THE TRANSFER OF ^{99}TC FROM SELLAFIELD TO THE ARCTIC

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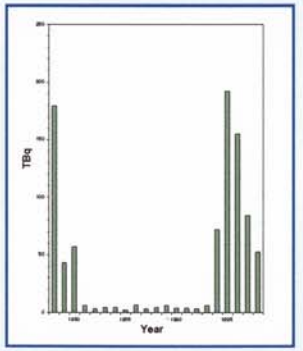
Background

The authorised release of low-level radioactive waste, from European reprocessing facilities, has provided the oceanographic community with a variety of tracers for use in long-distance transport studies. Substantial increases in the discharge of ^{99}Tc occurred in the mid-1990's, from the Sellafield nuclear fuel reprocessing plant in the UK, against the overall trend of most other radionuclides. This was due to the commissioning of the Enhanced Actinide Removal Plant (EARP), which allowed the treatment of medium-level stored liquors containing actinides but which was not designed to remove technetium. ^{99}Tc is long-lived radionuclide (half-life 2.1*10^5 years) which normally is considered to behave conservatively in seawater and has been used as an oceanographic tracer in the North Sea and beyond. It is readily taken up by the brown seaweed *Fucus* (concentration factor ~ 1*10^5) which both concentrates and time-integrates the water signal, over the life-span of the plant (2-4 years). The 'pulsed' release of ^{99}Tc in 1994-1998 has provided a new opportunity to study transport pathways and transit times from the Irish Sea to other parts of the NE Atlantic and Arctic Oceans.

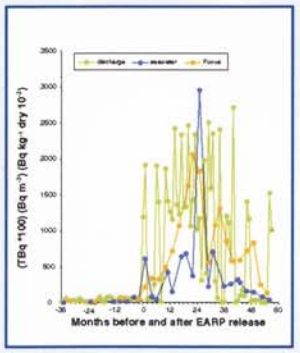


Sellafield Discharge of ^{99}Tc

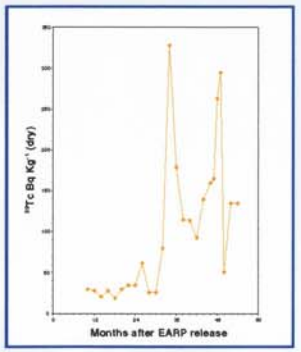
Distribution of ^{99}Tc in F. vesiculosus

The concentration of ^{99}Tc in *F. vesiculosus* within the Irish Sea responded rapidily to the increased discharge (within 1 month at St Bees, 15 km north of the outfall) and continued to accumulate despite fluctuations in the discharge. The quarterly sampling frequency revealed a striking periodicity at St Bees, with lower concentrations in the spring and summer due to rapid growth and dilution. This was also apparent at Utsira, a small island off SW Norway, within the Norwegian Coastal Current (NCC). The passage of the EARP-related 'pulse', as recorded by *Fucus* analysis, could be traced from the Irish Sea to the north coast of Norway. It had reached Ingøy by the summer of 1997, ~ 3 years after the release commenced. Transport along the NCC appears to have been rapid during the winter of 1996/97, on the basis of the concentration gradients observed in *Fucus* along the Norwegian coast. New growth has resulted in the progressive decrease in overall concentrations in *Fucus* at St Bees, in response to the lower discharge rates since 1995. This pattern should be repeated at more distant sites in the coming years.

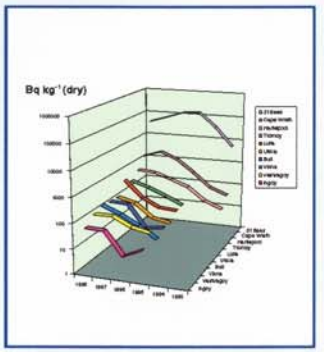
Fucus vesiculosus *sampling locations*



*Variation in discharge (TBq *100), seawater (Bq m^{-3}) and* Fucus *(Bq kg^{-1} dry 10^{-3}) concentrations at St Bees*



Utsira (F. versiculosus)



Changes in ^{99}Tc with location and time (F. vesiculosus)

Distribution of ^{99}Tc in seawater

The distribution of ^{99}Tc has been mapped from a series of RV cruises, extending from the Irish Sea to Fram Strait and the Barents Sea. The initial near-field distribution was influenced by the episodic nature of the releases and the timing of the observations. The leading edge of the first 'pulse' (March/April 1994) had been advected out of the Irish Sea, north via the North Channel, within 3 months and within 9 months had reached the North Sea via the Pentland Firth. These transit times were significantly shorter than rates reported previously using other radiotracers, principally radiocaesium. This may be due a real change in transport rates in Scottish coastal waters since the 1970s (when many of the earlier studies were carried out). This is not inconceivable given the fluctuations in the NAO over the same period. However, the observed 1990's rates refer to the leading edge of a distinct plume, rather than a time-integrated signal. Further time-series analysis is required to resolve this uncertainty. A survey in December 1996 revealed an area of higher concentrations (> 10 Bq m^{-3}) in surface waters, off the Scottish east coast, corresponding to the main 'pulsed' discharge in 1995. By July 1997 concentrations in the NCC (58° 45' - 62° N) had increased to 0.9-1.2 Bq m^{-3}, a factor of 10 higher than 3 years earlier. The leading edge of the plume had reached latitude 73° 30' by July 1998, in the West Spitzbergen Current, with concentrations also a factor of 10 higher than in the previous survey in 1994.

Levels of ^{99}Tc (Bq m^{-3}) in surface water July 1998

Levels of ^{99}Tc (Bq m^{-3}) in surface water December 1996

Future

The magnitude of the observed increase in ^{99}Tc concentrations, in both seawater and *F. vesiculosus*, in response to the 1994-1998 EARP-related release, suggests that this long-lived tracer has considerable potential for use in circulation studies in the Nordic Seas and the Arctic Basin. Conventional analytical methods (based on chemical separation and beta counting) require up to 200 litre water samples. Current developments in Accelerator Mass Spectrometry (AMS) will permit ^{99}Tc determination on samples as small as 1-5 litres. This will allow greater flexibility in sample acquisition e.g. from 'ships-of-opportunity'. *Fucus* carries out its own pre-concentration step and only a few grams of dried sample are needed. *Fucus* species are found extensively in the Arctic and North Atlantic. One step to overcome may be access to intertidal sites in areas of sea-ice formation. The combination of ^{99}Tc measurements with other radiotracers, especially ^{129}I (AMS) and ^{137}Cs, offers a particularly powerful oceanographic tracer tool.

Further reading

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3. Kershaw, P.J., A.J., Baxter, 1995. The transfer of reprocessing wastes from north-west Europe to the Arctic. Deep Sea Res., 42, 1413-1448.
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