TECHNETIUM (^{99}Tc) AS A TRANSIENT TRACER OF CIRCULATION FROM UK WATERS TO THE NORDIC SEAS

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Introduction

Technetium-99 (^{99}Tc) is a man-made radionuclide, a fission product with a long half-life ($t_{1/2} = 2.1 \times 10^5$ years), occurring as the highly soluble pertechnetate form in oxic seawater. It is ubiquitous in surface waters as a result of global fallout from nuclear weapons testing, giving a background concentration of about 5 mBq m^{-3} (Dahlgaard *et al.*, 1995; 1 Bq (becquerel) = 1 disintegration s^{-1}). Wastes arising from nuclear fuel reprocessing in NW Europe (Sellafield, UK; La Hague, near Cherbourg, France) have provided a substantial addition of this tracer to the NE Atlantic. Earlier studies have shown the transport of ^{99}Tc from the Irish Sea and English Sea into the Nordic Seas via the North Sea and the Norwegian Coastal Current (NwCC), and a return flow in the East Greenland Current (EGC) (Dahlgaard, 1995).

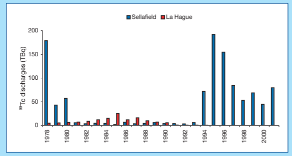
Throughout the 1980s ^{99}Tc discharges were dominated by La Hague, and several integrated tracer and modelling studies were completed in the Channel and southern North Sea (e.g. Salomon *et al*, 1995).

In March 1994 a new waste treatment plant came into operation at Sellafield, the Enhanced Actinide Removal Plant (EARP). This was designed to remove actinides (e.g. plutonium) and particulate activity but not ^{99}Tc. As a result of treating the back-log of medium-level stored waste, the discharge of ^{99}Tc increased substantially. There was a consequent increase in the inventory of ^{99}Tc in the Irish Sea, and a rapid transport of this EARP-related tracer via the Scottish Coastal Current (SCC) into the North Sea (Leonard *et al*, 1997). This presented a renewed opportunity to use this transient tracer to examine transport pathways and transport times from UK waters to the Arctic.

The discharges peaked in 1995 and decreased thereafter, partly in response to international pressure. Although the radiological impact is rather low there were concerns about the potential socio-economic impact on Norwegian fisheries (Defra, 2002).

Estimated seawater inventory (TBq) of ^{99}Tc in the Irish Sea (52.2°-55.1°N; 2.5°-6.5°W; from McCubbin et al, 2002)

Survey date	^{99}Tc inventory (TBq)
December 1992	6
December 1993	7
May 1994	34
December 1994	38
December 1995	94
December 1996	166
September 1998	40



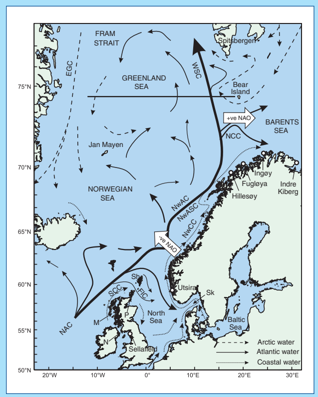
Discharges of ^{99}Tc from BNF-Sellafield and COGEMA-La Hague 1978-2001

Observations

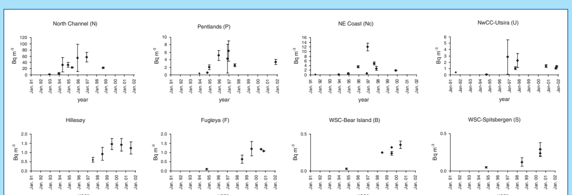
The EARP releases stimulated a number of new observing programmes (seawater and *Fucoid* seaweeds), particularly in the UK and Norway (e.g. Brown *et al.*, 2002; Kolstad & Lind, 2002). These observations enabled the 'leading edge' of the EARP-related ^{99}Tc to be followed. This 'post-EARP' signal (defined here as a factor of 2 increase in the pre-EARP 'background' concentration) progressed through the Irish Sea and exited via the North Channel to be advected north in the SCC. The initial transport was much more rapid than anticipated, with the signal reaching the North Channel within 3 months and the Pentlands within 9 months (Leonard *et al.*, 1997). The ^{99}Tc signal had reached Norwegian waters within 3 years (at Utsira, *Fucus* measurements) and was rapidly transported in the NwCC, reaching Ingøy on the north coast by August 1997, implying a mean advection of 0.07 m s^{-1} in the NwCC, for the first half of 1997, which is in good agreement with one other study, based on sea surface temperature anomalies.

The precision of the transport estimates is dependent of the spatial and temporal frequency of sampling. In most cases the estimates will be rather conservative. The leading edge of the EARP-related ^{99}Tc reached Arctic waters between 1998 and 1999. We were not able to follow this further east in the Barents Sea or north within the Fram Strait after 1998 due to the geographical limit of sampling. In July 1998, EARP-related ^{99}Tc was apparent southwest of Bear Island (~ 73° N) (^{99}Tc ~ 6 x background levels) and in one of three samples collected west of Spitsbergen at ~ 77° N (^{99}Tc ~2 x background). However, by May/June 2000 the leading edge had passed west of Spitsbergen (^{99}Tc ~ 6 x background), with concentrations about an order of magnitude higher than background levels in the northern section of the NwCC. Transport times for ^{99}Tc from Sellafield to west-southwest of Bear Island and west of Spitsbergen have been estimated to be 3 to 4 and 4 to 6 years, respectively.

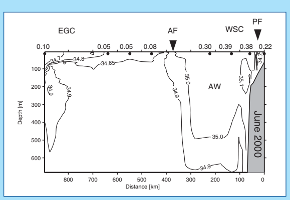
The initial releases in March 1994 were preceded by a period of intense storms, characteristic of a high +ve winter NAO index (North Atlantic Oscillation) with high volume transport estimated northwards through the North Channel (Young *et al.*, 2001). We anticipate the volume transport in the SCC would have been similarly affected in the winter of 1993/94 and the following winter 1994/95, when the NAO reached a record high +ve value.



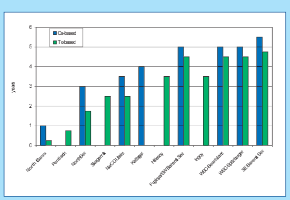
Circulation of surface waters of the North, Norwegian, Greenland and Barents Seas: East Greenland Current (EGC), Fair Isle Current (FIC), North Atlantic Current (NAC), North Cape Current (NCC), Norwegian Atlantic Current (NwAC), Norwegian Atlantic Slope Current (NwASC), Norwegian Coastal Current (NwCC), Scottish Coastal Current (SCC) and West Spitsbergen Current (WSC). The Bear Island West section together with referred locations are also indicated. The broad arrow labelled '–ve NAO' represents the increasing westward extent of the NwAC and NwASC reported during negative phases of the NAO. The broad arrow labelled '+ve NAO' represents the increased influx of AW into the Barents Sea reported during positive phases of the NAO (from: Kershaw et al., in press).



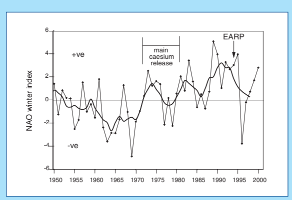
Time-series of ^{99}Tc concentrations (Bq m^{-3}) in surface waters at Hillesøy and in a set of demarcated regions, showing the progress of the EARP-related ^{99}Tc. Error bars represent the std. dev. of the variability in observed concentrations, taken from the following sources: Leonard et al., 1997; McCubbin et al., 2002; Hermann et al., 1995; Brown et al., 1998; Kershaw et al, 1997; Kolstad & Lind, 2002; G Christensen pers comm.; NRPA, 2001; Kershaw et al., in press)



Section west from Bear Island along 74° 30' N, showing salinity and surface ^{99}Tc concentrations (Bq m^{-3}), and the location of the EGC, WSC, Polar Front (PF) and Arctic Front (AF) in May/June, 2000. The pre-EARP concentrations in AW at this latitude in 1994 were <0.1 Bq m^{-3} (from: Kershaw et al., in press).



Transit times from Sellafield, based on observed environmental concentrations, consistently indicate more rapid transit of EARP-related ^{99}Tc compared with the main discharge of ^{137}Cs in the mid- to late-1970s. The pattern is particularly marked for the North Channel, central North Sea and Utsira.

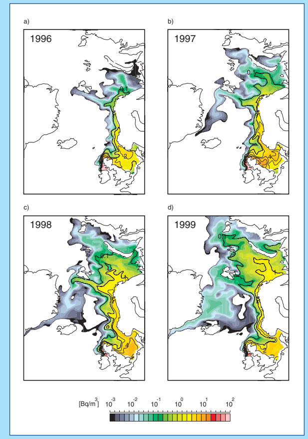


The initial EARP-related ^{99}Tc releases coincided with 2 winters exhibiting high +ve NAO indices, when transport through the North Channel, SCC and NwCC would be expected to be enhanced. This contrasts with a period of relatively lower indices in the mid- to late-1970s when the main main ^{137}Cs releases occurred.

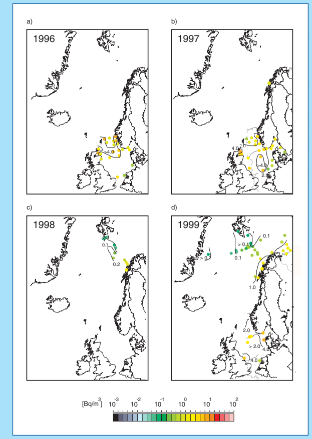
Modelling

The dispersion of ^{99}Tc was simulated using the 3D NAOSIM model (North Atlantic-Arctic Ocean Ice Model) developed at the Alfred Wegener Institute for Polar and Marine Research (Karcher *et al.*, in press). The model was driven with realistic daily atmospheric forcing from 1979 to 1999, using data supplied by the European Centre for Medium-Range Weather Forecasts (ECMWF). The release of ^{99}Tc was initiated in 1990, following a simulation of the period 1979-1989, into the uppermost model layer (20m depth) in the North Channel (the Irish Sea is not resolved sufficiently to simulate the release from Sellafield directly).

The model reproduced the overall pattern of artificial radionuclide distributions reported previously, but provided additional insight into temporal and spatial variability. For example, the surface velocity fields showed significant seasonal and interannual variability. Both the simulation and observations indicate increase concentrations in the summer of 1997 at Hillesøy. The model results suggest that the transit time from Sellafield to the Barents Sea of the initial EARP-related ^{99}Tc was approximately 3.5 years, significantly shorter than that estimated for ^{137}Cs (4-5 years). We suggest that a critical factor was the variation in the inflow of Atlantic Water across the Faroe-Shetland Gap, with a more intense inflow in 1994/95 compared to the mid-1970s when the peak releases of ^{137}Cs occurred.



Distribution of ^{99}Tc (Bq m^{-3}) in surface water in September (a) 1996, (b) 1997, (c) 1998 and (d) 1999, from model results (lines 0.1, 1.0, 2.0 and every 2.0 Bq m^{-3}).



Distribution of ^{99}Tc (Bq m^{-3}) in surface water in (a) 1996, (b) 1997, (c) 1998 and (d) 1999, from observations.

Conclusions

- The increased discharge of ^{99}Tc from BNFL Sellafield has provided a renewed opportunity to investigate the relationships between current and previous distributions of radioactive tracers, annual/decadal changes in the circulation of the North Atlantic and adjoining coastal waters, and climate indicators (i.e. NAO).
- These observations have provided an additional dataset with which a new generation of coupled ocean-ice models can be validated (NAOSIM).
- The propagation speed of the leading edge of the EARP-related ^{99}Tc signal appears to be linked to fluctuations in the NAO. A high positive winter index and increased westerly winds during the 1993/94 and 1994/1995 winters caused a larger inflow of AW across the Faroe-Shetland Gap, increased transport from the Irish Sea to the North Sea and led to more rapid transport of ^{99}Tc to Norwegian waters. The estimated transport times were significantly shorter, in this period, than those based on ^{137}Cs transport in the mid- to late-1970s.
- A combination of the pronounced –ve NAO winter index in 1995/96 and lower positive NAO winter indexes in the following years appears to have led to a slower propagation rate for ^{99}Tc along the NwCC and into Arctic waters, and is likely to have resulted in the increased westward excursion of the ^{99}Tc signal in the NwCC and NwASC.

Acknowledgements

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