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

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Introduction

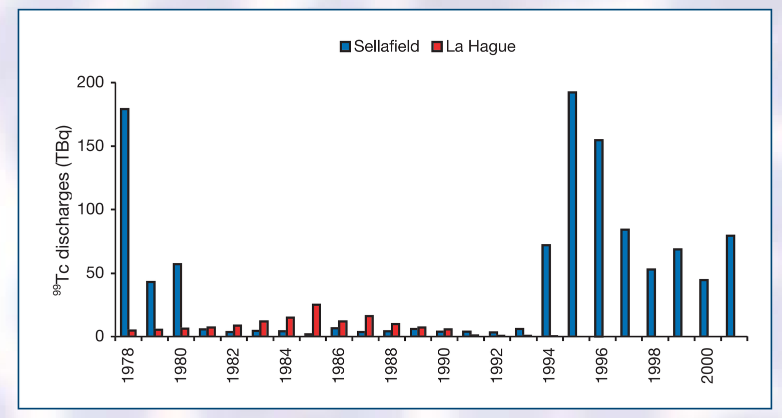
Technetium-99 (^{99}Tc) is a man-made radionuclide, a fission product with a long half-life ($t½ = 2.1x10^5$ years), occurring as the highly soluble pertechnetate form in oxic seawater. It is ubiquitous in surface waters as a result of global fallout from nuclear weapons testing, giving a background 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).

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



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

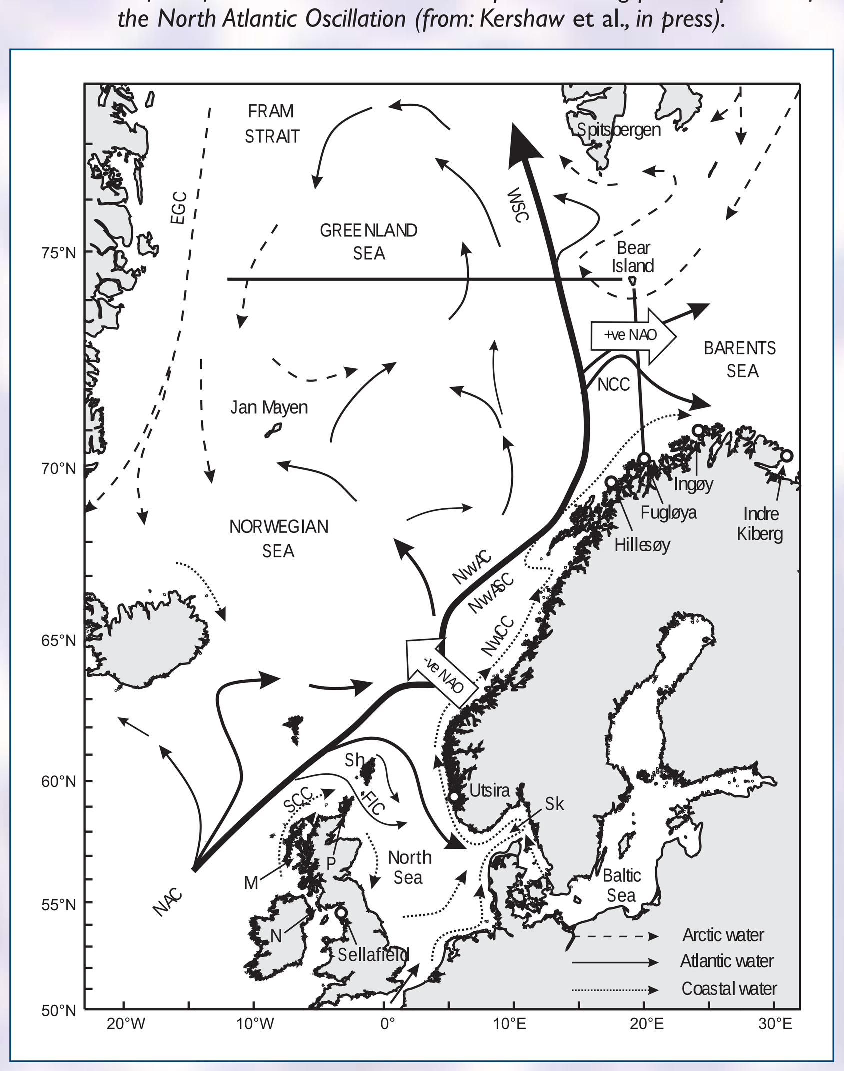
Observations

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 Scottish Coastal Current (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 Norwegian Coastal Current (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.

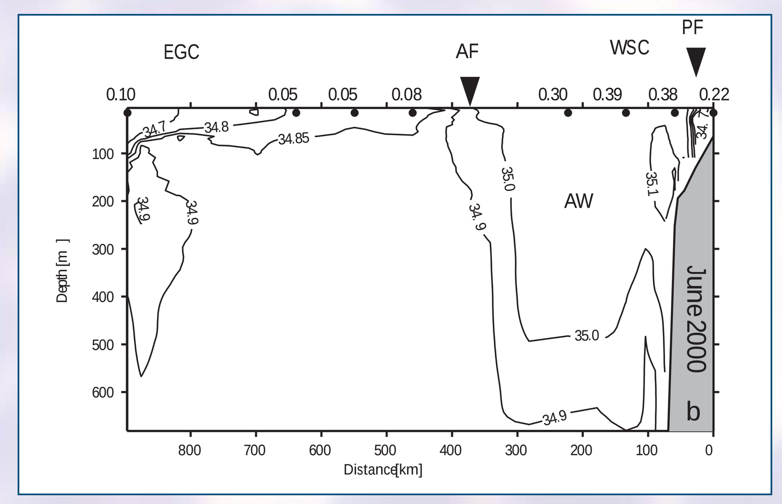
The leading edge of the EARP-related ^{99}Tc reached Arctic waters between 1998 and 1999. We were not able to define follow this further east in the Barents Sea or north within the Fram Strait after 1998 due to the geographical limit of sampling. In July 1998, EARP-related ^{99}Tc was apparent southwest 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 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.

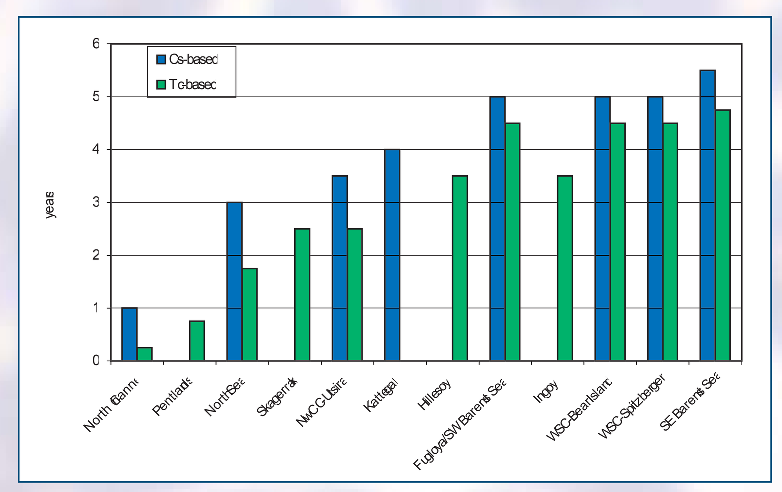
Circulation of surface waters of the North, Norwegian, Greenland and Barents Seas: East Greenland Current (EGC), Fair Isle Current (FIC), North Atlantic Current (NAC), North Cape Current (NCC), Norwegian Atlantic Current (NwAC), Norwegian Atlantic Slope Current (NwASC), Norwegian Coastal Current (NwCC), Scottish Coastal Current (SCC) and West Spitsbergen Current (WSC). The Fugløya - Bear Island and Bear Island West sections 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 North Atlantic Oscillation. The broad arrow labelled '+ve NAO' represents the increased influx of AW into the Barents Sea reported during positive phases of the North Atlantic Oscillation (from: Kershaw et al., in press).



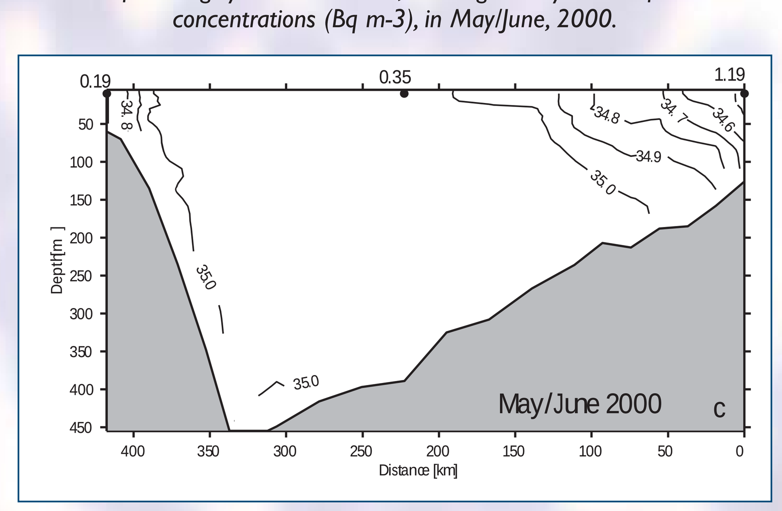
Section west from Bear Island along 74° 30' N, showing salinity and surface ^{99}Tc concentrations (Bq m^{-3}), and the location of the EGC, WSC, Polar Front (PF) and Arctic Front (AF) in May/June, 2000. The pre-EARP concentrations in AW at this latitude in 1994 were <0.1 Bq m^{-3} (from: Kershaw et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



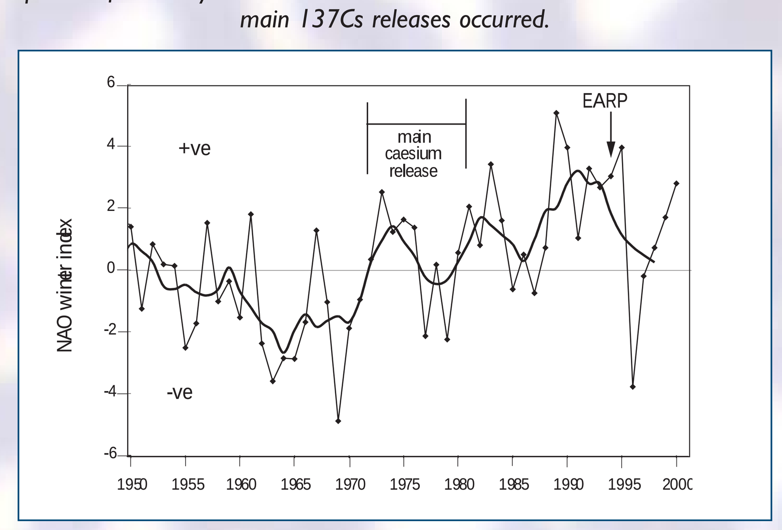
Transit times from Sellafield, based on observed environmental concentrations, consistently indicate more rapid transit of EARP-related ^{99}Tc compared with the main discharge of ^{137}Cs in the mid- to late-1970s. The pattern is particularly marked for the North Channel, central North Sea and Utsira.



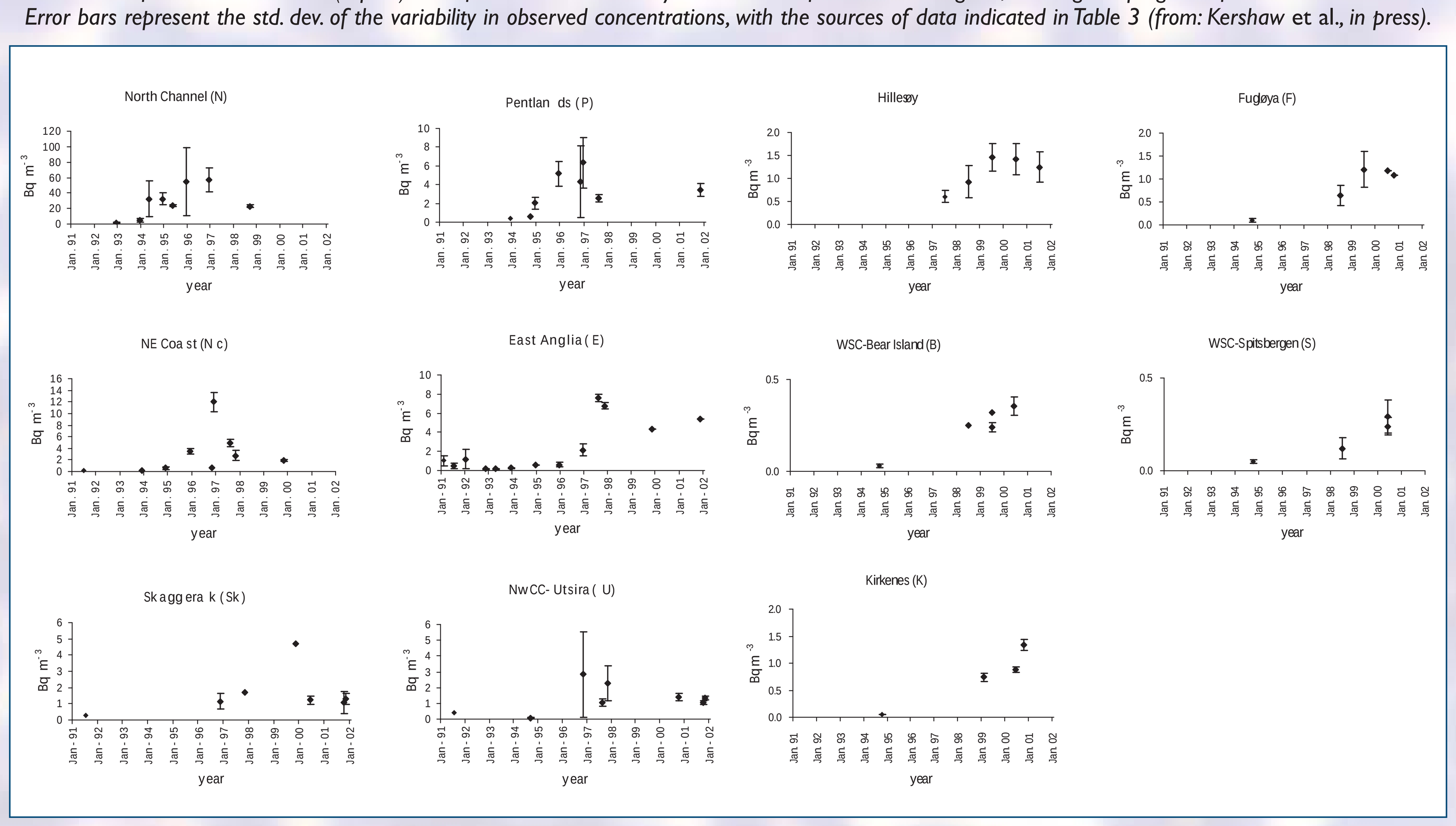
Section from Fugløya to Bear Island, showing salinity and surface 99Tc concentrations (Bq m-3), in May/June, 2000.



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 137Cs releases occurred.



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, with the sources of data indicated in Table 3 (from: Kershaw et al., in press).



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).

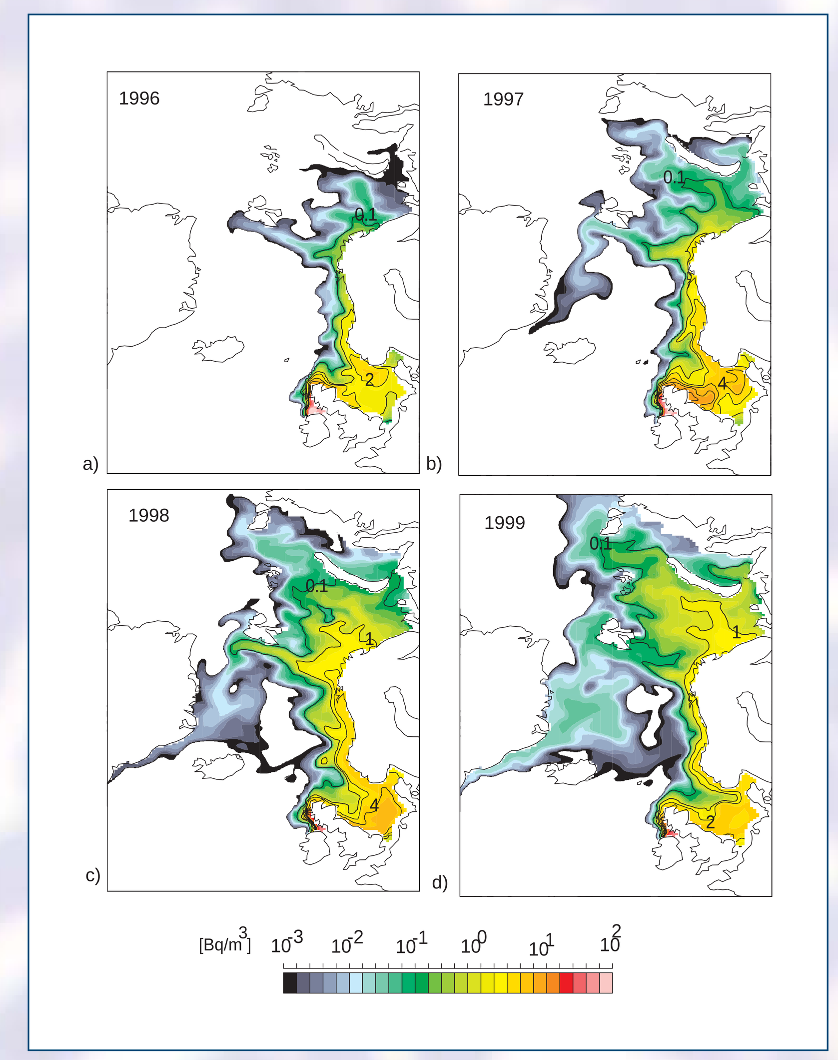
The model reproduced the overall pattern of artificial radionuclide distributions reported previously, but provided additional insight into temporal and spatial variability. For example, the surface velocity fields showed significant seasonal and interannual variability. The model results suggest that the transit time from Sellafield to the Barents Sea of the initial EARP-related ^{99}Tc was approximately 2.5-3.5 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.

First observed occurrence of EARP-related ^{99}Tc in a series of seawater boxes and implied transport times

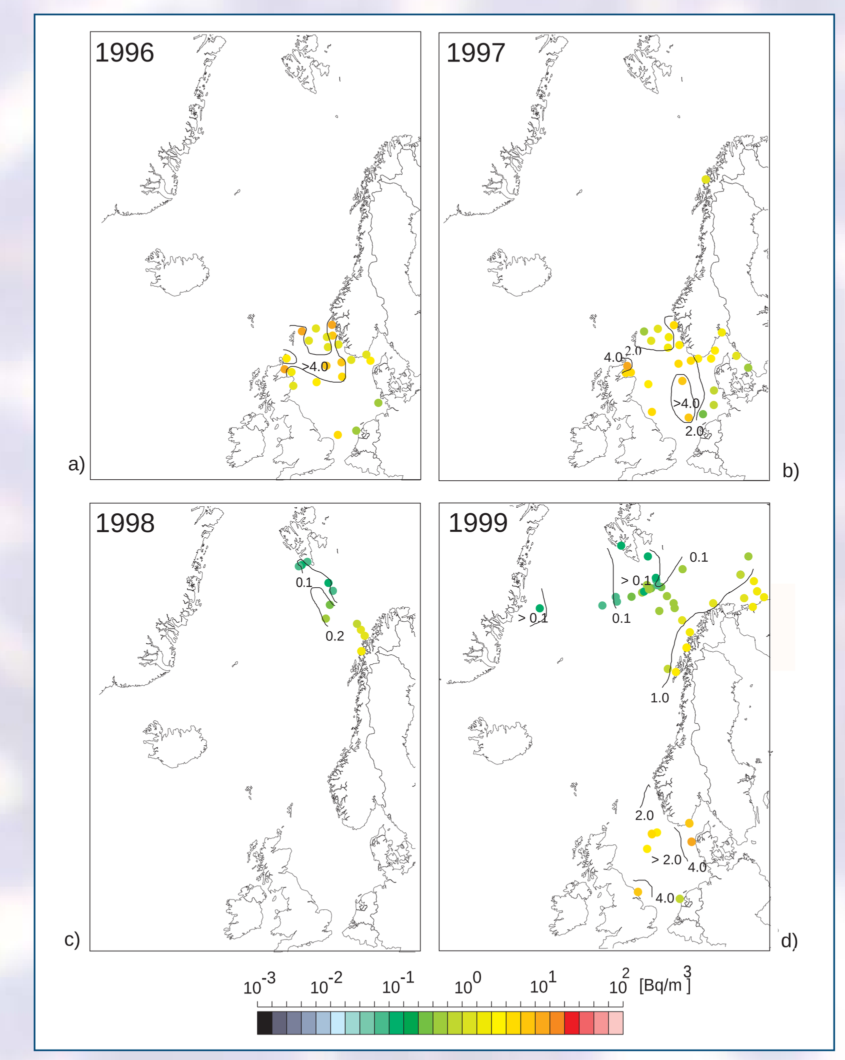
Box	EARP signal observed	Approximate Transport time	References
North Channel	May 1994	≤ 3 months	1,2
Pentlands	December 1994	< 9 months	1,2
NE Coast	December 1994	≤ 9 months	1,2
East Anglia	December 1996	≤ 2.5 years	1,2,3
NwCC-Utsira	November 1996	≤ 2.5 years	4,5
Skagerrak	November 1996	≤ 2.5 years	4
Hillesøy	July 1997	≤ 3.5 years	6
Ingøy – shoreline *Fucus*	August 1997	≤ 3.5 years	7
Fugløya	July 1998	≤ 4.5 years	8,9
WSC-Bear Island	July 1998	≤ 4.5 years	8,9
WSC-Spitsbergen	July 1998	≤ 4.5 years	8,9

References: 1 Leonard *et al.*, 1997; 2. McCubbin *et al.*, 2002; 3. Herrmann *et al.*, 1995; 4. Brown *et al.*, 1998; 5. Kershaw *et al.*, 1997; 6. Kolstad & Lind, 2002; 7. G Christensen pers comm.; 8. NRPA, 2001; 9. Kershaw *et al.*, in press

Distribution of ^{99}Tc (Bq m^{-3}) in surface water in (a) 1996, (b) 1997, (c) 1998 and (d) 1999, from observations (same colour code as Fig. 2, isolines in Bq m^{-3}).



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}).



Conclusion

We conclude that the leading edge of the EARP-related ^{99}Tc, identified as a factor of 2 increase over the 1994 'background', reached Arctic waters between 1998 and 1999. We have not been able to define the leading edge of the EARP-^{99}Tc 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 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. Transfer factors, relating environmental concentration with discharge rate, are shown to be unreliable when dealing with a rapidly-changing input.

The propagation speed of the leading edge of the EARP-^{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 from the Irish Sea to the North Sea and more rapid transport of ^{99}Tc. In contrast to the rapid transport of ^{99}Tc from the Irish Sea into the North Sea, the further transport along the NwCC and beyond was in this study found to have slowed down. A combination of the pronounced –ve NAO winter index in 1995/96 and lower positive NAO winter indexes in the following years 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. We estimated a mean propagation speed for the leading edge along the Norwegian coast of 0.07 m s^{-1}, for the first half of 1997, which is in good agreement with one other study, based on sea surface temperature anomalies. The propagation of the leading edge of a rapidly increasing tracer signal will more readily respond to intra- and inter-annual variability due to atmospheric forcing and the presence of preferred pathways, such as the inner branches of the SCC and NwCC. In contrast, mixing with AW along the boundary with the SCC and NwCC, including the formation of meso-scale eddies, leads to a lag in the propagation rate of a portion of the tracer signal, thus increasing the mean transport time.

The existence of EARP-related ^{99}Tc has provided an opportunity to investigate the relationships between climate indicators such as the North Atlantic Oscillation (NAO), the current and previous distributions of radioactive tracers, and annual/decadal changes in the circulation of the North Atlantic and adjoining coastal waters, and an additional dataset with which a new generation of coupled ocean-ice models can validated.

Acknowledgements

We would like to thank the crew and cruise participants onboard RV 'Cirolana', RV 'Johan Hjort', RV 'Michael Sars' and RV 'Polarstern' and our colleagues at our home institutes for support and encouragement. This work was funded partly or jointly by Defra (Department of Environment, Food & Rural Affairs, UK), the Norwegian Research Council, NRPA (Norwegian Radiation Protection Authority), the EU (FP5-REMOTRANS: Processes controlling the remobilisation and relocation of radionuclides in the marine environment) and the Alfred Wegener Institute.

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