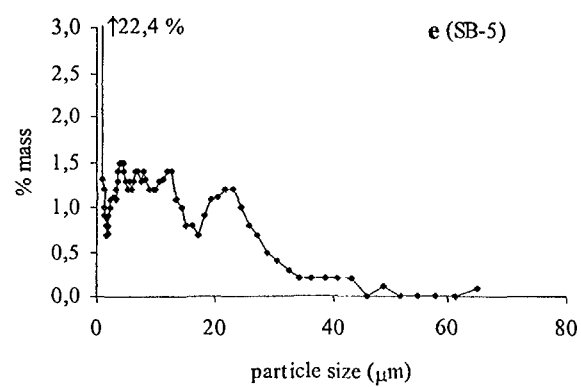
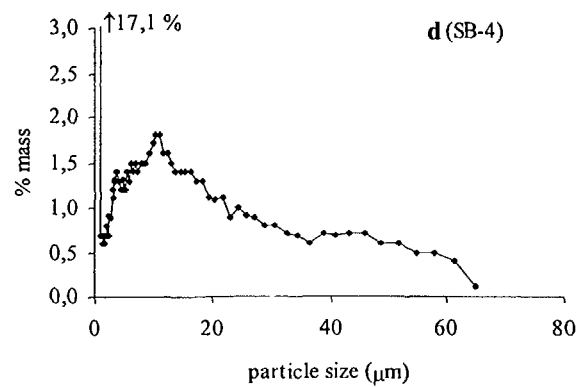
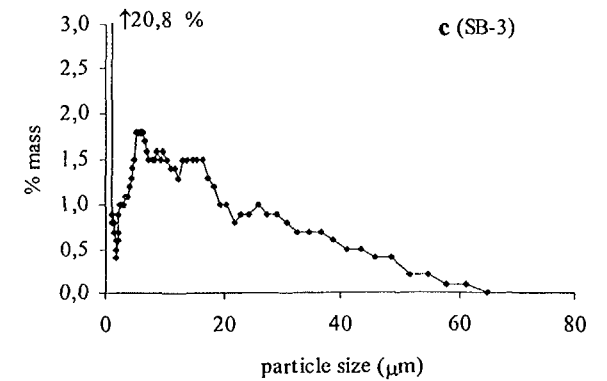
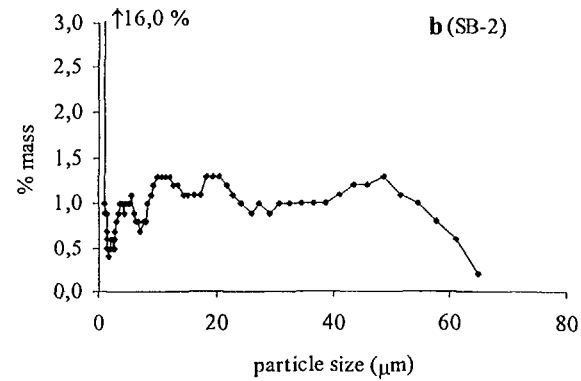
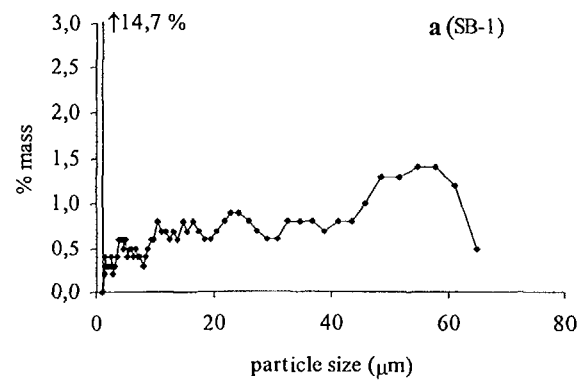


Figure 4. The particle size distribution in the silt/clay fraction of surface sediments. Particles  $> 250 \mu\text{m}$  was removed from each sample prior to analysis.

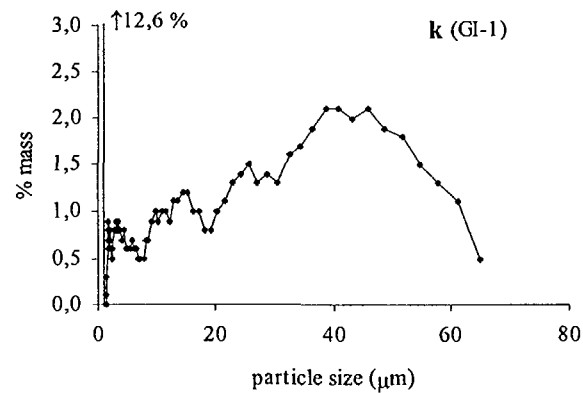
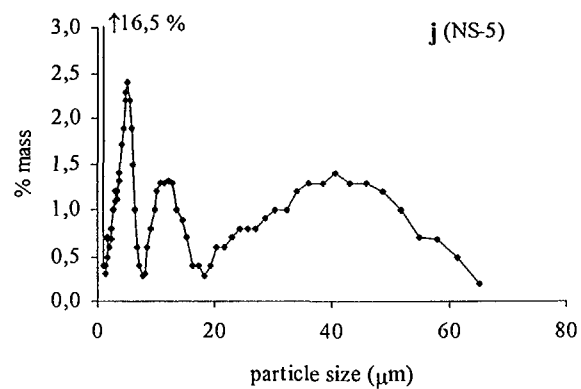
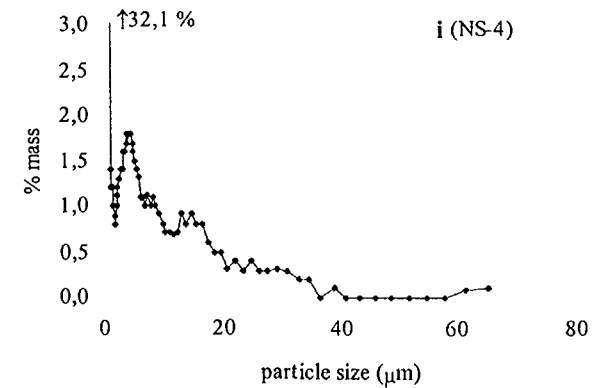
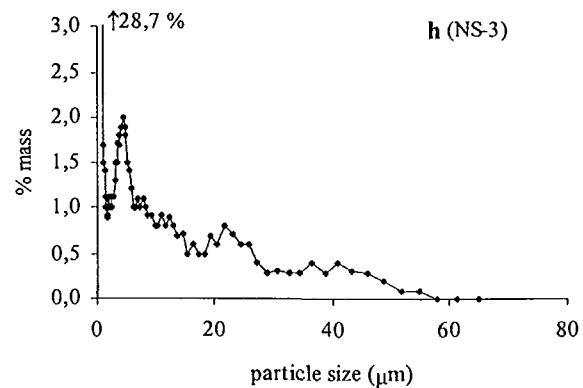
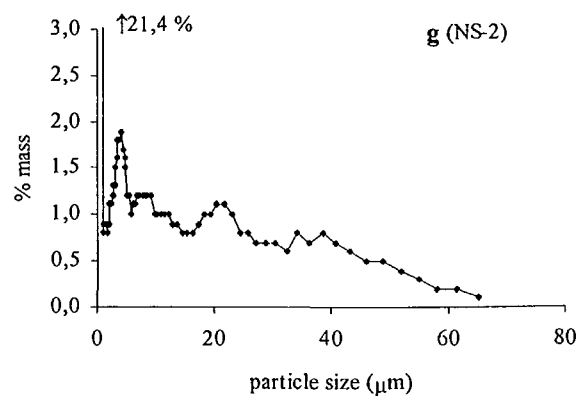


Figure 4 (cont.). The particle size distribution in the silt/clay fraction of surface sediments. Particles  $> 250 \mu\text{m}$  was removed from each sample prior to analysis.

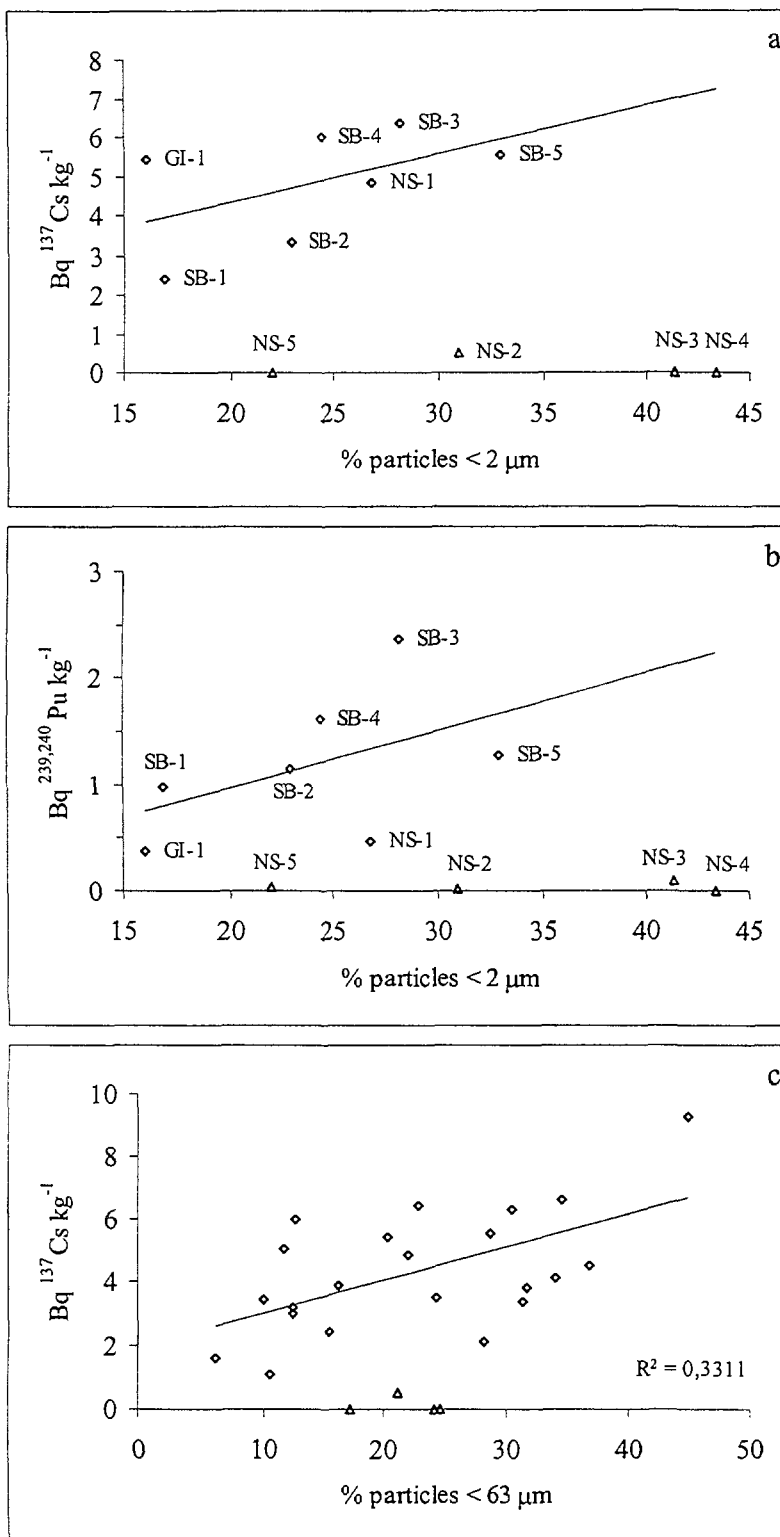


Figure 5. a) Fraction (%) of particles  $< 2 \mu\text{m}$  (clay) in surface sediments plotted against levels of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$ ). b) Fraction (%) of particles  $< 2 \mu\text{m}$  (clay) in surface sediments plotted against levels of  $^{239,240}\text{Pu}$  ( $\text{Bq kg}^{-1}$ ). c) Fraction (%) of particles  $< 63 \mu\text{m}$  (clay and silt) in surface samples plotted against levels of  $^{137}\text{Cs}$  ( $\text{Bq kg}^{-1}$ ). Triangles ( $\Delta$ ) are low-levelled samples collected in the deep Norwegian Sea deviating from the linear correlation pattern. The labels in a) and b) are Sample IDs.

## **PAPER II**

# Bioaccumulation of $^{137}\text{Cs}$ and $^{57}\text{Co}$ by five marine phytoplankton species

Hilde Elise Heldal<sup>a,\*</sup>, Ian Stupakoff<sup>b</sup>, Nicholas S. Fisher<sup>b</sup>

<sup>a</sup> Institute of Marine Research, PO Box 1870 Nordnes, N-5817 Bergen, Norway

<sup>b</sup> Marine Sciences Research Center, State University of New York, Stony Brook, NY 11794-5000, USA

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## Abstract

Under controlled laboratory conditions, we have examined the bioaccumulation of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  in three prymnesiophytes, the coccolithophorid *Emiliania huxleyi* and the non-calcareous species *Isochrysis galbana* and *Phaeocystis globosa*, and two diatoms *Skeletonema costatum* and *Thalassiosira pseudonana*. We measured the uptake in growing and non-growing cells and determined concentration factors on both volume and dry weight bases. For uptake of  $^{57}\text{Co}$  in non-growing cells, volume concentration factors (VCF) at equilibrium ranged from  $0.2 \times 10^3$  for *E. huxleyi* to  $4 \times 10^3$  for *T. pseudonana*. For uptake of  $^{137}\text{Cs}$  in non-growing cells, the VCFs were low for all species and the uptake pattern seemed unsystematic. The results suggest that, in contrast to Co, the cycling and bioaccumulation of Cs in marine animals are unlikely to be affected by Cs accumulation in primary producers. © 2001 Elsevier Science Ltd. All rights reserved.

**Keywords:** Cesium; Cobalt; Bioaccumulation; Marine phytoplankton

## 1. Introduction

The artificial radionuclides  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  are important components of radioactive wastes discharged into the coastal waters of Northwest Europe. The European reprocessing plants at Sellafield and La Hague discharge radioactive

\*Corresponding author. Tel.: +47-55-23-63-34; fax: +47-55-23-85-84.

E-mail address: hilde.elise.heldal@imr.no (H.E. Heldal).



wastes into the Irish Sea and the English Channel, respectively. Ocean currents transport these wastes into the North Sea where they mix with waters from the Baltic Sea containing significant amounts of  $^{137}\text{Cs}$  from Chernobyl fallout (Herrmann, Kershaw, Bailly du Bois, & Guegueniat, 1995). The transport continues northwards along the Norwegian coast by the Norwegian Coastal Current; this splits into two branches, one that enters the Barents Sea and the other that passes west of Spitsbergen (Kershaw & Baxter, 1995; Guegueniat, Kershaw, Herrmann, & Bailly du Bois, 1997). These areas are of major importance for Norwegian fisheries both as nursery and fishing grounds. Knowledge of bioaccumulation and various transport routes of radionuclides through marine food webs in these waters is therefore of special interest. Phytoplankton lie at the base of most marine food webs and could serve to introduce radionuclides into these food webs. This paper presents the results of laboratory experiments in which we studied the bioaccumulation of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  (as an analog of  $^{60}\text{Co}$ ) in five phytoplankton species, all of which can occur in the Barents Sea phytoplankton community.

## 2. Materials and methods

We used monocultures of *Emiliania huxleyi* (Prymnesiophyceae), *Isochrysis galbana* (Prymnesiophyceae), *Phaeocystis globosa* (Prymnesiophyceae), *Skeletonema costatum* (Bacillariophyceae) and *Thalassiosira pseudonana* (Bacillariophyceae). All cultures were maintained aseptically in f/2 medium (Guillard & Ryther, 1962) minus EDTA, Cu and Zn, prepared with sterile-filtered Southampton (Long Island, NY) surface water (0.2  $\mu\text{m}$  pore size filter). To determine the bioaccumulation of the radioisotopes, we basically followed experimental protocols described by Fisher, Bjerregaard, and Fowler (1983). Briefly, we performed two experiments under identical experimental conditions, one on non-growing cells and one on growing cells. Growing cells were exposed to cyclic light (L:D 14:10) provided by cool-white fluorescent lamps (170  $\mu\text{E m}^{-2} \text{s}^{-1}$ ). Non-growing cells were kept in darkness to prevent growth. Both non-growing and growing cells were maintained without shaking at  $12 \pm 1^\circ\text{C}$ . In both experiments, growth was checked by counting cells microscopically with a hemacytometer. To calculate volume–volume concentration factors ( $\text{VCF} = \text{radioactivity } \mu\text{m}^{-3} \text{ cell divided by radioactivity dissolved } \mu\text{m}^{-3} \text{ seawater}$ ), population densities were measured microscopically by counting cells in a hemacytometer while cell volumes were measured with a Coulter Counter (Coulter Multisizer II electronic particle counter, Coulter Electronics). For calculations of dry weight concentration factors ( $\text{DCF} = \text{radioactivity } \text{g}^{-1} \text{ dry cell divided by radioactivity dissolved } \text{g}^{-1} \text{ seawater}$ ), the dry weights of the cells were determined as follows: cells were filtered on a membrane filter that had already been washed in an isotonic solution of  $\text{NH}_4\text{COOH}$ , dried and preweighed (Fisher & Schwarzenbach, 1978). After filtration, the cells were washed, dried at  $60^\circ\text{C}$  and weighed. Radioisotope additions were added, via Eppendorf micropipette, as microliter quantities of 0.1 N HCl ( $^{57}\text{Co}$ ) and 1.0 N HCl ( $^{137}\text{Cs}$ ). The pH in the seawater solutions was unaffected by the additions.

The concentrations of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  in the experimental solution were about 300 and 40 Bq ml $^{-1}$ , respectively. Over a six-day period (non-growing cells) and ten-day period (growing cells), cells were periodically filtered onto Nuclepore polycarbonate membranes and the fraction of radioactivity associated with cells was determined for each isotope at each sample time (Fisher et al., 1983). Blanks that contained only media and isotopes were treated in the same manner and corrections were made for isotope sorption to the filters.

### 3. Results and discussion

The results show that  $^{57}\text{Co}$  was systematically taken up by all the species investigated, as illustrated in Figs. 1–3. For growing cells, the uptake of  $^{57}\text{Co}$  was highest in *E. huxleyi*, for which it reached 1 mBq cell $^{-1}$  (Fig. 1a). For non-growing cells, however, the uptake of  $^{57}\text{Co}$  was highest in *T. pseudonana* (Fig. 1b), and *E. huxleyi* in this experiment took up the lowest amounts of  $^{57}\text{Co}$ . Fig. 1b also shows that saturation was reached in the uptake for non-growing cells after approximately 100 h. The initial uptake in non-growing cells was slightly higher than the uptake in growing cells for some of the species. For  $^{137}\text{Cs}$ , however, the uptake was generally much lower in both growing and non-growing cells (Figs. 1c and d) and for some of the species it was not significantly different from the blanks. No significant differences in the uptake of  $^{137}\text{Cs}$  between the species were evident from these data. The volume–volume concentration factors (VCFs) for  $^{57}\text{Co}$  varied from approximately  $0.2 \times 10^3$  for *E. huxleyi* to  $4 \times 10^3$  for *T. pseudonana* for non-growing cells

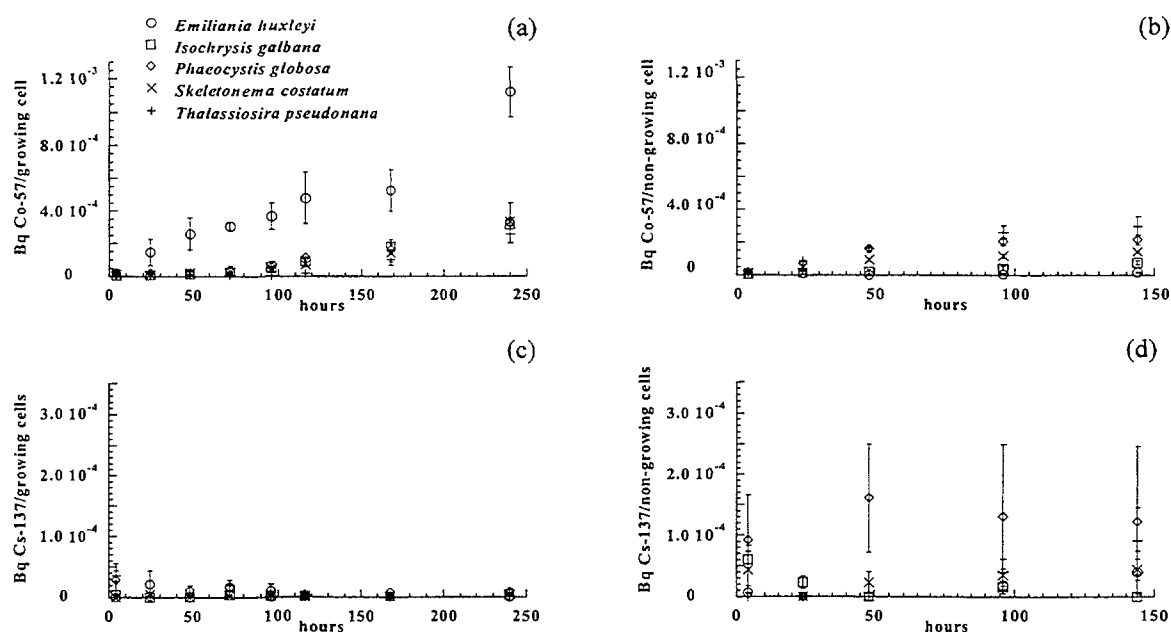


Fig. 1. (a) Bq  $^{57}\text{Co}$ /cell, growing cells, (b) Bq  $^{57}\text{Co}$ /cell, non-growing cells, (c) Bq  $^{137}\text{Cs}$ /cell, growing cells, (d) Bq  $^{137}\text{Cs}$ /cell, non-growing cells.

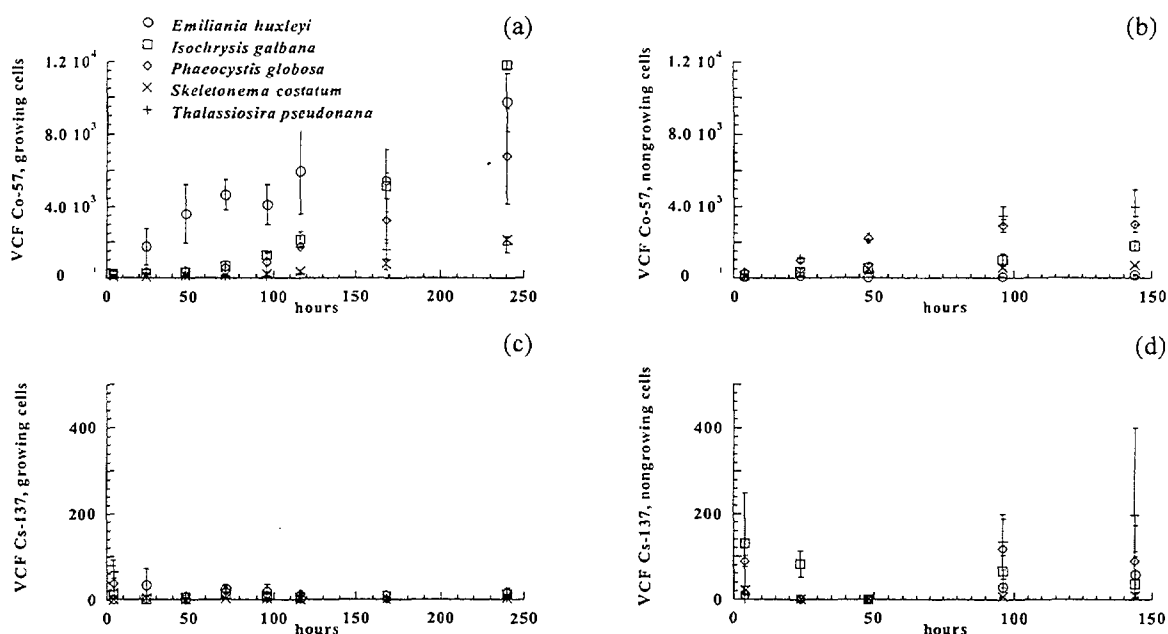


Fig. 2. (a) VCF  $^{57}\text{Co}$  (volume concentration factors, radioactivity  $\mu\text{m}^{-3}$  cell divided by radioactivity dissolved  $\mu\text{m}^{-3}$  seawater), growing cells, (b) VCF  $^{57}\text{Co}$  non-growing cells, (c) VCF  $^{137}\text{Cs}$ , growing cells, (d) VCF  $^{137}\text{Cs}$ , non-growing cells.

(Fig. 2b). The VCFs for  $^{57}\text{Co}$  uptake in the growing cells generally reached higher values, from  $2 \times 10^3$  for *S. costatum* and *T. pseudonana* to  $12 \times 10^3$  for *I. galbana* (Fig. 2a).

The VCF values for  $^{57}\text{Co}$  uptake did not increase after approximately 100 h for the non-growing cells, whereas the VCFs of the growing cells continued to increase through 240 h. This may be due to different uptake mechanisms. While the uptake mechanism in non-growing cells may be a simple surface sorption, the uptake mechanism in growing cells may be an active process, where the growing cells continue to accumulate  $^{57}\text{Co}$ , perhaps internally. The DCF values for all species of growing cells ranged from  $2.5 \times 10^4$  to  $5 \times 10^5$  for  $^{57}\text{Co}$ , and they reached  $1 \times 10^3$  for  $^{137}\text{Cs}$  (Figs. 3a and c). For non-growing cells, the DCF values reached  $1.5 \times 10^4$  and  $2.5 \times 10^3$  for  $^{57}\text{Co}$  and  $^{137}\text{Cs}$ , respectively (Figs. 3b and d). Overall, the concentration factors are comparable with previously published values for these metals (IAEA, 1985; Fisher & Reinfelder, 1995).

Cs, an alkali metal, is an analog of potassium and could be thought to exchange with K and in this way enter the food chain. However, K concentrations in seawater (10 mM) far exceed Cs concentrations (2 nM) and consequently Cs uptake through K uptake channels is negligible. Although the concentration of stable Cs in water is small, it is large enough to reduce the specific activity of radioactive Cs. This can further reduce the uptake of radioactive Cs in marine phytoplankton. Our results indicate that phytoplankton are unlikely to influence the Cs build-up in marine food chains and Cs flux to deep waters. The bioconcentration of Co in phytoplankton is comparable with earlier findings;

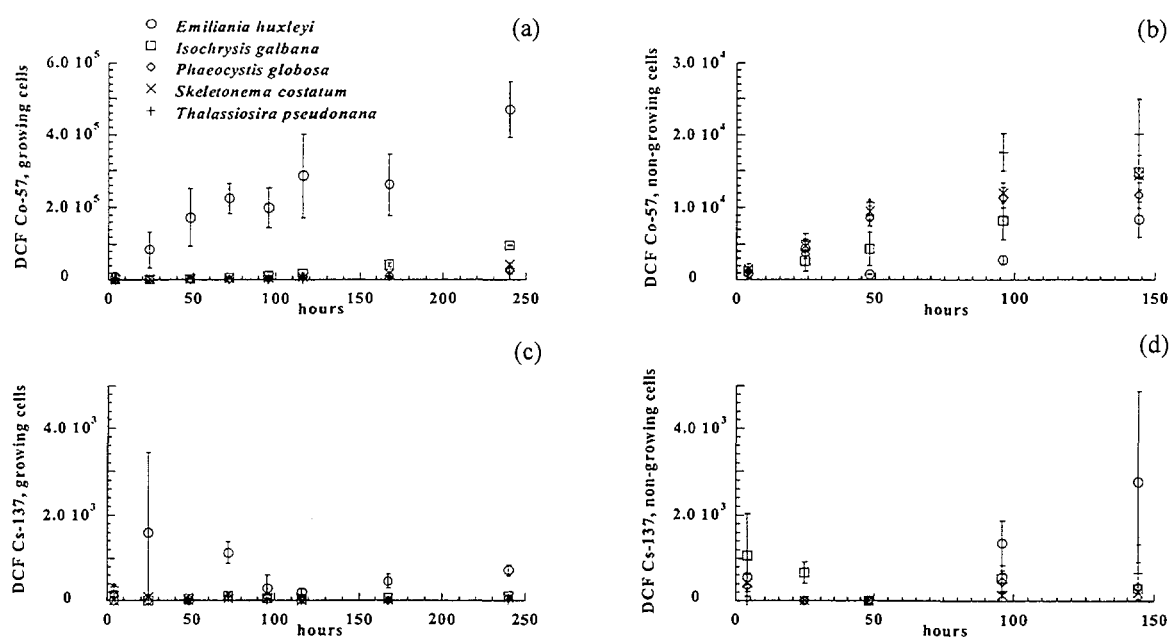


Fig. 3. (a) DCF  $^{57}\text{Co}$  (dry weight concentration factors, radioactivity  $\text{g}^{-1}$  dry cell divided by radioactivity dissolved  $\text{g}^{-1}$  seawater), growing cells, (b) DCF  $^{57}\text{Co}$  non-growing cells, (c) DCF  $^{137}\text{Cs}$ , growing cells, (d) DCF  $^{137}\text{Cs}$ , non-growing cells.

overall, Co shows moderate enrichment in phytoplankton relative to other metals (Fisher, 1986).

## Acknowledgements

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## References

- Fisher, N. S. (1986). On the reactivity of metals for marine phytoplankton. *Limnology and Oceanography*, 31, 443–449.
- Fisher, N. S., Bjerregaard, P., & Fowler, S. W. (1983). Interactions of marine plankton with transuranic elements. I. Biokinetics of neptunium, plutonium, americium, and californium in phytoplankton. *Limnology and Oceanography*, 28, 432–447.
- Fisher, N. S., & Reinfelder, J. R. (1995). The trophic transfer of metals in marine systems. In A. Tessier, & D. R. Turner (Eds.), *Metal Speciation and Bioavailability in Aquatic Systems* (pp. 363–406). Chichester: Wiley.
- Fisher, N. S., & Schwarzenbach, R. P. (1978). Fatty acid dynamics in two clones of a marine diatom: Implications for physiological ecology. *Journal of Phycology*, 14, 143–150.
- Guegueniat, P., Kershaw, P., Herrmann, J., & Bailly du Bois, P. (1997). New estimation of La Hague contribution to the artificial radioactivity of Norwegian waters (1992–1995) and Barents Sea (1992–1997). *Science of the Total Environment*, 202, 249–266.

- Guillard, R. R., & Ryther, J. H. (1962). Studies on marine planktonic diatoms 1. *Cyclotella nana* Hustedt and *Detonula confervacea* (Cleve) Gran. *Canadian Journal of Microbiology*, 8, 229–239.
- Herrmann, J., Kershaw, P. J., Bailly du Bois, P., & Guegueniat, P. (1995). The distribution of artificial radionuclides in the English Channel, southern North Sea, Skagerrak and Kattegat 1990–1993. *Journal of Marine Systems*, 6, 427–456.
- IAEA (1985). *Sediment K<sub>d</sub>'s and concentration factors for radionuclides in the marine environment*. Technical Report Series No. 247. Vienna: International Atomic Energy Agency.
- Kershaw, P., & Baxter, A. (1995). The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep-Sea Research*, 42, 1413–1448.

## **PAPER III**

# Bioaccumulation of $^{137}\text{Cs}$ in pelagic food webs in the Norwegian and Barents Seas

Hilde Elise Heldal<sup>1\*</sup>, Lars Føyn<sup>1</sup>, Per Varskog<sup>2</sup>

<sup>1</sup>Institute of Marine Research

PO Box 1870 Nordnes

N-5817 Bergen, Norway

Telephone: +47 55 23 63 34

Fax: +47 55 23 85 84

\*Contact e-mail: hilde.elise.heldal@imr.no

<sup>2</sup>Institute for Energy Technology,

PO Box 40,

N-2007 Kjeller, Norway

## Abstract

Knowledge and documentation of the levels of radioactive contamination in fish stocks important to Norwegian fisheries is of major importance to Norwegian consumers and fish export industry. In the present study, the bioaccumulation of caesium-137 ( $^{137}\text{Cs}$ ) has been investigated in marine food webs in the Barents and Norwegian Seas. The contents of  $^{137}\text{Cs}$  in these food webs were generally low ( $< 1 \text{ Bq kg}^{-1}$  wet weight), but a marked bioaccumulation was apparent. The concentration of  $^{137}\text{Cs}$  was about 10-fold higher in the harbour porpoise *Phocoena phocoena*, representing the upper level of the food web, than in the amphipod *Themisto* sp., representing the lower level of the food web. The Concentration Factors ( $\text{CF} = \text{Bq kg}^{-1} \text{ wet tissue} / \text{Bq L}^{-1} \text{ seawater}$ ) increased from  $10 \pm 3$  for a mixed sample of krill and amphipods to  $167 \pm 5$  for harbour porpoises.

Key words: Caesium-137; Bioaccumulation; Marine food webs; The Barents Sea; The Norwegian Sea

## 1. Introduction

The Barents Sea and the polar front in the Norwegian Sea have some of the richest fishing grounds in the world. Knowledge and documentation of the levels of radioactive contamination in fish inhabiting these areas is of major importance to Norwegian consumers and fish export industry. Several authors have reported levels of anthropogenic radionuclides in fish, crustaceans and seaweed in the Barents and Norwegian Seas (e.g. Aarkrog, Baxter, Bettencourt, Bojanowski, Bologna, Charmasson et al., 1997; Brungot, Sickel, Bergan, Bøe, Hellstrøm & Strand, 1997; Rissanen, Ikaheimonen, Matishov & Matishov, 1997; Brungot, Føyn, Carroll, Kolstad, Brown, Rudjord et al., 1999; Fisher, Fowler, Boisson, Carroll, Rissanen, Salbu et al., 1999). There are, however, few, if any, published studies that describe the variation in radionuclide concentrations between different trophic levels in food webs in these areas. The purpose of the present study was to measure the concentration of caesium-137 ( $^{137}\text{Cs}$ ) in different trophic levels of the Barents and Norwegian Seas food webs, and to determine whether the concentration increases from low to high trophic levels, i.e. determine if  $^{137}\text{Cs}$  is bioaccumulated in these food webs.

The main sources of  $^{137}\text{Cs}$  are the Baltic Sea outflow, which contains significant amounts of  $^{137}\text{Cs}$  originating from the Chernobyl accident (Herrmann, Kershaw, Bailly du Bois & Guegueniat, 1995; Kershaw & Baxter, 1995; Kershaw, Gurbutt, Woodhead, Leonard & Rees, 1997) and the two nuclear reprocessing plants Sellafield (UK) and La Hague (France), which discharge radioactive wastes into the Irish Sea and the English Channel, respectively. Other potential sources of radioactive contamination in the Norwegian and Barents Seas include global fallout, dumping of radioactive material in the Barents and Kara Seas off northern Russia, fallout from nuclear weapons testing by the former Soviet Union near Novaya Zemlya, and discharges into Russian rivers draining into the Barents and Kara Seas (e.g. Aarkrog, 1998).

The first trophic level in a pelagic food web consists of phytoplankton. Due to difficulties in collecting large enough phytoplankton samples in the field for quantification of  $^{137}\text{Cs}$ , uptake experiments of  $^{137}\text{Cs}$  in five marine phytoplankton species representative for the phytoplankton communities of the Barents and Norwegian Seas were performed in the laboratory. Results from these experiments have been published by Heldal, Stupakoff & Fisher (2001).

Species investigated in the present study have been assigned to different trophic levels according to their sizes and feeding habits (Figure 1). These assignments must be regarded as



rough approximations, as, for example, one particular species may belong to one trophic level at young age and a higher trophic level at older age. The second trophic level (herbivores) is represented by the copepod *Calanus finmarchicus*, amphipods and krill. Amphipods and krill may, in some cases, be carnivores, but that is neglected in this study. Several species represent the third trophic level (planktivores): fry of saithe (*Pollachius virens*), herring larvae, juvenile and adult herring (*Clupea harengus*), capelin (*Mallotus villosus*), mackerel (*Scomber scombrus*), redfish (*Sebastes marinus*), the cephalopod *Gonatus fabricii* and the shrimp *Pandalus borealis*. These species represent different levels in the community of planktivores as their sizes determine the size of their prey items. For example, juvenile herring feeds mostly on the early copepodit stages while adult herring feeds on krill, adult copepods and fish larvae. Capelin and mackerel feed on krill, amphipods and fish larvae. *G. fabricii* feeds mainly on amphipods and copepods (Bjørke, 1995). The fact that *G. fabricii* may feed on lantern fish (*Myctophiformes* sp.) and juvenile *G. fabricii* (cannibalism), i.e. the organism is piscivore in some cases, is neglected in this study. *P. borealis* prey on pelagic copepods and krill and benthic organisms as polychaetes and echinoderms (Hopkins, Sargent & Nilssen, 1993). The fourth trophic level (piscivores) is represented by cod (*Gadus morhua*). Important prey items to cod are capelin, herring, shrimp and juvenile cod (cannibalism). Harbour porpoises (*Phocoena phocoena*) represent the fifth and upper trophic level in this study. Harbour porpoises generally prey on small coastal fishes such as herring, capelin and cod (Bjørge et al., 1991). The benthic organisms polychaetes and echinoderms, belonging to the second trophic level, are represented in this study by a brittle worm belonging to the Scalibregmidae family and the sea star *Hippasteria phrygiana*, respectively. Echinoderms may also be carnivores. Their main predators are gadoid fish, in particular cod.

## 2. Materials and methods

All biological samples, with the exception of harbour porpoises, were collected during four cruises conducted by the Institute of Marine Research (IMR): aboard R/V 'Michael Sars' in July 1998 and aboard R/V 'Johan Hjort' in June/July 1999, September 1999 and May/June 2000. The samples were collected in the Norwegian Sea and western Barents Sea within the geographical boundaries 66° 00' N - 76° 00' N and 14° 35' W - 37° 00' E. Zooplankton were collected using vertical tows at selected depth intervals by means of a WP-2 plankton net (56 cm opening, 180 µm mesh size). Fish and benthic organisms were collected by means of pelagic and bottom trawls. Harbour porpoises were collected opportunistically by commercial

bottom-set gillnet fisheries along the northern coast of Norway between April and June 1999. The methodical details are given in Tolley & Heldal (2001). All samples were stored deep-frozen and transported to IMR where they were subsequently ground, freeze-dried and homogenized. Fry and juvenile fish were ground whole, while skin and bone were removed from the larger fish. The sample sizes varied from 20 g to 150 g dry weight. A selection of the samples was ashed at 450° C at Institute for Energy Technology (IFE). Non-ashed and ashed samples were filled into polyethylene counting boxes of appropriate size and analysed for  $^{137}\text{Cs}$  using high-resolution gamma spectrometry at IMR and IFE, respectively. The detection limits ranged from 0.02 Bq kg<sup>-1</sup> ww (wet weight) to 0.10 Bq kg<sup>-1</sup> ww, depending on sample size. The counting times varied from 24 to 72 hours.

Seawater samples were collected aboard R/V 'Johan Hjørt' in May/June 2000. 100-150 L surface (5 m) seawater was collected with a shipboard pump for each sample. Caesium- $^{137}$  was extracted by pumping the samples, acidified to pH ~ 2 with nitric acid, through cartridges (50 ml plastic syringes) containing the Cs-sorbent ASG (ammonium-duodecamolybdophosphate) on silica gel. The cartridges were gamma-counted on NaI well detectors at CEFAS (The Centre for Environment, Fisheries & Aquaculture Science), UK. Details of the method and its efficiency are described by Baker (1975) and the method's quality assurance procedures are reported by Steele (1989).

The Concentration Factor (CF) for each species was calculated by dividing the concentration of  $^{137}\text{Cs}$  in each species by the mean  $^{137}\text{Cs}$  concentration found in seawater (CF = Bq kg<sup>-1</sup> wet tissue / Bq L<sup>-1</sup> seawater).

### 3. Results and discussion

The concentrations of  $^{137}\text{Cs}$  were low in all investigated species (Table 1). The lowest concentrations were found in copepods, amphipods and krill (varying from below the detection limit to  $0.06 \pm 0.02$  Bq kg<sup>-1</sup> ww), and the highest concentrations were found in harbour porpoises (varying from  $0.29 \pm 0.01$  Bq kg<sup>-1</sup> ww to  $0.69 \pm 0.01$  Bq kg<sup>-1</sup> ww). Two benthic organisms, a bristle worm belonging to the Scalibregmidae family and the sea star *H. phrygiana*, had higher  $^{137}\text{Cs}$  concentrations than expected (Table 1). These species feed on sedimentary particles. As the  $^{137}\text{Cs}$  concentrations are higher in sediments (ranging from < 1.0 Bq kg<sup>-1</sup> to 8.6 Bq kg<sup>-1</sup> in the Barents Sea; Føyn & Sværen, 1997) than in seawater, it is likely that benthic filter feeders would contain elevated  $^{137}\text{Cs}$  concentrations.

The mean concentration of  $^{137}\text{Cs}$  in seawater was  $3.0 \pm 0.2 \text{ mBq L}^{-1}$  (Table 2). There was no apparent relation between the concentrations of  $^{137}\text{Cs}$  in seawater and salinities ( $r = 0.2$ ); the arithmetic mean for all seawater samples was therefore used as an approximate for the concentration of  $^{137}\text{Cs}$  in seawater throughout the areas investigated. CFs were calculated for all species except the two benthic organisms, as they probably have received substantial amounts of  $^{137}\text{Cs}$  from the sediments. As shown in Figure 2, there is a marked increase in the CFs (and thus the  $^{137}\text{Cs}$  concentration) from low to high trophic levels. The CFs increased from  $10 \pm 3$  for a mixed sample of krill and amphipods to  $167 \pm 5$  for harbour porpoises. Further, the CFs vary within each trophic level (Figure 2), probably as a result of different feeding habits within each trophic level. Variations in CFs and  $^{137}\text{Cs}$  concentrations within one species may be due to age-dependent variations in feeding habits, as described in the introduction. This is not particularly pronounced for any of the species investigated.

The concentrations of  $^{137}\text{Cs}$  found in marine organisms in the present study are generally comparable or slightly lower than those found by others. Rissanen et al. (1997) found  $^{137}\text{Cs}$  concentrations in arctic cod and redfish collected in the eastern Barents Sea in 1994 of  $0.9$  and  $0.4 \text{ Bq kg}^{-1} \text{ ww}$ , respectively. Brungot et al. (1997) found  $^{137}\text{Cs}$  concentrations in cod meat collected in the Barents Sea in 1995-1996 to vary from  $0.1 \text{ Bq kg}^{-1} \text{ ww}$  to  $0.6 \text{ Bq kg}^{-1} \text{ ww}$ . It may be noted that, in contrast to low concentrations of  $^{137}\text{Cs}$  in marine organisms, the concentrations of the natural radionuclide  $^{40}\text{K}$  are orders of magnitude higher (e.g. ranging from  $70$  to  $130 \text{ Bq kg}^{-1} \text{ ww}$  in fish collected in the eastern Barents Sea; Rissanen et al., 1997). Likewise, CFs found in the present study are comparable or slightly lower than those found by others. Rissanen et al. (1997) found CFs of  $150$  and  $70$  in arctic cod and redfish, respectively, collected in the eastern Barents Sea in 1994 (calculated for a seawater concentration of  $^{137}\text{Cs}$  of  $6 \text{ mBq l}^{-1}$ ). Fisher et al. (1999) report a mean CF for  $^{137}\text{Cs}$  of  $146 \pm 68$  for various pelagic fish species collected in the Kara and Barents Seas. Polikarpov (1966) reports a whole range of CFs, mostly experimental, for radioactive and stable elements in various organisms. For  $^{137}\text{Cs}$ , he reports levels of  $1-2$  and  $5-35$  for diatoms (phytoplankton) and muscle tissue from fish, respectively.

It is not possible from the present data to determine if the accumulation of  $^{137}\text{Cs}$  is caused by direct uptake from seawater, uptake via dietary sources or both pathways. Experimental uptake studies of  $^{137}\text{Cs}$  in marine phytoplankton representative for the Barents and Norwegian Seas showed that the uptake of  $^{137}\text{Cs}$  was low or even negligible (Heldal et al., 2001). The Volume Concentration Factors ( $\text{VCF} = \text{Bq } \mu\text{m}^{-3} \text{ cell} / \text{Bq } \mu\text{m}^{-3} \text{ seawater}$ ) at equilibrium were found to be  $11 \pm 5$  for  $^{137}\text{Cs}$  in growing phytoplankton cells. Heldal et al.

(2001) concluded that phytoplankton are unlikely to contribute to build-up of  $^{137}\text{Cs}$  in marine food webs. In the present study,  $^{137}\text{Cs}$  has been quantified in zooplankton. The question that arises is whether the  $^{137}\text{Cs}$  concentrations in phytoplankton are, although low, high enough to introduce  $^{137}\text{Cs}$  to higher trophic levels of the food web via zooplankton, and how important this mechanism is compared to direct uptake of  $^{137}\text{Cs}$  from seawater. The pathways and mechanisms for uptake of radiocaesium into food webs are a subject of controversy (Rowan & Rasmussen, 1994). There is, however, a general agreement that food is the major route of  $^{137}\text{Cs}$  uptake by fish (Rowan & Rasmussen, 1994). Several authors have also investigated uptake from seawater. Baptist & Price (1962) showed with the aid of laboratory experiments that  $^{137}\text{Cs}$  was accumulated readily from both seawater and dietary sources. Although these experiments were performed in the laboratory, it is likely that fish in the Barents and Norwegian Sea also may accumulate  $^{137}\text{Cs}$  via both pathways.

Caesium is chemically similar to potassium (K). K is a major component in cell metabolism. Uptake of  $^{137}\text{Cs}$  in fish may take place via K uptake channels. K concentrations in seawater with salinity  $\sim 35$  (10 mM) far exceed stable Cs concentrations (2 nM), and consequently Cs uptake through K uptake channels is negligible. Although the concentration of stable Cs in water is small compared to the concentration of K, it is large enough to reduce the specific activity of  $^{137}\text{Cs}$ . This further reduces the uptake of  $^{137}\text{Cs}$  in marine organisms. In contrast to the low levels of  $^{137}\text{Cs}$  in marine fish, levels above  $1500 \text{ Bq kg}^{-1} \text{ ww}$  were measured in freshwater fish from lakes in Norway and Sweden after the Chernobyl accident (Håkanson & Andersson, 1992; Hessen, Skurdal, Hegge & Andersen, 2000).

The  $^{137}\text{Cs}$  concentration in marine organisms appears to be consistent with the  $^{137}\text{Cs}$  concentration in seawater. The concentrations of  $^{137}\text{Cs}$  in seawater vary depending on distance from the sources (Herrmann et al., 1995; Kershaw & Baxter, 1995). In Norwegian and adjacent waters, the southern part of the North Sea, Skagerrak and coastal areas have the highest degree of contamination due to the nearby locations of point sources for  $^{137}\text{Cs}$  (Kershaw et al., 1997). The northern North Sea is less contaminated because the distance from these sources is greater, while the Norwegian and Barents Seas have a low degree of contamination due to the large distance from Sellafield and the Baltic Sea (Føyn & Sværen 1997; Kershaw et al., 1997). Sources in northern Russia do not appear to have a large effect on contamination levels in the Norwegian and Barents Seas because of the current systems transporting contamination from these sources eastwards. The  $^{137}\text{Cs}$  concentration in fish is higher in more contaminated areas such as the Irish and Baltic Seas. For example, the mean  $^{137}\text{Cs}$  concentration in cod collected in the Sellafield coastal area in the Irish Sea in 1999 was

14 Bq kg<sup>-1</sup> ww (FSA & SEPA, 2000). The concentrations of <sup>137</sup>Cs in Baltic fish collected in the early 1990s were approximately 12-22 Bq kg<sup>-1</sup> ww (Holm, 1994). The concentration of <sup>137</sup>Cs in fish from the Skagerrak is also higher (~ 1 Bq kg<sup>-1</sup> ww; Brungot et al., 1997) than the <sup>137</sup>Cs concentration in fish from the Norwegian and Barents Sea. A study of the <sup>137</sup>Cs concentration in harbour porpoises along the Norwegian coast showed that the concentrations of <sup>137</sup>Cs were approximately 5 times higher along the southwestern coast of Norway than along the northern coast of Norway (Tolley & Heldal, 2001).

It can be concluded that the concentrations of <sup>137</sup>Cs in all areas important to Norwegian fisheries are low at present, and the lowest levels occur in the Barents and Norwegian Seas. A marked bioaccumulation of <sup>137</sup>Cs was, however, observed in food webs inhabiting these ocean areas. The sinking of the Russian nuclear submarine 'Kursk' in the Barents Sea in August 2000 renewed the public concern about possible radioactive contamination of the marine environment, and reminded us of how exposed Norwegian fisheries are to rumours of possible radioactive contamination. It is therefore of great importance to Norwegian fisheries to be able to document the levels of contamination at any time. It is also important to understand various mechanisms for accumulation of radioactive contaminants in food webs on a broad level.

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## References

- Aarkrog, A., Baxter, M. S., Bettencourt, A. O., Bojanowski, R., Bologna, A., Charmasson, S., Cunha, I., Delfanti, R., Duran, E., Holm, E., Jeffree, R., Livingston, H. D., Mahapanyawong, S., Nies, H., Osvath, I., Pingyu, Li, Povinec, P. P., Sanchez, A., Smith, J. N., & Swift, D. (1997). A Comparison of Doses from  $^{137}\text{Cs}$  and  $^{210}\text{Po}$  in Marine Food: A Major International Study. *Journal of Environmental Radioactivity*, 34 (1), 69-90.
- Aarkrog, A. (1998). A retrospect of anthropogenic radioactivity in the global marine environment. *Radiation Protection Dosimetry*, 75, 23-31.
- Baker, C. W. (1975). The determination of radiocaesium in sea and freshwater. *Fisheries Research Technical Report, No. 16*, MAFF, Directorate of Fisheries Research, Lowestoft, UK, 7 pp.
- Baptist, J. P., & Price, T. J. (1962). Accumulation and retention of cesium $^{137}$  by marine fishes. *US Fish and Wildlife Service, Fishery Bulletin*, 206, 177-187.
- Bjørge, A., Aarefjord, H., Kaarstad, S., Kleivane, L., & Øien, N. (1991). Harbour porpoise *Phocoena Phocoena* in Norwegian waters. *ICES CM/N:16*, 24 pp.
- Bjørke, H. (1995). Norwegian investigations on *Gonatus Fabricii* (Lichtenstein). *ICES CM/K:12*, 13 pp.
- Brungot, A. L., Sickel, M. A. K., Bergan, T. D., Bøe, B., Hellstrøm, T., & Strand, P. (1997). Radioactive contamination of the marine environment. *Report no. 2 from the national surveillance programme*, Norwegian Radiation Protection Authority, NRPA Report 1997:3.
- Brungot, A. L., Føyn, L., Carroll, J. L., Kolstad, A. K., Brown, J., Rudjord, A.L., Bøe, B., & Hellstrøm, T. (1999). Radioactive contamination in the marine environment. *Report no. 3 from the national surveillance programme*, Norwegian Radiation Protection Authority, NRPA Report 1999:6.
- Fisher, N. S., Fowler, S. W., Boisson, F., Carroll, J., Rissanen, K., Salbu, B., Sazykina, T. G., & Sjöblom, K.-L. (1999). Radionuclide Bioconcentration Factors and Sediment Partition Coefficients in Arctic Seas Subject to Contamination from Dumped Nuclear Wastes. *Environmental Science & Technology*, 33 (12), 1979-1982.
- FSA & SEPA (2000). *Radioactivity in Food and the Environment, 1999 RIFE-5*. Food Standards Agency & Scottish Environment Protection Agency, London (176 pp).
- Føyn, L., & Sværen, I. (1997). Distribution and sedimentation of radionuclides in the Barents Sea. *ICES Journal of Marine Sciences*, 54, 333-340.

- Heldal, H. E., Stupakoff, I., & Fisher, N. S. (2001). Bioaccumulation of  $^{137}\text{Cs}$  and  $^{57}\text{Co}$  by five marine phytoplankton species. *Journal of Environmental Radioactivity*, 57 (3), 231-236.
- Herrmann, J., Kershaw, P. J., Bailly du Bois, P., & Guegueniat, P. (1995). The distribution of artificial radionuclides in the English Channel, southern North Sea, Skagerrak and Kattegat, 1990-1993. *Journal of Marine Systems*, 6, 427-456.
- Hessen, D. O., Skurdal, J., Hegge, O., & Andersen, T. (2000). Modelling ecological half-lives for radiocaesium in Norwegian brown trout populations. *Journal of Applied Ecology*, 37, 109-116.
- Holm, E. (1994). Polonium-210 and radiocaesium in muscle tissue of fish from different Nordic marine areas. In H. Dahlgaard (Ed.), *Nordic Radioecology: The transfer of radionuclides through Nordic ecosystems to man* (pp. 119-126). Amsterdam: Elsevier.
- Hopkins, C. C. E., Sargent, J. R., & Nilssen, E. M. (1993). Total lipid content and lipid and fatty acid composition of the deep-water prawn *Pandalus borealis* from Balsfjord, northern Norway: growth and feeding relationships. *Marine Ecology Progress Series*, 96, 217-228.
- Håkanson, L., & Anderson, T. (1992). Remedial measures against radioactive Caesium in Swedish lake fish after Chernobyl. *Aquatic Sciences*, 54 (2), 141-164.
- Kershaw, P. J., & Baxter, A. (1995). The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep-Sea Research Part II*, 42, 1413-1448.
- Kershaw, P. J., Gurbutt, P., Woodhead, D., Leonard, K., & Rees, J. (1997). Estimates of fluxes of  $^{137}\text{Cs}$  in northern waters from recent measurements. *The Science of the Total Environment*, 202, 211-223.
- Polikarpov, G. G. (1966). *Radioecology of aquatic organisms. The accumulation and biological effect of radioactive substances* (314 pp.). Amsterdam: North-Holland Publishing Company.
- Rissanen, K., Ikaheimonen, T. K., Matishov, D., & Matishov, G.G. (1997). Radioactivity levels in Fish, Benthic Fauna, Seals and Sea Birds Collected in the Northwest Arctic of Russia. *Radioprotection-Colloques*, 32, C2-323-331.
- Rowan, D. J., & Rasmussen, J. B. (1994). Bioaccumulation of Radiocaesium by fish: the Influence of Physicochemical Factors and Trophic Structure. *Canadian Journal of Fisheries and Aquatic Sciences*, 51, 2388-2410.
- Steele, A. K. (1989) Laboratory intercomparison of  $^{137}\text{Cs}$  in sea water. *Fisheries Technical Note, No. RL1/89*, MAFF, Directorate of Fisheries Research, Lowestoft, UK, 25 pp.

Tolley, K. A., & Høidal, H. E. (2001). Inferring ecological separation from regional differences in radioactive caesium in harbour porpoises (*Phocoena phocoena*). *Marine Ecology Progress Series*. In press.



Table 1. Concentration of  $^{137}\text{Cs}$  in marine organisms in the Norwegian and Barents Seas. Samples were taken in 1998, 1999 and 2000. Analytical uncertainties are given as follows: n = 1: total analytical error; n = 2, 3: rooted square sum of individual sample errors; n > 3: Standard Error of the Mean.

Species	Trophic level	$^{137}\text{Cs}$ (Bq kg <sup>-1</sup> (wet weight))			n
		Min	Max	Mean	
Copepods ( <i>Calanus finmarchicus</i> )	2	bd	bd	-	3
Amphipods ( <i>Themisto</i> sp.)	2 (3)	bd	0.06 ± 0.02	-	2
Krill (Euphausiacea); Amphipods ( <i>Themisto</i> sp.) <sup>a</sup>	2 (3)	bd	0.03 ± 0.01	-	2
Polychaetes (Scalibregmidae)	2	-	0.19 ± 0.04	-	1
Echinoderms ( <i>Hippasteria phrygiana</i> )	2 (3)	-	0.28 ± 0.08	-	1
Cephalopods ( <i>Gonatus fabricii</i> )	3 (4)	bd	0.08 ± 0.04	0.05 ± 0.02	7
Decapods ( <i>Pandalus borealis</i> )	3	-	0.10 ± 0.05	-	1
Fry of Saithe ( <i>Pollachius virens</i> )	3	bd	0.15 ± 0.02	0.09 ± 0.01	13
Herring larvae ( <i>Clupea harengus</i> )	3	bd	0.14 ± 0.04	0.12 ± 0.01	5
Juvenile Herring ( <i>Clupea harengus</i> )	3	0.11 ± 0.07	0.13 ± 0.08	0.12 ± 0.11	2
Herring ( <i>Clupea harengus</i> )	3	bd	0.20 ± 0.08	0.16 ± 0.02	5
Capelin ( <i>Mallotus villosus</i> )	3	0.04 ± 0.01	0.06 ± 0.01	0.05 ± 0.01	2
Mackerel ( <i>Scomber scombrus</i> )	3	bd	0.24 ± 0.08	0.19 ± 0.09	3
Redfish ( <i>Sebastes marinus</i> )	3	-	0.09 ± 0.05	-	1
Cod ( <i>Gadhus morhua</i> )	4	0.21 ± 0.04	0.22 ± 0.02	0.22 ± 0.04	2
Harbour porpoise ( <i>Phocoena phocoena</i> )	5	0.29 ± 0.01	0.69 ± 0.01	0.50 ± 0.02	19

bd – below detection limit

<sup>a</sup> – mixed sample

Table 2. Concentration of  $^{137}\text{Cs}$ , temperature and salinity in seawater at five locations in the Norwegian Sea. Errors quoted are  $\pm$  one  $\sigma$  (sigma) percentage counting propagated errors only. The uncertainty estimate for the mean value is given as the Standard Error of the Mean.

Sample ID	Position		Temperature	Salinity	$^{137}\text{Cs}$ (mBq L <sup>-1</sup> )
	Latitude	Longitude			
389	74° 30' N	11° 43' W	-0.6528	34.3273	4.4 $\pm$ 0.4
396	74° 30' N	00° 00'	0.4109	34.8295	3.5 $\pm$ 0.4
404	74° 30' N	14° 00' E	6.0124	35.0742	3.1 $\pm$ 0.4
412	74° 30' N	18° 30' E	0.3922	34.6277	2.0 $\pm$ 0.4
426	75° 46' N	16° 35' E	0.8477	34.0228	2.4 $\pm$ 0.4
428	75° 39' N	14° 35' E	0.5375	33.6898	2.6 $\pm$ 0.4
Mean value					3.0 $\pm$ 0.2

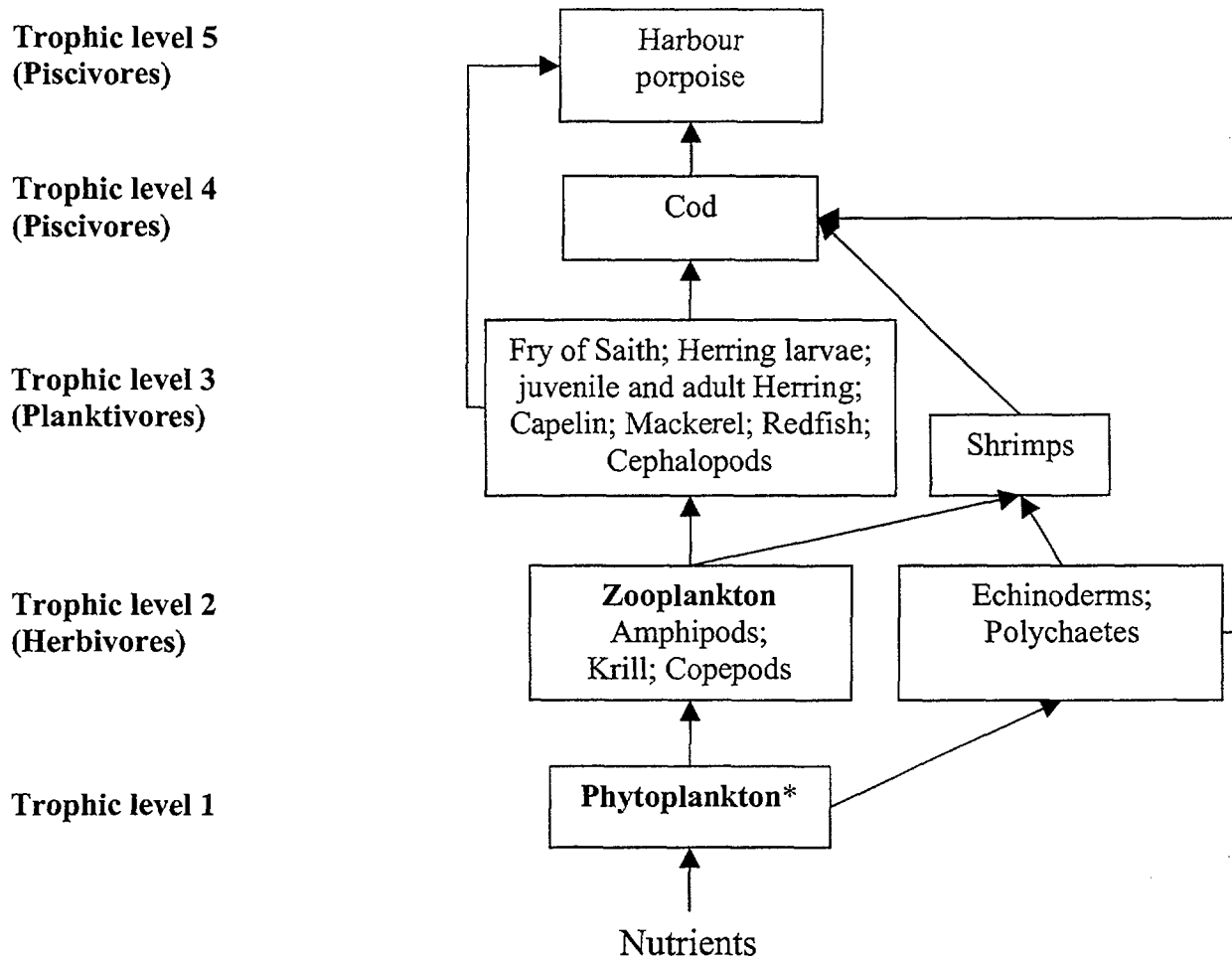


Figure 1. Investigated species assigned to different trophic levels. \*Phytoplankton data have been taken from Høidal et al. (2001).

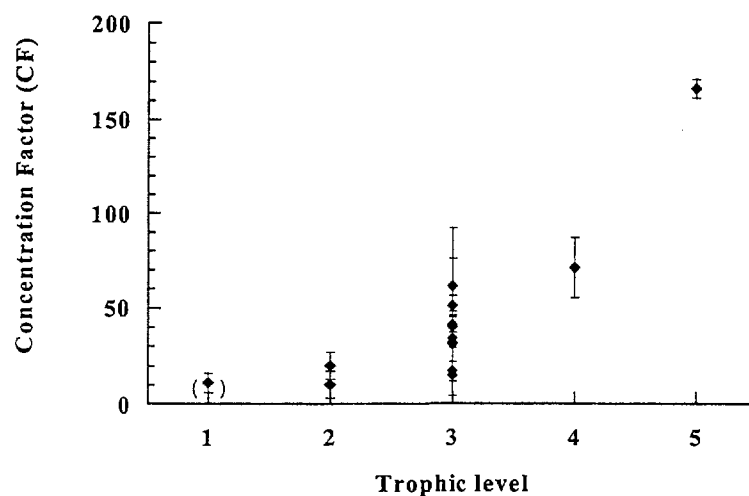


Figure 2. Concentration Factors ( $CF = \text{Bq kg}^{-1} \text{ wet tissue} / \text{Bq kg}^{-1} \text{ seawater}$ ) for various trophic levels of the Barents and Norwegian Seas food webs. The CFs were calculated individually for each biotic sample. The CFs shown are mean values. Uncertainty estimates for each species are calculated the same way as for the  $^{137}\text{Cs}$  concentrations in Table 1. ( ): experimental data from Heldal et al. (2001).

## **PAPER IV**

Inferring ecological separation from regional differences in radioactive caesium in harbour porpoises (*Phocoena phocoena*).

Krystal A. Tolley<sup>1</sup> and Hilde Elise Heldal<sup>2</sup>

<sup>1</sup>Marine Mammal Division and <sup>2</sup>Marine Chemistry Division

Institute of Marine Research

P.O. Box 1870 Nordnes

5817 Bergen, Norway

Telephone: +47-55 23 8619

Fax: +47-55 23 8617

Contact e-mail: [krystal@imr.no](mailto:krystal@imr.no)

**Abstract:** Geographic variation in the level of radioactive caesium-137 (<sup>137</sup>Cs) was examined from axial muscle of 36 bycaught harbour porpoises from five locations along the coast of Norway, ranging from southern Norway (North Sea) to northern Norway (Barents Sea). Levels of <sup>137</sup>Cs in seawater and sediments have been found to differ along the coast of Norway due to distance from point sources, such as the Sellafield nuclear fuel reprocessing plant in Cumbria, UK, discharging into the Irish Sea, and outflow from the Baltic Sea, which contains significant amounts of Chernobyl originated <sup>137</sup>Cs. Radioactive caesium is accumulated in muscle in a manner related to ambient levels in the environment through exposure and/or ingestion of contaminated prey items. Hence, we hypothesised that if harbour porpoises are philopatric, their muscle tissue would reflect concentrations of <sup>137</sup>Cs in their area of residence. The levels found in harbour porpoise muscle were consistent with that found in the area from which they were caught. Porpoises from the Barents Sea had significantly lower levels of <sup>137</sup>Cs than porpoises from the west and south coasts of Norway, reflecting concentration patterns of <sup>137</sup>Cs in the waters of those areas. This suggests ecological separation of harbour porpoises in these regions, and that porpoises are limited in their movements along the coast of Norway.

**KEY WORDS:** population structure. harbour porpoise. caesium-137.

## INTRODUCTION

Elucidating population structure for species threatened by human activities is critical for formulating conservation and management plans. However, it is often difficult to create *a priori* hypotheses regarding population structure especially for species whose distribution is not delimited by obvious natural zoogeographic barriers. Further, many marine species have the potential for high dispersal and gene flow, making population structure difficult to detect (Palumbi 1994, Hoelzel 1998, Waples 1998) even if hypotheses of their structure can be formulated. Despite such homogenising qualities, marine species inhabiting geographically different areas can be evolutionarily and/or demographically independent, and depending upon management objectives, may require separate management plans (Taylor & Dizon 1999). Consequently, information on population structure is essential to manage such species effectively to ensure that any one particular population does not sustain disproportionately high anthropogenic impacts.

There appears to be no universally held definition for a 'population' (sometimes called a 'biological stock'), but generally a population can be characterised as a group of interbreeding individuals living in a limited geographic area (see review in Carvalho & Hauser 1994, Hartl & Clark 1997). If gene flow between populations is minimal, individual populations will be on separate demographic and evolutionary tracks (Waples 1998). Eventually, such populations will become genetically distinct and genetic markers can be useful to identify population structure. However, genetic markers may fail to detect population structure when separation is too recent for genetic differences to have accumulated between the populations, or if gene flow is high enough to prevent detectable differences from accumulating (Slatkin 1987, Waples 1998). In such instances, ecological markers may be useful for making inferences regarding contemporary population structure. However, caution in the interpretation of ecological separation must be used. Such markers are acquired rather than inherited, and therefore do not necessarily indicate the presence of populations but instead suggest where an individual has spent a portion of its lifetime (A vise 1994). Hence, such information reflects contemporary spatial distribution of the individuals. Regardless, ecological markers can be used to assist in creating hypotheses regarding population structure when genetic markers fail to detect differences.

The harbour porpoise (*Phocoena phocoena*) suffers heavy mortality from bycatch in commercial gillnet fisheries. In some regions, it has been suggested that bycatch rates are higher

than intrinsic rates of population growth (Jefferson & Curry 1994, Tregenza *et al.* 1997, Anonymous 2000a, Anonymous 2000b), the effect of which would be declining populations. In many other areas, bycatch is known to exist but has not been quantified (IWC 1996), hence the effects of bycatch in these areas are virtually unknown. Although conservation actions are in order for this species, without a full understanding of the population structure, bycatch mitigation plans are unlikely to be effective because such measures may not be applied to units that are biologically meaningful. For harbour porpoises, the difficulty in identifying such units lies in the spatial distribution of this species and the high potential for dispersal. Harbour porpoises are found in the coastal waters of the North Pacific, North Atlantic, and the Black Sea. In the North Atlantic, they are distributed in patches of varying density from Senegal in West Africa, northward into European waters and westward to Iceland, Greenland, and North America as far south as North Carolina. Their distribution is concentrated around landmasses and islands, but they are occasionally observed in the deep oceanic expanses dividing these landmasses (Donovan & Bjørge 1995). Hence, there is a potential for these concentrations of porpoises to form distinct populations, but remain connected by corridors through which gene flow may occur.

In the North Atlantic, harbour porpoises have been divided into thirteen putative populations by the International Whaling Commission (IWC 1996). The designations were based on the distribution and abundance of porpoises, plus oceanographic features that could potentially restrict gene flow (Donovan & Bjørge 1995). Recently, ecological and genetic markers have been used to gain insight into the structuring of the North Atlantic populations. These studies have supported some of the IWC designations, especially in the western North Atlantic (Wang *et al.* 1996, Read & Westgate 1997, Rosel *et al.* 1999, Westgate & Tolley 1999, Tolley *et al.* In press). Conversely, morphometric, genetic, and ecological data have lent support to the presence of fine-scale population structure in the eastern North Atlantic that is not captured by the IWC definitions (*e.g.* Andersen 1993, Kleivane *et al.* 1995, Tiedemann *et al.* 1996, Börjesson & Berggren 1997, Walton 1997, Wang & Berggren 1997, Berrow *et al.* 1998, Tolley *et al.* 1999).

Radionuclides can be useful biomarkers for indicating ecological separation in high trophic level marine species. Radioactive caesium-137 ( $^{137}\text{Cs}$ ) is useful in this respect, because this contaminant accumulates in muscle tissue due to its chemical similarity to potassium (Dahlgard *et al.* 1994).  $^{137}\text{Cs}$  (half-life = 30 years) is a fission product and is an important component of radioactive wastes discharged into coastal waters of Northwest Europe. The main



sources are the nuclear reprocessing plants at Sellafield (UK), which has discharged into the Irish Sea since 1952, and La Hague (France), which has discharged into the English Channel since 1966 (Kershaw & Baxter 1995, Kershaw *et al.* 1999). These discharges are transported from their sources into the North Sea, where they mix with waters from the Baltic Sea containing significant amounts of  $^{137}\text{Cs}$  originating from fallout of the Chernobyl nuclear reactor accident (Hermann *et al.* 1995, Kershaw & Baxter 1995, Føyn & Sværen 1997, Kershaw *et al.* 1997). The contamination is transported further northwards in the Norwegian Coastal Current, which splits into two branches off northern Norway: the North Cape Current, which enters the Barents Sea to the east, and the West Spitsbergen Current, which enters the Fram Strait to the west (Kershaw & Baxter 1995). Other potential sources for radioactivity in Norwegian and adjacent seas are global fallout, dumping of radioactive material in the Barents and Kara Seas off northern Russia, fallout from nuclear weapons testing near Novaya Zemlya by the former Soviet Union, and discharge into Russian rivers draining into the Barents and Kara Seas (Aarkrog 1998).

Radioactive caesium in the water column is useful for labelling water masses (Herrmann *et al.* 1995) as its concentration varies depending on distance from the source and the oceanographic forces that spread the contamination. In Norway, the southern coastal waters have the highest levels of contamination due to the nearby locations of point sources for  $^{137}\text{Cs}$  (Kershaw *et al.* 1997). The west coast of Norway is less contaminated because the distance from these sources is greater, while the northern coast of Norway has low levels of contamination due to the large distance from Sellafield and the Baltic Sea (Føyn & Sværen 1997, Kershaw *et al.* 1997). Sources in northern Russia do not appear to have a large effect on contamination levels along the coast of northern Norway because of the eastward-flowing current systems transporting contamination away from northern Norway.

In this study, we hypothesised that the level of  $^{137}\text{Cs}$  in muscle tissue of harbour porpoises should reflect the ambient concentration in the local region from which they were sampled, if porpoises are philopatric. The elimination rate of  $^{137}\text{Cs}$  from muscle is species-dependent, and has not been investigated in marine mammals. However, elimination proceeds at a slower rate with larger body size, and when at steady state with ambient levels of  $^{137}\text{Cs}$  for both poikilotherms (Mailhot *et al.* 1989, Rowan & Rasmussen 1995) and homeotherms (Mailhot *et al.* 1989). Given this general pattern, we assumed that elimination does not proceed rapidly for harbour porpoises due to their large body size and the potential for chronic exposure from the

environment. Given the distribution of  $^{137}\text{Cs}$  in the water column, we expected porpoises from the southern most regions of Norway to have the highest levels of  $^{137}\text{Cs}$ , while those from western Norway would have intermediate levels and those from the Barents Sea would have the lowest. Alternatively, if harbour porpoise movements are not restricted along the coast of Norway, levels of  $^{137}\text{Cs}$  in muscle should not show a high correspondence to geographic location.

## METHODS

Samples were collected opportunistically from porpoises caught incidentally by commercial bottom-set gillnet fisheries along the Norwegian coast between April - June 1999, and April 2000. Thirty-six porpoises were collected (13 females, 23 males) from 5 different counties in Norway (Vest-Agder ( $n = 3$ ), Rogaland ( $n = 5$ ), Hordaland ( $n = 2$ ), Nordland ( $n = 7$ ), Finnmark ( $n = 19$ ), spanning a distance of approximately 2200 kilometres (Fig. 1). Large quantities of muscle tissue are required for the analysis, therefore approximately 500-750 grams of epaxial muscle were obtained from each individual and frozen at  $-20^{\circ}\text{C}$ . The samples were subsequently ground, freeze dried, homogenized and filled into polyethylene counting boxes of appropriate size prior to analysis. The gamma-counting for  $^{137}\text{Cs}$  was performed on a Canberra HPGe-detector with 30 % efficiency, an electric cryostat cooling system, and a 10 cm lead shielding.

Three regional groupings were used to compare geographic variation of  $^{137}\text{Cs}$  levels in porpoise tissue. Due to small sample sizes in some of the regions, the 3 most geographically close counties (Vest-Agder, Rogaland, and Hordaland) were grouped together ( $n = 10$ ), and are hereafter referred to as the 'Southwest' group. Porpoises collected from the counties of Nordland ( $n = 7$ ) and Finnmark ( $n = 19$ ) were grouped separately.

The levels in tissues for some classes of contaminants have been shown to vary with age, sex, body size, and diet (Aguilar *et al.* 1999). Data on ages of individuals were not available for these samples, so the relationship of the level of  $^{137}\text{Cs}$  (Bq/kg ww; Becquerel per kilogram, wet weight) with body size was investigated by regression against standard length separately for each region. Although length is not a flawless proxy for age, length and age are positively related up to at least 5 years of age in male harbour porpoises, and 7 years of age in females (Lockyer 1995, Read & Tolley 1997). Because sample sizes were small, regressions were stratified only according to region and not by sex.

To investigate the possible implications of combining both sexes within each region, a Mann-Whitney U test was conducted between the sexes for the Finnmark sample. The comparison was limited to a single region so as not to introduce any bias due to regional differences in the level of  $^{137}\text{Cs}$ . This comparison was run only for the Finnmark group because sample sizes were largest (8 females, 11 males).

For the regional comparisons of  $^{137}\text{Cs}$  levels in porpoises, raw and log transformed data showed heterogeneity of variances among groups (Levene's test of equality of variance). Hence, a non-parametric test was used to compare levels of  $^{137}\text{Cs}$  among the three regions (Kruskal-Wallis test), and pairwise comparisons were made using a Mann-Whitney U test.

The concentration factor (CF) for each porpoise was calculated by dividing the level of  $^{137}\text{Cs}$  in each porpoise by the  $^{137}\text{Cs}$  concentrations in seawater from the region where the porpoise was caught. The  $^{137}\text{Cs}$  concentrations used were median values of those available in the literature (Brungot *et al.* 1997, Kershaw *et al.* 1997, Grøttheim 1998, Brungot *et al.* 1999). Values used were 0.020 Bq/l (Vest-Agder), 0.012 Bq/l (Rogaland), 0.012 Bq/l (Hordaland), 0.008 Bq/l (Nordland) and 0.004 Bq/l (Finnmark).

## RESULTS

Levels of  $^{137}\text{Cs}$  in porpoises from the Norwegian coast ranged from 0.3 – 3.8 Bq/kg ww (Table 1). Overall, there was a decreasing trend for  $^{137}\text{Cs}$  level and standard length, but this was due to the high levels found in the tissue of porpoises from Rogaland (Fig. 2). Within each region, there was no significant relationship between standard length of porpoises and levels of  $^{137}\text{Cs}$  (Finnmark;  $r^2 = 0.04$ ,  $p = 0.42$ ; Nordland;  $r^2 = 0.36$ ,  $p = 0.21$ ; Southwest,  $r^2 = 0.15$ ,  $p = 0.24$ ; Fig. 2).

There was no significant difference between the sexes within the Finnmark region (Mann-Whitney U test,  $p = 0.457$ ). Among the 3 geographic regions, there was a significant difference in  $^{137}\text{Cs}$  level in the muscle of porpoises (Kruskal-Wallis test,  $p = 0.001$ ; Table 2). Pairwise comparisons indicated that porpoises from Finnmark had significantly lower levels than those in the Southwest region ( $p = 0.002$ ), and those from Nordland ( $p = 0.001$ ). There was no significant differences in  $^{137}\text{Cs}$  levels between porpoises from the Southwest and Nordland ( $p = 0.961$ ). To investigate whether the high levels observed in several of the porpoises from Vest-Agder (Fig. 3) could have biased the results of the regional comparison, an additional comparison was run with

all Vest-Agder porpoises excluded from the Southwest group. Pairwise comparisons showed the same regional differences even when these Vest-Agder porpoises were excluded (Mann-Whitney U test; Finnmark vs. Southwest,  $p = 0.029$ ; Nordland vs. Southwest,  $p = 0.302$ ).

Fine-scale regional differences for all 5 counties were investigated by comparing the mean level of  $^{137}\text{Cs}$  in porpoises from each county with the ambient concentrations of  $^{137}\text{Cs}$  in seawater (Fig. 3). The levels of  $^{137}\text{Cs}$  in porpoises generally decreased from south to north (highest in Vest-Agder and lowest in Finnmark), which mirrored the concentrations found in seawater (Fig. 3). The exception was that the seawater concentration off Nordland was lower than some of the other locations in western Norway, yet the porpoises from this region did not show a corresponding lower concentration of  $^{137}\text{Cs}$  (Fig. 3). Concentration factors ranged from 29 to 176 with a mean of 105. The average is consistent with that reported for other cetaceans, although the range is wider (Calmet *et al.* 1992, Berrow *et al.* 1998, Watson *et al.* 1999).

## DISCUSSION

The levels of  $^{137}\text{Cs}$  in porpoises collected from the five geographic regions along the coast of Norway generally reflected ambient water concentrations in these same regions. Levels were highest in porpoises from southern Norway, intermediate in those from the west coast, and lowest in those from northern Norway. The exception was for Nordland, where seawater concentrations were lower than in other west coast regions (Kershaw *et al.* 1997), but the  $^{137}\text{Cs}$  concentrations in porpoise tissue were not lower (Fig. 3). However, because seawater concentrations from this region were only available from measurements taken at sea and not in the inshore waters that porpoises inhabit, there is a potential for bias in the results. In Nordland, there is a substantial amount of  $^{137}\text{Cs}$  that enters the coastal waters as run-off from the northern fjords, as this region received high levels of Chernobyl fall-out (Dahlgaard *et al.* 1994). Furthermore, this is a region where porpoises inhabit near-coastal waters because the continental shelf that they prefer is very narrow. Therefore, they could receive higher doses than expected, due to their being restricted to more contaminated coastal waters. Although the level of  $^{137}\text{Cs}$  has not yet been quantified in these near-coastal waters, the level of  $^{137}\text{Cs}$  in seaweed and sediments from the Nordland region has also been found to be slightly higher than in other regions along the west coast (Brungot *et al.* 1997). This suggests that higher concentrations than predicted based on the available seawater concentrations may be expected in porpoises from this region.

The International Whaling Commission (IWC) has proposed the presence of several populations of harbour porpoises along the Norwegian coast: (1) the Kattegat and adjacent waters, (2) the North Sea, and (3) North Norway/Barents Sea (Fig. 1). The putative division between the North Sea population and the North Norway/Barents Sea population is located on the west coast of Norway at 66°N (Fig. 1; Donovan & Bjørge 1995). Of the five collection areas in the present study, three fall into the North Sea population (Vest-Agder, Rogaland, Hordaland), two into the North Norway/Barents Sea population (Finnmark, Nordland), and none into the Kattegat and adjacent waters. The differing levels of  $^{137}\text{Cs}$  in porpoises from the Norwegian coast provide some information on the biological significance of the populations proposed by the IWC. Given the difference in  $^{137}\text{Cs}$  concentrations between Finnmark and Nordland, a single North Norway/Barents Sea population is not supported. Nordland porpoises showed significantly higher levels of  $^{137}\text{Cs}$  than those from the Barents Sea suggesting that there is little movement of porpoises between the northwest coast (Nordland) and the Barents Sea (Finnmark). Hence, the IWC's North Norway/Barents Sea population may encompass a region that is too broad to reflect population structure on an ecological scale; combining porpoises from the Barents Sea with those south to 66°N may be inappropriate.

There was no significant difference detected between the Southwest region (Vest-Agder, Rogaland, Hordaland) and Nordland. However, ambient  $^{137}\text{Cs}$  concentrations in seawater from these regions are similar, which limits the ability of this technique to detect differences between these regions. Within the Southwest group, porpoises from Vest-Agder averaged higher levels of  $^{137}\text{Cs}$  than the other regions (Fig. 3), but sample sizes in that region were too small to treat Vest-Agder as a separate group. The higher levels for Vest-Agder porpoises possibly suggests they are part of a Kattegat population to the east, an area which is more heavily contaminated with  $^{137}\text{Cs}$  than both the North Sea and the west coast of Norway (Brungot *et al.* 1999).

Porpoises from Rogaland had somewhat lower levels than those in Hordaland and Nordland (Fig. 3). Near Rogaland, the Norwegian Coastal Current (NCC), which transports  $^{137}\text{Cs}$  northwards, is relatively narrow (Sætre & Ljøen 1971). Porpoises off southwest Norway (*i.e.* Rogaland) could be moving outside the narrow NCC and into more saline North Sea or Atlantic waters, as they are known to utilise the relatively shallow offshore waters of the North Sea (Hammond *et al.* 1995). As a result, they may spend a substantial amount of time in less contaminated waters, resulting in lower levels of  $^{137}\text{Cs}$  in their tissue. At higher latitudes, the

NCC undergoes mixing with Atlantic water (Sætre & Ljøen 1971), and the zone of  $^{137}\text{Cs}$  contamination tends to extend westwards (Casso & Livingston 1984). Thus, variability associated with the NCC will be less of a factor in northern regions.

As contaminants are acquired by the individual rather than inherited, these data measure population structure on an ecological time scale, and are suggestive of ecological separation. This differs from the time scale at which genetic data measures population structure. For groups that have become isolated, it may take hundreds or thousands of generations for genetic differences to accumulate, even when highly sensitive markers such as mitochondrial DNA or microsatellites are used to quantify this variation (Palumbi *et al.* 1991). In a study of the sequence variation in the mitochondrial DNA control region, harbour porpoises from the Barents Sea and southwestern coast of Norway could not be differentiated, suggesting that porpoises in these two regions are genetically similar (Tolley *et al.* 1999). These genetic data stand in contrast to the differences found for  $^{137}\text{Cs}$  levels in porpoises from these two regions. One explanation for genetic similarity may be a recent recolonisation of the Barents Sea by harbour porpoises since the end of the last Pleistocene glaciation, approximately 10,000 years before present (Tolley *et al.* 1999, Tolley *et al.* In press). The genetic similarity of southern and northern porpoises could be the result of the short time span since recolonisation, perhaps coupled with some contemporary gene flow. However, the differences in  $^{137}\text{Cs}$  indicate ecological separation does exist, despite the absence of genetic differentiation.

Fine-scale population structure in the eastern North Atlantic has been suggested previously, and porpoises from the Kattegat-Skagerrak Seas have been found to be genetically different than those from the west coast of Norway (Wang & Berggren 1997). Furthermore, geographic variation in organochlorine levels has been detected in male harbour porpoises from the Barents Sea and the west coast of Norway (Kleivane *et al.* 1995), which suggests that porpoises do not regularly move throughout the coast of Norway, but may be restricted to relatively limited geographic regions. Further support comes from monitoring the movements of individual porpoises in the eastern North Atlantic. Satellite telemetry data from Denmark indicates that porpoises do not regularly move out of the Kattegat-Skagerrak region (Teilmann 2000). In that study, seventeen porpoises were satellite tagged off the coast of Denmark, and these individuals restricted themselves to the Kattegat throughout the study period which spanned the breeding season. Only two of the seventeen porpoises moved northwards toward the

Skagerrak Sea early in the breeding season (Teilmann 2000). Although the telemetry data cannot confirm that the Kattegat contains a separate population, it does suggest there is limited movement between the Kattegat and other regions. The situation may be similar in the Barents Sea, where four satellite tagged porpoises did not move west of 29°E throughout the breeding season (K.A. Tolley unpublished data). This suggests there is limited exchange between the Barents Sea and the northwest coast of Norway.

The results of the present study are based on a single tracer, and therefore must be interpreted with some caution. Firstly, elimination rates associated with  $^{137}\text{Cs}$  exposure have not been conducted for this species. Although it is not possible to generalise across species, experimental studies of elimination rates from fishes suggest this contaminant could remain present for years if exposure was chronic rather than acute, and body size is large (Rowan and Rasmussen 1995, Forseth *et al.* 1998). Further, some types of pollutants are known to be influenced by age, sex, and/or reproductive status (Kleivane *et al.* 1995, Westgate *et al.* 1997, Aguilar *et al.* 1999). Because sample sizes of several of the southern regions were small, it was necessary to combine individuals of all sizes and both sexes in the analysis, perhaps confounding the results. In the present study, there was no indication of a significant relationship between the level of  $^{137}\text{Cs}$  and body size suggesting that for these data, patterns of accumulation due to size (as a proxy for age) may not be sufficient to bias the analysis. Similarly, Berrow *et al.* (1998) found no strong relationship between levels of  $^{137}\text{Cs}$  and age, length, or weight in harbour porpoises from British and Irish waters. Furthermore, there was no difference between females and males from Finnmark for  $^{137}\text{Cs}$  loads, suggesting that any bias due to differences between the sexes would be limited for this sample.

The movements of prey items should be considered in the interpretation of the results because the uptake of  $^{137}\text{Cs}$  could be due to either direct ingestion of seawater, or ingestion of contaminated prey items. In the regions investigated, harbour porpoises generally prey on small coastal fishes such as herring (*Clupea harengus*), capelin (*Mallotus villosus*), pollack (*Pollachius pollachius*), cod (*Gadus morhua*), saithe (*Pollachius virens*), haddock (*Melanogrammus aeglefinus*), and whiting (*Merlangius merlangus*) (Bjørge *et al.* 1991). Although the data are limited, many of these fishes from the Barents, Norwegian, and North Seas have similar levels of  $^{137}\text{Cs}$  in their tissues, generally on the order of 0.2-0.5 Bq/kg ww in adult fish, and 0.1 Bq/kg ww or less in juvenile fish (Brungot *et al.* 1999, H.E. Høidal unpub. data). The similarities in the

levels of  $^{137}\text{Cs}$  in fishes from these regions may be due to similar levels of  $^{137}\text{Cs}$  in seawater between some of the areas and the movements associated with spawning migrations between regions of varying ambient levels of  $^{137}\text{Cs}$ , as individuals moving between regions of varying contamination will have homogeneous levels of  $^{137}\text{Cs}$  in their tissues. It is possible that some of the regional similarities in porpoise tissue are due to predation upon these fishes with homogeneous  $^{137}\text{Cs}$  levels that have migrated in from another region. Conversely, it is possible that porpoises in southern Norway have preyed upon fishes that had moved out of the more contaminated Baltic Sea. In particular, the Rügen herring spawns in the Baltic Sea but is known to move through the Skagerrak (Knijn *et al.* 1993), and the levels of  $^{137}\text{Cs}$  in both Skagerrak fishes (*ca.* 1 Bq/kg ww; Brungot *et al.* 1997) and Baltic fishes (*ca.* 12-22 Bq/kg ww; Holm 1994) are higher than fishes from the other regions. Ingestion of prey fishes containing higher levels of  $^{137}\text{Cs}$ , rather than direct exposure from the water, could have caused the elevated levels observed in the Vest-Agder porpoises.

The present results, when taken together with the differences in organochlorine loads among porpoises from the same regions (Kleivane *et al.* 1995), indicate there is ecological separation of harbour porpoises along the coast of Norway. This separation is in contrast to the findings based on mtDNA where no differences were found between porpoises from southern and northern Norway (Tolley *et al.* 1999). Although the results from ecological and genetic markers are apparently in contrast, these methods assess population structure on two very different time scales. Presumably, porpoises in the Barents Sea have only recently recolonised that region and have not substantially diverged from porpoises in southern Norway at a level that is detectable using the available genetic markers (Tolley *et al.* 1999). However, the lack of genetic differences does not necessarily indicate panmixia along the coast of Norway, as ecological separation could indicate that porpoises are philopatric within a region. Hence, there is a need to consider the time scale relevant to management. If the objective of management is to preserve only evolutionarily independent units, management must proceed based solely on genetic differences, yet this criteria is extremely strict. In the present case, it would be precautionary to give credence to the results as shown by ecological markers when making assumptions regarding population structure.



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#### LITERATURE CITED

- Aarkrog A (1998) A retrospect of anthropogenic radioactivity in the global marine environment. *Radiat Prot Dosim* 75:23-31
- Aguilar A, Borrell A, Pastor T (1999) Biological factors affecting variability of persistent pollutant levels in cetaceans. In: Reijnders PJH, Aguilar A, Donovan GP (eds) *Chemical Pollutants and Cetaceans*. *J Cetacean Res Manage* (Special Issue 1):83-116
- Andersen LW (1993) The population structure of the harbour porpoise, *Phocoena phocoena*, in Danish waters and part of the North Atlantic. *Mar Biol* 6:1-7
- Anonymous (2000a) Annual Report to Congress – 1999. Marine Mammal Commission, 4340 East-West Highway, Bethesda, MD 20814, USA
- Anonymous (2000b) A UK conservation strategy for the harbour porpoise (*Phocoena phocoena*). Department of the Environment, Transport and the Regions. DETR, PO Box 236, Wetherby, N Yorks, LS23 7NB, UK.
- Awise JC (1994) Conservation genetics. In: Awise JC (ed) *Molecular Markers, Natural History and Evolution*. Chapman & Hall, Inc. New York, p 511
- Berrow SD, Long SC, McGarry AT, Pollard D, Rogan E, Lockyer C (1998) Radionuclides ( $^{137}\text{Cs}$  and  $^{40}\text{K}$ ) in harbour porpoises *Phocoena phocoena* from British and Irish Coastal waters. *Mar Pollut Bull* 36:569-576
- Bjørge A, Aarefjord H, Kaarstad S, Kleivane L, Øien N (1991) Harbour porpoise *Phocoena phocoena* in Norwegian waters. ICES Working Paper C.M. 1991/N:16
- Brungot AL, Sickel MAK, Bergan TD, Bøe B, Hellstrøm T, Strand P (1997) Radioactive contamination in the marine environment. Report no. 2 from the National Surveillance Programme, Norwegian Radiation Protection Authority, NRPA Report 1997:3

- Brungot AL, Føyn L, Carroll JL, Kolstad AK, Brown J, Rudjord AL, Bøe B, Hellstrøm T (1999) Radioactive contamination in the marine environment. Report no. 3 from the National Surveillance Programme, Norwegian Radiation Protection Authority, NRPA Report 1999:6
- Börjesson P, Berggren P (1997) Morphometric comparisons of skulls of harbour porpoises (*Phocoena phocoena*) from the Baltic, Kattegat, and Skagerrak seas. *Can J Zool* 75:280-287
- Calmet D, Woodhead D, André JM (1992)  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in three species of porpoises caught in the Eastern Tropical Pacific Ocean. *J Environ Radioact* 15:153-169
- Carvahlo GR, Hauser L (1994) Molecular genetics and the stock concept in fisheries. *Rev Fish Biol Fisher* 4:326-350
- Casso SA, Livingston HD (1984) Radiocaesium and other nuclides in the Norwegian-Greenland Seas 1981-1982. Technical Report, No. WHOI-84-40, Woods Hole Oceanographic Institution, Woods Hole, MA, U.S.A.
- Dahlgaard H, Notter M, Brittain JE, Strand P, Rantavaara A, Holm E (1994) Nordic Radioecology 1990-1993. In: Dahlgaard H (ed) *Nordic Radioecology: The transfer of radionuclides through Nordic ecosystems to man*. Elsevier, p 7-20
- Donovan GP, Bjørge A (1995) Harbour porpoises in the North Atlantic: edited extract from the Report of the IWC Scientific Committee, Dublin 1995. In: Bjørge A, Donovan GP (eds) *Biology of the Phocoenids*. Rep int Whal Commn Spec Issue 16:3-25
- Forseth T, Ugedal O, Naesje TF, Jonsson B (1998) Radiocaesium elimination in fish: variation among and within species. *J Appl Ecol* 35:847-856
- Føyn L, Sværen, I (1997) Distribution and sedimentation of radionuclides in the Barents Sea. *ICES J Mar Sci* 54:333-340
- Grøttheim S (1998) A preliminary report on radioactive contamination in the northern marine environment. Studies on distribution of radiocaesium, plutonium and americium in seawater and sediments (M.Sc. Thesis). Norwegian Radiation Protection Authority and Biological Institute, University of Oslo, Norway
- Hammond PS, Benke H, Berggren P, Borchers DL, Buckland ST, Collet A, Heide-Jørgensen MP, Heimlich-Boran S, Hiby AR, Leopold MF, Øien N (1995) Distribution and abundance of the harbour porpoise and other small cetaceans in the North Sea and adjacent waters. European Commission Directorate General for the Environment, Nuclear Safety and Civil Protection. LIFE 92-2/UK/027

- Hartl DL, Clark AG (1997) Principles of Population Genetics. Third Edition. Sinauer Associates, Inc., Sunderland, Massachusetts
- Herrmann J, Kershaw PJ, Bailly du Bois P, Guegueniat P (1995) The distribution of artificial radionuclides in the English Channel, southern North Sea, Skagerrak and Kattegat, 1990-1993. *J Mar Syst* 6:427-456
- Holm E (1994) Polonium-210 and radiocaesium in muscle tissue of fish from different Nordic marine areas. In: Dahlgard H (ed) *Nordic Radioecology: The Transfer of Radionuclides through Nordic Ecosystems to Man*. Elsevier, Amsterdam, p 119-126
- Hoelzel AR (1998) Genetic structure of cetacean populations in sympatry, parapatry, and mixed assemblages: implications for conservation policy. *J Hered* 89:451-458
- IWC (International Whaling Commission) (1996) Report of the Sub-committee on small cetaceans. *Rep int Whal Commn*, Annex H 46:160-179
- Jefferson TA, Curry BE (1994) A global review of porpoise (Cetacea: Phocoenidae) mortality in gillnets. *Biol Conserv* 67:167-183
- Kershaw PJ, Baxter A (1995) The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep-Sea Res Pt II* 42:1413-1448
- Kershaw PJ, Gurbutt P, Woodhead D, Leonard K, Rees J (1997) Estimates of fluxes of  $^{137}\text{Cs}$  in northern waters from recent measurements. *Sci Tot Environ* 202:211-223
- Kershaw PJ, McCubbin D, Leonard KS (1999) Continuing contamination of north Atlantic and Arctic waters by Sellafield radionuclides. *Sci Tot Environ* 237/238:119-132
- Knijn R, Boon TW, Heessen HJL, Hislop JRG (1993) *Atlas of North Sea Fishes*. ICES Cooperative Research Report, 194:1-268
- Kleivane L, Skaare JU, Bjørge A, de Ruiter E, Reijnders P (1995) Organochlorine pesticide residue and PCBs in harbour porpoise (*Phocoena phocoena*) incidentally caught in Scandinavian waters. *Environ Pollut* 89:137-146
- Lockyer C (1995) Investigation of aspects of the life history of the harbour porpoise, *Phocoena phocoena*, in British waters. In: Bjørge A, Donovan GP (eds) *Biology of the Phocoenids*. *Rep Int Whal Commn* (Special Issue 16):189-197
- Mailhot H, Peters RH, Cornett RJ (1989) The biological half-time of radioactive Cs in poikilothermic and homeothermic animals. *Heath Phys*: 56:473-484

- Palumbi SR (1994) Genetic divergence, reproductive isolation, and marine speciation. *Annu Rev Ecol Syst* 25:547-572
- Palumbi SR, Martin AP, Kessing B, McMillan WO (1991) Detecting population structure using mitochondrial DNA. In: Hoelzel AR (ed) *Genetic Ecology of Whales and Dolphins*. Rep int Whal Commn Spec Issue 13:271-278. Cambridge
- Read AJ, Tolley KA (1997) Postnatal growth and allometry of harbour porpoises from the Bay of Fundy. *Can J Zool* 75:122-130
- Read AJ, Westgate AJ (1997) Monitoring the movements of harbour porpoises (*Phocoena phocoena*) with satellite telemetry. *Mar Biol* 130:315-322
- Rosel PE, France SF, Wang JY, Kocher TD (1999) Genetic structure of harbour porpoise *Phocoena phocoena* populations in the Northwest Atlantic based on mitochondrial and nuclear markers. *Mol Ecol* 8: S41-S54
- Rowan DJ, Rasmussen JB (1995) The elimination of radiocaesium from fish. *J Appl Ecol* 32:739-744
- Slatkin M (1987) Gene flow and the geographic structure of natural populations. *Science* 236:787-792
- Sætre R, Ljøen R (1971) The Norwegian Coastal Current. In: *Proceedings of the first international conference on port and ocean engineering under Arctic conditions*, Vol. 2, Technical University of Norway, Trondheim, Norway, pp. 514-535
- Taylor BL, Dizon AE (1999) First policy then science: why a management unit based solely on genetic criteria cannot work. *Mol Ecol* 8:S11-S16
- Teilmann J (2000) The behaviour and sensory abilities of harbour porpoises (*Phocoena phocoena*) in relation to bycatch in gillnet fishery. Ph.D. Thesis. Odense University, Campusvej 55, DK-5230, Odense, Denmark
- Tiedemann R, Harder J, Gmeiner C, Haase E (1996) Mitochondrial DNA sequence patterns of harbour porpoises (*Phocoena phocoena*) from the North and Baltic Sea. *Z Säugetierkd* 61:104-111
- Tolley KA, Vikingsson GA, Rosel PE (In press) Mitochondrial DNA sequence variation and phylogeographic patterns in harbour porpoises *Phocoena phocoena* from the North Atlantic. *Conserv Gen*

- Tolley KA, Rosel PE, Walton MJ, Bjørge A, Øien N (1999) Genetic population structure of harbour porpoises (*Phocoena phocoena*) in the North Sea and Norwegian waters. *J Cetacean Res Manage* 1:265-274
- Tregenza NJC, Berrow SD, Hammond PS, Leaper R (1997) Harbour porpoise (*Phocoena phocoena* L.) by-catch in set gillnets in the Celtic Sea. *ICES J Mar Sci* 54:896-904
- Walton MJ (1997) Population structure of harbour porpoises *Phocoena phocoena* in the seas around the UK and adjacent waters. *Proc R Soc Lond B* 264:89-94
- Wang JY, Gaskin DE, White BN (1996) Mitochondrial DNA analysis of harbour porpoise, *Phocoena phocoena*, sub-populations in North American waters. *Can J Fish Aquat Sci* 5:1632-1645
- Wang JY, Berggren P (1997) Mitochondrial DNA analysis of harbour porpoises (*Phocoena phocoena*) in the Baltic Sea, the Kattegat-Skagerrak Seas and off the west coast of Norway. *Mar Biol* 127:531-537
- Waples RS (1998) Separating the wheat from the chaff: patterns of genetic differentiation in high gene flow species. *J Hered* 89:438-450
- Watson WS, Sumner DJ, Baker JR, Kennedy S, Reid R, Robinson I (1999) Radionuclides in seals and porpoises in the coastal waters around the UK. *Sci Tot Environ* 234:1-13
- Westgate AJ, Muir DCG, Gaskin DE, Kingsley MCS (1997) Concentrations and accumulation patterns of organochlorine contaminants in the blubber of harbour porpoises, *Phocoena phocoena*, from the coast of Newfoundland, the Gulf of St. Lawrence and the Bay of Fundy/Gulf of Maine. *Environ Poll* 95:105-119
- Westgate AJ, Tolley KA (1999) Regional differences in organochlorine contaminants in harbour porpoises (*Phocoena phocoena*) from the western North Atlantic. *Mar Ecol Prog Ser* 177:243-254

Table 1. Sampling locations and levels of  $^{137}\text{Cs}$  (Bq/kg wet weight) in skeletal muscle from porpoises collected along the coast of Norway. Counting error for each measurement (error) and concentration factor (CF) are also given.

Location	ID#	Sex	Bq/kg	Error	CF
<i>Vest-Agder</i>	N9922	M	3.8	0.1	190.8
	N9925	M	2.5	0.1	123.6
	30-03-00	F	1.2	0.1	58.6
<i>Rogaland</i>	N9908	F	0.6	0.1	50.1
	N9905	M	0.4	0.1	32.9
	N9906	M	0.5	0.1	44.9
	N9907	M	0.5	0.1	41.4
	N9921	M	1.1	0.1	91.8
<i>Hordaland</i>	N9901	F	0.9	0.1	74.7
	N9902	F	1.1	0.1	89.4
<i>Nordland</i>	N9932	F	0.8	0.1	94.4
	N9918	M	1.2	0.1	142.4
	N9919	M	0.8	0.1	93.3
	N9920	M	0.9	0.1	107.6
	N9929	M	0.7	0.1	76.5
	N9930	M	0.5	0.1	63.12
	N9931	M	1.5	0.1	176.0
<i>Finnmark</i>	N9904	F	0.4	0.1	96.5
	N9909	F	0.5	0.1	130.5
	N9910	F	0.4	0.1	104.0
	N9911	F	0.6	0.1	148.1
	N9913	F	0.3	0.1	73.1
	N9914	F	0.7	0.1	171.2
	N9916	F	0.7	0.1	171.3
	VF06	F	0.5	0.1	132.7
	N9903	M	0.6	0.1	142.2
	N9912	M	0.4	0.1	110.2
	N9915	M	0.5	0.1	136.62
	N9917	M	0.5	0.1	127.22
	N9923	M	0.4	0.1	92.4
	N9924	M	0.5	0.1	127.7
	N9926	M	0.3	0.1	87.4
	N9927	M	0.4	0.1	96.5
	N9928	M	0.6	0.1	143.2
	VF03	M	0.5	0.1	125.1
	VF04	M	0.6	0.1	147.7

Table 2. Sample sizes ( $n$ ), means, and standard deviations ( $sd$ ) for levels of  $^{137}\text{Cs}$  (Bq/kg ww) in harbour porpoises from three regions. Significant differences between groups are also given (Sig. Diff.).

	$n$	Mean	$sd$	Sig. Diff.
Southwest	10	1.22	1.030	Finnmark
Nordland	7	0.93	0.361	Finnmark
Finnmark	19	0.50	0.110	Southwest, Nordland

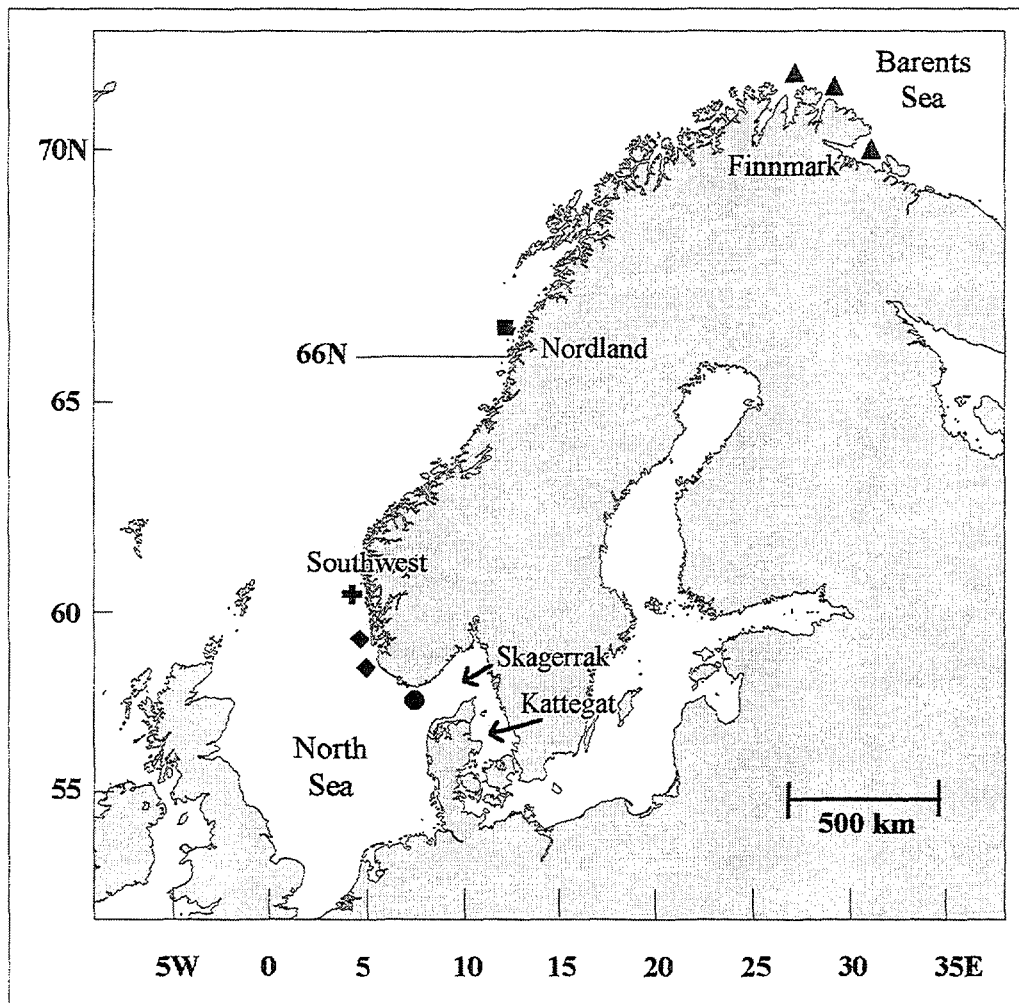


Fig. 1. Sampling locations of harbour porpoises from the coast of Norway. Vest-Agder (●); Rogaland (◆); Hordaland (+); Nordland (■); Finnmark (▲). The dividing line (at 66°N) between the North Sea and the North Norway/Barents Sea putative populations defined by the International Whaling Commission is shown.



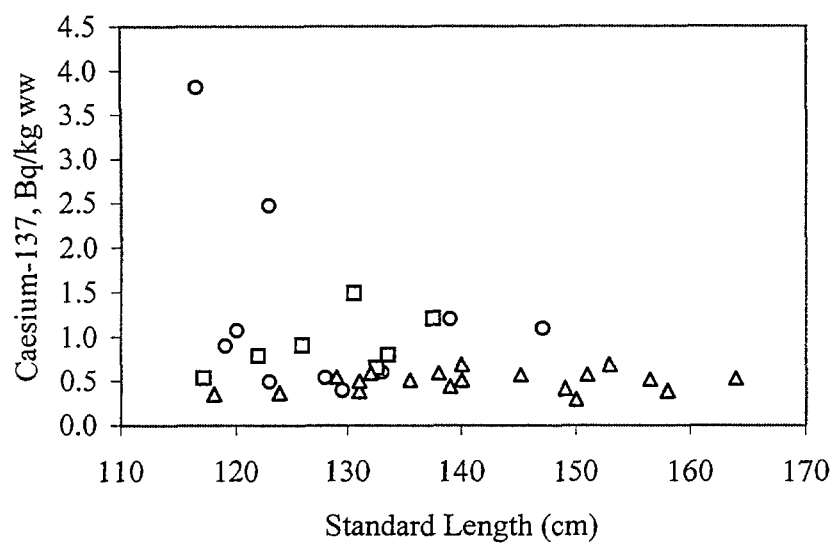


Fig. 2. Standard lengths and levels of  $^{137}\text{Cs}$  (Bq/kg ww) in harbour porpoises from three regions in Norway. Southwest (○), Nordland (□), and Finnmark (Δ).

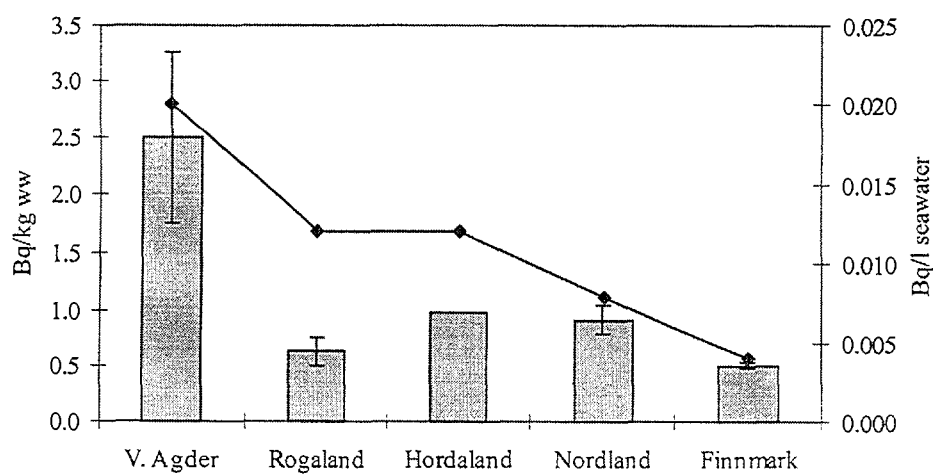


Fig. 3. Mean levels and standard errors of  $^{137}\text{Cs}$  (Bq/kg ww) in harbour porpoises from five locations in Norway (bars). The 'Southwest' region consists of Vest-Adger, Rogaland, and Hordaland. Median concentrations of  $^{137}\text{Cs}$  in seawater within each area are shown by the solid line.

## **PAPER V**

## **Technetium-99: a renewed transient tracer of Nordic Seas and Arctic circulation**

Hilde Elise Heldal<sup>1</sup>, Peter John Kershaw<sup>2\*</sup> and Kjell Arne Mork<sup>1</sup>

<sup>1</sup>Institute of Marine Research, PO Box 1870 Nordnes, N-5817 Bergen, Norway

<sup>2\*</sup>The Centre for Environment, Fisheries & Aquaculture Science,

Pakefield Road, Lowestoft, Suffolk NR33 0HT, UK

Telephone: +44-1502-524455

Fax: +44-1502-513865

E-mail: [P.J.Kershaw@CEFAS.co.uk](mailto:P.J.Kershaw@CEFAS.co.uk)

### **Abstract**

In April 1994, a new waste treatment plant, the Enhanced Actinide Removal Plant (EARP), began operation at the nuclear reprocessing facilities Sellafield in Cumbria, UK. EARP was introduced primarily to reduce the discharges of plutonium and americium, and does not remove technetium-99 (<sup>99</sup>Tc) from the effluents. The discharges of <sup>99</sup>Tc from Sellafield into the Irish Sea have therefore increased significantly since 1994. Early studies of the propagation of the '<sup>99</sup>Tc-front' from the Irish Sea into the North Sea showed much more rapid transport than reported for other radionuclides prior to 1994. The purpose of the present study was twofold: to extend the geographical range of sampling to allow the continuing spread of EARP-related <sup>99</sup>Tc to be followed, and to place the results in an appropriate oceanographic setting. Our results show that the <sup>99</sup>Tc contamination has reached Arctic waters. The <sup>99</sup>Tc concentration was ~9 and ~6 times higher than background levels in the Norwegian Coastal Current (NWCC) outside Troms county and in the West Spitsbergen Current (WSC) in May/June 2000, respectively. However, the propagation speed of <sup>99</sup>Tc appears to have slowed down on route to Arctic waters. Based on results from other studies, a mean propagation speed for EARP-related <sup>99</sup>Tc along the Norwegian coast is estimated in the present study to be approximately 7 cm s<sup>-1</sup>. We propose that a particularly high NAO index during the 1994/1995 winter, which correlates with strong westerly winds and a large inflow from the Irish Sea to the North Sea, caused the initial rapid transport of <sup>99</sup>Tc from the Irish Sea to the North Sea. Lower NAO

winter indexes and reduced westerly winds towards the late 1990's have most likely 'normalised' the transport rate for  $^{99}\text{Tc}$ .

**Keywords:** Technetium-99; Transport times; Transfer factors; Norwegian Coastal Current; West Spitsbergen Current; NAO winter index

## 1. Introduction

Artificial radionuclides introduced to the marine environment - principally global fallout from weapons testing and releases from nuclear fuel reprocessing operations - have provided useful tracers of ocean circulation in the Nordic Seas and the Arctic (Aarkrog et al., 1987; Kautsky, 1988; Livingston, 1988; Dahlgaard, 1995; Guegueniat et al., 1997; Smith et al., 1998). The radionuclides  $^3\text{H}$ ,  $^3\text{He}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and isotopes of Pu have been used at different times for different applications. Technetium-99 ( $^{99}\text{Tc}$ ) is a fission and activation product with a half-life of  $2.13 \cdot 10^5$  years. It is ubiquitous in the oceans as a consequence of global fallout from nuclear weapons detonations. In oxic seawater, technetium is found as the highly soluble pertechnetate anion ( $\text{TcO}_4^-$ ). In such circumstances it behaves conservatively and can be used as an oceanographic tool for circulation studies, from measurements of both seawater and biota (e.g. brown seaweed *Fucus* sp.; Aarkrog et al., 1987).

Reprocessing operations at BNFL-Sellafield (UK) and COGEMA-La Hague (F) have provided an additional source of  $^{99}\text{Tc}$  to the North Atlantic. Sellafield (formerly Windscale) started operations in 1952 and La Hague in 1966. Precise discharge data have been available only since 1978 for Sellafield and 1979 for La Hague. However, estimates have been made of the magnitude of the releases prior to this based on environmental measurements. For example, Aarkrog et al. (1987) estimated discharges from Sellafield in the period 1970-1977 to be  $\sim 40 \text{ TBq a}^{-1}$  ( $1 \text{ TBq} = 10^{12} \text{ Bq}$ ) based on measurements of  $^{99}\text{Tc}$  in brown seaweed (*Fucus* sp.). Releases from Sellafield declined significantly after 1978 (179 TBq) and remained at a relatively uniform level throughout the 1980s ( $2\text{-}7 \text{ TBq a}^{-1}$ ) up until the first quarter of 1994. Releases from La Hague rose to a maximum in 1985 (25 TBq) and exceeded those from Sellafield from 1981 to 1990.

In April 1994, a new waste treatment plant, the Enhanced Actinide Removal Plant (EARP), began operation at Sellafield. EARP was introduced primarily to

reduce the release of plutonium and americium – significant contributors to human dose – using a combination of chemical treatments and ultra filtration. This allowed stored medium-level liquors, and other waste streams, to be treated resulting in the removal of most alpha and particulate beta activity. However,  $^{99}\text{Tc}$  was not removed by EARP. Consequently, the increased waste throughput resulted in an increased discharge of  $^{99}\text{Tc}$ , in early 1994, to a level significantly higher than the releases reported throughout the 1980s. Fig. 1 shows annual discharges of  $^{99}\text{Tc}$  from Sellafield and La Hague in the years 1978-1999.

The concentrations of  $^{99}\text{Tc}$  in seawater and biota (e.g. lobster *Homarus gammarus*, nephrops *Nephrops norvegicus*, and brown seaweed *Fucus* sp.) have increased significantly in the latter part of the 1990's accompanied with a small increase in the radiation dose received by the local critical group of seafood consumers (Smith et al., 1997; Brown et al., 1999; MAFF and SEPA, 1999). The discharge was close to the authorised limit ( $200 \text{ TBq a}^{-1}$ ) in 1995 but decreased for a variety of reasons thereafter, including socio-political concerns. The authorised limit has since been reduced to  $90 \text{ TBq a}^{-1}$ . As a result of these factors the nuclear industry has provided a substantial pulsed source-term to European coastal waters, which is being propagated through the Nordic Seas to the Arctic. The progress of the ' $^{99}\text{Tc}$ -front' has provided an excellent tracer for studying the circulation of the NE Atlantic and Nordic Seas from the mid-1990s onwards.

The generalised transport pathways of conservative radionuclides released from Sellafield are well established (e.g. Dahlgaard et al., 1986; Kershaw and Baxter, 1995). The greatest proportion exits the Irish Sea via the North Channel and is transported into the North Sea via the Scottish Coastal Current. Sellafield radionuclides are carried into the southern North Sea and eventually merge with water flowing through the English Channel, labelled with La Hague radionuclides, in Danish waters near the entrance of the Skagerrak. This water becomes incorporated with the Baltic Sea outflow and fresh water run off from Norway and forms the Norwegian Coastal Current (NWCC), which flows northwards along the Norwegian coast. At around  $63^{\circ} 30' \text{ N}$ , the NWCC splits in two branches: one narrow branch along the coastal side and another further offshore which follows the shelf break northwards parallel to the Norwegian Atlantic Current (NWAC) (e.g. Ljøen and Nakken, 1969; Poulain et al., 1996; Sætre, 1999). The inner branch of the NWCC has

higher drift speed while the outer branch mixes with NWAC northwards and loses its identity (Sætre, 1999). Atlantic Water (AW) progressively dilutes the radiotracer signal in the NWCC, leading to contamination of the NWAC. At the western boundary of the Barents Sea (at about 70° N) the NWAC splits into two currents: the northwards flowing West Spitsbergen Current (WSC) and the eastwards flowing North Cape Current (NCC). The generalised distribution of water masses and circulation of surface waters are summarised in Fig. 2.

Initial observations of EARP-related  $^{99}\text{Tc}$  indicated much more rapid transit times through UK waters than had been anticipated, on the basis of previous estimates (Leonard et al., 1997; Brown et al., 1999). The latter were based on radiotracer distributions (e.g.  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ ,  $^{125}\text{Sb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ) and model simulations, reported during the 1980s and 1990s. For example, transport times have been reported from Sellafield to: the North Channel: 1 year (Jefferies et al., 1982; Kershaw and Baxter, 1995); the central North Sea: 2-3 years (Dahlgaard, 1995; Dahlgaard et al., 1986); the NWCC: 4 years (Dahlgaard, 1995; Dahlgaard et al., 1986; Kershaw and Baxter, 1995); and to the entrance of the Barents Sea: 4-6 years (Kershaw and Baxter, 1995). In contrast, the leading edge of the ' $^{99}\text{Tc}$ -front' reached the North Channel in < 3 months and the northern North Sea within 9 months (Leonard et al., 1997). The ' $^{99}\text{Tc}$ -front' had reached the southwestern coast of Norway within less than 3 years (Brown et al., 1999; Christensen et al., 2001) and by August 1997, only a little more than 3 years post-EARP, it could be detected in brown seaweed (*F. vesiculosus*) collected at Ingøy (71° N 26° E) on the north coast of Norway (G. Christensen, pers. comm.).

The purpose of the present study was twofold: to extend the geographical range of the sampling to allow the continuing spread of EARP-related  $^{99}\text{Tc}$  to be followed; and to place the results in an appropriate oceanographic setting.

## **2. Materials and methods**

Sample collection took place during four cruises conducted by the Institute of Marine Research (IMR): in July 1998 and June 2000 aboard R/V 'Michael Sars' and in June/July 1999 and May/June 2000 aboard R/V 'Johan Hjørt'. Hydrographic measurements were made both underway and using CTD casts. Discrete near-surface (5m) water samples (100-150 litre) were collected with shipboard pumps for  $^{99}\text{Tc}$

analysis.  $^{99}\text{Tc}$  is a pure beta-emitter ( $E_{\text{max}} = 292 \text{ keV}$ ) decaying from  $^{99}\text{Mo}$ , and a radiochemical separation of this radionuclide is required for quantitative analysis. The detailed analytical method adopted is described by Harvey et al. (1992). Briefly, after adding rhenium in the form of  $\text{KReO}_4^-$  as a yield monitor, a preliminary extraction of  $^{99}\text{Tc}$  (and Re) based on anion-exchange separation was performed. After iron hydroxide scavenging,  $^{99}\text{Tc}$  and Re were further extracted by a second anion-exchange and subsequent sulphide precipitation. Finally, their tetraphenyl arsonium salts were isolated.  $^{99}\text{Tc}$  was beta-counted, and the yield of the rhenium tetraphenyl arsonium salt was determined gravimetrically. The samples were not filtered prior to analysis, as they were collected in waters with low concentrations of suspended materials. All the chemical treatment and beta-counting were carried out at the Centre for Environment, Fisheries and Aquaculture Science (CEFAS), UK, with the exception that the first radiochemical separation step (addition of yield monitor and preliminary anion-exchange) in June/July 2000 was performed aboard R/V 'Johan Hjord' during the cruise. The analytical method has been quality assured through internal accreditation (UKAS-NAMAS) and international intercomparison exercises (Dahlgard et al., 1995a).

During both R/V 'Johan Hjord' cruises, hydrographic data were collected using a SBE 911 plus CTD water sampling package from SeaBird Inc. At all stations, water samples were collected for calibration of the conductivity sensors at all depths below 1000 db or from the deepest sampling level. During the R/V 'Johan Hjord' cruise in May/June 2000, temperature and salinity were continuously monitored in surface water from the ship's water intake at 5 meters depth using a SBE 21 Thermosalinograph, SeaBird Inc. The Thermosalinograph was calibrated against CTD temperature and salinity records at the subsurface about 2-3 meters distance from the water intake of the Thermosalinograph, prior to the initiation of the downcast (Rey, 2001). During both R/V 'Michael Sars' cruises, a CTD NBIS Mk III was used to collect hydrographic data.

Results from a UK survey of the distribution of  $^{99}\text{Tc}$  in the Norwegian, Greenland and the Barents Seas in September/October 1994 are used to provide the pre-EARP background concentrations of  $^{99}\text{Tc}$  (Kershaw et al., 1999a).

### 3. Results and discussion

The results of all  $^{99}\text{Tc}$  determinations carried out on samples collected in 1998, 1999 and 2000 are presented in Table 1. Concentrations were significantly above the detection limit ( $0.018 \text{ Bq m}^{-3}$  in seawater) in all samples except one. The analytical error due to the combined uncertainty in the chemical yield and the counting statistics were generally below 5%, but reached above 20% for a few low-level samples.

#### 3.1. Spatial and temporal distribution of $^{99}\text{Tc}$

The spatial distribution of  $^{99}\text{Tc}$  in 1998, 1999 and 2000 is shown in Figs. 3 a-d. Highest concentrations in all years occurred in the NWCC, characterised with relatively low salinity. Concentrations within the core of the NWCC were relatively uniform within-year, but significant gradients were observed with distance from the coast, paralleling the gradients in salinity and temperature. Mixing of NWCC water with NWAC has resulted in a clear labelling of the WSC.

The distribution of SST (Sea Surface Temperature), based on continuous thermosalinograph measurements, provides useful contextual information about the distribution of water masses and thus explanation of the observed variations in  $^{99}\text{Tc}$  concentrations. Continuous SST data, which were available for parts of the study areas in 1994 and 2000, are shown in Figs. 4 a and b, with  $^{99}\text{Tc}$  concentrations overlain. The spatial coverage in 1998 and 1999 was insufficient to allow reliable contouring of the SST. It is evident from the SST distribution in 1994 that the background concentrations of  $^{99}\text{Tc}$  do not vary significantly between different water masses, with exception of within the NWCC, where a slightly elevated concentration of  $^{99}\text{Tc}$  occurred (Fig. 4 a). The position of the Polar Front is evident from the SST distribution in 2000 (Fig. 4 b), extending from the Bear Island northwards along the west coast of Spitsbergen, separating colder Polar Water ( $S < 34.5$  and  $T < 1^\circ\text{C}$ ) from AW in the WSC. AW contains higher concentrations of  $^{99}\text{Tc}$  than PW. Samples from within the AW-PW transition zone had lower  $^{99}\text{Tc}$  concentrations than in adjacent AW at the same latitude (Fig. 4 b, latitude  $\sim 74^\circ 30' \text{ N}$ ) due to dilution. Lower concentrations also occurred in the West Spitsbergen Coastal Current (WSCC). Further west lies the Arctic Front, separating AW from the colder and less saline



waters of the Greenland Sea.  $^{99}\text{Tc}$  concentrations in the Greenland Sea, in 1999 and 2000, were significantly lower than in AW and in the WSC at the same latitude (Figs. 3 b and c), and of similar levels to 1994 background concentrations (Kershaw et al., 1999a).

CTD sections were worked along Fugløy (70° 30' N; 20° 00' E) – Bear Island (74° 15' N; 19° 10' N) (1998, 1999 and 2000) and west of Bear Island along 74° 30' N (1999 and 2000). Salinity transects for the two sections are presented in Figs. 5 a-c and Figs. 6 a-b, respectively, together with an overlay of the  $^{99}\text{Tc}$  concentrations. The Fugløy – Bear Island transect was dominated by AW, with lower salinity NWCC spreading out from the Norwegian coast. The  $^{99}\text{Tc}$  concentration is inversely proportional to the salinity, with highest levels found within the NWCC, and a decrease along the section towards Bear Island. Along the 74° 30' N sections west of the Bear Island, the positions of the Polar and Arctic fronts and the core of AW are clearly defined. The within-year  $^{99}\text{Tc}$  concentrations in AW were relatively uniform.

In July 1998, the  $^{99}\text{Tc}$  concentration near Fugløy was approximately 6 times higher than the 1994 background concentration ( $0.14 \text{ Bq m}^{-3}$ ; Kershaw et al. 1999a). In June/July 1999, the  $^{99}\text{Tc}$  concentration was an order of magnitude higher than 1994 levels at this location, and it had increased to approximately double the 1998 value. However, in May/June 2000, the  $^{99}\text{Tc}$  concentration at this location was reduced to a factor of 9 higher than the 1994 level.

The progress of the ' $^{99}\text{Tc}$ -front' in the WSC is illustrated in Fig. 7, in which  $^{99}\text{Tc}$  concentrations have been plotted against latitude for all stations falling within the boundaries 70-78° N and 10-20° E, for the years 1994, 1998, 1999 and 2000. Samples with PW characteristics ( $S < 34.5$  and  $T < 1^\circ\text{C}$ ) were excluded, but samples within the AW-PW transition zone were included. Higher concentrations, indicative of an EARP origin, were apparent in the WSC at 73° 30' N in 1998, but not at the next station at 74° 20' N. However, one of three samples collected further north at ~ 77° the same year contained slightly elevated levels of  $^{99}\text{Tc}$  ( $0.16 \text{ Bq m}^{-3} \pm 10\% \sim$  a factor of 2 higher). This may be an early influence of EARP-related  $^{99}\text{Tc}$ . Samples collected north of ~ 74° in 1998 are characterised with relatively low temperatures and salinities, and may look like they are influenced by PW. However, a great deal of sea ice was present in the study area in 1998. Melting sea ice may have locally lowered the temperature and salinity in AW, and  $^{99}\text{Tc}$  concentrations may have been diluted.

In 1999, EARP-related  $^{99}\text{Tc}$  was evident west of Bear Island at  $74^{\circ} 30' \text{ N}$ .

Unfortunately no samples were collected further north this year. The leading edge of the ' $^{99}\text{Tc}$ -front' was evident in the WSC west of Spitsbergen by May/June 2000 and had advanced beyond the limit of sampling ( $77^{\circ} 20' \text{ N}$ ).

### *3.2 Mixing of EARP-related $^{99}\text{Tc}$ with AW*

$^{99}\text{Tc}$  concentrations in surface waters of the NWCC, NWAC and WSC, for 1998-2000, are plotted against salinity in Fig. 8. Samples with PW characteristics ( $S < 34.5$  and  $T < 1^{\circ}\text{C}$ ) are excluded, but samples from within the transition zone are included. The result represents a mixing curve defined by two end-members: AW characterised by a mixture of  $^{99}\text{Tc}$  originating from global fallout ( $\sim 5 \text{ mBq m}^{-3}$ ; Dahlggaard et al., 1995b) and  $^{99}\text{Tc}$  originating from earlier reprocessing wastes; and NWCC water, contaminated by a time-varying EARP reprocessing signal. The global fallout background concentration was obtained from surface water samples near the Azores, in waters which have not been contaminated by reprocessing wastes. In contrast, all the waters sampled in the present study, including PW, will have been contaminated to varying extents by releases of  $^{99}\text{Tc}$  in the 4 decades of reprocessing operations. The highest concentrations, determined in the present study, occurred in the NWCC in 1999. The degree of variability in  $^{99}\text{Tc}$  concentrations appears to have decreased with time, and this has been accompanied by a progressive increase in the  $^{99}\text{Tc}$  signal in AW as a result of continuing mixing and transport. The mixing curve is very similar to that obtained for a larger dataset of  $^{137}\text{Cs}$  measurements in 1989, covering a similar geographic region (Kershaw and Baxter, 1995), with the exception that the  $^{137}\text{Cs}$  concentrations were a factor of 10 higher.

### *3.3 Transport times and Transfer factors*

A compilation of transport times from Sellafield to various locations based on EARP-related  $^{99}\text{Tc}$ , including data from the present study, is presented in Table 2. The ' $^{99}\text{Tc}$ -front' had reached the southwestern coast of Norway in November 1996 (Brown et al., 1999), and Utsira ( $59^{\circ} 19' \text{ N}$ ) in January 1997 (Christensen et al., 2001), 2.5-3 years post-EARP. The ' $^{99}\text{Tc}$ -front' had reached Ingøy by August 1997, a little more

than 3 years post-EARP (G. Christensen, pers. comm.). The present study shows that the  $^{99}\text{Tc}$ -front' had passed southwest of Bear Island in the WSC in July 1998,  $\sim 4$  years post-EARP (concentrations  $\sim 0.3 \text{ Bq m}^{-3}$ ,  $\sim 6$  times 1994 background levels), indicating a transport time from Sellafield to this location of 3-4 years. The sample with elevated  $^{99}\text{Tc}$  concentration collected in the WSC west of Spitsbergen ( $76^{\circ} 55' \text{ N}$ ) in July 1998, 4 years post-EARP, may represent an early influence of EARP-related  $^{99}\text{Tc}$ . The  $^{99}\text{Tc}$ -front' had clearly passed west of Spitsbergen in the WSC in May/June 2000 (concentrations  $\sim 0.3 \text{ Bq m}^{-3}$ ), 6 years post-EARP. Thus, the initial  $^{99}\text{Tc}$ -front' must have passed west of Spitsbergen in the WSC 4-6 years post-EARP.

Transfer factors (TFs) represent an effective method of describing the transport and dilution of a contaminant from a well-defined point-source, such as the pipeline discharges at Sellafield and La Hague (Dahlgaard, 1995). A TF is the relationship between the environmental concentration at a given remote location and the average quantity discharged  $t$  years earlier, where  $t$  is the mean transport time to the location. The units for radionuclide TFs are usually given as  $\text{Bq m}^{-3}/\text{PBq a}^{-1}$ . TFs have been calculated for  $^{99}\text{Tc}$  for 1994, 1998, 1999 and 2000 at three locations: Near Fugløy (NWCC) ( $70^{\circ} 30' \text{ N}$ ), west-southwest of Bear Island (WSC) ( $73^{\circ} 30' \text{ N} - 74^{\circ} 30' \text{ N}$ ) and west of Spitsbergen (WSC) ( $\sim 77^{\circ} 00' \text{ N}$ ) (Table 3). The TFs have been calculated by dividing the environmental concentration of  $^{99}\text{Tc}$  with the time-dependent discharge. Samples with temperature and salinity characteristics of PW ( $S < 34.5$  and  $T < 1^{\circ}\text{C}$ ) have been excluded, but samples from within the transition zone are included. Discharges of  $^{99}\text{Tc}$  from La Hague were accounted for in calculations of TFs for 1994, but neglected in calculations of TFs for 1998, 1999 and 2000, due to the overwhelming influence of the Sellafield discharge from the late 1990s onwards (Fig. 1). In the calculations of TFs for 1994, we assumed a two-year relative time lag for the Sellafield discharges in relation to the La Hague discharges (Dahlgaard et al., 1997). For 1994, we based the calculations of TFs at the three locations on average discharge rates from La Hague (Sellafield) of 2 (4), 3 (5) and 4 (6) years earlier, respectively. These transport times are based mainly on measurements of  $^{137}\text{Cs}$ . The calculations of TFs at the three locations for 1998-2000 were based on average discharges from Sellafield of 3, 3-4 and 4-5 years earlier. These transport times are based on measurements of EARP-related  $^{99}\text{Tc}$ . The TFs varied from 4 (in 1998) to 66 (in 1994) at the location near Fugløy, from 1 (in 1999) to 9 (in 1994) at the location

west-southwest of Bear Island, and from 2 (in 2000) to 10 (in 1994) at the location west of Spitsbergen (Table 3). This means, for example, that a  $^{99}\text{Tc}$  discharge from Sellafield of 200 TBq will give a seawater concentration near Fugløya 3 years later of approximately  $1 - 13 \text{ Bq m}^{-3}$ , estimated on the basis of TFs of 4 and 66, respectively. The actual environmental  $^{99}\text{Tc}$  concentration one might expect lies probably in between these extremities.

Dahlgaard (1995) presented transport times and TFs for Sellafield discharges to various locations, largely based on Cs observations from a number of authors. These are reproduced in Table 4 together with other reported values. Dahlgaard (1995) reported a transport time from Sellafield to the NW Norwegian Current of 3-4 years. The appearance of the ' $^{99}\text{Tc}$ -front' at Ingøy in the NWCC at the north coast of Norway in August 1997 (G. Christensen, pers. comm.)  $\sim 3$  years post-EARP is, in fact, in accordance with Dahlgaard's (1995) previous reported transport time. Kautsky (1987) reported transport times to north Norway (North Cape) and the Bear Island-Spitsbergen area of 5-6 and 6-7 years, respectively; these are somewhat longer than transport times found in most other studies. The transport time found in this study to the WSC west of Spitsbergen (4-6 years) is also in accordance with that reported by Dahlgaard (1995) ( $\sim 5$  years). Raisbeck et al. (1997) reported a transport time of 2 years from Utsira at the southwestern coast of Norway to Indre Kiberg at the northeastern coast of Norway. This is based on  $^{129}\text{I}/^{99}\text{Tc}$  and  $^{129}\text{I}/^{137}\text{Cs}$  ratios in brown seaweed (*F. vesiculosus*) collected at these locations between 1980 and 1995. In contrast, the transport time from Utsira to Ingøy was less than a year, based on  $^{99}\text{Tc}$  (Christensen, pers. comm.). Although the distance Utsira-Indre Kiberg is somewhat longer than the distance Utsira-Ingøy, the transport time reported by Raisbeck et al. (1997) appears to be long compared to the transport time based on  $^{99}\text{Tc}$ . Nevertheless, the present study suggests that the transport of EARP-related  $^{99}\text{Tc}$  northwards in the NWAC and WSC slowed down compared to the fast transport from the Irish Sea into the North Sea. We propose that the transport time for EARP-related  $^{99}\text{Tc}$  from Sellafield to Arctic waters was shortened by approximately half a year compared to 'normal' transport times, due to the initial rapid transport from Sellafield into the North Sea. Variations in transport times of radionuclides from point-sources are closely related to variations in physical oceanographic conditions. This is discussed in more detail below.

The TFs calculated for 1994, using transport times based mainly on  $^{137}\text{Cs}$ , were higher at all three locations than the TFs calculated for 1998-2000 using transport times based on EARP-related  $^{99}\text{Tc}$  (Table 3). This is contrary to what one should expect.  $^{99}\text{Tc}$  behaves more conservatively in seawater than Cs, and one should therefore find larger fractions of discharged  $^{99}\text{Tc}$  at a given distance from the source compared to Cs. The result may be a consequence of using the ' $^{99}\text{Tc}$ -front' instead of the main  $^{99}\text{Tc}$  pulse in the calculation of TFs. The low temporal resolution of our data is likely to bias the results in such a manner. The calculated TFs are higher in the NWCC near Fugløya than in the WSC west-southwest of Bear Island and west of Spitsbergen. This is expected, as a large part of the  $^{99}\text{Tc}$  contamination travels eastwards with the NWCC and NCC into the Barents Sea instead of travelling northwards with the WSC. TFs found in this study are comparable to earlier findings, although the range is larger. Dahlgaard (1995) reported TFs in the NW Norwegian Coastal Current and west of Spitsbergen of 10-50 and  $\sim 5$ , respectively. Aarkrog et al. (1987) reported a TF NW of Spitsbergen of 2-3 (Table 4).

Radionuclides with relatively long half lives (relative to the transit time) and, to a first approximation, similar 'conservative' chemical behaviour in seawater can be expected to have similar TFs. This applies to  $^{99}\text{Tc}$  and  $^{129}\text{I}$ , both released as a result of reprocessing operations. However, radiocaesium has been used more often as a conservative tracer in marine waters, due to the relatively large quantities discharged and ease of analysis. The greatest releases of radiocaesium occurred in the 1970s and many studies were reported of the progress of this substantial tracer pulse through the Nordic Seas, into the Arctic Basin and in the return flow in the East Greenland Current (EGC) and deeper waters (Dahlgaard et al., 1986; Aarkrog et al., 1987; Kautsky, 1987; Dahlgaard, 1994). However, a significant fraction of the discharged caesium became associated with the sediments of the Irish Sea. Following substantial reductions in the discharge, radiocaesium started to be released from the seabed back into the water column, resulting in a decrease in the decay-corrected environmental inventory of  $^{137}\text{Cs}$  in the sub-tidal sediments (Poole et al., 1997). Further evidence of significant remobilisation had been reported previously based on TFs (in a range of environmental materials, including biota) within the Irish Sea and compartment modelling (Hunt and Kershaw, 1990; Nicholson and Hunt, 1995; Kershaw et al., 1999b). The seabed sediments have represented a more significant source of  $^{137}\text{Cs}$  for export from the Irish Sea than direct discharges since the mid-1980s. This has also

been true for plutonium, certainly throughout the 1990s (Leonard et al., 1997; Kershaw et al., 1999b). In these circumstances the apparent TF for  $^{137}\text{Cs}$ , with respect to the Sellafield discharge, can be expected to have increased. From 1986 the Baltic Sea outflow, labelled predominantly by Chernobyl deposition, has progressively dominated the supply of  $^{137}\text{Cs}$  to the NWCC. Estimates of TFs for  $^{137}\text{Cs}$  in the Nordic Seas from the mid-1980s onwards should take account of the remobilised Sellafield  $^{137}\text{Cs}$  and the Chernobyl accident. The supply of  $^{137}\text{Cs}$  from La Hague amounted to approximately 2.3% of the Sellafield discharge, decay-corrected up to 1992 (Kershaw and Baxter, 1995). The shorter-lived radionuclide  $^{125}\text{Sb}$  ( $t_{1/2} = 2.8$  years), although released in relatively large quantities from La Hague ( $\sim 2$  PBq) decays too rapidly to be used as a tracer beyond the NWCC, despite its utility as a tracer in the English Channel and North Sea (Guegueniat et al., 1997).

### *3.4 Variability of transport and flux estimates*

The North Atlantic Oscillation (NAO) has become accepted as the most reliable indicator of large-scale wind forcing in the North Atlantic and Nordic Seas (Hurrell, 1995). The NAO winter index is the normalised gradient in sea level air pressure between two fixed points at Stykkisholmur (Iceland) and Lisbon or the Azores (Portugal). It exhibits fluctuations between ‘high’ (positive) and ‘low’ (negative) phases. A ‘high’ (positive) NAO winter index is associated with intense low mean pressure over Iceland. This leads to increased wind forcing and precipitation over the NW European shelf, an increase in volume transport of the North Atlantic Current (Orvik et al., 2001), inflow of relatively warm AW into the Arctic Ocean (Dickson et al., 2000; Furevik, 2001) and a reduction in the westwards extent of the NWAC (Blindheim et al., 2000). The NAO winter index correlates with the strength of westerly winds, where high values correspond to more westerly winds and storms. The NAO winter index shows significant inter-annual and decadal fluctuations. From the mid-1960s the index has increased progressively with relatively high values in the early- to mid-1990s. Fig. 9 shows the NAO winter index from 1950 through 2000, updated from Hurrell (1995).

The largest difference between the transport times before and after EARP began operation are for the signal to travel from the Irish Sea to the North Sea. The NAO winter index was particularly high during the 1994/1995 winter, the first year

after EARP began operation. Current meter observations indicate that increased westerly winds result in a larger inflow to the North Sea (Furnes, 1980; Dooley and Furnes, 1981). The increased wind forcing during this particular period (1994/1995 winter), and to a lesser extent during the three previous winters, is likely to have resulted in a larger inflow into the North Sea from the Irish Sea and thus explain the difference in the pre-EARP and post-EARP transport times for water masses from the Irish Sea into the North Sea.

The mean monthly sea surface temperature (SST) of the North Sea showed both extreme cold temperatures in the winter of 1995/96, marked by a substantial negative NAO winter index, and extreme warm temperatures in the summer of 1996 (Loewe, 1996), with a cold core of deeper water persisting beneath the seasonal thermocline in the north-central North Sea. This is likely to have enhanced the seasonal development of a jet-like circulation along the Flamborough Front (separating stratified water and the tidally-mixed shallower central-southern North Sea), in which core velocities have been reported as exceeding  $0.1 \text{ m s}^{-1}$  (Brown et al., 1999; Brown et al., 2001). A reduced wind forcing throughout this period will have minimised the transport of Sellafield-labelled water to the southern North Sea. In such circumstances, Scottish coastal waters entering the North Sea in the latter part of 1995, labelled with  $^{99}\text{Tc}$  released in 1994/1995, would be expected to reach the southern North Sea in reduced quantities compared with winters with 'normal' wind forcing. A proportion may have become entrained in the frontal jets and transported relatively rapidly across the central North Sea in the summer of 1996, but this cannot be quantified. Once the EARP-related  $^{99}\text{Tc}$  became entrained in the NWCC its northwards transport would be expected to correlate increasingly with the volume transport of the NWAC. The volume and heat transport of the NWCC also is highly seasonal and correlates with wind forcing and freshwater inputs (Blindheim, 1993). Mixing with AW occurs as a result of the formation of mesoscale frontal waves and associated eddies and the mean salinity of the NWCC increases northwards (Sætre and Ljøen, 1971; Mork, 1981).

The high levels of  $^{99}\text{Tc}$  in the coastal water along the coast of northern Norway suggests that a large part of the EARP-related  $^{99}\text{Tc}$  has propagated with the inner branch of the NWCC which, as mentioned in the introduction, has a higher speed than the outer branch (Sætre, 1999). Based on data indicating that the ' $^{99}\text{Tc}$ -front' reached Utsira in January 1997 (Christensen et al., 2001) and Ingøy in August 1997 (G.

Christensen, pers. comm.), we can roughly calculate the mean propagation speed of  $^{99}\text{Tc}$ . Following the NWCC close to the coast from Utsira to Ingøy gives an average speed of  $7 \text{ cm s}^{-1}$  when a transport time of 7 months is used. This propagation velocity can be compared with results from other studies. The Great Salinity Anomaly of the 1960s and 1970s was advected at a speed of  $3 \text{ cm s}^{-1}$  (Dickson et al., 1988) but that was in the Subpolar Gyre. A more comparable study, which is in agreement with our results, is that of Furevik (2000). He investigated the propagation of the sea surface temperature anomalies in the Nordic Seas with the complex principal component analysis. The leading mode propagated along the Norwegian coast with speeds of  $3\text{-}7 \text{ cm s}^{-1}$  where the highest speed ( $7 \text{ cm s}^{-1}$ ) was found at the northern coast of Norway.

#### 4. Conclusions

We conclude that the leading edge of the ' $^{99}\text{Tc}$ -front' has reached Arctic waters. The  $^{99}\text{Tc}$  concentration was  $\sim 9$  times higher than background levels in the north Norwegian Coastal Current in May/June 2000. We have not been able to define the leading edge of the ' $^{99}\text{Tc}$ -front' further east in the Barents Sea or within the Fram Strait after 1998 due to the geographical limit of sampling. In July 1998, EARP-related  $^{99}\text{Tc}$  was apparent southwest of Bear Island ( $\sim 73^\circ \text{ N}$ ) ( $^{99}\text{Tc}$  concentrations  $\sim 6$  times background levels) and one of three samples collected west of Spitsbergen at  $\sim 77^\circ \text{ N}$  contained slightly elevated levels of  $^{99}\text{Tc}$ . However, the leading edge of the ' $^{99}\text{Tc}$ -front' had passed west of Spitsbergen in May/June 2000 ( $^{99}\text{Tc}$  concentrations  $\sim 6$  times background levels). Transport times for  $^{99}\text{Tc}$  from Sellafield to west-southwest of Bear Island and west of Spitsbergen have been estimated to be 3-4 and 4-6 years, respectively. A transfer factor (TF) of  $\sim 5\text{-}65$  was calculated for  $^{99}\text{Tc}$  from Sellafield to Fugløy within the NWCC and of  $\sim 1\text{-}10$  to both west-southwest of Bear Island and Spitsbergen within the WSC.

In contrast to the rapid transport of  $^{99}\text{Tc}$  from the Irish Sea into the North Sea, the further transport along the NWCC and beyond was in this study found to have slowed down. We estimated a mean propagation speed for the ' $^{99}\text{Tc}$ -front' along the Norwegian coast of  $5 \text{ cm s}^{-1}$ , which is in good agreement with other studies, based on e.g. sea surface temperature anomalies. The variation of the propagation speed for  $^{99}\text{Tc}$  during the mid- and late-1990's is most likely due to variations in the NAO



winter index. A high NAO winter index and relatively increased westerly winds during the 1994/1995 winter caused a larger inflow from the Irish Sea to the North Sea and a more rapid transport time for  $^{99}\text{Tc}$ . The lower NAO winter indexes the following years led to a slower propagation rate for  $^{99}\text{Tc}$  along the NWCC and into Arctic waters.

The existence of EARP-related  $^{99}\text{Tc}$  has provided an excellent opportunity to investigate the relationships between climate indicators such as the North Atlantic Oscillation (NAO), the current and previous distributions of radioactive tracers, and annual/decadal changes in the circulation of the North Atlantic.

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### **References**

- Aarkrog, A., Boelskifte, S., Dahlgaard, H., Duniec, S., Hallstadius, L., Holm, E., Smith, J.N., 1987. Technetium-99 and Cesium-134 as long distance tracers in Arctic waters. *Estuarine, Coastal and Shelf Science* 24, 637-647.
- Aarkrog, A., Dahlgaard, H., Hansen, H., Holm, E., Hallstadius, L., Rioseco, J., Christensen, G., 1995. Radioactive tracer studies in the surface waters of the northern North Atlantic including the Greenland, Norwegian and Barents Seas. *Rit Fiskideildar* 9, 37-42.
- Blindheim, J., 1993. Seasonal variations in the Atlantic inflow to the Nordic Seas. ICES, C.M. 1993/C:39.

- Blindheim, J., Borokov, V., Hansen, B., Malmberg, S.-Aa., Turrell, W.R., Østerhus, S., 2000. Upper layer cooling and freshening in the Norwegian Sea in relation to atmospheric forcing. *Deep-Sea Research Part I* 47, 655-680.
- Brown, J., Hill, A.E., Fernand, L., Horsburgh, K.J., 1999. Observations of a seasonal Jet-like Circulation at the Central North Sea Cold Pool Margin. *Estuarine, Coastal and Shelf Science* 48, 343-355.
- Brown, J., Fernand, L., Horsburgh, K.J., Hill, A.E., Read, A.J., 2001. Paralytic shellfish poisoning on the east coast of the UK in relation to seasonal density-driven circulation. *Journal of Plankton Research* 23 (1), 105-116.
- Brown, J.E., Kolstad, A.K., Brungot, A.L., Lind, B., Rudjord, A.L., Strand, P., Føyn, L., 1999. Levels of  $^{99}\text{Tc}$  in Seawater and Biota Samples from Norwegian Coastal Waters and Adjacent Seas. *Marine Pollution Bulletin* 38 (7), 560-571.
- Christensen, G.C., Strålberg, E., Holm, E., 2001. Time trends of anthropogenic radionuclides at Utsira. The Eight Nordic Seminar on Radioecology, 25.-28. February 2001, Rovaniemi, Finland.
- Dahlgaard, H., 1994. Sources of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{99}\text{Tc}$  in the East Greenland Current. *Journal of Environmental Radioactivity* 25, 37-55.
- Dahlgaard, H., 1995. Transfer of European Coastal Pollution to the Arctic: Radioactive Tracers. *Marine Pollution Bulletin* 31 (1-3), 3-7.
- Dahlgaard, H., Aarkrog, A., Hallstadius, L., Holm, E., Riosco, J., 1986. Radiocesium transport from the Irish Sea via the North Sea and the Norwegian Coastal Current to East Greenland. *Rapport et Procès – Verbaux Réunion Conseil Internationale Exploration de Mer* 186, 70-79.
- Dahlgaard, H., Herrmann, J., van Weers, A.W., Masson, M., Ibbett, R.D., Chen, Q., 1995a. Quality assurance of the oceanographic tracers Technetium-99 and Antimony-125: Intercomparisons and recovery tests. *Journal of Marine Systems* 6, 391-396.
- Dahlgaard, H., Chen, Q., Herrmann, J., Nies, H., Ibbett, R.D., Kershaw, P.J., 1995b. On the background level of  $^{99}\text{Tc}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the North Atlantic. *Journal of Marine Systems* 6, 571-578.
- Dahlgaard, H., Bergan, T.D.S., Christensen, G.C., 1997. Technetium-99 and Caesium-137 Time Series at the Norwegian Coast Monitored by the Brown Algae *Fucus Vesiculosus*. *Radioprotection-Colloques* 32 (C2), 353-358.

- Dickson, R.R., Meincke, J., Malmberg, S.-A., Lee, A.J., 1988. The “Great Salinity Anomaly” in the northern North Atlantic 1968-1982. *Progress in Oceanography* 20, 103-151.
- Dickson, R.R., Osborn, T.J., Hurrell, J.W., Meincke, J., Blindheim, J., Adlandsvik, B., Vinje, T., Alekseev, G., Maslowski, W., 2000. The Arctic Ocean Response to the North Atlantic Oscillation. *Journal of Climate* 13, 2671-2696.
- Dooley, H.D., Furnes, G.K., 1981. Influence of wind field on the transport of the northern North Sea. In: *The Norwegian Coastal Current*. Sætre, R., Mork, M. (Eds), University of Bergen, Norway, pp. 57-71.
- Furevik, T., 2000. On anomalous sea surface temperatures in the Nordic Seas. *Journal of Climate* 13, 1044-1053.
- Furevik, T., 2001. Annual and interannual variability of Atlantic Water temperatures in the Norwegian and Barents Seas: 1980-1996. *Deep-Sea Research Part I* 48, 383-404.
- Furnes, G., 1980. Wind Effects in the North Sea. *Journal of Physical Oceanography* 10, 978-984.
- Guegueniat, P., Kershaw, P., Hermann, J., Bailly de Bois, P., 1997. New estimation of La Hague contribution to the artificial radioactivity of Norwegian waters (1992-1995) and Barents Sea (1992-1997). *The Science of the Total Environment* 202, 249-266.
- Harvey, B.R., Williams, K.J., Lovett, M.B., Ibbett, R.D., 1992. Determination of technetium-99 in environmental materials with rhenium as a yield monitor. *Journal of Radioanalytical and Nuclear Chemistry* 158, 417-436.
- Hunt, G.J., Kershaw, P.J., 1990. Remobilisation of artificial radionuclides from the sediments of the Irish Sea. *Journal of Radiological Protection* 10 (2), 147-151.
- Hurrell, J.W., 1995. Decadal Trends in the North Atlantic Oscillation: Regional Temperatures and Precipitation. *Science* 269, 676-679.
- Jefferies, D.F., Steele, A.K., Preston, A., 1982. Further studies on the distribution of  $^{137}\text{Cs}$  in British coastal waters – I. Irish Sea. *Deep-Sea Research Part A* 29 (6), 713-738.
- Kautsky, H., 1987. Investigations of the Distribution of  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$  and  $^{90}\text{Sr}$  and the Water Mass Transport Times in the Northern North Atlantic and the North Sea. *Deutsche hydrographische Zeitschrift* 40 (2), 49-69.

- Kautsky, H., 1988. Artificial radioactivity in the North Sea and the northern North Atlantic during the years 1977-1986. TECDOC, 481, International Atomic Energy Agency, Vienna, Austria, 71-102.
- Kershaw, P.J., Baxter, A., 1995. The transfer of reprocessing wastes from north-west Europe to the Arctic. *Deep-Sea Research Part II* 42 (6), 1413-1448.
- Kershaw, P.J., McCubbin, D., Leonard, K.S., 1999a. Continuing contamination of north Atlantic and Arctic waters by Sellafield radionuclides. *The Science of the Total Environment* 237/238, 119-132.
- Kershaw, P.J., Denoon, D.C., Woodhead, D.S., 1999b. Observations on the redistribution of plutonium and americium in the Irish Sea sediments, 1978 to 1996: concentrations and inventories. *Journal of Environmental Radioactivity* 44, 191-221.
- Leonard, K.S., McCubbin, D., Brown, J., Bonfield, R., Brooks, T., 1997. Distribution of Technetium-99 in UK Coastal Waters. *Marine Pollution Bulletin* 34 (8), 628-636.
- Livingston, H.D., 1988. The use of Cs and Sr isotopes as tracers in the Arctic Mediterranean Seas. *Philosophical Transactions of the Royal Society of London* A325, 161-176.
- Ljøen, R., Nakken, O., 1969. On the hydrography of the shelf waters off Møre and Helgeland. *Fiskeridirektoratets Skrifter, Serie Havundersøkelser* 15, 285-294.
- Loewe, P., 1996. Surface temperatures of the North Sea in 1996. *Deutsche hydrographische Zeitschrift* 48 (2), 175-184.
- MAFF and SEPA, 1999. Ministry of Agriculture, Fisheries and Food and Scottish Environment Protection Agency. *Radioactivity in Food and the Environment*, 1998. RIFE-4. MAFF and SEPA, London, pp 176.
- Mork, M., 1981. Circulation phenomena and frontal dynamics of the Norwegian coastal current. *Philosophical Transactions of the Royal Society of London* A302, 635-647.
- Nicholson, M.D., Hunt, G.J., 1995. Measuring the availability to sediments and biota of radionuclides in wastes discharged to the sea. *Journal of Environmental Radioactivity* 28 (1), 43-56.
- Orvik, K.A., Skagseth, Ø., Mork, M., 2001. Atlantic inflow to the Nordic Seas: current structure and volume fluxes from moored current meters, VM-ADCP and SeaSoar-CTD observations, 1995-1999. *Deep-Sea Research Part I* 48, 937-957.

- Poole, A.J., Denoon, D.C., Woodhead, D.S., 1997. The distribution and inventory of  $^{137}\text{Cs}$  in sub-tidal sediments of the Irish Sea. *Radioprotection-Colloques 32 (C2)*, 263-270.
- Poulain, P.-M., Warn-Varnas, A. Niiler, P.P., 1996. Near-surface circulation of the Nordic Seas measured by Lagrangian drifters. *Journal of Geophysical Research* 110 (C8), 18237-18258.
- Raisbeck, G.M., Yiou, F., Martin, F.X., Dahlgaard, H., Bergan, T.D.S., Christensen, G.C., 1997.  $^{129}\text{I}/^{99}\text{Tc}$  and  $^{129}\text{I}/^{137}\text{Cs}$  in the Norwegian Coastal Current from 1980-1995. The Third International Conference on Environmental Radioactivity in the Arctic, Tromsø, a:7.
- Rey, F., 2001. Havforskningsinstituttet, Interne Notat Nr. 01, 2001. Interne Toktrapporter Nr. 01, 2001. (Cruise Report). Institute of Marine Research, Bergen, Norway.
- Smith, J.N., Ellis, K.M., Kilius, L.R., 1998.  $^{129}\text{I}$  and  $^{137}\text{Cs}$  tracer measurements in the Arctic Ocean. *Deep-Sea Research Part I*, 45, 959-984.
- Smith, V., Ryan, R.W., Pollard, D., Mitchell, P.I., Ryan, T.P., 1997. Temporal and geographical distributions of  $^{99}\text{Tc}$  in inshore waters around Ireland following increased discharges from Sellafield. *Radioprotection-Colloques 32 (C2)*, 71-77.
- Sætre, R., Ljøen R., 1971. The Norwegian Coastal Current. In: *Proceedings of the first international conference on port and ocean engineering under Arctic conditions*, Vol. 2, Technical University of Norway, Trondheim, Norway, pp. 514-535.
- Sætre, R., 1999. Features of the central Norwegian shelf circulation. *Continental Shelf Research* 19, 1809-1831.

Table 1. Sampling dates, sampling positions, water depths (m), sampling depths (m), salinities, temperatures and  $^{99}\text{Tc}$  concentrations ( $\text{Bq m}^{-3}$ ). The error includes a statistical counting error and uncertainties in the chemical procedure. bd = below detection limit.

Sampling date	Latitude	Longitude	Depth (m)	Sampling depth (m)	Salinity	Temperature ( $^{\circ}\text{C}$ )	$^{99}\text{Tc}$ ( $\text{Bq m}^{-3}$ )	Error (%)
03.07.98	72° 48' N	13° 51' E	1133	5	34.776	8.444	0.28	10.5
03.07.98	73° 34' N	16° 07' E	452	5	35.005	7.266	0.25	7.3
04.07.98	74° 21' N	18° 25' E	73	5	34.616	1.406	0.08	15.8
04.07.98	74° 59' N	18° 18' E	94	5	34.077	3.172	0.13	10.4
05.07.98	76° 20' N	16° 11' E	77	5	32.647	-0.366	bd	-
06.07.98	76° 55' N	12° 30' E	746	5	34.371	5.233	0.08	19.7
06.07.98	76° 55' N	13° 30' E	101	5	32.348	2.746	0.16	9.8
06.07.98	76° 58' N	15° 30' E	150	5	32.626	1.567	0.06	24.9
09.07.98	71° 30' N	19° 48' E	236	5	34.661	9.150	0.42	5.4
09.07.98	71° 00' N	19° 54' E	190	5	34.462	9.897	0.65	5.0
09.07.98	70° 30' N	20° 00' E	126	5	33.863	10.979	0.86	4.5
09.07.98	70° 30' N	20° 00' E	126	30	34.554	7.161	0.80	4.5
18.06.99	68° 26' N	14° 01' E	110	5	33.616	8.214	1.61	4.2
18.06.99	68° 51' N	12° 48' E	660	5	34.912	9.245	0.52	4.9
24.06.99	74° 30' N	14° 37' W	267	5	32.823	-0.041	0.13	10.6
24.06.99	74° 30' N	14° 37' W	267	30	33.925	0.077	0.09	18.3
26.06.99	74° 30' N	03° 00' E	3320	5	34.738	2.164	0.09	20.6
27.06.99	74° 30' N	11° 00' E	2390	5	35.100	6.020	0.21	8.5
28.06.99	74° 30' N	14° 00' E	2106	5	35.103	6.328	0.26	6.5
28.06.99	74° 30' N	16° 40' E	193	5	35.090	6.927	0.25	6.4
01.07.99	70° 30' N	20° 00' E	146	5	33.812	9.212	1.49	4.1
01.07.99	71° 30' N	19° 48' E	236	5	34.663	9.431	0.93	4.4
02.07.99	73° 30' N	19° 20' E	480	5	35.054	6.680	0.26	7.3
02.07.99	74° 15' N	19° 10' E	58	5	34.920	3.605	0.22	7.5
03.07.99	76° 35' N	20° 00' E	205	5	33.536	1.888	0.11	13.2
28.05.00	69° 29' N	10° 56' E	2954	5	35.132	7.329	0.27	5.12
31.05.00	74° 05' N	03° 40' W	3494	5	34.835	-0.067	0.05	11.1
01.06.00	74° 30' N	11° 43' W	2955	5	34.327	-0.652	0.10	7.15
02.06.00	74° 30' N	03° 00' W	3601	5	34.842	0.487	0.05	12.4
03.06.00	74° 30' N	00° 00' E	3740	5	34.830	0.411	0.05	16.2
03.06.00	74° 30' N	03° 00' E	3354	5	34.823	1.360	0.08	10.0
04.06.00	74° 30' N	11° 00' E	2396	5	35.087	5.558	0.30	4.65
04.06.00	74° 30' N	13° 59' E	2112	5	35.074	6.012	0.39	4.50
05.06.00	74° 30' N	16° 30' E	252	5	35.072	5.952	0.38	4.44
05.06.00	74° 30' N	18° 30' E	64	5	34.628	0.392	0.22	5.40
07.06.00	75° 49' N	17° 35' E	247	5	34.023	0.848	0.20	5.61
07.06.00	75° 43' N	15° 35' E	382	5	33.690	0.538	0.22	5.02
08.06.00	75° 30' N	12° 00' E	1986	5	35.091	5.277	0.27	4.75
08.06.00	76° 20' N	10° 25' E	2168	5	35.050	4.466	0.25	4.86
08.06.00	76° 20' N	16° 00' E	102	5	33.858	-0.021	0.20	5.67
09.06.00	77° 20' N	13° 45' E	73	5	33.567	0.128	0.21	5.08
09.06.00	77° 20' N	10° 55' E	1076	5	34.802	4.000	0.30	4.63
10.06.00	74° 15' N	19° 10' E	60	5	34.697	0.224	0.19	5.27
11.06.00	72° 30' N	19° 34' E	389	5	35.087	6.275	0.35	4.61
12.06.00	70° 30' N	20° 00' E	126	5	34.464	7.101	1.19	4.09

Table 1. (contd.) Sampling dates, sampling positions, water depths (m), sampling depths (m), salinities, temperatures and  $^{99}\text{Tc}$  concentrations ( $\text{Bq m}^{-3}$ ). The error includes a statistical counting error and uncertainties in the chemical procedure. bd = below detection limit.

Sampling date	Latitude	Longitude	Depth (m)	Sampling depth (m)	Salinity	Temperature ( $^{\circ}\text{C}$ )	$^{99}\text{Tc}$ ( $\text{Bq m}^{-3}$ )	Error (%)
14.06.00	71° 00' N	26° 30' E	279	5	33.561	6.347	0.72	4.47
14.06.00	71° 10' N	26° 30' E	165	5	34.553	6.232	0.63	4.49
14.06.00	71° 20' N	25° 00' E	301	5	34.543	6.421	0.72	4.43
14.06.00	71° 10' N	25° 00' E	155	5	33.759	5.774	0.69	4.45
16.06.00	71° 20' N	28° 00' E	412	5	33.559	6.331	0.70	4.44
16.06.00	71° 10' N	28° 00' E	112	5	34.106	5.546	0.71	4.48
16.06.00	70° 50' N	30° 00' E	381	5	33.910	5.906	0.88	4.29
16.06.00	71° 00' N	30° 00' E	299	5	33.757	5.828	0.90	4.32
23.06.00	70° 30' N	31° 13' E	187	5	33.869	5.961	0.82	4.35
23.06.00	70° 24' N	31° 13' E	131	5	33.456	6.136	0.94	4.35

Table 2. Transport times from Sellafield to various North Atlantic and Arctic regions based on  $^{99}\text{Tc}$ .

Location	Transport time	Data source
North Channel, UK	6 months	Leonard et al. 1997
Northern North Sea	9 months	Leonard et al. 1997
SW Norwegian coast	~ 2.5 years	Brown et al. 1999; Christensen et al. 2001
Ingøy, NWCC North	~ 3 years	G. Christensen (pers. comm.)
W Bear Island, WSC	3-4 years	This study
W Spitsbergen, WSC	4-6 years	This study

Table 3. Transfer factors (TFs) calculated at three locations by dividing the environmental concentration of  $^{99}\text{Tc}$  with the discharge of  $^{99}\text{Tc}$   $t$  years earlier, where  $t$  is the transport time. The transport times used are based on  $^{137}\text{Cs}$  (1994) and  $^{99}\text{Tc}$  (1998, 1999 and 2000).

Location	Latitude	Longitude	TF ( $\text{Bq m}^{-3}/\text{PBq y}^{-1}$ )			
			1994	1998	1999	2000
North Norway, NWCC	70° 30' N	20° 00' E	66	4	10	14
W Bear Island, WSC	73° 30' N - 74° 30' N	11° 00' E - 17° 30' E	9	2	1	3
W Spitsbergen, WSC	76° 55' N - 77° 35' N	09° 20' E - 15° 30' E	10	3	-	2

Table 4. Transport times and Transfer factors from Sellafield to various North Atlantic and Arctic regions, mainly based on  $^{137}\text{Cs}$ .

Location	Transport time (y)	Transfer Factor (TF) ( $\text{Bq m}^{-3}/\text{PBq y}^{-1}$ )	Data source
North Channel, UK	1		Kershaw and Baxter (1995)
North Sea	3		Dahlgaard (1995)
North Sea		17	Aarkrog et al. (1995)
Kattegat	4	15	Dahlgaard (1995)
NW Norwegian Coastal Current	3–4	10–50	Dahlgaard (1995)
North Norway (North Cape)	5–6		Kautsky (1987)
SW Barents Sea	4–6		Kershaw and Baxter (1995)
S-SE Barents Sea	5–6	5–10	Dahlgaard (1995)
Kara Sea	5–6	5–10	Dahlgaard (1995)
Bear Island-Spitsbergen area	6–7		Kautsky (1987)
West Spitsbergen	~5	~5	Dahlgaard (1995)
NW Spitsbergen		2–3	Aarkrog et al. (1987)
Arctic Ocean	~6–10	~1–10	Dahlgaard (1995)
East Greenland Current	7–10	1–2	Dahlgaard (1995)
North Atlantic Current, Faroe Islands	~14–17	~0,3	Dahlgaard (1995)



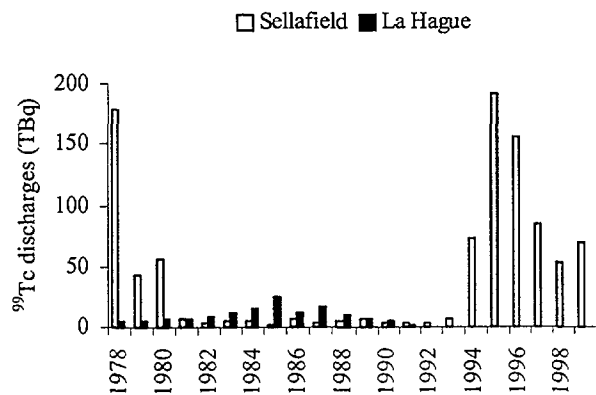


Figure 1. Discharges of  $^{99}\text{Tc}$  from BNF-Sellafield and COGEMA-La Hague 1978-1999

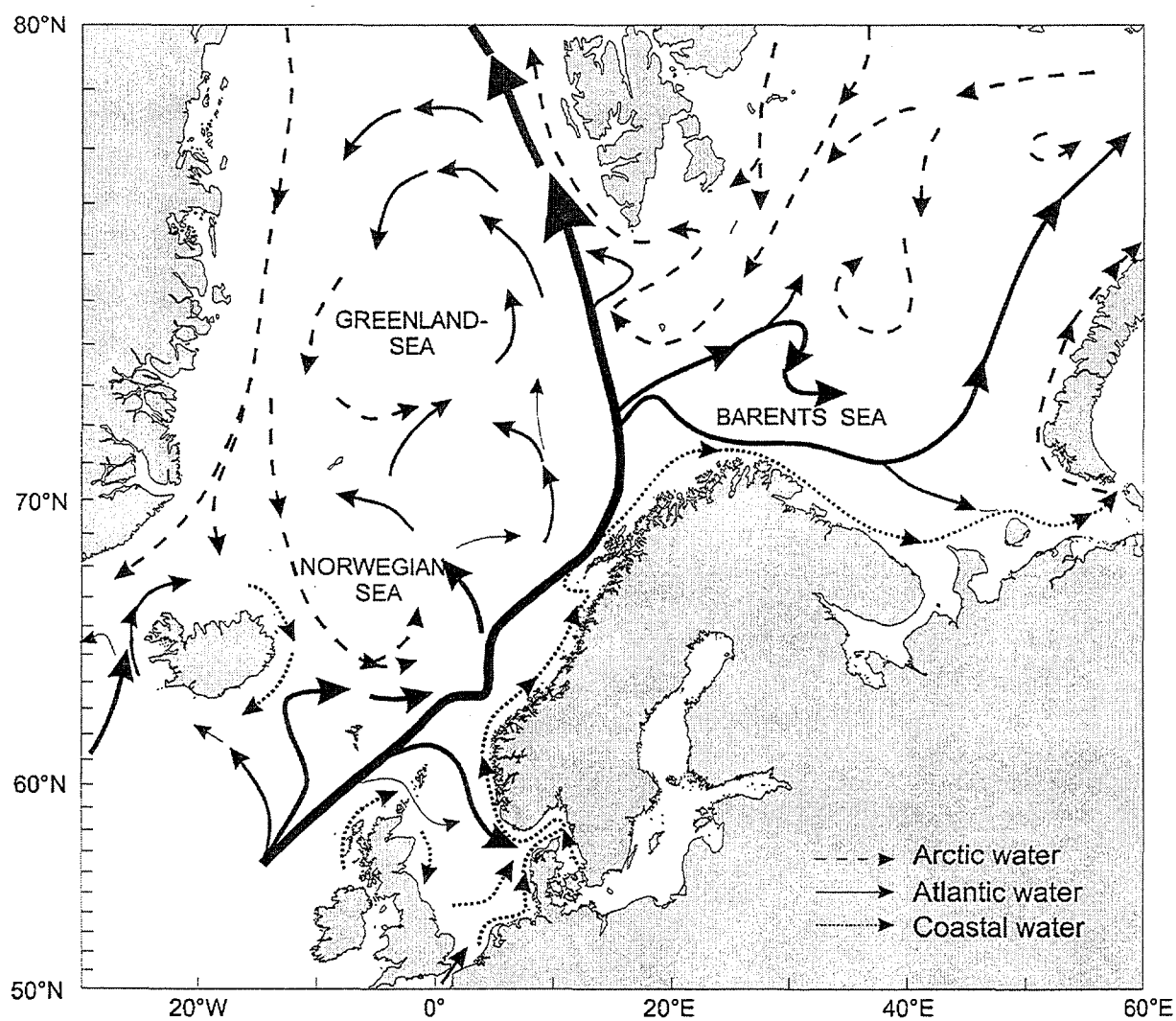
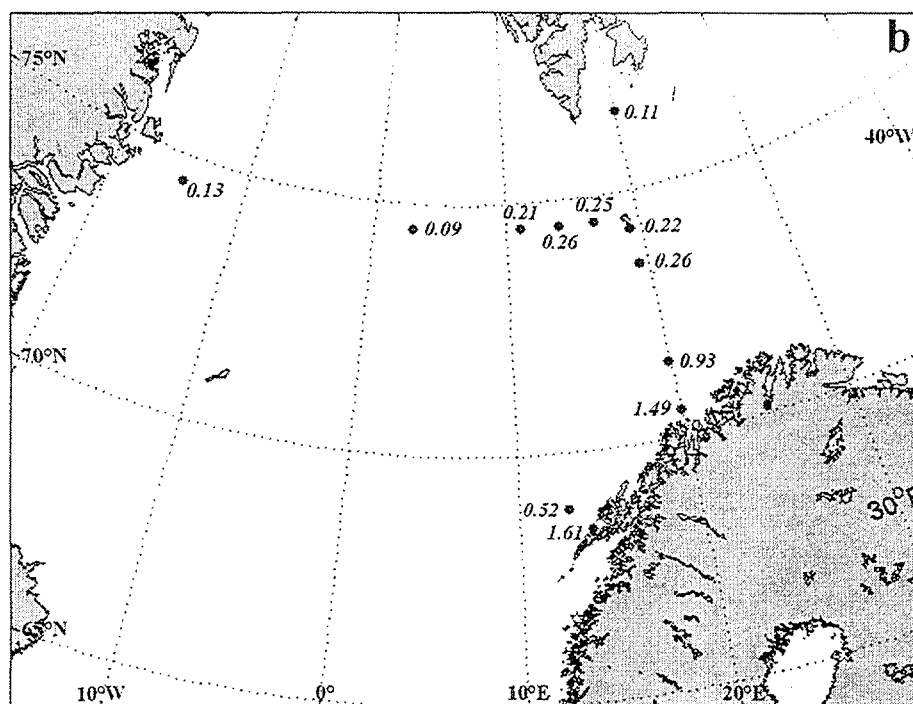
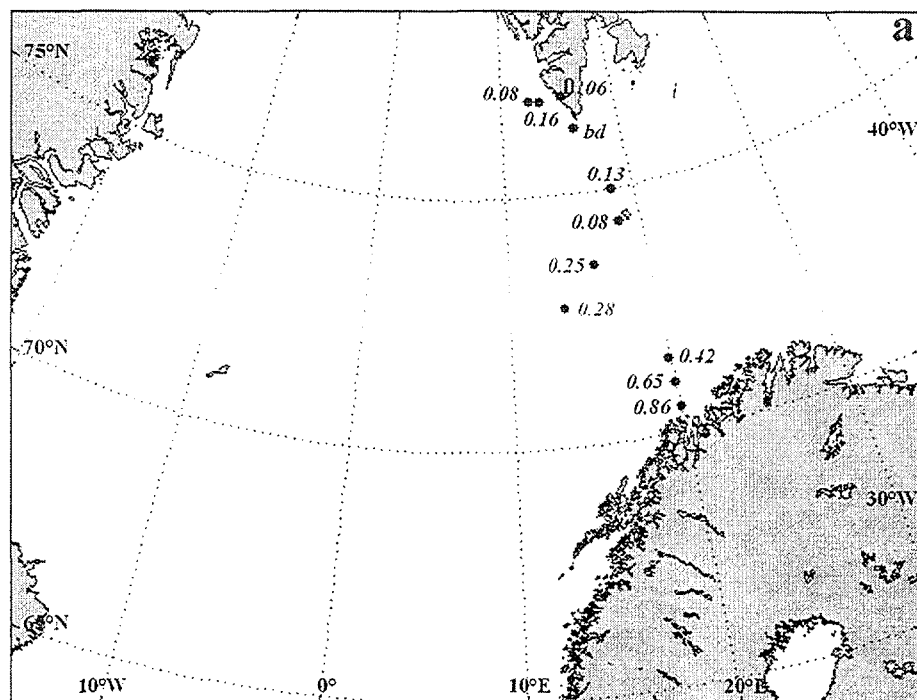


Figure 2. Circulation of surface waters of the North, Norwegian, Greenland and Barents Seas.



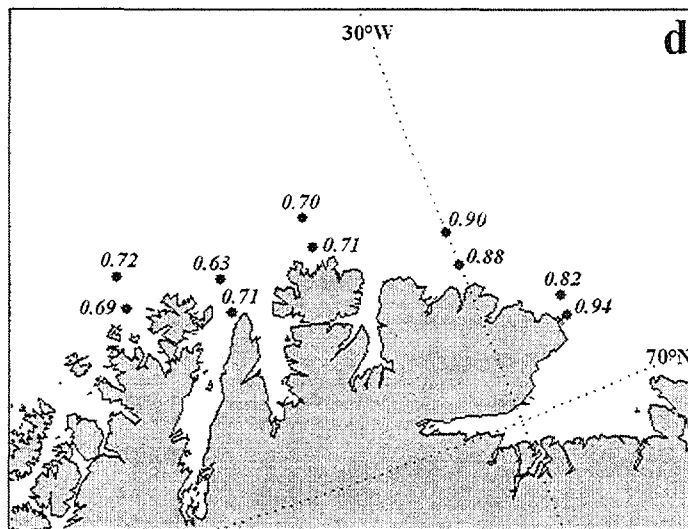
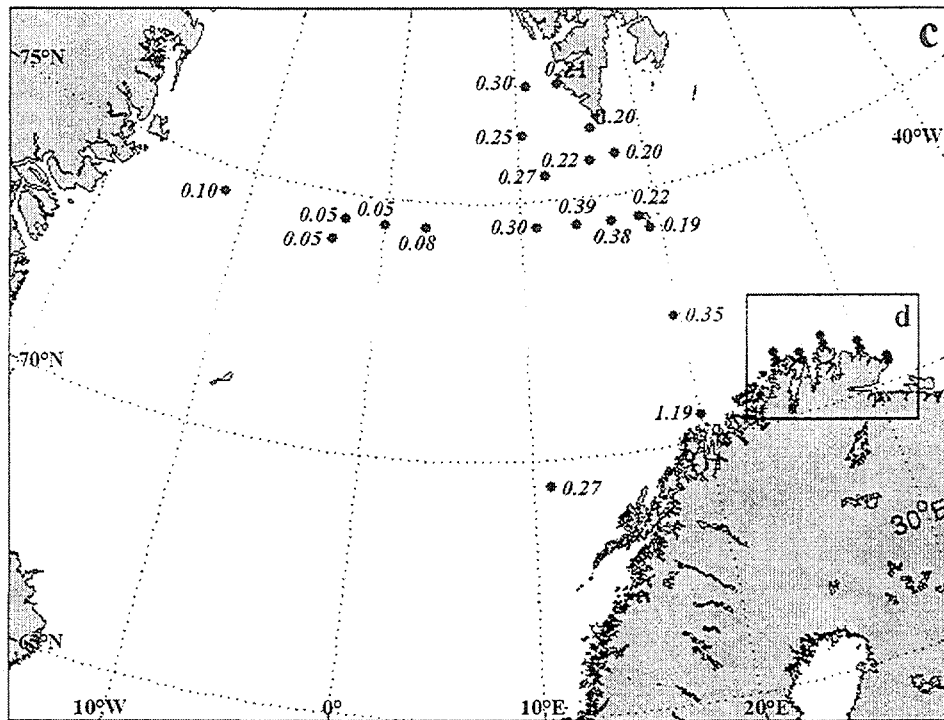


Figure 3. Spatial distribution of  $^{99}\text{Tc}$ , in (a) 1998; (b) 1999; (c) 2000; (d) 2000

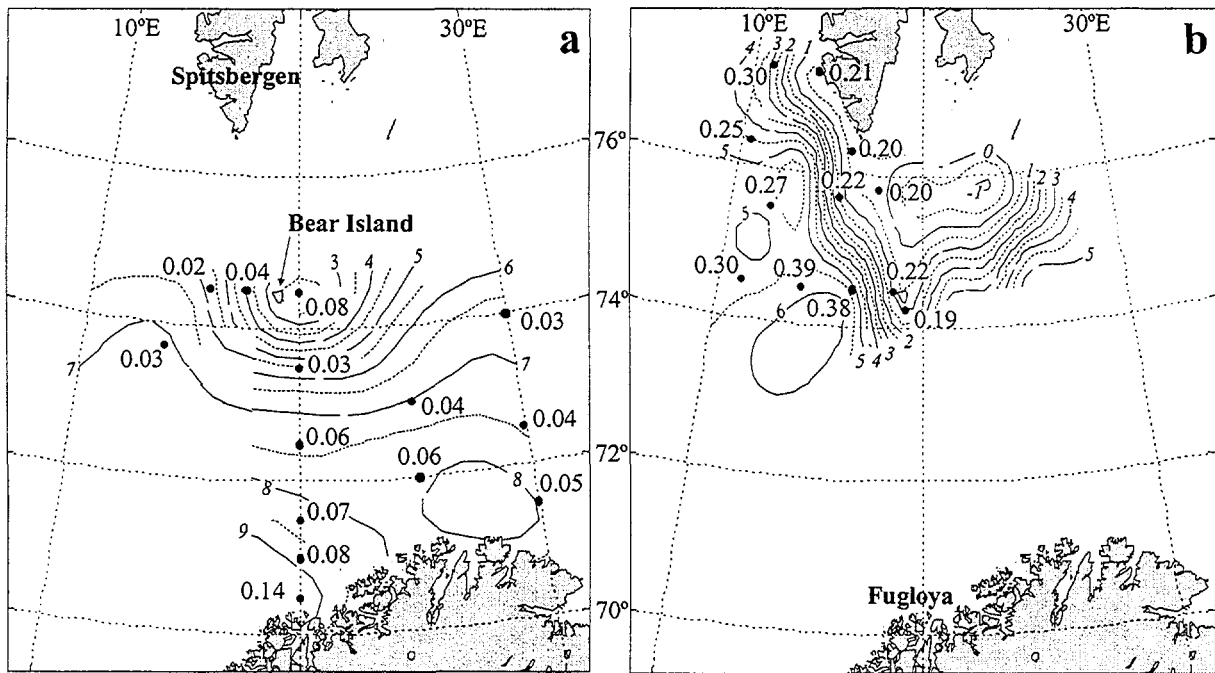


Figure 4. Spatial distribution of  $^{99}\text{Tc}$  overlay on near-surface temperatures in (a) 1994; (b) 2000



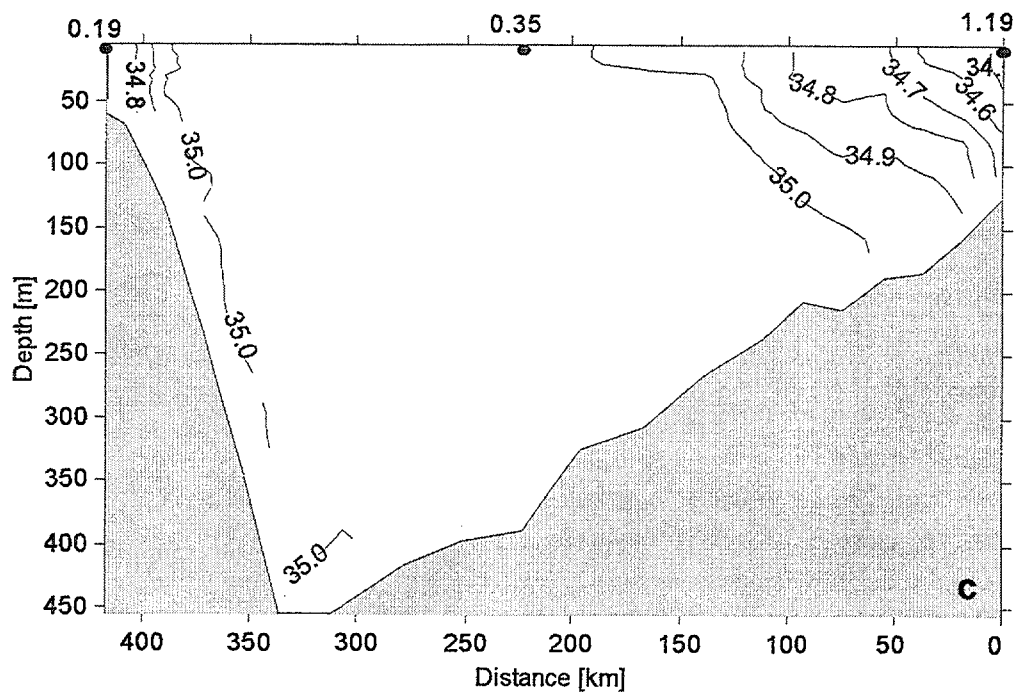


Figure 5. The Fugløy (70° 30' N; 20° 00' E) - Bear Island (74° 15' N; 19° 10' N) section (Bear Island to the left), showing salinity and  $^{99}\text{Tc}$  concentrations, (a) 1998; (b) 1999; (c) 2000

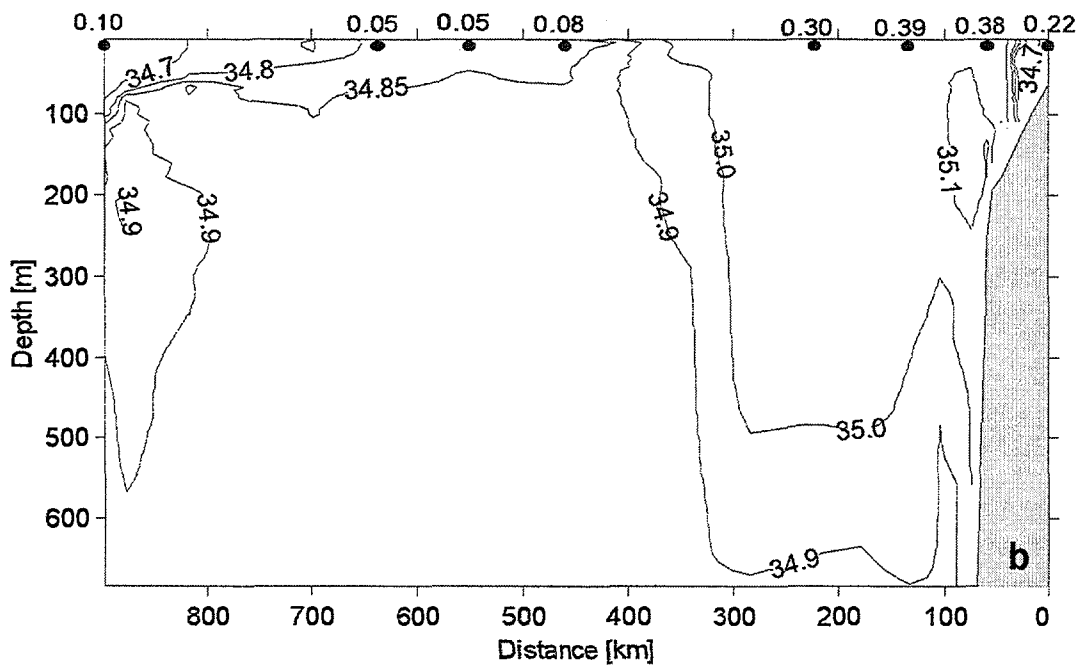
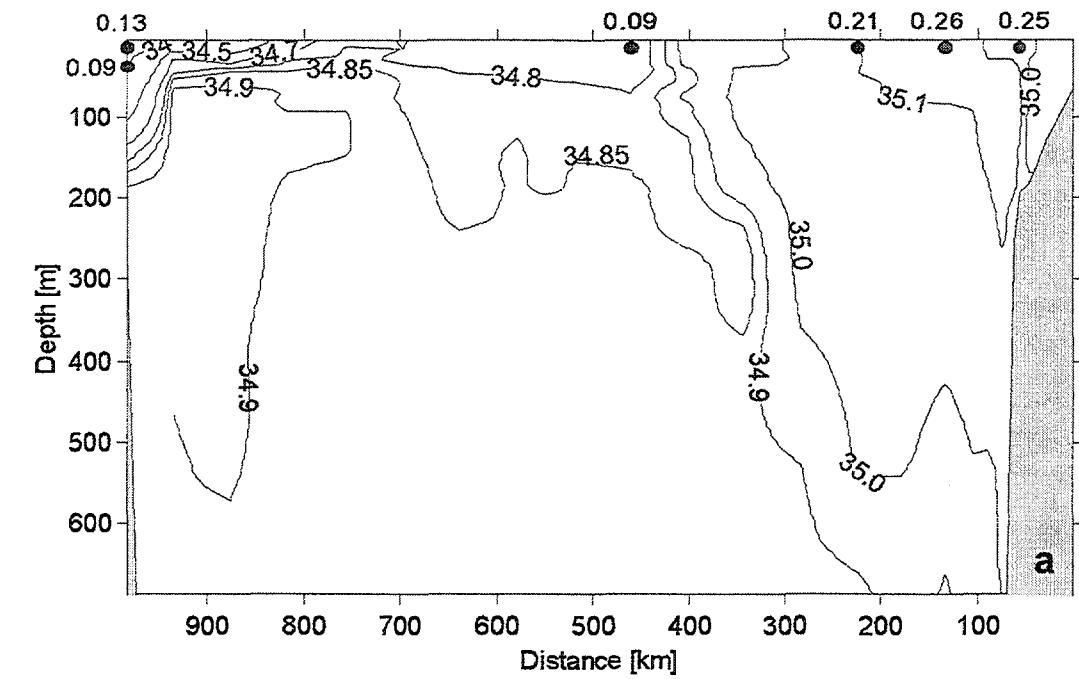


Figure 6. The Bear Island West section (along  $74^{\circ} 30' \text{ N}$ ), showing salinity and  $^{99}\text{Tc}$  concentrations, (a) 1999; (b) 2000

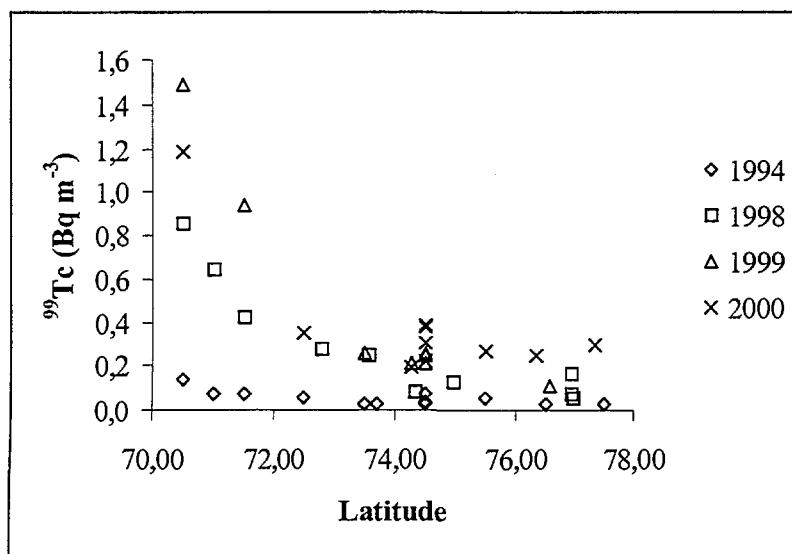


Figure 7. The progress of the  $^{99}\text{Tc}$  plume in the WSC; 1994, 1998, 1999 and 2000. Samples with PW characteristics ( $S < 34.5$  and  $T < 1^\circ\text{C}$ ) are excluded.

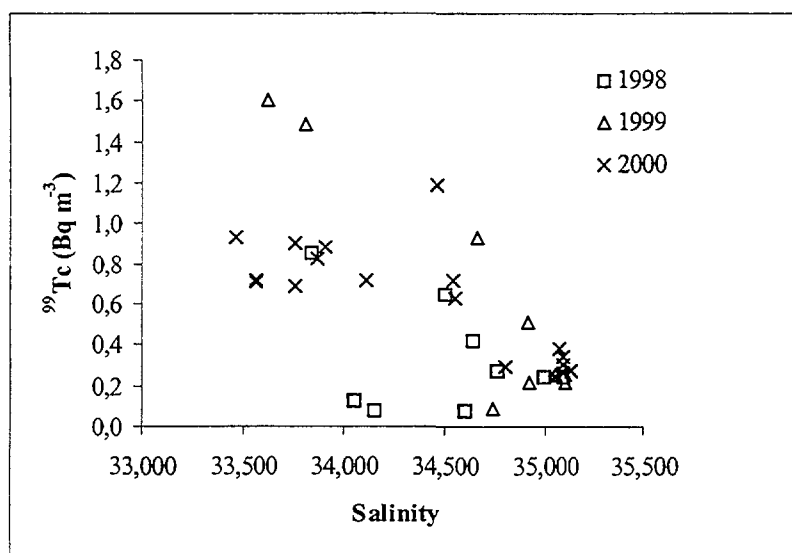


Figure 8.  $^{99}\text{Tc}$  concentrations in surface waters of the NWCC, NWAC, NCC and WSC for 1998-2000 plotted against salinity. Samples with PW characteristics ( $S < 34.5$  and  $T < 1^\circ\text{C}$ ) are excluded.



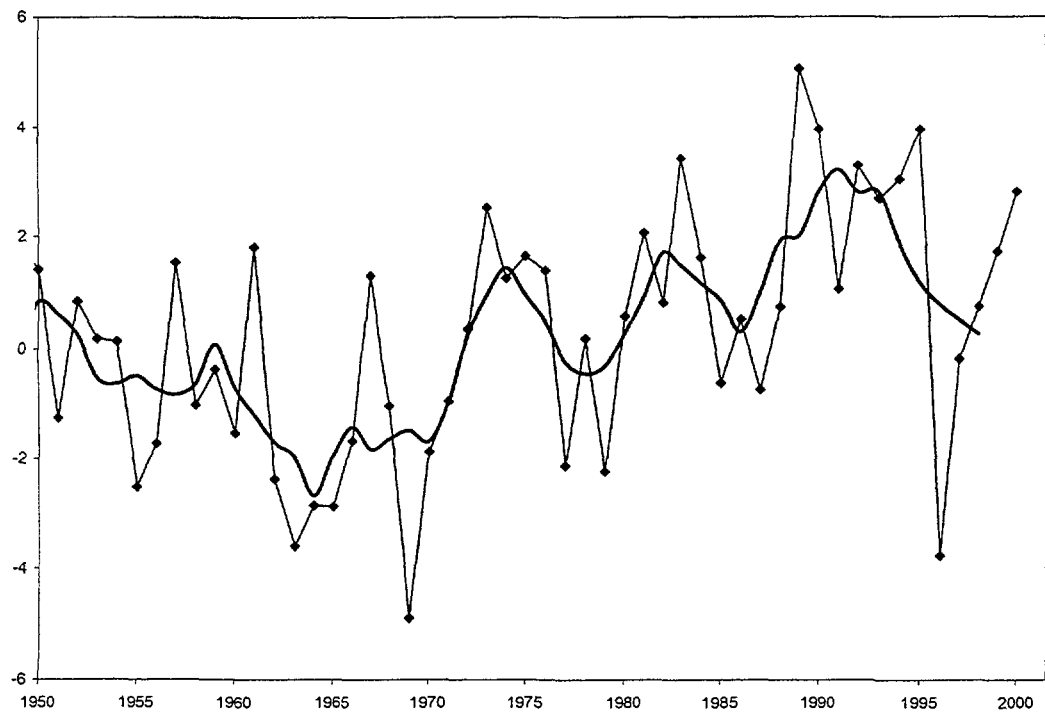


Figure 9. The NAO winter index 1950-2000, updated from Hurrell (1995).

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**Department of Chemistry,  
University of Bergen, Norway**