

Monitoring of ^{99}Tc in the Norwegian Arctic marine environment



Reference:

Dowdall M., Gwynn J. P., Selnæs Ø. G. and Lind B. Monitoring of ⁹⁹Tc in the Norwegian Arctic marine environment. StrålevernRapport 2003:5
Østerås: Norwegian Radiation Protection Authority, 2003.

Keywords:

Technetium-99, Arctic, radioactivity, marine, radiochemical

Abstract:

This report details the monitoring of ⁹⁹Tc in the Norwegian Arctic by the Norwegian Radiation Protection Authority and a discussion of technical considerations for the production of high quality ⁹⁹Tc monitoring data. The results of monitoring indicate that ⁹⁹Tc levels in Norwegian Arctic waters and biota are low compared to the Norwegian Coastal environment, but that continued monitoring is required to assess future levels of this radionuclide within Arctic waters.

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Stikkord:

Technetium-99, arktisk, radioaktivitet, marint, radiokjemisk

Sammendrag:

Denne rapporten gir en redegjørelse for overvåkningen av ⁹⁹Tc i norsk arktisk område gjort ved Statens strålevern og diskuterer tekniske overveielser ved fremskaffelsen av overvåkingsdata av god kvalitet for ⁹⁹Tc. Resultatene av overvåkningen antyder at ⁹⁹Tc-nivåer i norske arktiske havområder og biota er lave sammenlignet med norske kystmiljøer, men at fortsatt overvåking er nødvendig for å vurdere fremtidige nivåer til denne radionukliden i arktiske havområder.

The project is funded by the Ministry of the Environment
Head of project: Anne Liv Rudjord

Approved:



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25 pages.

Published 2003-08-15.

Printed number 200 (03-08).

Cover design: Lobo Media as, Oslo.

Printed by Lobo Media as, Oslo

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www.nrpa.no

ISSN 0804-4910

Preface

Monitoring of radioactivity in the Arctic marine environment is of special significance due to the particular vulnerability of the environment and its constituent ecosystems. In recent years much attention has been devoted to the presence of the radioactive contaminant ^{99}Tc in the Norwegian marine environment, the main source of this contamination being activities conducted at European nuclear reprocessing facilities. The discovery of ^{99}Tc in the Norwegian Arctic marine environment precipitated the initiation of monitoring activities by the Norwegian Radiation Protection Authority, within the framework of the national programme for monitoring of Radioactivity in the Marine Environment (RAME), to assess and elucidate the levels and behaviour of this contaminant in the Norwegian Arctic marine environment. This report presents and discusses the results of those monitoring activities to provide an overview of ^{99}Tc levels in this vulnerable environment.

The provision of accurate data pertaining to the levels of ^{99}Tc in the Arctic marine environment presents special problems to the monitoring authority with respect to both logistical and analytical aspects. This report also serves to provide information regarding considerations that must be made in the analysis of samples exhibiting low levels of ^{99}Tc , such as those found in the Arctic areas.

Forord

Overvåkning av radioaktivitet i arktisk marint miljø er spesielt viktig på grunn av den særlige sårbarheten til miljøet og de involverte økosystemene. I de siste årene har det blitt gitt mye oppmerksomhet til tilstedeværelsen av det forurensende radioaktive stoffet ^{99}Tc i det norske marine miljøet, og den hovedsakelige kilden til denne forurensningen er aktiviteter som utføres ved europeiske gjenvinningsanlegg for radioaktivt avfall. Oppdagelsen av ^{99}Tc i norsk arktisk marint miljø fremskyndet oppstart av overvåkningsaktiviteter ved Statens strålevern, innenfor rammen til det nasjonale programmet for overvåkning av radioaktivitet i det marine miljøet (RAME), for å vurdere og forklare nivåene og oppførselen til denne forurensningen i det norske arktiske marine miljøet. Denne rapporten fremlegger og diskuterer resultatene fra denne overvåkingen og gir en oversikt over ^{99}Tc -nivåer i dette sårbare miljøet.

Fremskaffelsen av nøyaktige data angående nivåene av ^{99}Tc i det arktiske marine miljøet byr på spesielle problemer for overvåkningsmyndighetene av både logistisk og analytisk art. Denne rapporten gir også informasjon angående hensyn som må tas i analysen av prøver som fremviser lave ^{99}Tc -nivåer, slik som de som stammer fra de arktiske områdene.

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1. Introduction

The marine environment of the Norwegian Arctic is often perceived as relatively pristine but is nonetheless extremely vulnerable to a wide range of contaminants. Radioactive contamination of the region has received a lot of attention in recent years due to a number of factors. Perhaps the most important of these factors is the extent of public concern with respect to current and future levels of radioactive contaminants in Norwegian waters. This concern has been heightened with the discovery that radioactive contamination from the Irish Sea was being transported by ocean currents through the North Sea, along the Norwegian coast and into Arctic waters. The particular sensitivity of society to radioactive contaminants can have significant and far-reaching impacts.

The importance of maritime industries to the Norwegian economy and the vulnerability of these industries to the public perception of their products, both in Norway and internationally, mean that

rumours regarding radioactive contaminant levels can have serious and long-lasting economic and social ramifications.

The long-range transport of radioactive contaminants to the Arctic region is also of some concern given the vulnerability of this environment to contamination and the relative lack of knowledge with respect to the long-term behaviour and impacts of introduced contaminants on the peoples, flora and fauna of the region. It is these facets of the problem of radioactive contamination that impart particular importance to the monitoring of radionuclide levels in Arctic waters and the provision of high-quality data regarding these levels.

1.1 Technetium-99 (^{99}Tc)

^{99}Tc is a fission product of uranium and is formed in kilogram amounts during nuclear reactions by the beta decay of ^{99}Mo as presented in Figure 1.

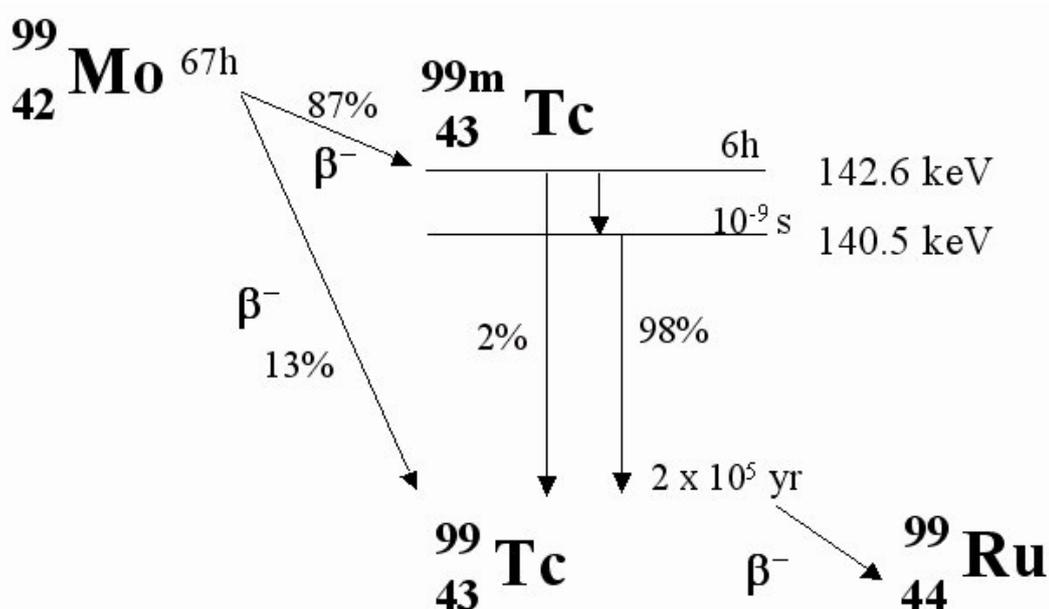


Figure 1. Decay scheme of ^{99}Mo

^{99}Tc , with a half-life of 213000 years and a specific activity of 630 kBq/mg, undergoes beta decay to form the stable isotope ^{99}Ru . Since its discovery, artificially created by the bombardment of molybdenum with deuterons (Perrier and Segre, 1947), 21 isotopes of the element have been prepared with atomic mass numbers between 90 and 110 (Long and Sparkes, 1988). All known isotopes of technetium are radioactive and although miniscule amounts of ^{99}Tc have been found as a result of naturally occurring processes, namely the spontaneous fission of ^{238}U (Kenna and Kuroda, 1964), by far the greatest source of ^{99}Tc is the operation of nuclear reactors and associated facilities in the nuclear industry. An estimated 25-30 tonnes of the isotope have been produced worldwide with ~1% of that amount (150-200 TBq) having been released to the environment (Aarkrog et al., 1986, Beasley and Lorz, 1986). Of the total amount released to the environment, ~85% has been released as a result of nuclear fuel reprocessing plants and the remaining ~15% has resulted from the testing of nuclear weapons (Dahlgaard, 1995).

The long half-life of ^{99}Tc , its high environmental mobility as the poorly sorbed, chemically stable, anionic pertechnetate species (TcO_4^-) and its infiltration of food chains as an analogue of sulphate (Cataldo et al., 1989, Bondietti and Francis, 1979) make the contamination of the environment with this isotope a matter of some concern.

1.2. ^{99}Tc discharges from nuclear reprocessing facilities

Past and continued sources of ^{99}Tc in the Arctic marine environment are the nuclear fuel reprocessing facilities at Sellafield in the UK and Cap la Hague in France. Both plants are engaged in the reprocessing of spent fuel from nuclear reactors. Although some nations have chosen to store the spent nuclear fuel from their reactors, the UK

and France reprocess the spent fuel produced by their own reactors and the reactors of some other countries. The reprocessing process involves dissolution of the nuclear fuel in acids with subsequent isolation of the reusable uranium and plutonium by a variety of chemical means. Other radioactive isotopes, formed during usage of the fuel, are liberated from the solid fuel by the dissolution process and are regarded in general as waste products. Such isotopes include ^{90}Sr , ^{137}Cs and ^{99}Tc . Much of this waste can be concentrated and stored on-shore for future disposal or continued storage. However a proportion of this waste is discharged to the oceans.

1.3. Historical discharges of ^{99}Tc from the Sellafield and Cap la Hague facilities

Discharges of ^{99}Tc from these facilities have varied in magnitude over the past decades (Figure 2). During the 1980's and into the early 1990's, the French facility at Cap la Hague dominated in terms of total ^{99}Tc discharged. In the period 1982-1991, Sellafield discharged a total of 42.4 TBq of ^{99}Tc (BNFL, 1982-1991) compared to a discharge of 102 TBq by the la Hague facility during the same period (Herrmann et al., 1995).

In the period prior to the early 1980's, ^{99}Tc discharges were largely dominated by the Sellafield contribution although estimation of the discharges are complicated due to the fact that ^{99}Tc was not reported as an individual radionuclide prior to 1978. Leonard et al. (1997a) estimate discharges of ^{99}Tc between 1952 and 1968 to have been between 5 and 10 TBq per annum rising to 40 TBq per annum between 1970 and 1977. Between 1980 and 1994, ^{99}Tc was stored on-site at Sellafield whilst the Enhanced Actinide Removal Plant (EARP) was commissioned and discharges of ^{99}Tc were of the order of 4-6 TBq per annum. The EARP plant was designed to treat high

level nuclear wastes arising from the operation of BNFL's Magnox reprocessing plant by removing plutonium, cesium and strontium isotopes from the waste. ^{99}Tc in the waste is present as the pertechnetate ion and cannot easily be removed from the waste by the processes employed at the EARP plant (Busby et al., 1997). The net result of this fact has been that since 1994, ^{99}Tc discharges from the EARP plant at Sellafield have increased 50-fold from the discharge levels of the early 1990's, to a level of 150-190 TBq per annum in 1995 and 1996.

northern reaches of the Irish Sea in a relatively short period (approx. 3 months). The ^{99}Tc signal was then lost due to inflows of water from the Atlantic Ocean but reappeared in the North Sea having taken ~9 months to traverse the distance between the point of discharge (Cumbria, UK) and the northern coast of Scotland.

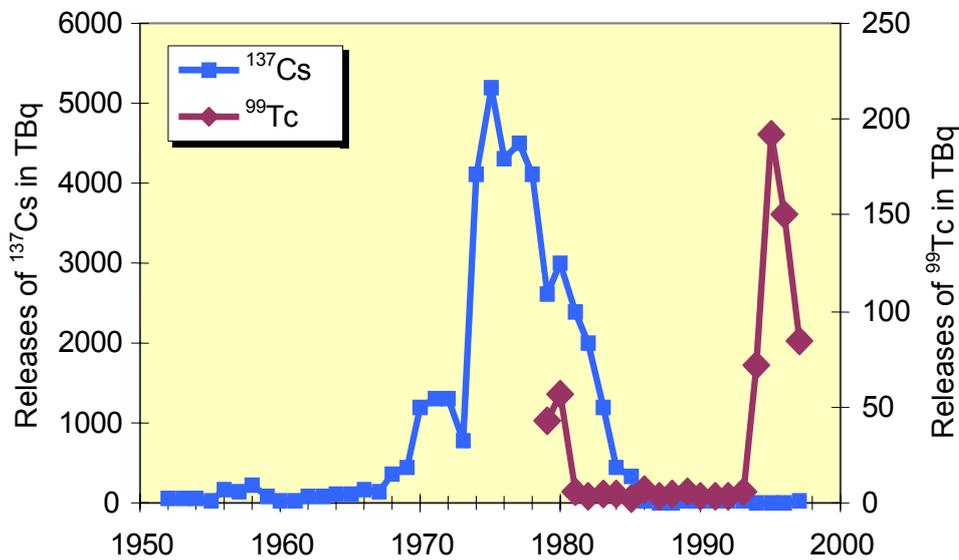


Figure 2. Annual discharges (TBq) of ^{99}Tc and ^{137}Cs from the Sellafield facility

1.4 Transport of ^{99}Tc to the Arctic

Leonard et al. (1997b) compared levels of ^{99}Tc in British coastal waters before and after the initiation of activities at the EARP plant and observed an increase in the levels of ^{99}Tc in the waters of the Irish Sea from 1-4 Bq/m³ before operations at EARP commenced to in excess of 30 Bq/m³ (northern Irish Sea) and 200 Bq/m³ (off western Scotland) after EARP began processing nuclear waste. Observations indicated that the ^{99}Tc had migrated to the

The findings of Brown et al. (1998) indicated that ^{99}Tc from the increased Sellafield discharges beginning in 1994 had reached Norwegian coastal waters some time before November 1996 and could be detected off Northern Norway by December 1997. ^{99}Tc is transported to Norwegian west coast waters by the Norwegian Coastal Current via the North Sea. This current continues along the Norwegian coast before flowing splitting

into the north flowing West Spitsbergen Current and the east flowing Nordkapp Current, transporting any contaminant load into the Norwegian Arctic marine environment (Figure 3).

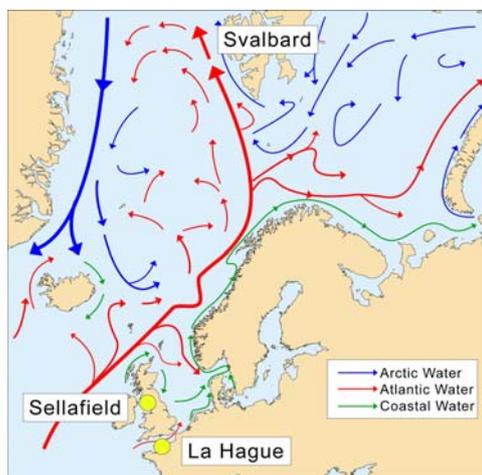


Figure 3. Schematic of the surface circulation of the Northern seas that are involved in the transport ^{99}Tc to the Norwegian Arctic

The most recent data regarding levels of ^{99}Tc in the waters and biota off mainland Northern Norway indicate an average level of $1.25 \pm 0.33 \text{ Bq/m}^3$ in seawater and $321 \pm 68 \text{ Bq/kg (d.w.)}$ in seaweed. These values continue to demonstrate the contemporary elevated levels of this isotope in the Norwegian marine environment.

2. Monitoring of ^{99}Tc in the Norwegian Arctic marine environment

In 1999, sampling and analysis of waters and biota along the Norwegian coast by the Norwegian Radiation Protection Authority increased under the new marine monitoring programme (RAME) funded by the Ministry of the Environment, the primary objectives of which are to monitor and document levels of and trends in radionuclide contamination in Norwegian waters. In 2000, an Environmental Unit was established by NRPA at the Polar Environmental Centre in Tromsø, part of its responsibilities being monitoring of the Norwegian Arctic marine environment for traces of radioactive contamination.

This monitoring is conducted via regular sampling at a number of sites within the Norwegian Arctic waters, participation in cruises and sampling during field expeditions on Svalbard. The aim of these activities is to obtain information regarding the occurrence and behaviour of ^{99}Tc and other radionuclides in the Norwegian Arctic marine environment.

2.1 Sampling

As part of NRPA's ongoing radionuclide marine monitoring programme (RAME), stations for the routine collection of sea water samples for monitoring the levels of ^{99}Tc within the Norwegian Arctic were established in 2001 at the Ny Ålesund settlement in Kongsfjorden (78°56' N 11°56' E), Svalbard and at the Norwegian Meteorological Institute's base stations on Bjørnøya (74°18' N 19°00' E), Hopen (76°24' N 25°00' E) and Jan Mayen (70°36' N 8°24' W). Additional sampling of seawater and marine biota was conducted during NRPA field campaigns to Svalbard in 2000, 2001 and 2002 and to Bjørnøya and Hopen in 2000 onboard the K.V. Senja.



Figure 4. Sampling of seawater by NRPA personnel in Kongsfjorden, Svalbard (Photo: S.Gerland).

2.2 Analytical Methods

^{99}Tc is a pure beta emitter and its quantitative analysis is conducted by its pre-concentration using ion-exchange, isolation using co-precipitation and solvent extraction and mounting on steel planchettes using an electrolytic procedure. A schematic of the method is presented in Figure 6.

Sample volumes are usually 50 l but sometimes may be as much as 100 l. The recovery of ^{99}Tc from the analytical procedure is determined via the use of $^{99\text{m}}\text{Tc}$, which is added at the start of the procedure.

The low levels of ^{99}Tc in Arctic waters necessitate special precautions with respect to the possible introduction of contaminant species in the tracer solution. A discussion of the analysis of environmental samples from Arctic areas for ^{99}Tc is presented in Section 4.

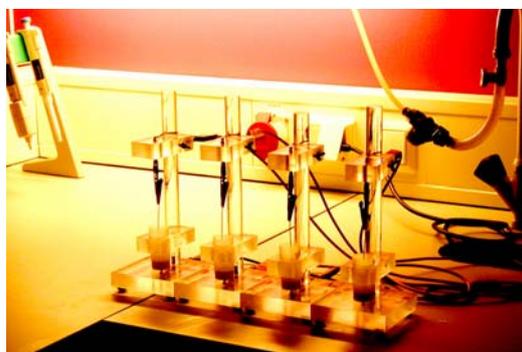


Figure 5. Electrodeposition of purified ^{99}Tc onto steel planchettes for final counting.

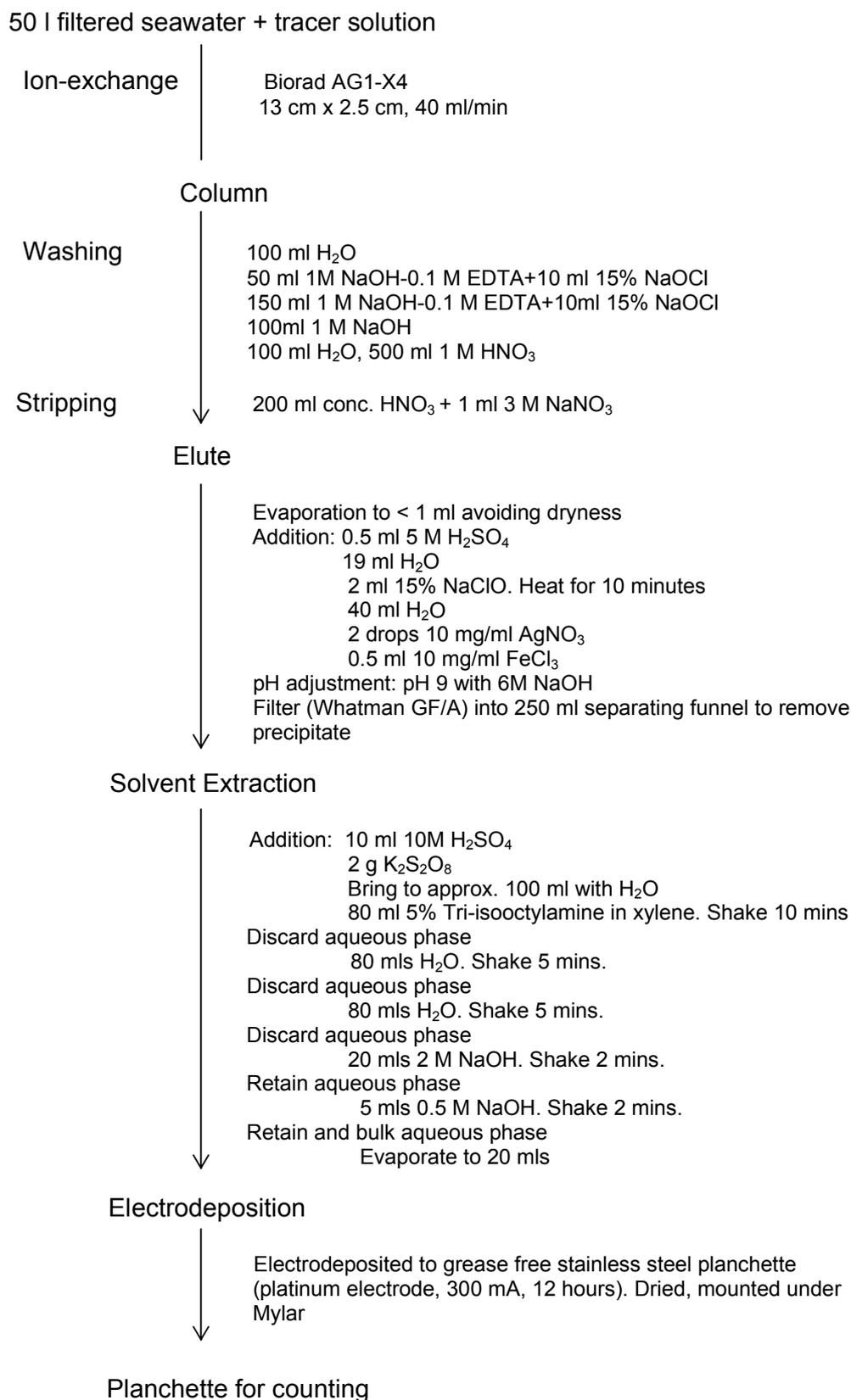


Figure 6. Radiochemical separation of ⁹⁹Tc (after Chen et al., 2001)

3. Results and Discussion

3.1 ⁹⁹Tc in seawater

Activity concentrations of ⁹⁹Tc in sea water from the Norwegian Arctic in 2001 ranged from 0.13 – 0.36 Bq/m³, a similar magnitude to that reported for the Norwegian Arctic in 2000, but a five fold increase on 1994 levels of 0.03 - 0.08 Bq/m³ (Kershaw et al., 1999). The highest concentrations of ⁹⁹Tc at the Svalbard monitoring stations were observed at Bjørnøya and Hopen (0.35 ± 0.05 and 0.32 ± 0.04 Bq/m³) whilst the highest concentration at Ny Ålesund was 0.25 ± 0.03 Bq/m³ (Figure 7). Average levels of ⁹⁹Tc at Jan Mayen (0.1 Bq/m³) were less than half the observed average levels at the three monitoring stations within the Svalbard area.

These compare with an average concentration for 2001 of 1.24 Bq/m³ (range 0.9 – 1.95 Bq/m³) for the NRPA monitoring station at Hillesøy on mainland Norway (Kolstad and Lind, 2002).

These values reflect the dominant oceanic surface circulation in the Northern seas and the concomitant transfer of EARP associated ⁹⁹Tc into the Norwegian Arctic. That lower levels of ⁹⁹Tc are observed in the Svalbard area compared to Norwegian coastal waters for the same period is due to the ingress of North Atlantic Water into the Norwegian Coastal Current (NCC) and the West Spitsbergen Current (WSC) with the resultant dilution of the EARP associated ⁹⁹Tc signal.

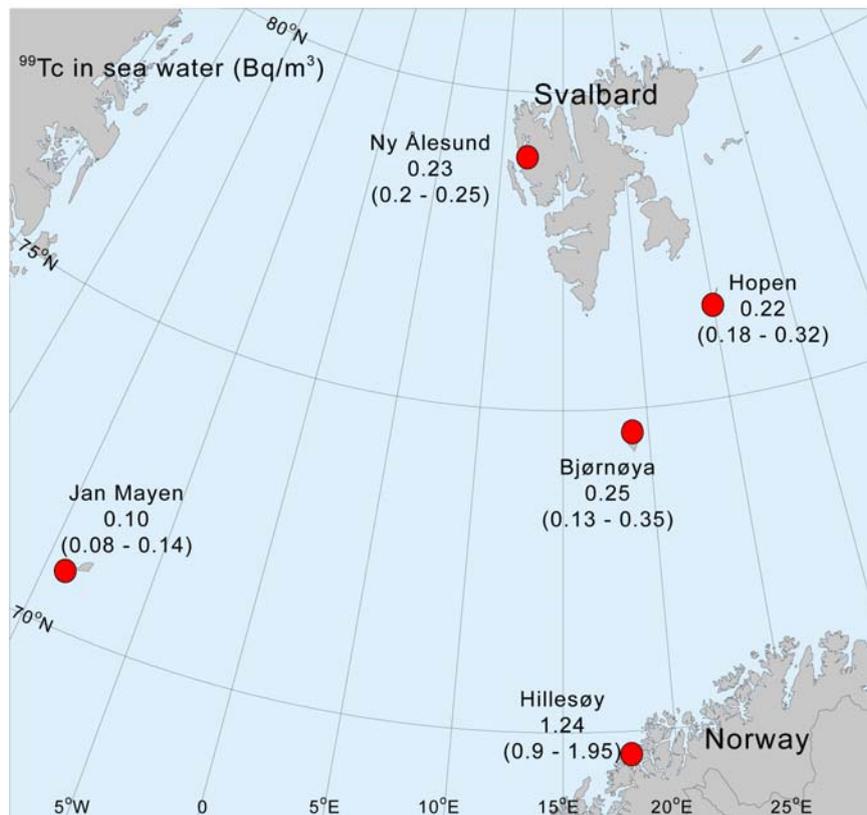


Figure 7. The average and range of activity concentrations of ⁹⁹Tc (Bq/m³) in sea water collected at monitoring stations within the Norwegian Arctic.

Nevertheless, levels of ^{99}Tc in the centre of the WSC for 2000 and 2001 were recorded at 0.39 ± 0.05 and $0.24 \pm 0.04 \text{ Bq/m}^3$ respectively, compared to 1994 pre-EARP levels of 0.04 Bq/m^3 (Kershaw et al., 1999). The lower levels of ^{99}Tc observed at Jan Mayen reflect its oceanographic setting and distance from the NCC and WSC. That levels of ^{99}Tc at Jan Mayen are higher than reported background levels for Atlantic water of 0.005 Bq/m^3 (Dahlgaard et al., 1995, may be a reflection of residual ^{99}Tc levels in southerly flowing Arctic Water resulting from earlier discharges.

The importance of the WSC influence on Svalbard can be seen in the associated increase in levels of ^{99}Tc within the WSC and within fjords along the west coast of Spitsbergen. In 2001, the concentration of ^{99}Tc in two water samples from Isfjorden (~100km south of Kongsfjorden) was 0.28 ± 0.04 and $0.29 \pm 0.05 \text{ Bq/m}^3$ compared to a single measurement of $0.13 \pm 0.03 \text{ Bq/m}^3$ in 1999. That ^{99}Tc levels in the inner part of Kongsfjorden are of a similar magnitude to levels observed in the outer fjord, is an indication of the importance and scale of the WSC in exchange processes throughout the entirety of this fjord.

In 2001, sub-surface samples taken in addition to surface samples at stations in Kongsfjorden, Isfjorden and in the WSC, showed relatively uniform levels of ^{99}Tc (Table 1). However, sub-surface samples taken in the WSC in 2000 at greater depths (465 - 1000m) showed levels of ^{99}Tc of 0.17 and 0.04 Bq/m^3 , 2 to 5 fold lower than surface levels. That uniform distributions of ^{99}Tc were observed in the top 55 – 125m of Kongsfjorden, Isfjorden and the WSC is a reflection of the conservative nature of ^{99}Tc in sea water and the apparent depth of the mixed layer at the time of sampling.

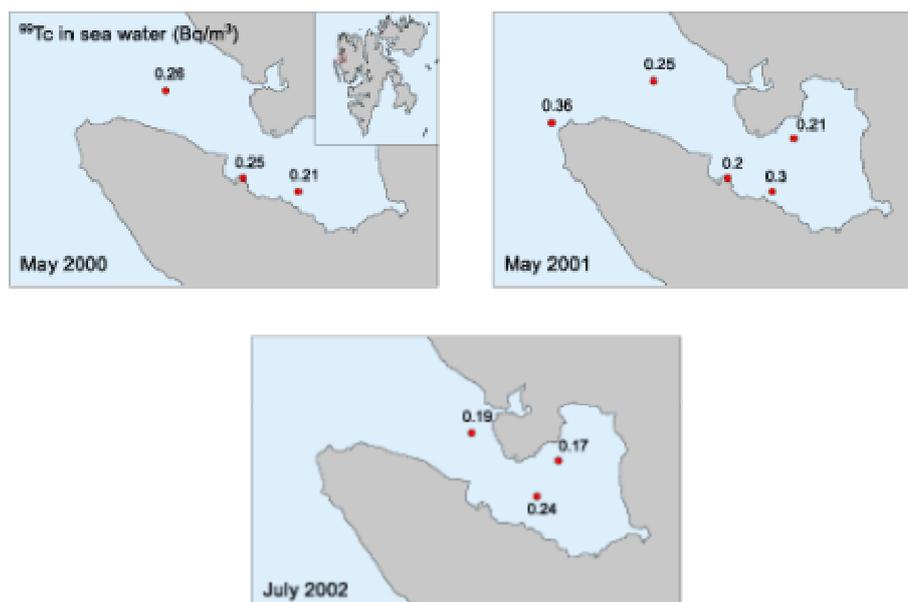


Figure 8. Activity concentrations of ^{99}Tc (Bq/m^3) in surface seawater from Kongsfjorden, collected in May 2000, 2001 and July 2002.

Table 1. Activity Concentrations of ^{99}Tc (Bq/m^3) in surface and sub-surface seawater samples collected from the Svalbard area in 2001.

Location	Depth (m)	^{99}Tc (Bq/m^3)	Salinity (‰)
Kongsfjorden	0	0.25 ± 0.04	34.4
	125	0.29 ± 0.03	34.6
Isfjorden	0	0.28 ± 0.04	34.1
	55	0.35 ± 0.04	34.1
WSC	0	0.24 ± 0.04	34.5
	100	0.22 ± 0.04	34.1

The observed variation in sample salinity may partly explain the variation in surface ^{99}Tc levels both within fjords and between consecutive years. Due to the conservative behaviour of ^{99}Tc in sea water, mixing dynamics between ^{99}Tc bearing sea water from the WSC and fresh water input from terrestrial runoff and glacial meltwater should have proportional effects on both

^{99}Tc levels and sample salinity. In Kongsfjorden, average ^{99}Tc and salinity levels for May 2001 were $0.24 \text{ Bq}/\text{m}^3$ and 34.5‰ , whilst average ^{99}Tc levels for July 2002 were $0.2 \text{ Bq}/\text{m}^3$ and 28.9‰ . A relationship between salinity and ^{99}Tc concentrations in seawater has been observed previously at Hillesøy (Kolstad and Lind, 2002) and is further suggested by

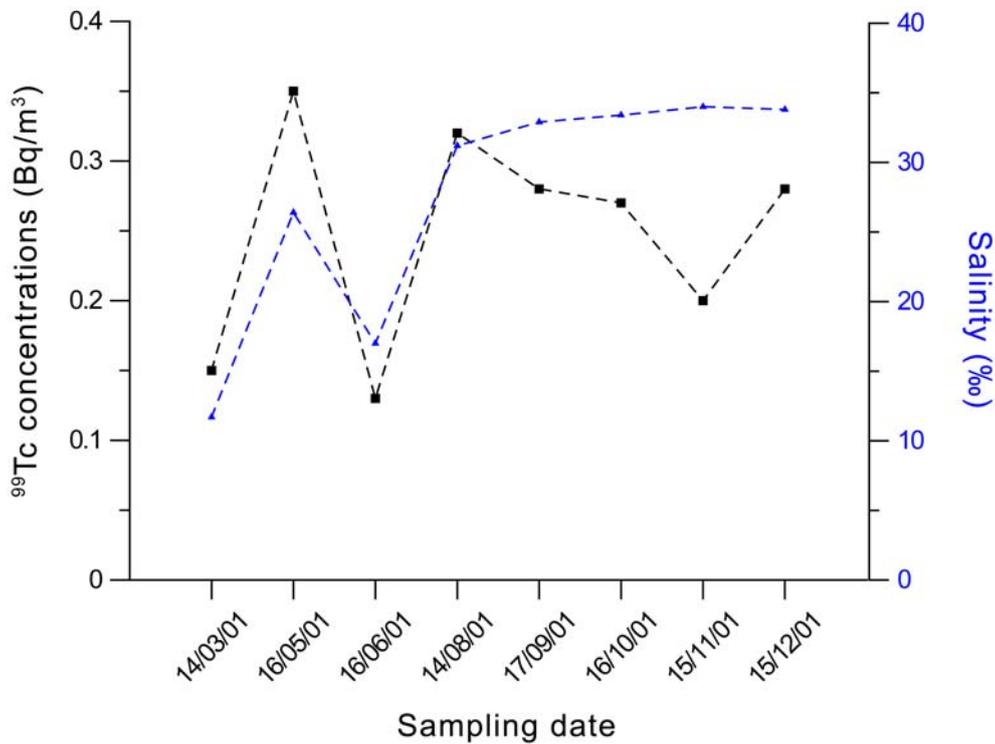


Figure 9. Activity concentrations of ^{99}Tc (Bq/m^3) and salinity (‰) of surface seawater samples collected at Bjørnøya in 2001

the 2001 time series for Bjørnøya (Figure 9). When considered together, these observations suggest that ^{99}Tc levels may in fact be unrepresentative in situations where the salinity of a seawater sample is below the normal range for that sampling location. Alternatively, fluctuations in ^{99}Tc concentrations in the short term may simply reflect variations in actual discharges from Sellafield (Brown et al., 2002).

3.2 ^{99}Tc in marine algae

Seaweed samples were collected from Kongsfjorden in 2000, 2001 and 2002 and from Bjørnøya and Hopen in 2001. Data for fucoid seaweeds (*Fucus distichus* and *F. vesiculosus*) represents bulked samples, while data for kelp seaweeds (*Laminaria* spp. and *Alaria esculenta*) represents individual specimens.

The average ^{99}Tc concentrations in different species of seaweed collected from Kongsfjorden in 2000, 2001 and 2002 are summarised in Table 2.

Over the period 2000 to 2002, higher ^{99}Tc levels were generally observed in the fucoid seaweeds (25.7 – 40.3 Bq/kg) than the kelp seaweeds (5.8 – 28.9 Bq/kg) as has been reported for mainland Norway (Kolstad and Lind, 2002).

It is difficult to draw any firm conclusions with regard to any temporal trends due to the limited number of samples that were taken, although there is a suggestion that levels in the fucoid seaweeds maybe decreasing, whilst levels in the kelp seaweeds are increasing.

The variation in ^{99}Tc concentrations between species during the sampling period are probably related to species specific metabolic processes governing uptake and depuration rates.

Table 2. Average activity concentrations (Bq/kg d.w.) and concentration factors (CF) of ^{99}Tc in marine algae from Kongsfjorden collected in 2000, 2001 and 2002.

Species	2000		2001		2002	
	^{99}Tc (Bq/kg)	CF	^{99}Tc (Bq/kg)	CF	^{99}Tc (Bq/kg)	CF
<i>Laminaria</i> spp.	6.8 ±1.4 (n=3)	32000	5.8 ±0.8 (n=2)	16000	28.9 ±2.8 (n=1)	120000
<i>Fucus</i> <i>distichus</i>	34.3 ±3.3 (n=1)	137000	40.3 ±7.1 (n=3)	145000	25.7 ±2.5 (n=1)	151000
<i>Fucus</i> <i>vesiculosus</i>	-	-	46.7 ±6.6 (n=2)	172000	31.5 ±3.1 (n=1)	185000
<i>Alaria</i> <i>esculenta</i>	8.2 ±0.8 (n=1)	40000	13.2 ±1.3 (n=1)	37000	-	-

For seaweeds collected from Bjørnøya and Hopen in 2001, higher ^{99}Tc concentrations were seen in fucoid seaweeds (26.6 – 58.7 Bq/kg) than kelp seaweeds (15.1 – 25.1 Bq/kg) as was observed for samples collected from Kongsfjorden (Table 3). However, levels of ^{99}Tc were in general slightly higher compared to those observed in Kongsfjorden for the same year, especially with regard to the kelp seaweed *L. hyperborea*. This is probably a reflection of the higher levels of ^{99}Tc in the marine environment around Bjørnøya and Hopen as compared to Kongsfjorden. Levels of ^{99}Tc in fucoid seaweeds from the Svalbard area from 2000 and 2001 were 7-8 fold lower than reported average levels for the same period in *F. vesiculosus* from Hillesøy of 318 and 321 Bq/kg respectively (Kolstad and Lind, 2002). This is in keeping with the observed gradient in ^{99}Tc concentrations in sea water between the Svalbard area (lower ^{99}Tc levels) and mainland coastal sites in Norway (higher ^{99}Tc levels).

Average concentration factors (CF) for ^{99}Tc in seaweed samples from Kongsfjorden, Bjørnøya and Hopen were calculated using single or where possible, average sea water levels.

Since the inferred CF represent snapshot and not equilibrium values, care must be taken in their use. However, CF for ^{99}Tc in fucoid seaweeds from the Svalbard area during the period 2000 to 2001 of 1.4×10^5 to 2.8×10^5 are in good agreement with previously reported values of 1.5×10^5 and 2.6×10^5 for Norwegian waters in 1998 and 2001 respectively (Kolstad and Lind, 2002). Likewise, ^{99}Tc CF for kelp seaweeds from the Svalbard area, in the sampling period are, with the exception of two samples, of the same order of magnitude compared to a 2001 value of 1.4×10^4 for *L. digitata* from Lista in Southern Norway (Kolstad and Lind, 2002).

Table 3. Average activity concentrations (Bq/kg d.w.) and concentration factors (CF) for ^{99}Tc in marine algae from Hopen and Bjørnøya collected in 2001.

Location	Species	^{99}Tc (Bq/kg)	CF
Hopen	<i>Laminaria hyperborea</i>	15.1 ±1.5 (n=1)	72000
	<i>Fucus distichus</i>	58.7 ±5.8 (n=1)	280000
Bjørnøya	<i>Laminaria hyperborea</i>	25.1 ±4.6 (n=3)	132000
	<i>Fucus vesiculosus</i>	26.6 ±2.6 (n=1)	140000

4. Measurement of ^{99}Tc in Arctic waters: Analytical considerations

The low levels of ^{99}Tc in Arctic waters necessitate special considerations with respect to the accurate determination of the quantities of the nuclide present. This discussion will concern itself with radiochemical procedures for the determination of the nuclide in low level samples with particular reference to the use of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators for the production of $^{99\text{m}}\text{Tc}$ yield tracers.

4.1 Radiochemical procedures for the determination of ^{99}Tc

The radiochemical separation and analysis of ^{99}Tc in environmental matrices is relatively straightforward. However a difficulty is encountered when attempting to accurately determine the chemical yield of the radiochemical procedure. The chemical yield may be determined by a number of methods:

1. Gravimetric measurement of a rhenium carrier (Harvey et al., 1991)
2. Simultaneous analysis of a sample carrying a known amount of ^{99}Tc
3. Addition of an isotopic tracer such as $^{99\text{m}}\text{Tc}$, ^{97}Tc , $^{97\text{m}}\text{Tc}$ or $^{95\text{m}}\text{Tc}$

All of these methods have concomitant advantages and disadvantages. Rhenium is often used as tracer for ^{99}Tc recovery. As a stable element, the use of rhenium is convenient for field operations as its use places no restraints on the time between tracer addition and preconcentration and

the final analysis sequence. The possibility of preconcentration of samples in the field also exists when employing rhenium which can result in an increase in the maximum size of samples. However, as with all non-isotopic tracers, problems exist in relation to slight differences in the chemistry of Tc and Rh with particular reference to anion-exchange behaviour, differences in the volatilities of the two elements, and differences in the ease with which they are reduced. Although such problems are significant, modifications to the chemical procedures can be made to accommodate these differences.

Simultaneous analysis of samples with known amounts of ^{99}Tc presents problems in relation to the amount of work and expense required to conduct such analysis and ensuring similar behaviours between a "natural" sample and one containing added ^{99}Tc . The use of isotopic tracers has been discussed extensively in the literature. ^{97}Tc , $^{95\text{m}}\text{Tc}$ and $^{97\text{m}}\text{Tc}$ have all been proposed as isotopic tracers. ^{97}Tc with a half life of 2.6×10^6 years appears to offer a viable solution and has been used as a tracer in the mass spectrometric determination of ^{99}Tc (Anderson and Walker, 1980), however ^{97}Tc solutions of a satisfactory purity have yet to be produced. This problem is not so significant in the production of $^{95\text{m}}\text{Tc}$ which can be prepared by bombardment of ^{93}Nb with helium nuclei but low yields make the cost prohibitive. $^{97\text{m}}\text{Tc}$ has been reported as a successful tracer (Kaye et al., 1982) but requires the use of absorbers during beta counting to eliminate beta emissions from the nuclide.

These problems have focussed attention on $^{99\text{m}}\text{Tc}$ as a suitable tracer. $^{99\text{m}}\text{Tc}$ is perhaps the pre-eminent radionuclide in the field of diagnostic nuclear medicine. This is largely due to the diversity of the chemical reactions in which Tc can be involved and the nuclear and physical properties of the $^{99\text{m}}\text{Tc}$ isotope (Siegel and Deutsch, 1975, Clarke and Fackler, 1982). However the

major factor in the growth in usage of the isotope has been the wide availability of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators. The production of such generators involves the irradiation of Mo or its compounds, which are typically on an alumina column, within a nuclear reactor to produce ^{99}Mo which decays to form $^{99\text{m}}\text{Tc}$ according to Figure 1.

The $^{99\text{m}}\text{Tc}$ so formed can be washed or eluted out of the generator, as the pertechnetate species, using sodium chloride solutions.

4.2 Radiochemical purity of $^{99\text{m}}\text{Tc}$ solutions from $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators

A major concern regarding the use of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators is the purity of the eluate solution (Vesely and Cifka, 1970). The possible contaminants that may occur may be conveniently divided into two groups; isotopes of elements other than Tc and isotopes of Tc other than $^{99\text{m}}\text{Tc}$. The possible isotopes and their properties are listed in Table 4.

As can be seen, some of the possible non-Tc impurities can be detected by gamma spectrometry if present in the eluate. Determination of ^{131}I is complicated by the continuous production of $^{99\text{m}}\text{Tc}$ by any contaminant ^{99}Mo present in the eluate. This is easily remedied however by the addition of potassium iodide carrier to an acidified aliquot of the eluate, addition of ferric chloride and extraction of elemental iodine into chloroform which can then be re-purified and counted on a gamma spectrometer. A second problem in the quality control of eluates for impurities is the presence of high activities of $^{99\text{m}}\text{Tc}$, which is a strong gamma emitter. The high activity of the eluate can place constraints on the gamma spectrometry system but the major limitation on using gamma spectrometry for the determination of

radiochemical impurities is the inclusion of sum peaks in the gamma spectrum. These sum peaks may preclude the assessment of ^{131}I in the 300 keV region and very high activities may even affect ^{99}Mo in the 700 keV region. Use of lead absorbers can mitigate the problem somewhat as the reduction in the count rate for $^{99\text{m}}\text{Tc}$ is much more severe than for ^{131}I or ^{99}Mo . The presence of ^{99}Mo is of potential concern as it will constitute a source of $^{99\text{m}}\text{Tc}$ within the tracer solution. Chemical yield calculations in ^{99}Tc analysis rely on the comparison of the signal produced in a gamma detector by an unadulterated mass of tracer and a mass of tracer that has gone through the analytical sequence.

Assuming removal of ^{99}Mo at an early stage in the analysis, a comparison of prepared sample and the unadulterated tracer solution will not yield correct results due to the continued ingrowth of $^{99\text{m}}\text{Tc}$ in the unadulterated tracer solution. Further contaminants in the eluates of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators are the isotopes ^{90}Sr , ^{89}Sr and ^{90}Y (Sodd and Fortman, 1976) although reported levels are so low as to be insignificant with respect to the current discussion.

Consideration of the isotopes so far discussed that may be present in eluates indicates a number of pertinent points. The half-lives of the two iodine isotopes are so short that it is unlikely in the extreme that any significant amounts of these isotopes could be present in a generator two to three weeks after manufacture. Although the Sr and Y isotopes reported as having been found in some eluates are of long enough half-life to have a presence after some weeks, it appears from the literature that levels of these isotopes are, if present, too low to be of significance. The Ru isotopes reported as contaminants are of sufficient half-life to be present in eluates after some time but one of these is easily detected if present.

Table 4. Potential isotopic impurities in eluates from $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators

Isotope	$t^{1/2}$	γ Energies (keV)	β_{max} Energy (MeV)
^{99}Mo	65.9 hours	140.5 (4.5%) 739.5 (12.2%) 777.9 (4.2%)	1.350
^{99}Tc	211100 years		0.294
$^{99\text{m}}\text{Tc}$	6.01 hours	140.5 (89.6%)	0.437
^{131}I	8.02 days	364.4 (81.7%)	0.971
^{132}I	2.95 hours	522.6 (16.0%)	3.577
^{106}Ru	373.59 days		0.039
^{90}Sr	28.74 years		0.546
^{90}Y	64.1 hours		2.282
^{89}Sr	50.53 days		1.495
^{103}Ru	39.26 days	497.1 (91%)	0.763

With these considerations in mind, it is unlikely that any of these isotopes presented could be present in enough quantities to constitute an interferent after the isolation of Tc during the radiochemical separation. ^{99}Mo could constitute an interference via its presence in the unadulterated reference tracer solution.

Although the presence of non-Tc isotopic impurities appears to be a matter of little concern regarding the usage of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ as a source of tracer in low level samples, more significant is the presence of Tc isotopes, namely ^{99}Tc .

A cursory examination of the decay scheme of ^{99}Mo (Figure 1.) indicates that there are in fact two routes by which ^{99}Tc can be produced within a $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generator. Any ^{99}Tc within the eluate produced by such a generator will pass through the analytical sequence and contribute to the final analytical signal. The most obvious solution to predicting how much ^{99}Tc is generated by the generator to use the ^{99}Mo decay scheme, information regarding the amount of Mo irradiated and the elution history of the generator to theoretically predict the activity of $^{99\text{m}}\text{Tc}$ in the generator eluate, the elution efficiency and the total mass of technetium present in the

eluate. This is impracticable due to the difficulties in obtaining such information and certain practical points that must be considered. Holland et al. (1986) assessed commercial $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators in the United States and reached a number of important conclusions;

1. That the amount of ^{99}Tc produced by generators is often underestimated by up to 300%
2. The amount of ^{99}Tc produced is extremely variable between generators from different manufacturers
3. That theoretical calculations are not sufficient to calculate the ratio of $^{99\text{m}}\text{Tc}/^{99}\text{Tc}$ in eluates from generators.

A number of possibilities are proposed as to account for the observed anomalies. These include differences in the chemical behaviour of the two isotopes in the generator due to the way the isotopes are formed within the generator and excess loading of Tc on the alumina column during the manufacture of the generator unit. Although it is often assumed that adopting certain milking procedures can reduce the ^{99}Tc levels in the generator to such a level that they do not represent a cause for concern, the points as are presented previously may have significant impacts for the use of such generators for the provision of $^{99\text{m}}\text{Tc}$ tracer solution for the analysis of ^{99}Tc in low level samples.

4.3 Assessment of radiochemical purity and ^{99}Tc contamination

Current procedures at NRPA for the radiochemical assay of ^{99}Tc in seawater are applied to samples whose ^{99}Tc activity is up to one order of magnitude higher than levels observed in high Arctic waters.

Assessment of the analytical procedures in relation to the use of $^{99\text{m}}\text{Tc}$ as a tracer in the analysis of Arctic samples indicated that it was necessary to establish if and to what extent possible isotopic contamination could have an impact on the analysis of low level samples such as those found in the Arctic marine environment whilst maintaining practicable sample sizes of the order of 50 l.

$^{99\text{m}}\text{Tc}$ tracer solution is currently obtained at NRPA from $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators whose nominal activity at time of manufacture is 25 GBq ^{99}Mo . At the time of use, some two to three weeks later, this activity has dropped by a factor of 1000 to levels of MBq ^{99}Mo . Procedures for the extraction of eluate from the generator involve an initial milking of the generator to remove such ^{99}Tc and $^{99\text{m}}\text{Tc}$ as may have built up within the generator followed by a second milking to produce $^{99\text{m}}\text{Tc}$ for use as a tracer. The period between the milking is determined by the age of the generator and is of sufficient time to allow the in-growth of an amount of $^{99\text{m}}\text{Tc}$ that constitutes a viable activity for use as a tracer.

In order to check the radiochemical purity of the generator eluates, bulk samples were prepared of both the 1st elutions and the 2nd elutions (used for tracers). These samples were prepared from a 2 week old generator and were collected over a 3 week period. The samples were then allowed to decay for 10 days to remove as much $^{99\text{m}}\text{Tc}$ as possible in order to be able to clearly see the higher energy region of the spectrum with a high resolution gamma ray spectrometer.

Analysis of the spectra of both the 1st and 2nd eluates indicates clearly (via the characteristic 497.1 keV emission) the presence of ^{103}Ru . Comparison with a soil sample containing usual background lines indicated that it was unlikely that the observed peak was a sum or derivative peak of any natural nuclides. No other gamma

lines were observed that might indicate the presence of other contaminants.

However, should ^{106}Ru be present in the generator, it is likely to also contaminate the eluate as it has the same chemical behaviour as ^{103}Ru but it will not be detected by the gamma spectrometric analysis so it cannot be ruled out as a potential contaminant. No lines from ^{99}Mo were observed. Of all the possible contaminants, only ^{103}Ru and ^{106}Ru were likely to be present given the length of time between manufacture of the generator and usage in the laboratory.

The presence of ^{99}Tc as a result of decay of other nuclides present in the generator was assessed in eluates used as tracer solutions from a number of different generators of different ages.

**High resolution gamma spectrum of generator eluate
(4-5 weeks old)**

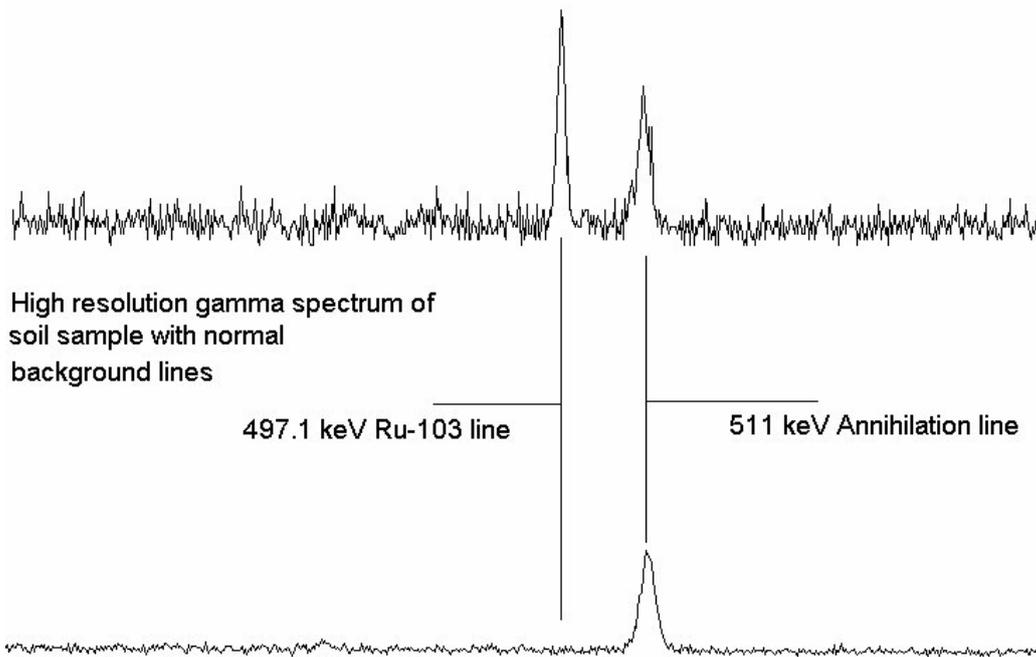


Figure 10. Gamma ray spectrum (400 – 600 keV) of tracer solutions exhibiting evidence of ^{103}Ru contamination. A soil sample exhibiting normal background lines is shown for comparative purposes.

A double approach was adopted, with known amounts of the same tracer, from different generators, electroplated directly onto steel planchettes and added to 50 l aliquots of distilled water and processed as per normal chemical procedures. All planchettes were then stored for up to 2 weeks to allow for decay of ^{99m}Tc and were then counted using a low background anti-coincidence detector. The counts per unit time provided by the activity on the planchettes were then used to calculate “apparent” ^{99}Tc concentrations (Table 5). Results indicate that the presence of ^{103}Ru and other possible contaminants have a dramatic effect on the apparent ^{99}Tc contaminant activity of the tracer solutions (via the emission of beta particles), thus eliminating the possibility of checking for ^{99}Tc contamination using straight forward electroplating and counting of tracer eluates as a means of assessing ^{99}Tc levels in the tracers. The presence of ^{103}Ru on the planchettes containing unadulterated tracer solution was confirmed by high resolution gamma spectrometry.

Although it would appear that the chemical separation procedure is sufficient to remove at least some of the non-Tc contaminants, the presence of ^{99}Tc in the tracer solution presents a continuing problem for measurement of low level samples. The impact of ^{99}Tc contamination in tracer solutions on samples with varying levels of ^{99}Tc is graphically shown in Figure 11.

Table 5. Influence of ^{103}Ru contamination on the apparent ^{99}Tc activity of tracer solutions.

Age of Generator (days)	Apparent ^{99}Tc activity mBq/g	Actual ^{99}Tc activity mBq/g
23	223.8	0.87
28	174.0	2.91
35	123.8	4.11
21	331.4	7.13
23	202.4	8.21
30	102.1	1.8
32	84.0	4.13
35	68.0	4.13

A series of experiments were run to attempt to quantify the amount, if any, of ^{99}Tc present in the eluates. Quantities of tracer were processed as per environmental samples and similar quantities were electroplated directly to steel planchettes. A number of distilled water samples were also run without tracer for comparison purposes. Tracer solutions that were plated directly produced anomalous beta activity relative to purified tracer solutions, presumably due to the presence of ^{103}Ru as described previously.

However, all planchettes containing tracer solution produced a positive response for ^{99}Tc , with levels of the nuclide in the tracer solutions of the order 1-5 mBq/g, no such signal being produced using distilled water samples. Typically, 0.3-0.8 grams of tracer solution is required to insure a detectable signal at the end of the purification process. This implies that for samples containing activities in the order of 8-10 mBq, interference from ^{99}Tc produced by the generator is significant.

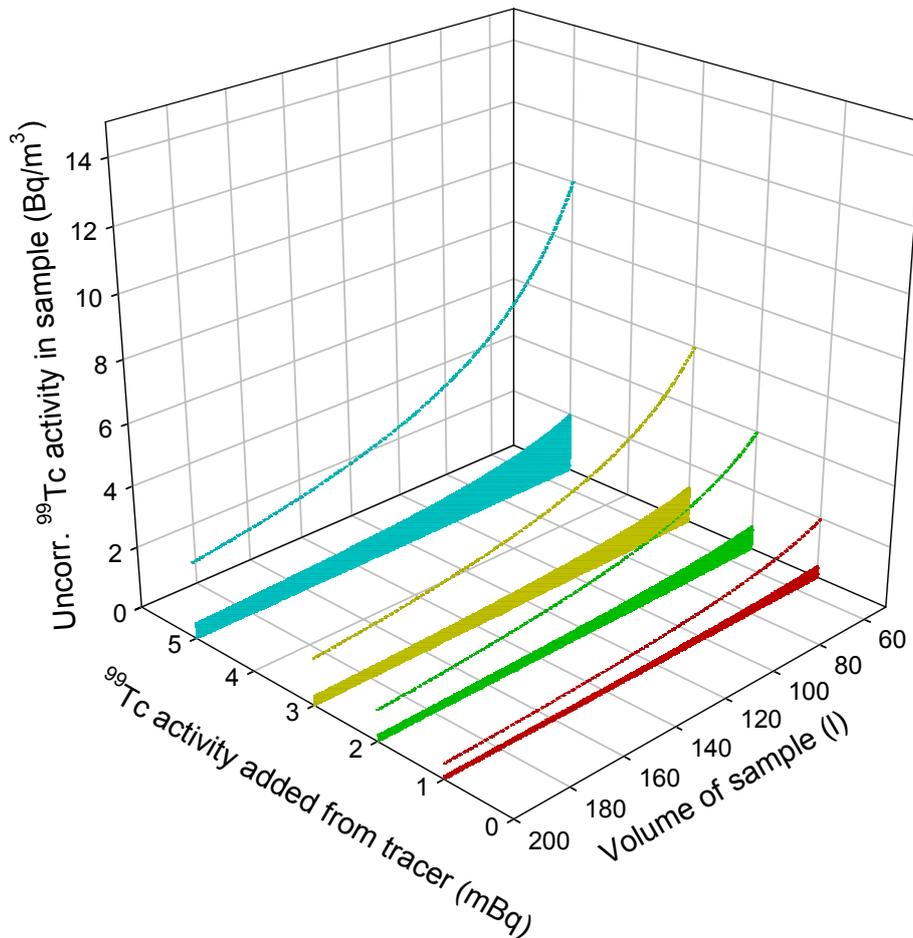


Figure 11. Contribution of tracer ^{99}Tc to total sample ^{99}Tc for varying sample volumes and tracer ^{99}Tc activity. Solid line: 5% or more, dashed line: 1%

The extent of this interference for a series of real samples indicates that ^{99}Tc contamination from the eluate can result in overestimation of activity in low level samples by factors of up to 30%. Some relation was observed between the age of the generator and the proportion of the total beta activity in the tracer solution that was constituted by the ^{99}Tc . Assuming that the excess beta activity was due to ^{103}Ru , this fact appears to be consistent with the decay of ^{103}Ru in the generator.

The presence of ^{99}Tc in the eluates is a significant interferent in the analysis of low level samples. A demonstration of this is provided in Table 6. As can be seen, levels of ^{99}Tc from the tracer solution can constitute up to 30% of the actual result in some cases. Correction for the interferent ^{99}Tc is possible but is not as straightforward as first seems. Simply depositing known masses of the tracer on a planchette to determine levels of ^{99}Tc in the tracer is not practicable, due to the presence of ^{103}Ru , as is trying to predict any amount of ^{99}Tc present in eluates using mathematical methods. Due to the variable nature of the extent of the interference from eluate to eluate, it is unwise to try and establish a correction factor for any single generator. The only practical solution to the problem is to run a "blank" sample with each batch of samples drawn from a single tracer solution.

Table 6. Extent of tracer ^{99}Tc interference for a set of low level seawater samples

Test Sample	^{99}Tc activity (Bq/m^3)	
	No correction	With correction
1	0.26 ± 0.03	0.25 ± 0.03
2	0.26 ± 0.03	0.23 ± 0.03
3	0.10 ± 0.01	0.08 ± 0.01
4	0.30 ± 0.03	0.24 ± 0.04
5	0.05 ± 0.01	< 0.05
6	0.28 ± 0.03	0.20 ± 0.04
7	0.12 ± 0.01	0.08 ± 0.02
8	0.31 ± 0.03	0.27 ± 0.04
9	0.10 ± 0.01	0.08 ± 0.01
10	0.30 ± 0.03	0.26 ± 0.04
11	0.29 ± 0.03	0.25 ± 0.04
12	0.18 ± 0.02	0.14 ± 0.03

5. Conclusions

5.1 Technetium-99 in the Norwegian Arctic marine environment

Discharges from Sellafield have led to enhanced levels of ^{99}Tc in the Norwegian Arctic marine environment. Concentrations of ^{99}Tc in the Svalbard area in 2001 were in the range of 0.13 – 0.36 Bq/m^3 , a five fold increase on pre-EARP levels in 1994. Average levels of ^{99}Tc were higher at the monitoring station on Bjørnøya (0.25 Bq/m^3) than at the Ny Ålesund Station (0.23 Bq/m^3) and Hopen (0.22 Bq/m^3). Levels in the Svalbard area for 2001 are up to one order of magnitude lower than levels reported in 2001 for the Norwegian coastal environment. Average levels from Jan Mayen (0.1 Bq/m^3) for 2001 were lower than the Svalbard area and may reflect residual levels in Arctic Water from earlier discharges.

The transfer and gradient in ^{99}Tc concentrations between the Norwegian coastal environment and the Norwegian Arctic is a reflection of the dominant oceanic circulatory processes in the Northern seas. The similarity between levels of ^{99}Tc in the West Spitsbergen Current and within fjords on the West of Svalbard, underlines the influence and importance of this current on the Svalbard marine environment.

The relationship between ^{99}Tc concentrations and salinity, may lead to unrepresentative ^{99}Tc levels, in situations where the salinity of a seawater sample is below the normal range for that sampling location.

Levels of ^{99}Tc in 2001 were generally higher in marine algae from Bjørnøya and Hopen than Kongsfjorden. Irrespective of location, higher levels of ^{99}Tc were

generally observed in furoid seaweeds in the Svalbard area than in kelp seaweeds. Levels of ^{99}Tc in furoid seaweeds from the Svalbard area from 2000 and 2001 were 7-8 fold lower than reported average levels for the same period in furoid seaweeds from the Norwegian coast. Concentration factors of ^{99}Tc in marine algae from the Svalbard area and mainland Norway are in good overall agreement. Levels of ^{99}Tc in marine algae in the Svalbard area reflect the lower in situ ^{99}Tc seawater concentrations as compared to the Norwegian coastal environment.

5.2 Measurement of ^{99}Tc in the Arctic marine environment: Considerations

The low levels of ^{99}Tc in the Arctic marine environment necessitate the implementation of special precautions in the determination of the isotope by radiochemical means. Not least of these are those related to the use of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators for the provision of $^{99\text{m}}\text{Tc}$ as a radiochemical yield tracer. The presence of impurities in the eluates of such generators has been well described previously and generally the levels of such impurities are low enough to be of little concern in the analysis of ^{99}Tc in environmental matrices. For samples exhibiting ^{99}Tc levels of less than 1 Bq/m^3 or kg however, the presence of impurities can have significant impacts on the accuracy of the analytical result.

Many of the reported impurities associated with $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators are either short lived, in which case they are unlikely to be present at the time of use of the generator for provision of tracer solution or are removed from the sample during the radiochemical isolation of ^{99}Tc . Other impurities present are in such low quantities as to be of no significant consequence.

Of more concern is the presence of ^{99}Tc in the tracer solution. The amount of this isotope in the generator and the eluates is difficult to predict by mathematical methods and has been shown to be variable between both generators and elutions.

Acknowledgements

The authors wish to acknowledge the support of the Norwegian Polar Institute, Tromsø, the staff of the Sverdrup Station, Ny Ålesund, the staff of DNMI Bjørnøya, Hopen, Jan Mayen, the staff of FTD Jan Mayen, the Norwegian Coast Guard (Kystvakt) and the Governor of Svalbard.

References

- Aarkrog, A., Dahlgaard, H., Hallstadius, L., Holm, E., Mattsson, S. and Rioseco, J. 1986. Time trend of ^{99}Tc in seaweed from Greenland waters. pp.69-78. *In*: G.Desmet and C.Myttenaere (Eds.), 'Technetium in the Environment'. Elsevier, London and New York, 419 p.
- Anderson, T.J. and Walker, R.L. 1980. Determination of picogram amounts of technetium-99 by resin bead mass spectrometric isotope dilution. *Analytical Chemistry*, 52:709-713.
- Beasley, T.M. and Lorz, H.V., 1986. A review of the biological and geochemical behaviour of technetium in the marine environment. *Journal of Environmental Radioactivity*, 3:1-22.
- BNFL. 1982-1991. BNFL Annual Reports on radioactive discharges and monitoring of the environment. British Nuclear Fuels Limited, Risley, UK. 1982-1991.
- Bondietti, E.A. and Francis, C.W. 1979. Geological migration potential of technetium-99 and neptunium-237. *Science*, N.Y., 203:1337-1340.
- Brown, J., Kolstad, A.K., Lind, B., Rudjord, A.L. and Strand, P. 1998. Technetium-99 contamination in the North Sea and Norwegian coastal areas 1996 and 1997. NRPA Report 1998:3, Østerås, Norwegian Radiation Protection Authority. 21 p.
- Brown, J., Iospje, M., Kolstad, A.K., Lind, B., Rudjord, A.L. and Strand, P. 2002. Temporal trends for ^{99}Tc in Norwegian coastal environments and spatial distribution in the Barents Sea. *Journal of Environmental Radioactivity*, 60:49-60.
- Busby, R., McCartney, M. and McDonald, P. 1997. Technetium-99 concentration factors in Cumbrian seafood. *Radioprotection-Colloques*, 32:311-316.
- Cataldo, D.A., Garland, T.R., Wildung, R.E. and Fellows, R.J. 1989. Comparative metabolic behaviour and inter-relationships of Tc and S in soybean plants. *Health Physics*. 57:281-288.
- Chen, Q., Aarkrog, A., Nielsen, P., Dahlgaard, H., Lind, B., Kolstad, A.K. and Yu, Y. 2001. Procedures for determination of $^{239,240}\text{Pu}$, ^{241}Am , ^{237}Np , $^{234,238}\text{U}$, $^{228,230,232}\text{Th}$, ^{99}Tc , ^{210}Pb and ^{210}Po in Environmental Materials. Riso National Laboratory, Roskilde, Denmark. Riso-R-1263 (EN).
- Clarke, M.J. and Fackler, P.H. 1982. Topics in Inorganic Physical Chemistry, 50. 57 p.
- Dahlgaard, H. 1995. Transfer of European coastal pollution to the Arctic: Radioactive tracers. *Marine Pollution Bulletin*, 31:353-358.
- Dahlgaard, H., Chen, Q., Herrmann, J., Nies, H., Ibbett, R.D. and Kershaw, P.J. 1995. On the background level of ^{99}Tc , ^{90}Sr and ^{137}Cs in the North Atlantic. *Journal of Marine Systems*, 6:571-578.

- Harvey, B.R., Ibbett, R.D., Williams, K.J. and Lovett, M.B. 1991. The determination of technetium-99 in environmental materials. Aquatic Environment Protection: Analytical Methods Number 8. MAFF, Directorate of Fisheries Research, Lowestoft, UK. 23 p.
- Herrmann J, Kershaw PJ, Dubois PB & 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, pp.427-456,
- Holland, M.E., Deutsch, E. and Heineman, W.R. 1986. Studies on commercially available $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ radionuclide generators – II. Operating characteristics and behaviour of $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators. *Applied Radiation and Isotopes*, 37(2):173-180.
- Kaye, J.H., Merrill, J.A., Kinnison, R.-R., Rapids, M.S. and Ballou, N.E. 1982. Radiochemical determination of technetium-99. *Analytical Chemistry*, 54:1158-1163.
- Kenna, B.T. and Kuroda, P.K. 1964. Technetium in nature. *Journal of Inorganic Nuclear Chemistry*, 2:493-499.
- Kershaw, P.J., McCubbin, D. and Leonard, K.S. 1999. Continuing contamination of north Atlantic and Arctic waters by Sellafield radionuclides. *Sci. Total Environment* 237/238:119-132.
- Kolstad, A.K. and Lind, B. 2002. Radioactivity in the marine environment 2000 and 2001, Technetium-99 concentrations in Norwegian coastal waters and biota. NRPA report 2002:6. Østerås: Norwegian Radiation Protection Authority, 19 p.
- Leonard, K.S., McCubbin, D., Brown, J., Bonfield, R. and Brookes, T. 1997a. Distribution of technetium-99 in UK coastal waters. *Marine Pollution Bulletin*, 37:628-636.
- Leonard, K.S., McCubbin, D., Brown, J., Bonfield, R. and Brookes, T. 1997b. A summary report of the distributions of ^{99}Tc in UK coastal waters. *Radioprotection-Colloques*, 32:109-114.
- Long, S.E. and Sparkes, S.T. 1988. A literature survey of techniques for the analytical determination of technetium-99. UKAEA, Harwell, Oxon., UK, AERE Report R 12742, 33 p.
- Perrier, C. and Segre, E. 1947. Technetium: the element of atomic number 43. *Nature*, London, 159:24.
- Rudjord, A.L., Føyn, L., Brungot, A.L., Kolstad, A.K., Helldal, H.E., Brown, J., Iospje, M. and Christensen, G. 2001. Radioactivity in the marine environment (RAME) 1999. NRPA Report 2001:9 Østerås: Norwegian Radiation Protection Authority, 39 p.
- Siegel, J.A. and Deutch, E. 1975. Annual Reports in Inorganic and General Synthesis, 311 p.
- Sodd, V.J. and Fortman, D.L. 1976. Analysis of the ^{89}Sr and ^{90}Sr content in eluates of fission produced $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators. *Health Physics*. 30:179-182.
- Vesely, P and Cifka, J. 1970. Some chemical and technical problems connected with technetium-99m generators. Nuclear Research Institute, Pez, Czechoslovakia. UJC 2414-Ch. 21 p.

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ISSN 0804-4910