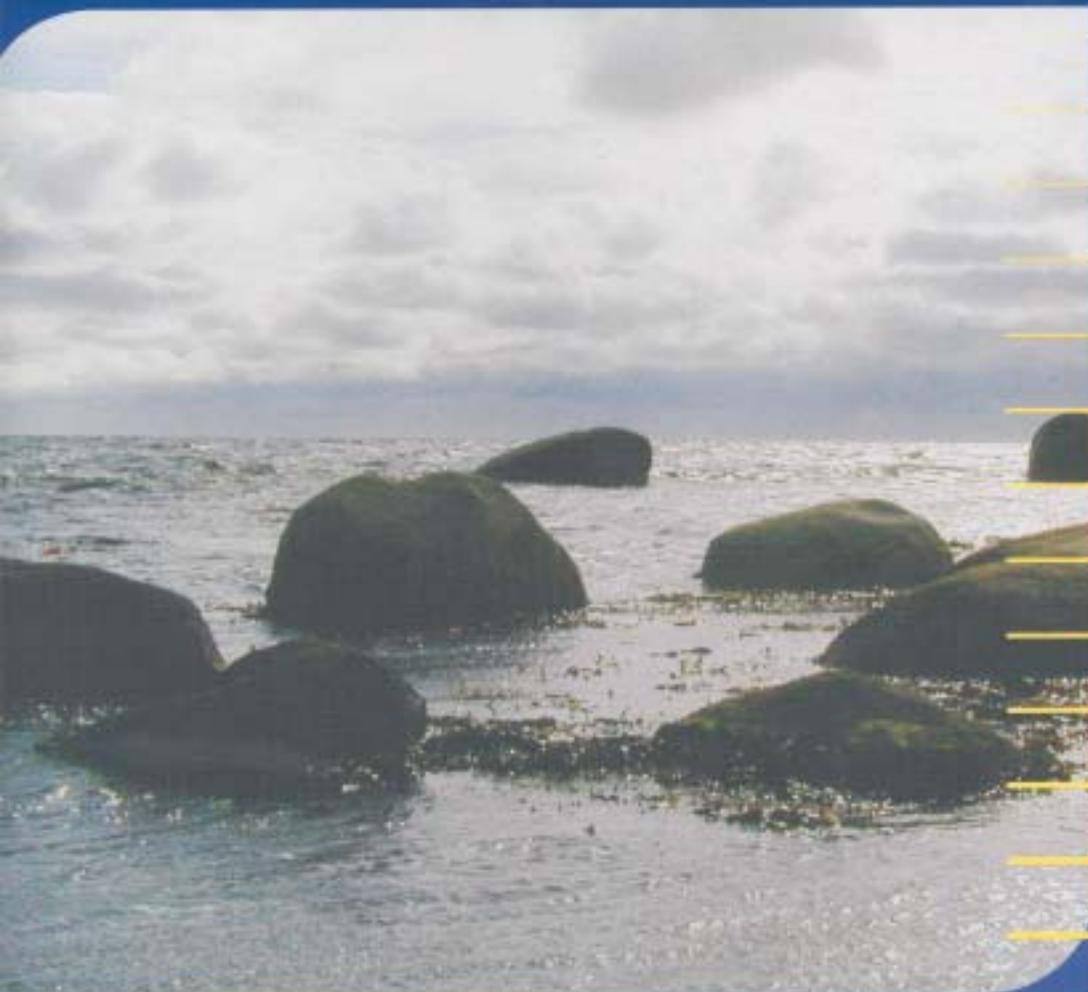


Radioactivity in the Marine Environment 2000 and 2001

Techneium-99 concentrations in Norwegian coastal waters and biota



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strålevern

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Protection Authority**

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Key words: Technetium-99, Sellafield, Technetium-99-discharges, radioactivity, sea water, seaweed, lobster.

Abstract: In this report, Technetium-99 results from the national monitoring programme, RAME, are presented. This includes data from coastal sampling stations and expeditions in adjacent seas in 2000 and 2001. Technetium-99 activity concentrations are measured in sea water, seaweed, lobsters and other types of biota. Discharges from Sellafield still lead to enhanced levels of Technetium-99 along the Norwegian coast.

Referanse: Kolstad Anne Kathrine and Lind Bjørn. Radioactivity in the Marine Environment 2000 and 2001, Technetium-99 concentrations in Norwegian coastal waters and biota. StrålevernRapport 2002:6. Østerås: Statens strålevern, 2002. Språk: engelsk.

Emneord: Technetium-99, Sellafield, Technetium-99-utslipp, radioaktivitet, havvann, tang, hummer

Resymé: I denne rapporten presenteres Technetium-99 resultater fra det nasjonale overvåkningsprogrammet, RAME. Den inkluderer prøvemateriale fra stasjoner langs norskekysten og tokt til nærliggende havområder. Technetium-99 konsentrasjoner i havvann, tang, hummer og annen biota blir presentert. Utslipp fra Sellafield anlegget fører fortsatt til forhøyede konsentrasjoner av Technetium-99 langs norskekysten.

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Index

| | | |
|----------|---|-----------|
| 1 | INTRODUCTION | 4 |
| 1.1 | ⁹⁹ Tc (TECHNETIUM-99) | 4 |
| 2 | MATERIALS AND METHOD | 5 |
| 2.1 | SAMPLING | 5 |
| 2.2 | METHOD | 6 |
| 3 | RESULTS AND DISCUSSION | 7 |
| 3.1 | ⁹⁹ Tc IN SEA WATER | 7 |
| 3.2 | ⁹⁹ Tc IN SEAWEED AND OTHER BIOTA | 11 |
| 3.2.1 | SEAWEED | 11 |
| 3.2.2 | CRUSTACEANS AND MOLLUSCS | 14 |
| 4 | CONCLUSIONS | 16 |
| | REFERENCES | 18 |

1 Introduction

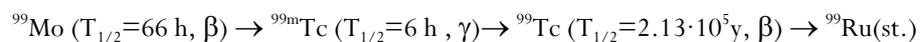
The issue of actual and potential radioactive contamination in the marine environment has received substantial attention in Norway in recent years. In 1994 and 1995, discharges of Technetium-99 from the reprocessing facilities at Sellafield in the United Kingdom increased sharply. There were public concerns about the consequences of such releases, because the radionuclides discharged to the Irish Sea in earlier times were known to be transported by ocean currents via the North Sea into the Norwegian Coastal Current (NCC) and the Barents Sea. In response to such concerns, programmes for monitoring radioactivity in the marine environment were established. Due to the economic importance of the fishing industry and its vulnerability to any rumours of radioactive contamination, one of the main objectives of such programmes is to document levels and trends of radionuclides in the Norwegian marine environment.

In 1999, the sampling programme along the Norwegian coast was extended under the new marine monitoring programme (RAME) funded by the Ministry of Environment. The main objectives of the programme are to document levels, distributions and trends of radionuclides in Norwegian coastal and sea areas.

In this report, Technetium-99 data from regular coastal sampling stations and sampling expeditions to adjacent seas are presented. Time series data pertaining to Technetium-99 in seaweed (*Fucus vesiculosus*) and sea water from Hillesøy in the period between July 1997 and January 2002 are presented. Finally, Technetium-99 data for a set of lobsters collected in 2001 are reported.

1.1 ⁹⁹Tc (Technetium-99)

The fission product ⁹⁹Tc (Technetium-99), is produced by decay of ⁹⁹Mo in nuclear reactors or nuclear explosions.



^{99}Tc is a soft beta emitter ($E_{\text{max}} = 292 \text{ keV}$) with a long half-life of 2.13×10^5 years. The long half-life of ^{99}Tc means that the radionuclide will persist in the environment for many generations.

In sea water, ^{99}Tc exists predominantly as the pertechnetate ion (TcO_4^-) which is highly soluble and available for transport with ocean currents.

2 Materials and Method

2.1 Sampling

Collection of samples takes place on an annual or monthly basis at coastal stations along the Norwegian coast and includes water, seaweed and other biota samples. Annually, NRPA collecting samples at Lista, see Figure 1.



Figure 1. Collection of samples at *Østhaselstrand, Lista (2001)*.

In northern Norway, at Hillesøy station in Troms, monthly sampling of sea water and seaweed is conducted. Each year, NRPA also takes part in expeditions to sea areas adjacent to Norway with the Institute of Marine Research in Norway. In October 2000, NRPA joined a cruise starting in Kirkenes

and ending a few weeks later in Bergen. ^{99}Tc results from approximately 20 stations were subsequently obtained. In November 2001, a total of 40 water samples were collected in the North Sea, covering the main part of the North Sea and Skagerrak between 53°00'N and 61°00'N. From these expeditions mainly sea water results are reported. In the years 2000 and 2001, NRPA conducted marine radioecological fieldwork in areas off Spitsbergen and in the Fram Strait. Marine radioecological surveys were undertaken in the Fram Strait by participating on board the research vessels “RV Polarstern” (summer 2000) and “RV Lance” (spring 2001). In December 2000, NRPA started the regular collection of water samples at Hopen (76°N, 25°E) and Bjørnøya (74°N, 18°E) and six months later at Jan Mayen (71°N, 8°W).

2.2 Method

NRPA normally analyses samples of 50 litres of filtered sea water (<1 μm) and 10 grams of dried biota (Kolstad et al., 1999). The analytical procedure is similar to the method used at Risø National Laboratory (Chen et al., 2001). Technetium is separated from the matrix by ion exchange chromatography using AG 1-X4 resin (BIO-RAD 100-200 mesh) and separation techniques such as precipitation and solvent extraction, see Figure 2. $^{99\text{m}}\text{Tc}$ is used as a yield monitor and the chemical recovery is achieved by gamma counting on a NaI well-detector prior to beta measurement on a low background anti-coincidence counter.

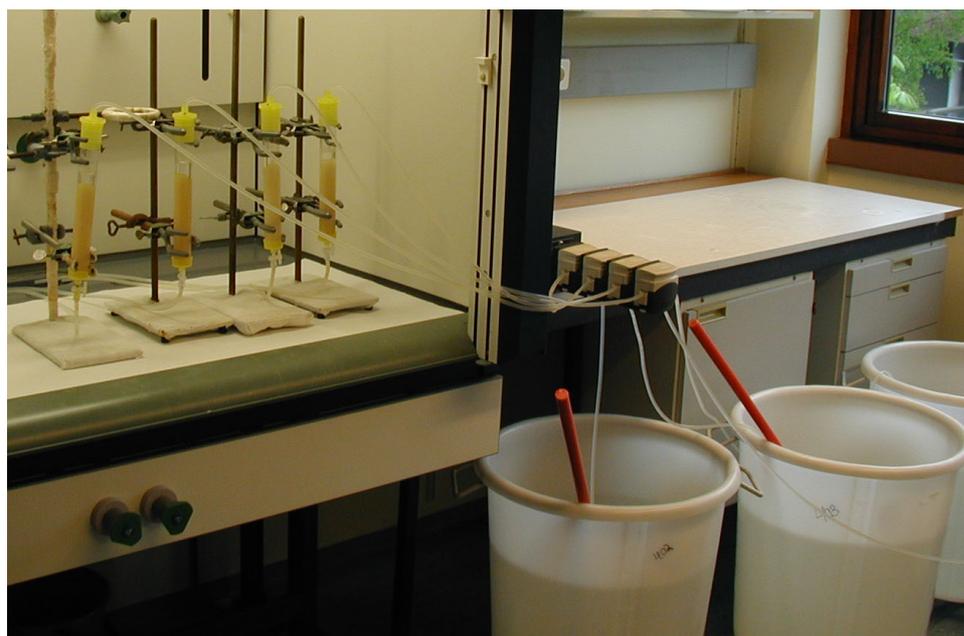


Figure 2. Technetium is separated from sea water by ion exchange chromatography.

3 Results and discussion

The surface circulation pattern around the Norwegian coast is shown in Figure 3. Discharges of radionuclides from the Sellafield reprocessing plant are transported from the Irish Sea and the English channel via the North Sea and into the Norwegian Coastal Current (NCC), to the Barents Sea and beyond.

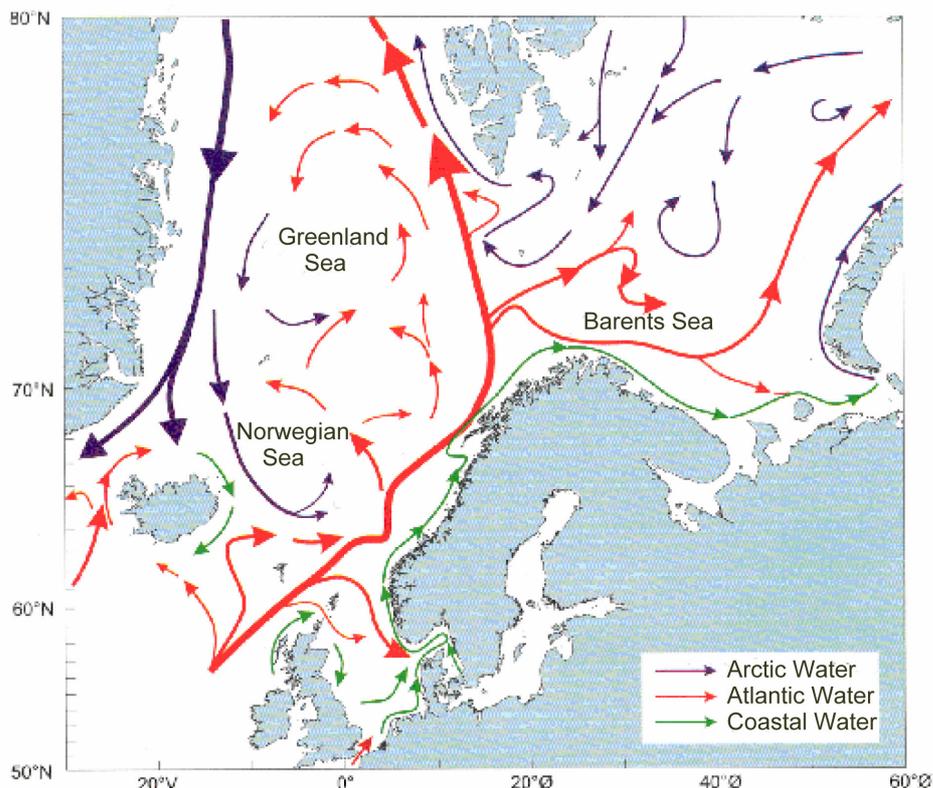


Figure 3. The surface circulation pattern of the Northern Seas (adapted from Aure et al. 1998).

3.1 ^{99}Tc in sea water

In October 2000, sea water samples were collected during an expedition along the northern Norwegian coast. The ^{99}Tc data are presented along with the results from permanent sampling stations in Figure 4. The concentrations of ^{99}Tc in surface waters are in the range $0.69\text{--}1.78\text{ Bq m}^{-3}$, average of 1.3 Bq m^{-3} ($1\sigma = 0.26$). All sea water concentrations refer to filtered water ($<1\ \mu\text{m}$). At three stations, the concentrations were below 1.0 Bq m^{-3} . This is probably due to dilution with Atlantic waters since the waters had salinities of about 35 ‰ .

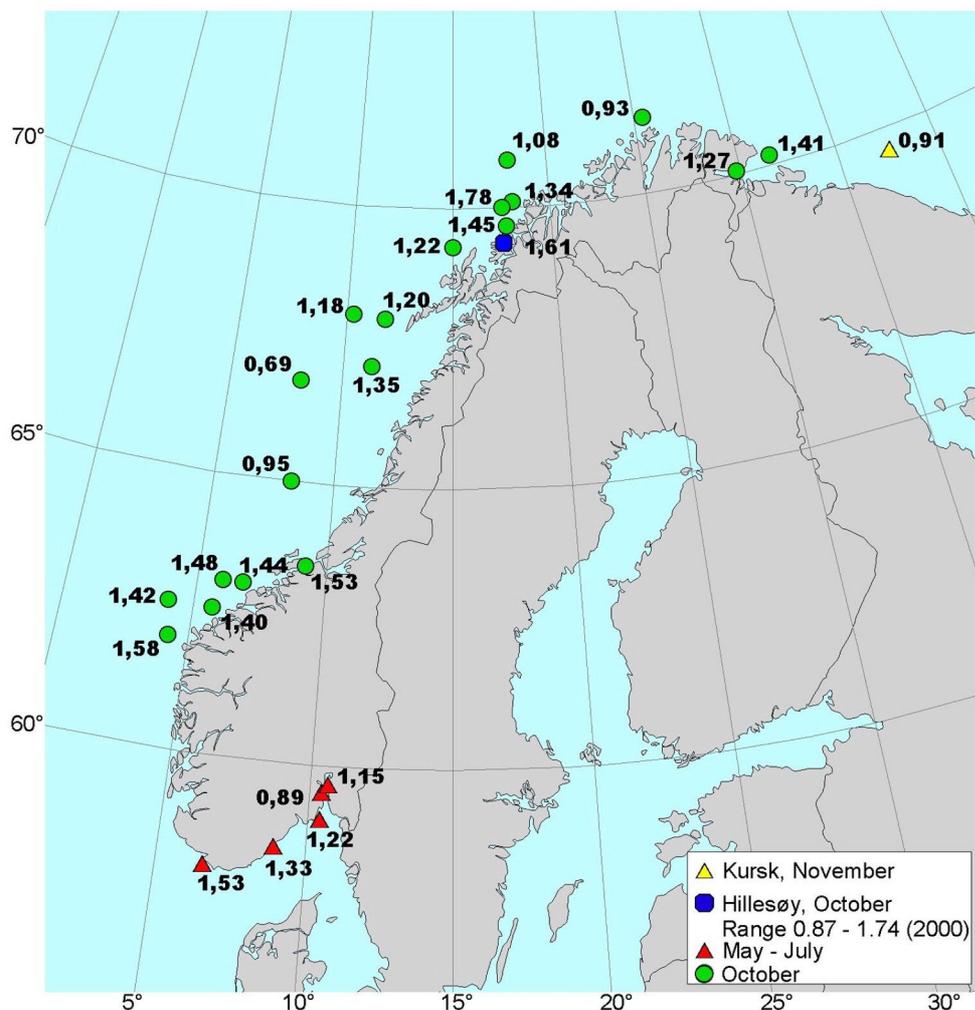


Figure 4. Concentrations of ^{99}Tc (Bq m^{-3}) in surface sea water samples collected in 2000.

The data from the North Sea expedition in 2001 are shown in Figure 5, along with the results from permanent sampling stations at the south coast. By considering concomitant salinity data, it is seen that the ^{99}Tc results reflect the general circulation of water masses in the North Sea (Brown et al., 1999). The concentrations of ^{99}Tc in surface waters are in the range 0.22-7.30 Bq m^{-3} . The highest concentrations are seen in western and southern parts of the North Sea, near the coast of Scotland and England. The very low concentrations observed at a few stations in the south eastern parts of the North Sea represent channel water or coastal waters unaffected by Sellafield discharges.

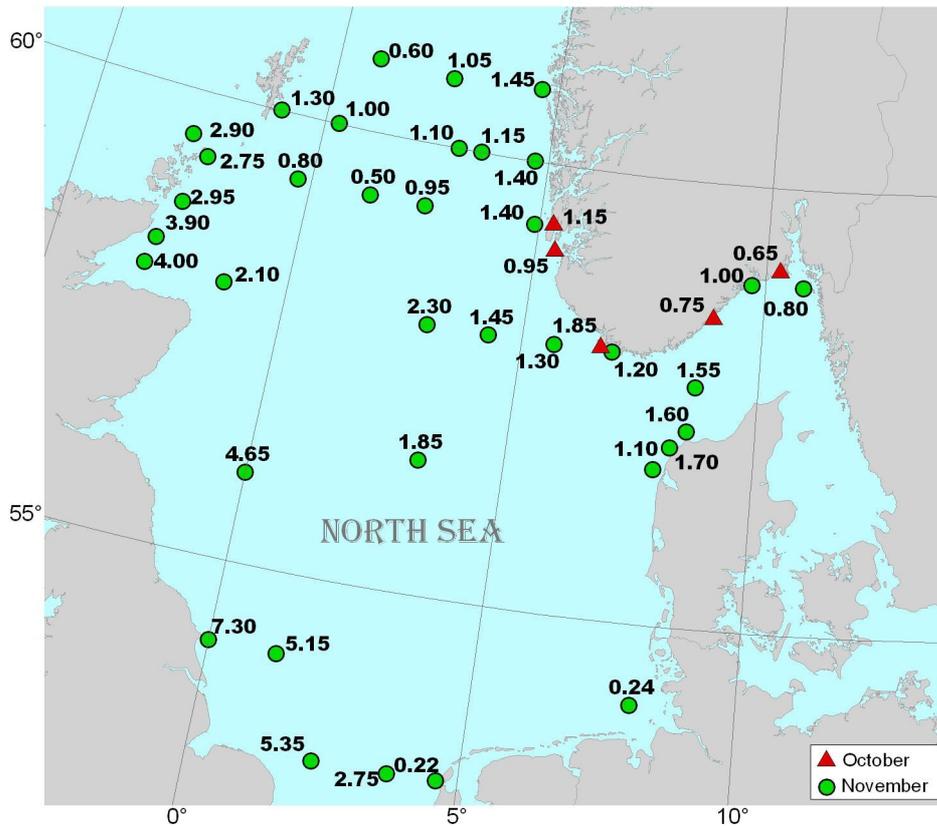


Figure 5. Concentrations of ⁹⁹Tc (Bq m⁻³) in surface sea water samples collected in the North Sea, 2001.

In the years 2000 and 2001, NRPA conducted water sampling in areas off Spitsbergen, in the Fram Strait, and at the islands Hopen, Bjørnøya and Jan Mayen (Gerland et al., 2002). The ⁹⁹Tc concentrations in the northern waters is shown in Figure 6. The highest ⁹⁹Tc level, 0.39 Bq m⁻³, was observed in the centre of the West Spitsbergen Current (WSC). Whereas earlier measurements of samples collected in the Norwegian Sea, Barents Sea and in the Fram Strait in 1994 were below 0.1 Bq m⁻³ (Kershaw et al. 1999). The enhanced ⁹⁹Tc levels in 2000 can be explained by increased discharges from Sellafield (Gerland et al., 2002).

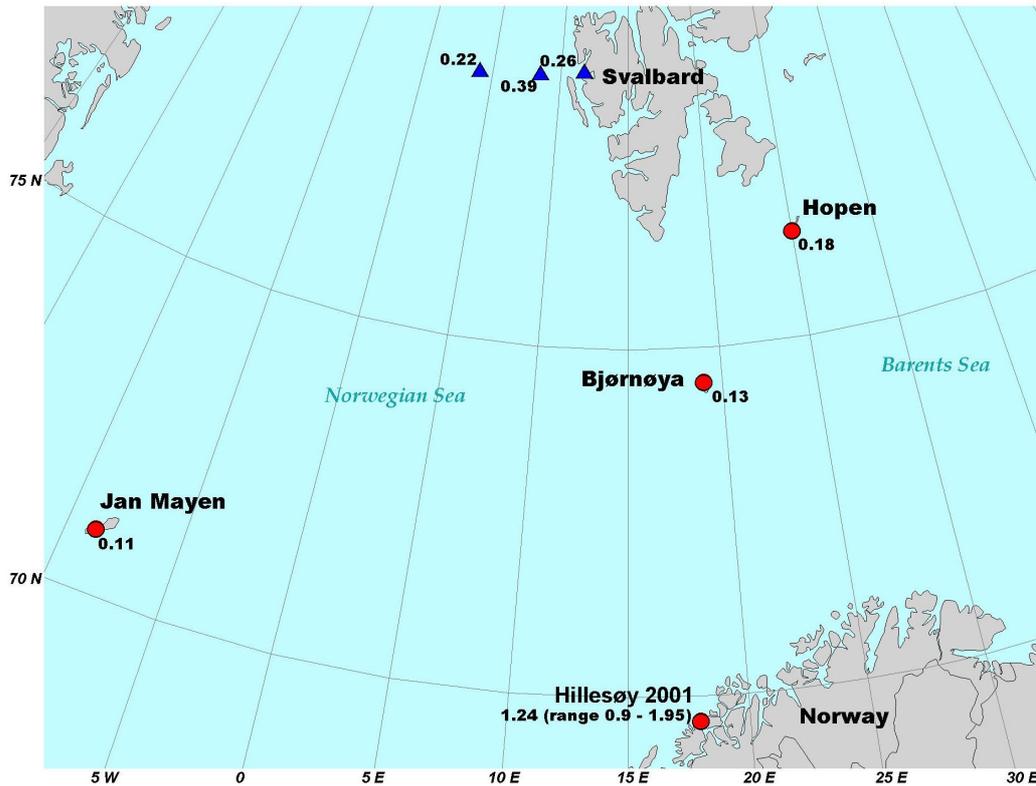


Figure 6. Concentrations of ^{99}Tc (Bq m^{-3}) in surface sea water samples collected in the Northern Seas, May-June 2000 (blue), April 2001 (red) and at Hillesøy sampling station an average value for 2001 (with range).

Monthly ^{99}Tc concentrations in surface sea water from Hillesøy as a function of time are shown in Figure 7. From July 1997 to January 2001, the ^{99}Tc concentration has increased from a level of 0.46 Bq m^{-3} to a maximum of 2.0 Bq m^{-3} . At Hillesøy, the concentrations of ^{99}Tc ranged between 0.87 Bq m^{-3} – 1.74 Bq m^{-3} and 0.9 Bq m^{-3} – 2.0 Bq m^{-3} respectively in 2000 and 2001. The levels are in the same range as in 1999 (range 1.0 Bq m^{-3} – 1.9 Bq m^{-3} , Strålevern Rapport 2001:9). The salinity is relatively high in all samples except for one sample collected in June 1998. The exceptionally low salinity measured at that time may be due to runoff of fresh water in connection with snow melting. A small dip in the salinity is also seen in the spring of 2000. There are indications of seasonal variations in concentrations of ^{99}Tc in sea water, with generally higher concentrations in the winter season. The reasons for this are not clear. It could be due to variations in local currents, resulting in periodic mixing with uncontaminated Atlantic water, or some other coastal effect. Another possibility is that it reflects the variations in actual discharges from Sellafield (see Brown et al., 2002).

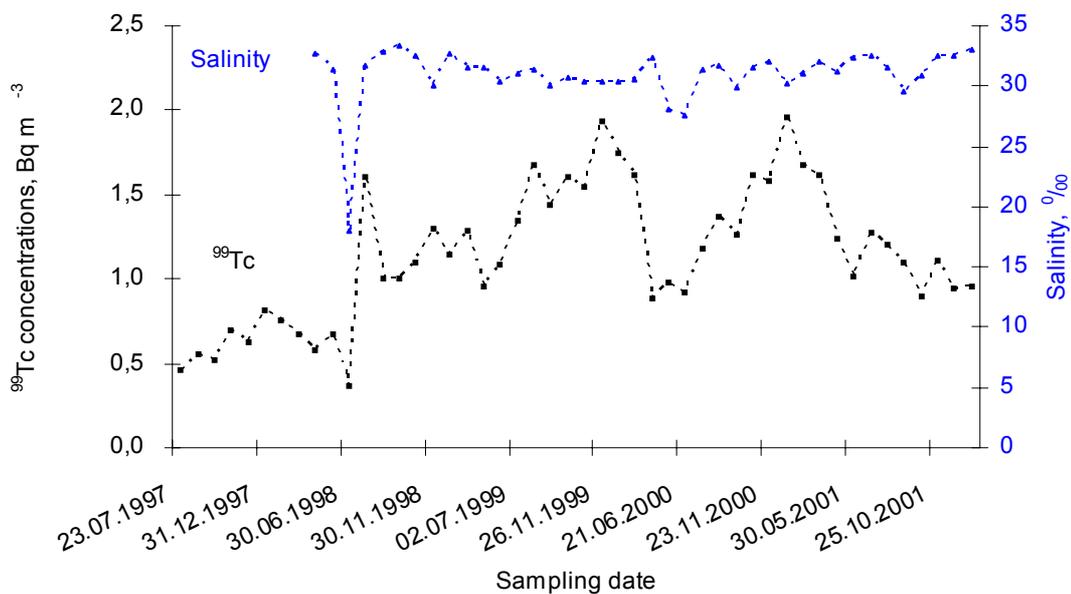


Figure 7. Concentrations of ^{99}Tc (Bq m^{-3}) in surface sea water samples collected at Hillesøy between July 1997 and January 2002.

3.2 ^{99}Tc in seaweed and other biota

3.2.1 Seaweed

The ^{99}Tc concentrations in *Fucus vesiculosus* collected at Hillesøy between July 1997 and January 2002 are shown in Figure 8. A trend of increasing concentrations is observed from 1997 and up to 2001. It seems that the concentrations increase in the winter seasons, and level off or decrease in late summer/autumn. The annual mean concentrations of ^{99}Tc in both sea water and seaweed are listed in Table 1. The standard deviations of the mean values are relatively large, reflecting the seasonal variations in concentrations. The highest average sea water concentration were observed in 1999, with similar values in 2000 and 2001, while the highest seaweed concentrations are observed in 2000 and 2001. In 2001, annual average ^{99}Tc concentration was 321 Bq kg^{-1} (d.w.) with a maximum value of 425 Bq kg^{-1} in January.

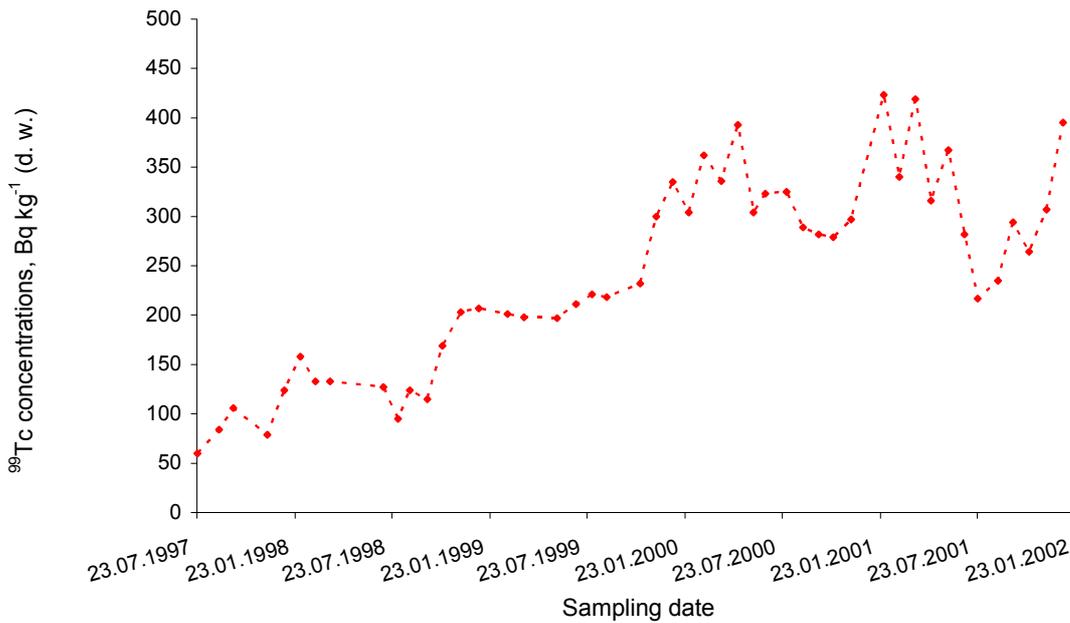


Figure 8. Concentrations of ^{99}Tc ($\text{Bq kg}^{-1} \text{d.w.}$) in *Fucus vesiculosus* collected at Hillesøy.

Concentration ratios were calculated for pairs of samples collected almost simultaneously (within three days). The ^{99}Tc concentration ratio, CR, was defined as the ratio between radionuclide concentrations in *Fucus vesiculosus* and in water, Bq kg^{-1} (dry) Fucus / Bq l^{-1} water. Since the system is not in equilibrium, a large variation is observed and annual average concentration ratios seems to increase with time, ranging from 1.5×10^5 for 1998 to 2.6×10^5 in 2001.

Table 1. Mean annual concentrations of ^{99}Tc in seawater and seaweed (*Fucus vesiculosus*).

The errors represent standard deviations of the mean values.

| Year | Sea water | | Seaweed | |
|-------|-------------------|--|-------------------|---|
| | Number of samples | Mean activity concentration (Bq m^{-3}) | Number of samples | Mean activity concentration ($\text{Bq kg}^{-1} \text{d.w.}$) |
| 1997* | 6 | 0.61 ± 0.13 | 4 | 82 ± 16 |
| 1998 | 11 | 0.93 ± 0.36 | 10 | 138 ± 31 |
| 1999 | 10 | 1.46 ± 0.30 | 10 | 232 ± 47 |
| 2000 | 9 | 1.42 ± 0.34 | 11 | 318 ± 36 |
| 2001 | 12 | 1.25 ± 0.33 | 12 | 321 ± 68 |

* Sampling commenced July 1997

When comparing the time trend of sea water concentrations with seaweed concentrations (Figure 9), there are indications of a delay of several months in the response of the seaweed in the years 1998 to 1999, while in 2000-2001, the two curves follow each other more closely.

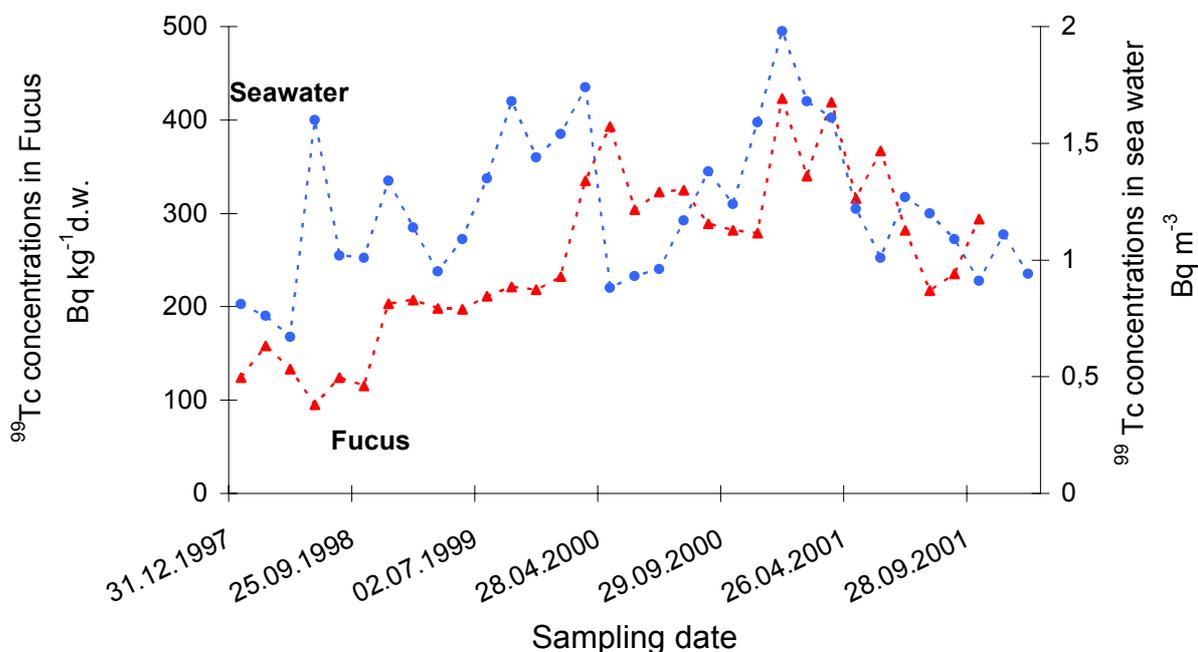


Figure 9. Variations in ⁹⁹Tc concentrations in sea water and *Fucus vesiculosus* collected at Hillesøy.

Concentrations of ⁹⁹Tc in seaweed collected from other coastal stations in 2000 and 2001 are shown in Table 2. The highest ⁹⁹Tc concentration in seaweed was found in *Ascophyllum nodosum* collected at Narestø (Arendal), 660 Bq kg⁻¹ and 435 Bq kg⁻¹ in 2000 and 2001, respectively.

Table 2. Concentrations of ^{99}Tc in seaweed (Bq kg^{-1} dry weight) in 2000 and 2001.

| Sample Id | Species | Sampling date | Location | ^{99}Tc concentration Bq kg^{-1} d.w. |
|-----------|---|---------------|------------|--|
| M-252 | Bladder wrack <i>Fucus vesiculosus</i> | 05.08.00 | Lista | 470 ± 46 |
| M-644 | Serrated wrack <i>Fucus serratus</i> | 04.10.01 | Lista | 150 ± 15 |
| M-641 | Sea girdle <i>Laminaria digitata</i> | 04.10.01 | Lista | $26,9 \pm 2,7$ |
| M-220 | Egg wrack <i>Ascophyllum nodosum</i> | 28.06.00 | Narestø | 660 ± 65 |
| M-646 | Egg wrack <i>Ascophyllum nodosum</i> | 05.10.01 | Narestø | 435 ± 44 |
| M-222 | Egg wrack <i>Ascophyllum nodosum</i> | 29.06.00 | Tjøme | 255 ± 25 |
| M-653 | Egg wrack <i>Ascophyllum nodosum</i> | 05.10.01 | Tjøme | 315 ± 31 |
| M-635 | Egg wrack <i>Ascophyllum nodosum</i> | 03.10.01 | Karmsundet | 340 ± 34 |
| M-180 | Bladder wrack <i>Fucus vesiculosus</i> | 15.05.00 | Rødtangen | 380 ± 37 |

3.2.2 Crustaceans and molluscs

In 2001, 5 female and 18 male European lobsters (*Homarus gammarus* L.) were analysed for ^{99}Tc . The lobsters were collected at Kvitsøy (Rogaland, 21 individuals) and in Stefjord (Tysfjord municipality 69°N, 2 individuals). Lobsters, ranged in size from 320 g to 3660 g fresh weight with mean values of 927 g and 767 g for male and female, respectively. The ^{99}Tc concentrations in lobsters from Kvitsøy and Stefjord are presented in Table 3.

Table 3. ^{99}Tc concentrations in lobsters from Kvitøy and Steffjord.

| Sex | Kvitøy | | Steffjord | |
|--------|-------------------|--|-------------------|--|
| | Number of samples | Mean ^{99}Tc concentration Bq kg ⁻¹ (range) wet weight | Number of samples | Mean ^{99}Tc concentration Bq kg ⁻¹ wet weight |
| Female | 4 | 34.2 ± 4.9 (31.1 – 41.5) | 1 | 20.2 ± 2.0 |
| Male | 17 | 6.6 ± 3.8 (2.2 – 12.7) | 1 | 2.8 ± 0.3 |

In 1997 and 1998, lobsters from the west coast of Norway and from the Outer Oslo Fjord had levels ranging from 35.0-42.0 Bq kg⁻¹ w. w. and 14.4-26.2 Bq kg⁻¹ w. w. respectively (Brown et al., 1998 and Kolstad et al., 2000). In 2001, the highest activity concentration recorded was 41.5 ± 4.1 Bq kg⁻¹ w. w. in a female lobster collected at Kvitøy. The ^{99}Tc concentrations in female lobsters (tail muscle) tended to be higher than those in males. In agreement with the observations of Swift and Nicholson (Swift and Nicholson, 2001).

Different body parts concentrate ^{99}Tc to varying degrees. The distribution of ^{99}Tc in tail muscle, left and right claw and spawn are given in Table 4.

Table 4. The distribution of ^{99}Tc in tail muscle, left and right claw and spawn

| Id/sex | ^{99}Tc concentration, Bq kg ⁻¹ w. w. | | | |
|--------|---|------------|-------------|------------|
| | Tail muscle | Claw, left | Claw, right | Spawn |
| 687 /M | 3.8 ± 0.4 | 5.9 ± 0.6 | 3.0 ± 0.3 | |
| 689 /M | 3.9 ± 0.4 | 6.3 ± 0.6 | 11.5 ± 1.1 | |
| 695 /M | 5.6 ± 0.6 | 10.0 ± 1.0 | 4.5 ± 0.5 | |
| 688 /F | 41.5 ± 4.1 | | | 9.1 ± 0.9 |
| 692 /F | 31.5 ± 3.1 | | | 12.6 ± 1.3 |

M=Male, F=Female

Sampling of sea water simultaneously with 6 lobster samples made it possible to estimate concentration ratios. Mean concentration ratio was calculated for 5 male lobsters to 6.5×10^3 activity per kg of biota wet weight/activity per litre of water. This is in the same order of magnitude as Smith et al. found in the Irish Sea in 1997 and 1998 (6.85×10^3). A higher concentration ratio of 3.5×10^4 activity per kg of biota wet weight/activity per litre of water was found for the female lobster (1 sample).

^{99}Tc concentrations in other seafood is shown in Table 5. Also Norway lobster *Nephrops Norwegicus* collected in the Outer Oslofjord had levels higher than 10 Bq kg⁻¹ w. w. The ^{99}Tc concentrations in other seafood were less than 3.0 Bq kg⁻¹ w. w.

Table 5. The distribution of ^{99}Tc in other seafood

| Sample Id | Species | Sampling locations | Sampling date | ^{99}Tc concentration Bq kg ⁻¹ d. w. | ^{99}Tc concentration Bq kg ⁻¹ w.w. |
|-----------|--|--------------------|---------------|--|---|
| 738/1 | Norway lobster (<i>Nephrops norwegicus</i>) | 56°59'N,12°11'E | 09.12.01 | 56.9 ± 5.6 | 11.6 ± 1.2 |
| 647/2 | Starfishes (<i>Asteroidea</i>) | Arendal | 05.10.01 | 1.0 ± 0.5 | 0.16 ± 0.09 |
| 832/2 | Starfishes (<i>Asteroidea</i>) | 57°59'N,04°26'E | 20.11.01 | 0.16 ± 0.10 | < 0.06 |
| 258/2 | Edible Crabs (<i>Cancer pagurus</i>) | Lista | 08.08.00 | 1.40 ± 0.15 | 0.18 ± 0.02 |
| 267/2 | Winkles | Tjøme | 09.08.00 | 11.9 ± 1.2 | 2.95 ± 0.30 |
| 654/2 | Winkles | Tjøme | 05.10.01 | 3.1 ± 0.4 | 0.95 ± 0.5 |
| 645/2 | Winkles | Lista | 05.10.01 | 2.0 ± 0.3 | 0.95 ± 0.5 |
| 820/2 | Sea urchin (<i>Echinus Eperlanus</i>) | 71°15'N,25°26'E | 18.10.00 | < 0.22 | |
| 837/2 | Sea urchin (<i>Echinus Eperlanus</i>) | 57°59'N,04°26'E | 20.11.01 | < 0.21 | |
| 833/2 | False octopus, (<i>Gonatus</i>) | 54°05'N,01°15'E | 28.11.01 | 0.40 ± 0.17 | <0.08 |
| 739/2 | Crab (<i>Lithodes maja</i>) | 57°59'N,04°26'E | 20.11.01 | 0.28 ± 0.16 | <0.06 |
| 210/2 | Mussels (<i>Mytilus edulis</i>) | Rødtangen | 15.05.00 | 6.0 ± 0,6 | 0.75 ± 0.08 |
| 228/2 | Mussels (<i>Mytilus edulis</i>) | Tjøme | 29.06.00 | 5.6 ± 0,6 | 0.73 ± 0.08 |

4 Conclusions

Discharges from Sellafield still lead to enhanced levels of ^{99}Tc in Norwegian waters. The time series from Hillesøy show seasonal variations in the ^{99}Tc concentrations and an increasing trend for seaweed continues up to the year 2000. The annual mean ^{99}Tc concentration in *Fucus vesiculosus* was 82 Bq kg⁻¹ d.w. in 1997 and 318 Bq kg⁻¹ and 321 Bq kg⁻¹ in 2000 and 2001, respectively, with a maximum ^{99}Tc concentration of 425 Bq kg⁻¹ in January 2001. Both in 2000 and 2001, the highest ^{99}Tc concentration in

seaweed was found in *Ascophyllum nodosum* collected at Narestø (Arendal) 660 Bq kg⁻¹ and 434 Bq kg⁻¹, respectively.

Between July 1997 and January 2001, the ⁹⁹Tc concentrations in sea water samples collected at Hillesøy have increased from a level of 0.46 Bq m⁻³ to a maximum of 2.0 Bq m⁻³. The ranges are, however, similar to results obtained in 1999 (range 1.0 Bq m⁻³-1.9 Bq m⁻³), 2000 (range 0.87 Bq m⁻³-1.74 Bq m⁻³) and 2001 (range 0.9 Bq m⁻³-2.0 Bq m⁻³). In 2000 and 2001, the ⁹⁹Tc concentrations in sea water collected in the Northern part of Norway and along the Norwegian coast were below 2 Bq m⁻³. In the North Sea, the concentrations of ⁹⁹Tc in surface waters were in the range of 0.22-7.30 Bq m⁻³ in November 2001. The highest concentrations are seen in western and southern parts of the North Sea, near the coast of Scotland and England.

In seafood, the highest ⁹⁹Tc concentrations were found in European lobsters, *Homarus gammarus* and Norway lobsters, *Nephrops Norwegicus*. The ⁹⁹Tc concentrations in other seafood were less than 3 Bq kg⁻¹ wet weight. In 2001, 5 females and 18 males European lobsters (*Homarus gammarus*) were analysed for ⁹⁹Tc. The concentrations ranged from 2.2 Bq kg⁻¹-41.5 Bq kg⁻¹ wet weight. In 1997 and 1998, lobsters from the west coast of Norway and from the Outer Oslo Fjord had levels ranging from 35.0-42.0 Bq kg⁻¹ w. w. and 14.4-26.2 Bq kg⁻¹ w. w. respectively. In 2001, ⁹⁹Tc concentrations in female lobsters tended to be higher than those in male lobsters (tail muscle).

In general, levels of ⁹⁹Tc in marine seafood collected along the Norwegian coastline are low. For lobsters, the concentrations are less than 4 % of the action level for a prospective nuclear accident established by the European Union (EU).

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