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Technetium-99 Contamination in the North Sea and in Norwegian Coastal Areas 1996 and 1997

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1. Summary

In 1994, liquid discharges of the radionuclide Technetium-99 from the British nuclear facilities in Sellafield were increased by approximately a factor 50. Technetium-99 is a long-lived, anthropogenic radionuclide formed in significant quantities by the fission processes that occur in nuclear reactors. It is a soft b -emitter and has a half-life of 213 000 years. The long half-life of ^{99}Tc means that the radionuclide will persist in the environment for thousands of generations. It is well known that such releases are transported by ocean currents to the Norwegian coast. Data after 1994 show that there are still uncertainties with regard to the uptake of ^{99}Tc to different types of biota. On the basis of this information, samples of sea water, seaweed, lobsters, mussels and shrimps from the North Sea and Norwegian coastal areas were collected for ^{99}Tc analysis. The data show that the ^{99}Tc plume reached the Norwegian south-west coast before November 1996, indicating a rapid travel time of less than 2.5 years for the arrival of the ^{99}Tc front. Analyses of seaweed samples from the coast of Troms suggests that the ^{99}Tc front reached this region before December 1997, but more data will be needed to verify this.

The highest concentrations of ^{99}Tc were observed in lobsters and seaweed (11.2 - 42 Bq/kg fresh weight)**.** The levels of ^{99}Tc recorded in lobster are about 30 times below the EC intervention level following future nuclear accidents of 1250 Bq/kg .Concentrations in mussels and shrimps were lower (0.54-0.68 Bq/kg fresh weight) In brown algae collected in the outer Oslofjord, concentrations of ^{99}Tc increased by a factor of 5 from1996 to 1997.

Individual radiation doses from human consumption of seafood from Norwegian waters are probably low due to the present low levels of contamination and the low dose conversion factor of ^{99}Tc. However, many uncertainties exists in relation to the uptake of ^{99}Tc in biota under equilibrium conditions and to the behaviour of ^{99}Tc in the Norwegian marine environment. Also, sufficient information regarding possible pathways of ^{99}Tc to man is lacking, in addition to the limited data available relating to activity levels in the environment. These questions need to be answered before a full dose assessement for man and biota can be implemented.

2. Sammendrag

I 1994 økte utslippene av technetium-99 fra Sellafield anlegget med en faktor 50. Technetium er et langlivet menneskeskapt radioaktivt stoff som dannes ved fisjonsprosessen i kjerne- reaktorer. Technetium er en b emitter og har en halveringstid på 213 000 år. Den lange halveringstiden betyr at stoffet vil forbli i miljøet i all overskuelig fremtid. Det er kjent at utslipp fra Sellafield blir transportert med havstrømmer til norskekysten. Nyere undersøkelser viser dessuten at det fortsatt er usikkerheter når det gjelder opptak av technetium-99 til ulike typer biota.

Basert på den ovenfornevnte informasjon, ble det igangsatt ekstraordinære analyser av technetium i innsamlede prøver av sjøvann, tang, hummer, skjell og reker. Resultatene viser at technetium-forurensningen nådde kysten av sør-vestlandet før november 1996. Dette tyder på en relativt rask transporttid på under 2½ år. Målinger av tang fra kysten av Tromsø tyder på at technetium-99 forurensningen nådde hit før desember 1997. Det er nødvendig med ytterligere målinger for å bekrefte dette.

De høyeste aktivitetsnivåene av technetium-99 ble observert i hummer og tang (11-42 Bq/kg fersk vekt. Nivåene av technetium-99 målt i hummer er ca 30 ganger under

EU's tiltaksgrense på 1250 Bq/kg som skal gjelde ved en fremtidig atomulykke. Aktivitetsnivåene i blåskjell og reker var lavere enn de for hummer (0,54-0,68 Bq/kg). Målinger av brunalger (*fucus serratus*) samlet i ytre Oslofjord, viste en femdobling i aktivitetsnivåene av technetium-99 fra 1996 til 1997. Individuelle stråledoser som skyldes konsum av sjømat fra norske havområder er lave, bl.a. på grunn av dagens lave technetium-99 nivåer i marin biota og den lave dosekonverteringsfaktor for technetium-99. Det eksisterer likevel en viss usikkerhet om stråledoser fra technetium-99 på grunn av manglende data og kunnskap om overførings-faktorer for technetium i ulike biota ved likevekt, og om hvordan technetium-99 vil oppføre seg i det norske marine miljø. Det er også manglende informasjon om den norske befolknings kostholdsvaner når det gjelder sjømat. Disse spørsmålene bør besvares før man kan etablere et fullstendig estimat av stråledoser til mennesker og miljø fra den nåværende og fremtidige technetium-forurensningen.

3. Introduction

In 1994, liquid discharges of the radionuclide ^{99}Tc from the British Nuclear Fuels (BNFL) reprocessing plant at Sellafield were increased by approximately a factor 50. It is well known that such releases are transported by ocean currents to the Norwegian coast. Preliminary studies have been undertaken at the Norwegian Radiation Protection Authority (NRPA) in order to investigate the impact of this radionuclide. This has been achieved by including water sampling for ^{99}Tc measurements in two cruises, organised regularly by the Marine Research Institute, in the North Sea and Norwegian coastal waters in 1996 and 1997 and the collection of seafood and other biota samples from Norwegian coastal waters over the last year. The first part of the report (Section 3) deals with relevant information relating to the sources and environmental behaviour of ^{99}Tc in the context of why there is renewed interest in this radionuclide and why ^{99}Tc may cause concern for Norway. In Section 4 of the report the sampling and analytical methods employed in this work are presented. The results and discussion are dealt with in Section 6 and conclusions are drawn in Section 7.

3.1 Background

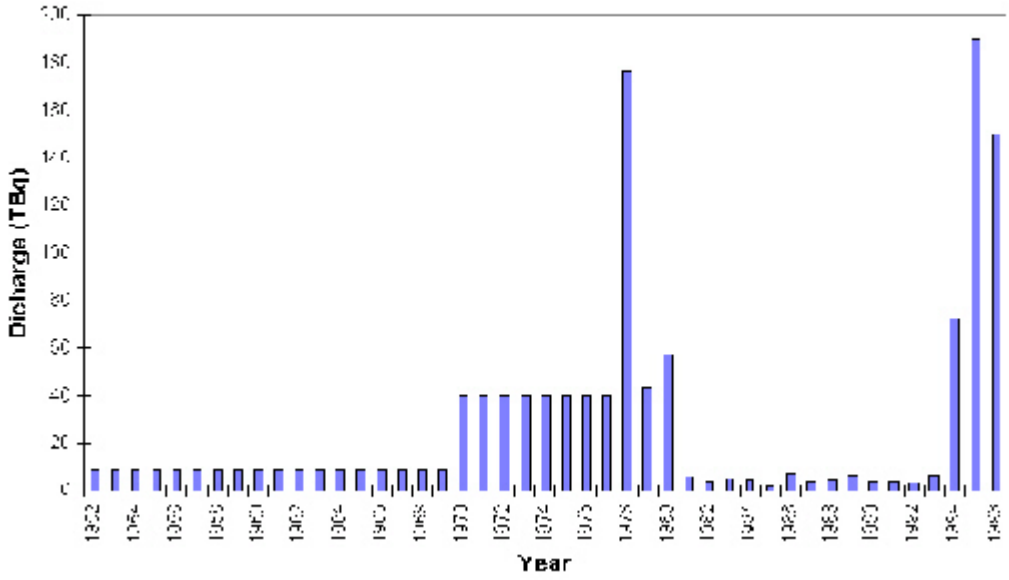
3.1.1 Technetium-99

Technetium-99 is a long-lived, anthropogenic radionuclide formed in significant quantities by the fission processes that occur in nuclear reactors. It is a soft b -emitter (Eb_{max}=293 keV) and has a half-life of 213 000 years. The long half-life of ^{99}Tc means that the radionuclide will persist in the environment for thousands of generations to come. In addition, the tendency of technetium to remain in the aqueous phase under the normal chemical conditions found in the open oceans, means that it is likely to remain available for uptake by biota and transfer to man over these long time periods. Until recently little was known with regard to certain aspects of the environmental behaviour (e.g. uptake by certain biota) of ^{99}Tc.

3.1.2 Sources

To put the relative sources of ^{99}Tc into an international context, around 85 % of the global inventory is associated with European coastal discharges, the remaining 15 % is associated with weapons tests (Dahlgaard, 1995). The two main sources of ^{99}Tc to

Norwegian coastal waters are the discharges from La Hague in France and Sellafield in the UK (Dahlgaard *et al*., 1997). During the 1980s and early 1990s, the discharges from La Hague dominated in terms of quantities discharged, i.e. Sellafield 1982-1991 inclusive = 42.4 TBq (BNFL, 1982-1991) compared to La Hague (in the decade since 1982) =102 TBq (Herrmann *et al*., 1995). Prior to this, Sellafield discharges dominated. Throughout the 1980s and early 1990s, ^{99}Tc was discharged in low levels from Sellafield at a rate of 4-6 TBq a^{-1}. The ^{99}Tc, that was present in medium active concentrate streams, was held on site during this period awaiting the commissioning of the Enhanced Actinide Removal Plant (EARP). This plant was designed to treat the backlog of stored concentrates together with the continuous production of these and other reprocessing effluents (Gray *et al*., 1995). Alpha activity is removed and beta activity reduced in these effluents before discharge to the Irish Sea. However, ^{99}Tc in the acidic effluent streams will be present as the highly soluble pertechnetate ion (TcO_4^-), and is not effectively removed by EARP (Busby *et al*., 1997). This has resulted in the fact that since operations began at EARP in 1994, discharges of ^{99}Tc have increased by about 50 times compared to levels in the early 1990s. Current discharges are below the discharge limit of 200 TBq a^{-1} permitted by the British authorities. The full discharge chronology from BNFL Sellafield (BNFL, 1979-1996) is illustrated in Figure



3.1.

Figure 3.1 : *Discharge chronology of ^{99}Tc from BNFL Sellafield. (Tc was not reported as a single radionuclide before 1978. Estimates of the Tc-99 discharges prior to this time are taken from Leonard et al. (1997b)).*

3.1.3 Behaviour of the initial releases of ^{99}Tc Sellafield (Irish Sea and UK coastal waters)

A comprehensive survey of ^{99}Tc levels in British coastal waters was undertaken by the Ministry of Agriculture, Fisheries and Food (MAFF), UK, before and after the operation of EARP (Leonard *et al*., 1997a). Pre-EARP levels (12/92) of ^{99}Tc in seawater were essentially uniform (1-4 Bq m^{-3}) in the central and western parts of the Irish Sea. Data from the post-EARP survey (6/94) showed a marked change in ^{99}Tc seawater concentrations (>30 Bq m^{-3} for large areas of the northern Irish Sea and

>200 Bq m^{-3} in the Solway Firth, western Scotland). The levels increased over a relatively large area and indicated a marked migration towards the southern Scottish coastline and North Channel of the Irish Sea. Results indicated that ^{99}Tc had been transported to this area in a relatively short period (» 3 months). Beyond the North Channel, the signature was temporarily lost owing to the inflow of Atlantic seawater. However, the ^{99}Tc signal re-emerged along the northern Scottish coastline, concentrations decreasing slowly with distance. It was concluded that ^{99}Tc had migrated to the northern North Sea within 9 months following the initial elevated discharge. The short transit times for ^{99}Tc to the North Sea suggest that transit times to the Arctic may need to be re-examined (Leonard *et al.*, 1997b).

3.1.4 Water movements in seas adjacent to Norwegian coastal waters

A review of transit times and transfer factors is discussed by Dahlgaard (1995). A transit time of 3-4 years is cited for the movement of a soluble (conservative) radionuclide from Sellafield to the north west Norwegian Current. Higher transfer of soluble radionuclides from La Hague, compared to Sellafield, were explained by the fact that there is a coastal current which leads directly from the English Channel, along the European coast, to the Kattegat. Sellafield discharges, on the other hand, are first transported to the North Sea *via* a coastal current around Scotland after which a fraction of the radionuclides crosses the North Sea before it enters the Kattegat (Figure 3.2).

Dahlgaard *et al*., 1997 calculated "Transfer Factors" (quotient of the observed concentration in seawater at a remote site to an average discharge rate from a source, ‘t’ years earlier) for ^{99}Tc from European reprocessing plants. Transfer factors of 55 Bq m^{-3} per PBq a^{-1} and 24 Bq m^{-3} per PBq a^{-1} were calculated from La Hague and Sellafield respectively. For two northern Norwegian sampling stations, a dilution of 0.45 was observed, relative to Utsira in western Norway. Transfer Factors for Sellafield were considered to be lower than those from La Hague owing to the dilution of ^{99}Tc in North Atlantic water.

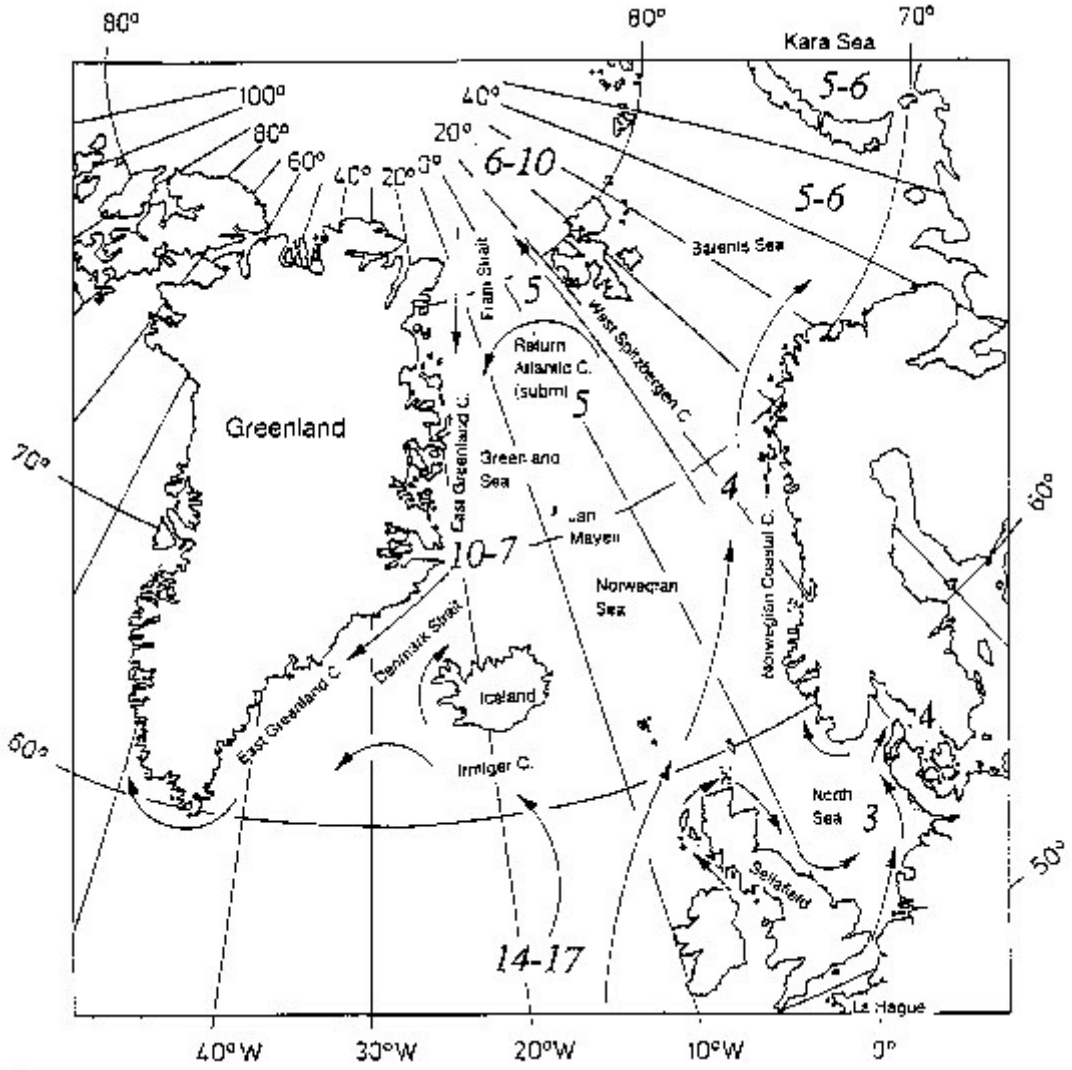


Fig. 3.2 : *Major surface currents and transit time in years from Sellafield to different sea areas from Dahlgaard (1995).*

3.1.5 Concentration factors

Concentration factors (CFs) are defined as shown in Equation 1.

$$CF = \frac{\text{Radionuclide concentration in biota (wet weight - Bq kg}^{-1})}{\text{Radionuclide concentration in water (Bq l}^{-1})}$$

Technetium-99 concentration factors are generally low for biota. However certain species exhibit high uptake of ^{99}Tc. The most important species, selected on the basis of their importance in the human diet and/or their high uptake of ^{99}Tc are listed in Table 3.1.

Table 3.1 : *Examples of Concentration Factors (activity per unit mass of biota/activity per unit mass of water) for selected biota.*

Organism	Description/species	CF	Notes	Reference
Seaweed	*Fucus vesiculosus*	1.19± 0.41x10^5	Irish Sea, Irish coastal	Smith *et al*. (1997)
Mussels	*Mytilus edulis*	500	Irish Sea, 1995	McCartney & Rajendran (1997)
Lobster	*Homarus gammarus* -abdomen muscle [2]	720	Irish Sea, 1995	Busby *et al*. (1997)
Lobster	*Homarus gammarus* -green gland [2]	6.5 x 10^5	Irish Sea, 1995	Busby *et al*. (1997)
Norwegian lobster	*Nephrops norvegicus* - abdomen muscle [2]	970	Irish Sea, 1995	Busby *et al*. (1997)
Norwegian lobster	*Nephrops norvegicus* -Hepatopancreas [2]	2300	Irish Sea, 1995	Busby *et al*. (1997)

2 : CF to body part

Certain types of brown seaweeds (e.g. *Fucus vesiculosus*) concentrate ^{99}Tc to a high degree. In the Irish sea CFs in excess of 1 x 10^5 have been measured which means that if the seawater concentration is 1 mBq l^{-1} (1 Bq m^{-3}) of ^{99}Tc in water you will observe a seaweed concentration of 100 Bq kg^{-1} (wet weight) at equilibrium. Uptake of ^{99}Tc by the common lobster and the Norwegian lobster is also significant. Some complication is introduced, however by the fact that different body parts concentrate ^{99}Tc to varying extents. For lobsters and Norwegian lobsters, although relatively high uptake (compared to other seafood species) is observed in edible meat (claw and tail muscle), the highest CFs are observed for the green gland and Hepatopancreas respectively (Busby *et al*., 1997), which are not normally consumed by people. Laboratory studies to consider the uptake of ^{99}Tc have met with mixed success. High levels of uptake by lobster (CFs > 1000) were recorded under laboratory conditions (in the early 1980s) by Swift (1985). These values broadly agree with those CFs recently measured in the Irish Sea. The uptake of ^{99}Tc by molluscs (e.g. mussels (*Mytilus edulis*)), on the other hand, has not been well represented under laboratory conditions. Concentration factors for mussels appear to be in the region of 500 for the Irish Sea (McCartney & Rajendran, 1997) whereas laboratory experiments have generally demonstrated uptake of < 20 (Fowler *et al*., 1981; Beasley *et al*., 1982). For other biota, such as *Nephrops norvegicus*, little was known in relation to the uptake of ^{99}Tc before the research of the last few years.

Data from laboratory studies (Pentreath, 1981a; Pentreath, 1981b) and from environmental monitoring (MAFF, 1996) suggest that the uptake of ^{99}Tc by marine fish is generally low. Levels of ^{99}Tc have, therefore, not been considered in the present study but will probably be determined in the future as part of a more comprehensive dose and impact assessment.

3.1.6 Doses

In 1995, the committed effective dose to the locally most exposed group of seafood consumers in UK coastal areas, for all "artificial" radionuclides was 0.12 mSv (MAFF, 1996). The increase from the previous year (0.08 mSv) was largely due to (i) the increase in consumption of molluscs by the most exposed group and (ii) the increased concentration of ^{99}Tc. The exposure from ^{99}Tc alone was 0.018 mSv for 1995. This compares to a 1 mSv limit (ICRP, 1991) which applies to the overall exposure of the general public from man-made radiation (including the effects of past and current discharges and summing across all relevant exposure pathways). Clearly, the increased discharges of ^{99}Tc have led to a slight increase in dose to members of the UK public. Although more remote from the source, it follows that consumers of seafood in Norway may also be exposed to a slight increase in doses. Some insight into the expected doses is provided by studies in the Irish Sea. Smith *et al.* (1997) assessed the radiological significance of the increased rate of ^{99}Tc discharge from the perspective of an Irish seafood consumer using literature derived CFs for marine fish (CF = 10) and crustaceans (CF = 100). The dose commitment for consumers of seafood from the western Irish Sea in 1995, based on ingestion statistics and a dose coefficient of 6.7 x 10^{-4} micro Sv Bq^{-1} (Phipps *et al*., 1991) was 0.006 microSv for a typical consumer and 0.025 microSv for a heavy consumer. This was considered to be of no radiological significance. It must be remembered, however, that significantly higher concentration factors than those used in these dose assessment calculations have been observed in some areas (e.g. CFs in excess of 1000 have recently been measured for some crustaceans in the Irish Sea).

3.1.7 Summary

To summarise, the key aspects leading to the renewed interest in ^{99}Tc from the point of view of Norway are :

- The fact that BNFL Sellafield has increased discharges of the radionuclide by about 50 times following the start of operation at EARP in 1994 (Current discharges from Sellafield, however, fall within the limit of 200 TBq permitted by British authorities).
- Previous research has shown that soluble radionuclides can be transported in significant quantities to Norwegian coastal waters in fairly short time periods.
- Certain biota concentrate ^{99}Tc to high levels in the environment. Some of these biota are consumed by members of the population.
- The long half-life of ^{99}Tc mean that it will persist in the environment for many thousands of generations.
- There are still uncertainties with regards to the environmental behaviour of ^{99}Tc.
- There are uncertainties relating to the effect Norwegian dietary habits will have on doses from ^{99}Tc.

4. Material and Methods

4.1 Sample collection

4.1.1 Seawater samples

During the expeditions arranged by the Institute of Marine Research , Norway in 1996 and 1997 in the North Sea and Skagerak, samples of water were collected (G.O. Sars 1996 and 1997). In total, 36 water samples were collected in 1996 and 45 collected in 1997 for ^{99}Tc analyses, covering the main part of the North Sea and Skagerak between 53°00'N and 61°00'N. The reported data also include water samples collected in the English Channel in 1996.
For ^{99}Tc determination , 50 litres of unfiltered surface water, from each station, were collected in 25-litre plastic containers and transported to the laboratory on shore.

4.1.2 Biota samples

Seaweed samples and different types of shellfish and crustaceans were collected at selected sites along the Norwegian coastline between 1996 and 1998. The shellfish and crustacean samples were collected by fishermen using traps and nets. The biota samples were transported in a fresh refrigerated or frozen condition to the laboratory. Seaweed, mussels, shrimp and lobsters from the outer part of Oslofjord, lobsters from the coast outside Sunnhordland and seaweed from Sommarøy in Troms have been analysed.

4.2 Methods of analysis

The 50-litre water samples from each site were filtered through a 1m m polypropylene cartridge to remove suspended matter and then stored in 70-litre tanks. Tc-99m was added to the samples for chemical recovery determination.
Fresh biota samples were dried, milled and homogenised. A 10-20 gram dried sample was transferred to a specially designed bottle and carbonised and dissolved by adding concentrated H_2SO_4 and then HNO_3. Tc-99m was added to the samples for chemical recovery determination.
Technetium is separated from the matrix by ion exchange chromatography and extraction with 5 % TIOA/Xylene solution. After back-extraction from the organic phase into 2M NaOH, Technetium is electro-deposited on stainless steel discs.
The chemical yields were determined by gamma counting from the ^{99m}Tc-tracer on a NaI well detector. The yield varied between 46 % and 85 %. The counting samples of ^{99}Tc were measured using a low background anti-coincidence beta counter.
Five aliquots of a seaweed sample were analysed to verify the precision of the radiochemical procedure. The results from individual replicates agreed closely indicating a high precision.
The uncertainty arising from calibrations and the chemical procedure is assumed to be approximately 10 %. The statistical errors from the counting of the samples are less than 5 %.
The detection limits are estimated to be in the range 0.2-0.5 Bq kg^{-1} for the biota samples and 0.03-0.06 Bq m^{-3} for the sea water samples.

5. Results and Discussion - Levels of ^{99}Tc in seawater and biota

5.1 ^{99}Tc in the North Sea and Norwegian coastal waters

Seawater samples have been collected in the North Sea and Skagerrak during two cruises of the research vessel G.O. Sars in November 1996 and 1997. The survey was also carried out in the Kattegat in 1997. The results from these expeditions are shown in Figures 5.1 & 5.2.

In the early 1990s (1990-1992), before operations began at EARP, levels of ^{99}Tc in seawater from the North Sea and Norwegian coastal areas were generally less than 1 Bq m^{-3} (Hermann *et al*., 1995). During the same period, levels of ^{99}Tc in northern French, Belgian, Dutch and German coastal waters (between Cherbourg and Denmark) were higher, in comparison to this, clearly showing the influence of the plume from the La Hague reprocessing plant in France. Levels, in these areas, were in the range of 1-4 Bq m^{-3}.

1996 Expedition : The ^{99}Tc data from the G.O. Sars 1996 expedition show a general increase in water activity levels, compared to the early 1990s, over large areas of the North Sea. The main points of interest are:

- Levels of ^{99}Tc in Norwegian coastal waters are generally in the range of 0.9-6.5 Bq m^{-3}.
- The highest level of ^{99}Tc recorded in the expedition (for the samples analysed to date) was 8.5 Bq m^{-3}, recorded to the north east of the Shetland Islands.
- The plume of ^{99}Tc from Sellafield has clearly reached the Norwegian coast. Unfortunately, the arrival date for the first inputs cannot be determined from our studies. However, results do indicate a rapid travel time (for the arrival of the first pulse of contamination) of under 2.5 years.
- Levels of ^{99}Tc in Norwegian coastal waters have increased (generally speaking) by almost an order of magnitude between 1991 and the end of 1996. Levels recorded in July 1991 (Hermann *et al*., 1995) were around 0.4 Bq m^{-3}. Levels in November 1996 had increased to a maximum of 6.5 Bq m^{-3}.

Figure 5.1 : *Levels of ^{99}Tc in seawater for the North Sea and Norwegian coastal waters in November 1996. Total uncertainties ranging from 10-15%.*

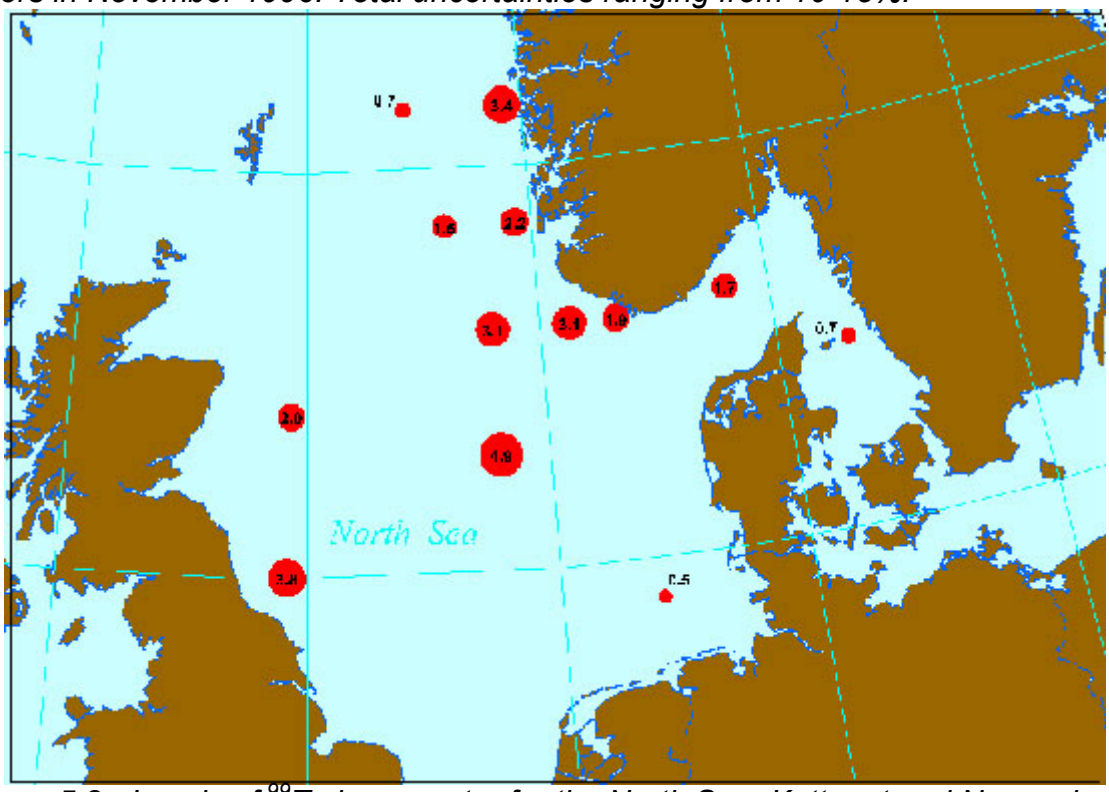


Figure 5.2 : *Levels of ^{99}Tc in seawater for the North Sea, Kattegat and Norwegian coastal waters in November 1997. Total uncertainties ranging from 10-15%.*

The results from this study generally agree with the conclusions from previous work (Dahlgaard, 1995; Dahlgaard *et al*., 1997) in relation to transfer factors and the route that contamination takes from its source to Norwegian coastal waters (i.e. a direct

route from Northern UK to Norway appears to be absent apart from an anomalous value close to the Shetland Islands. Contamination travels across the central and southern sections of the North Sea, before travelling northwards to the Norwegian coast). Transit times (for the occurrence of peak activities from a discharge event) may be shorter than anticipated, but further work is required to validate this.
The levels of ^{99}Tc in Norwegian coastal waters are low compared to levels in the Irish Sea. For example levels of ^{99}Tc in excess of 300 Bq m^{-3} were recorded in the NE Irish Sea by Leonard *et al.* (1997a) in 1994. Levels in excess of 1 000 Bq m^{-3} were recorded by Busby *et al.* (1997) in 1995 for sites close to Sellafield. Discharges become significantly diluted (i.e. concentrations decrease by approximately 100 times) between seas off Sellafield, and Norwegian coastal areas.
The NRPA also collected 5 seawater samples in the English Channel in December 1996. All the stations showed lower levels than those recorded in the North Sea. The concentrations of ^{99}Tc varied between 0.1 Bq m^{-3} and 0.3 Bq m^{-3} in the English Channel.

1997 Expedition : The data set for samples from 1997 is only partly complete but provides us with some interesting features. Levels of ^{99}Tc in Norwegian coastal waters are generally in the range of 1.7-3.4 Bq m^{-3}. Although peak activities are slightly lower than those measured in 1996, the average level of ^{99}Tc in seawater from Norwegian coastal waters is approximately the same. It may be that the "cells" of ^{99}Tc, evident in the 1996 survey, have now dispersed and become mixed with less-contaminated water.
The surveys of 1996 and 1997 show the dynamic nature of the spatial distribution of ^{99}Tc owing to the pulsed nature of the discharges from Sellafield and the variability in the prevailing hydrodynamic conditions (the wind-wave climate and the influence of different water masses in terms of their input into an area will change over time). Biota samples on the other hand will reflect the time-integrated, effectively "smoothed", effect of the ^{99}Tc inputs to the area.

5.2 ^{99}Tc in biota samples from Norwegian coastal waters

Samples of shrimp, lobster, mussel and seaweed have been analysed for their ^{99}Tc content (Table 5.1). The samples were collected from coastal areas of the Norwegian Sea, Outer Oslo Fjord and the North Sea.
In late 1997, early 1998, specific activity levels in seaweed were generally in the range of 79-170 Bq kg^{-1} (dry weight). The ^{99}Tc activity concentration in seaweed samples from Grundtvik in outer Oslofjord, appears to have increased significantly from 1996 to 1997, i.e. from 36 Bq kg^{-1}(dry weight) to 170 Bq kg^{-1} (dry weight).
In 1997, the lobsters from the west coast of Norway had higher levels of ^{99}Tc compared to samples from Outer Oslo Fjord. Specific activity levels were in the range of 56 - 270 Bq kg^{-1} (dry weight) in late 1997.
The content of ^{99}Tc in mussel samples varied from 2.4 Bq kg^{-1} (dry weight) to 7.6 Bq kg^{-1} (dry weight) for the two study sites, sampled in 1997.

Table 5.1 : *^{99}Tc levels in marine material in 1996, 1997 (and 1998 - 1 sample) The errors given represent one standard deviation of combined systematic and statistical errors.*

Sample	**Sample date**	**Sample place**	**^{99}Tc-concentration Bq kg^{-1} dry weight**	**Bq kg^{-1} wet weight**
Seaweed (*Fucus serratus)* No.96030/1-1 to 5	November 1996	Onsøy, Fredrikstad	36 ± 4	
Seaweed (*Fucus serratus)* No.97085/2-1 to 3	November 1997	Onsøy, Fredrikstad	170 ± 20	
Seaweed (*Fucus vesiculosus)* No.98086/1	December 1997	Hillesøy, Troms	79 ± 8	
Seaweed (*Fucus vesiculosus)* No.98086/2	January 1998	Hillesøy, Troms	124 ± 12	
Shrimps No.97085/1-1 and 2	November 1997	Outer Oslofjord	2.2 ± 0.2	0.54 ± 0.05
Lobster (*Homarus gammarus)* No.97085/5-1 and 2	November 1997	Outer Oslofjord	56 ± 6	11.2 ± 1.1
Lobster (*Homarus gammarus)* No.98087/1	December 1997	Sunnhordaland	180 ± 20	35 ± 4
Lobster (*Homarus gammarus)* No.98087/2	December 1997	Sunnhordaland	270 ± 30	42 ± 4
Mussels *(Mytilus edulis)* No.98086/3	July 1997	Hillesøy Troms	2.4 ± 0.2	0.53 ± 0.05
Mussels *(Mytilus edulis)* No.97085/3-1	November 1997	Onsøy Fredrikstad	7.6 ± 0.8	0.68 ± 0.07

The highest levels of ^{99}Tc were recorded in lobster and seaweed samples. Recent studies in the Irish Sea (see Section 3.1.5) have also shown high levels of uptake by these biota. Shrimp and mussel have concentrated ^{99}Tc to a much lower degree.

Although not strictly applicable to a non-accident situation, the observed levels of ^{99}Tc in sea food may be compared to the EC intervention level of 1250 Bq kg^{-1} (wet weight). The levels of ^{99}Tc recorded in our preliminary survey are 30 times below this level, i.e. a maximum of 42 Bq kg^{-1} (wet weight) for a lobster from Sunnhordland in December 1997.
The seaweed *Fucus vesiculosus*, has historically been used as an indicator species for ^{99}Tc. For this reason a large data set exists, containing data on ^{99}Tc levels in this algae, for several locations, spanning the last decade or so. Levels of ^{99}Tc at Ingøy (NW Norway) were relatively high (in the range of 50-70 Bq kg^{-1}, dry weight) during the early and mid 1980s (Dahlgaard *et al*., 1997) reflecting the elevated discharges from Sellafield and La Hague in the preceding years. In the late 1980s and early 1990s, a significant decrease in levels was observed corresponding to the low throughput of this radionuclide at Sellafield (before operations at EARP began) and the declining discharges from La Hague. Levels in *Fucus vesiculosus* were generally below 30 Bq kg^{-1} (dry weight) after 1990. As a comparison to this, samples at Hillesøy (Troms, NW Norway), taken for this survey, yield ^{99}Tc levels of 79 Bq kg^{-1} (December 1997) and 124 Bq kg^{-1} (January 1998) - higher than any of the levels observed previously. The increase in activity between December 1997 and January 1998 could be significant but may, alternatively, be caused by the random variation of levels, that is often observed, in individuals of the same species. However, a clear result from this work is that the effect of the increased discharges of ^{99}Tc from Sellafield can now be seen at locations as far north as Troms. It is highly likely that the initial plume of ^{99}Tc has now reached the Arctic seas.

5.3 Predictions for the levels of ^{99}Tc in seawater and biota

Ten-fold increases in ^{99}Tc in *Fucus vesiculosus* at two sites in Ireland were observed over a period of two years (Smith *et al*., 1997). This is shown in Figure 5.3. A similar, increasing trend is expected in Norway.
Further work is required to assess the exposure and consequences of the increased levels of ^{99}Tc on biota. Few studies have dealt with this in the past.

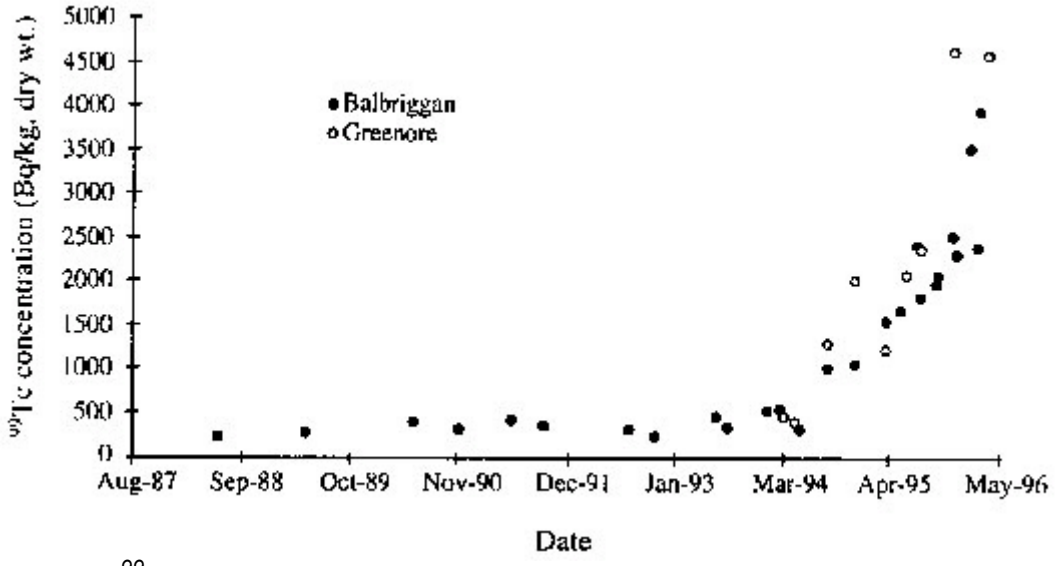


***Figure 5.3** : ^{99}Tc activity concentrations in Fucus vesiculosus sampled at Balbriggan and Greenore (Ireland) in the period 1988-1996. Taken from Smith et al., (1997).*
Owing to complex hydrographic transport patterns, the emissions from discrete discharge events will be dispersed to varying degrees when arriving at sampling

points, in the coastal currents, downstream from the discharge. This means that distinct peaks in the discharge will be smoothed over a certain time period. The larger distance from Sellafield to Norwegian coastal areas, as compared to sites in the Irish Sea, allow more dispersion to have taken place. This means that (a) the peak activities will tend to be significantly lower, (b) the maximum, for a given discharge event, will be broader and (c) the time-lag between the first increases and the maximum levels will be longer, than for locations close to the source. This suggests that we, still, may not have observed the maximum levels attributable to the initial, elevated related releases from EARP in 1994.
An estimated 1 260 TBq of ^{99}Tc will be removed from the back-log of stored waste and future reprocessing operations during the remainder of the Magnox programme, at Sellafield. The proportion of this activity that is discharged will depend on the development of abatement technologies, however, these techniques are not available for the present and it seems likely that the discharges will remain at a similar level for the next few years at least. It should be noted that the Environmental Agency (UK) is considering a new limit (possibly to a reduced level of 90 TBq a^{-1} from the current level of 200 TBq a^{-1}) for ^{99}Tc discharges.
Further monitoring will be necessary to assess whether the levels in seafood are increasing as the various environmental compartments attain equilibrium with respect to the new levels of ^{99}Tc in seawater.

5.4 Implication of ^{99}Tc levels in terms of doses

The study undertaken for this report is preliminary in nature and does not include a full dose and impact assessment owing to the limited size of the monitoring database and the lack of information about dietary habits. Superficially, the low committed effective dose factor for ^{99}Tc (Phipps *et al*., 1991), compared to other radionuclides such as ^{137}Cs, suggests that doses to the population in Norway are low. However, many questions remain with respect to the equilibrium conditions that will be attained in future years, the behaviour of ^{99}Tc in Norwegian coastal waters and the pathways leading to internal doses to man. These questions will need to be answered before a full dose assessment can be implemented.

6. Conclusions

- The levels of ^{99}Tc observed in seawater samples collected in the North Sea were in the range 0.9-8.5 Bq m^{-3} in November 1996 and in the range 1.7-3.4 Bq m^{-3} in 1997. This clearly shows that the plume(s) of ^{99}Tc from Sellafield reached the Norwegian south-west coast before November 1996. Data from the analysis of seaweed from the coast of Troms suggest that the ^{99}Tc-plume reached the Tromsø region some time before December 1997.

- In brown algae samples, collected in the outer Oslofjord, an increase in the ^{99}Tc concentration, from 1996 to 1997, by a factor of 5 was observed (36 Bq kg^{-1} and 170 Bq kg^{-1} (dry weight) respectively).

- The highest levels of ^{99}Tc were recorded in lobster and seaweed samples (maximum levels of 270 Bq kg^{-1} and 170 Bq kg^{-1} (dry weight) respectively) . Lower concentrations were found in shrimp and mussels (in the range 2.2 Bq kg^{-1} to 7.6 Bq kg^{-1}, dry weight).

- Further monitoring will be necessary to assess whether the ^{99}Tc levels in seaweed, lobsters and other seafood will increase further.

- Low levels of contamination and a low committed effective dose factor for ^{99}Tc, suggest that individual doses from ^{99}Tc in seafood are low. However, many uncertainties with respect to the uptake and concentration factors in equilibrium conditions, the behaviour of ^{99}Tc in Norwegian coastal waters and the pathways leading to internal doses to man need further investigation before a full dose assessment can be implemented.

- Work is required in order to assess the impact of ^{99}Tc on biota (doses and consequences). Few studies have dealt with this to date.

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