Contrasting behaviour of artificial radionuclides in the Pacific and other ocean basins: radionuclides as tracers of environmental change?

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Introduction

Ocean basins have received artificial radionuclides from a number of sources, including: global fallout from weapons testing, close-in fallout (e.g. Marshall Islands, Mururoa Atoll, Novaya Zemlya), dumped wastes (e.g. Sea of Japan, NE Atlantic, Kara Sea), accidental losses (e.g. SNAP-9A satellite, nuclear-powered vessels, nuclear weapons) and discharges into coastal regions (e.g. nuclear reprocessing facilities at Sellafield (UK) and La Hague (France)). In some cases the sources are well defined and in others it is more a case of estimating the size of the source from environmental measurements. These phenomena have provided a variety of tracers which can be used to describe the behaviour of artificial radionuclides in the water column and provide information on the underlying physics, biology and chemistry controlling their re-distribution.

This provides an opportunity to enquire whether artificial radiotracers can have a role in describing and quantifying the extent of environmental variability and change. In order to do this it is necessary to pose 2 fundamental questions:

What can observations of artificial radionuclide distributions tell us about ocean processes?

What can this information about observations and processes tell us about climate variability and environmental change?

To answer the first question we need to be able to clearly define the source terms and make adequate observations, both in space and time, of the radionuclide distributions. However, it is also essential to relate the observations to an adequate understanding of the underlying physics, biology and chemistry of the environment which are moderating the radionuclide behaviour. Some examples are presented below, from the Pacific and other ocean basins, to illustrate this point. To answer the second question we have to be able to define the variability, in space and time, of the observations and processes and link this to some index of environmental/climate variability. One example is presented, from the North Atlantic/ Arctic, where this approach is being attempted, comparing the distribution of mainly Sellafield-derived radionuclides with fluctuations in the North Atlantic Oscillation (NAO) as the index of environmental variability.

Radiotracers and ocean processes

Some datasets allow direct comparisons to be made between radionuclide distributions in different ocean basins. For example, Bourlat *et al.* (1996) presented ^{137}Cs, ^{90}Sr and ^{239,240}Pu concentrations in surface waters from the Pacific, Atlantic and Indian Oceans, from samples collected in 1992-1994. ^{137}Cs and ^{90}Sr both showed a latitudinal dependence but, in addition, concentrations in the Pacific were consistently higher (by ~ 0.5 Bq.m^{-3}) between 50° N and 30° S. The ^{137}Cs/^{90}Sr remained relatively constant (~ 1.7). The consistently higher surface concentrations in these Pacific latitudes most likely results from the residual effects of the additional close-in tropospheric testing from the U. S. Pacific weapons tests in the Marshall Islands. This difference was evident in the 1950's and 60's (Volchok *et al.*, 1971), in the 1970's (Livingston *et al.*, 1985) and still remains (IAEA, 1985). A decrease in ^{137}Cs concentrations in surface waters has also been observed in the Mediterranean Sea, with a consequent increase in deeper waters. In this case the transfer is intimately linked to convection and the seasonal formation of deep and intermediate water (Papucci *et al.*, 1996). The Mediterranean also received a significant additional input of ^{137}Cs as a consequence of the Chernobyl accident. The penetration of tritium to deeper layers in the Atlantic provided good evidence of the degree to which ventilation was taking place, following the contamination of surface waters by bomb-tritium and subsequent formation of intermediate and deep water in the Nordic Seas (Nyffeler *et al.*, 1996).

In oligotrophic regions, covering much of the world ocean, most of the plutonium has remained in the water column, unlike in coastal and shelf regions where there is significant removal on biological particles in highly productive shelf areas and through contact with higher shelf sediment particulates. The situation in the NW Pacific with respect to plutonium is less straightforward, both because of the more complex environmental behaviour of plutonium, compared with Cs and Sr, and because there are 2 significant sources. In addition to stratospheric global fallout (maximum input in the early 1960s) there was a significant contribution from tropospheric fallout, originating from the Marshall Islands proving grounds from tests conducted in the early 1950s (Bikini and Enewetak Atolls). Many of these tests were conducted at or near ground level. This source has a higher ^{240}Pu/^{239}Pu ratio (0.24 vs. 0.18) and appears to be more rapidly removed from surface waters (Buesseler, 1997). It has also contributed to an increase in ^{239,240}Pu concentrations in bottom waters, as well as the underlying sediments. These surface tests tended to produce relatively large calcium-rich particles, unlike the smaller iron-rich particles associated with stratospheric fallout. An overview of plutonium behaviour in the Pacific was recently presented by (Livingston *et al.*, 1999). They also concluded that tropospheric fallout had contributed to plutonium inventories in the NW Pacific. This study benefited from having access to the large GLO-

MARD database (Global Marine Radioactivity Database), under development at the IAEA-MEL (IAEA in press; www.iaea.org/monaco/glomard). Sediment and water column inventories, based on GLOMARD data, decreased with both latitude and longitude from the Marshall Islands, consistent with an additional localised source. In contrast, the French weapons programme conducted at Mururoa and Fangataufa did not produce a significant tropospheric footprint, beyond a few km from the atolls. This was as a result of the conditions under which the detonations took place (underground, under water, and at a height of several 100s m). The lagoons act as a local source of radioactivity but this is not significant on a regional scale.

Livingston *et al.* (1999) compared water column profiles of plutonium concentrations from common sampling locations visited in 1973 (GEOSECS), 1978, 1980, 1982 and 1997. Certain common features were revealed, in particular the persistence of sub-surface ^{239,240}Pu concentration maxima (Figure 1). However, profiles from the central NW Pacific showed a consistent trend, with the sub-surface maxima both deepening and becoming less intense with time. The authors took all available data within a defined region (20° – 40° N and 135° – 175° E) and established a doubling-depth of the ^{239,240}Pu maximum of about 40 years, and a half-value of the concentration of about 22 years. Although it is tempting to conclude that this re-distribution is entirely biologically mediated – i.e. remineralisation of sinking biogenic particles (Hirose 1997) – there is reason to believe that physical processes may have a significant influence. Profiles of ^{137}Cs and ^{90}Sr, which are not expected to be involved in biomediation, also revealed a decrease in concentrations and inventories, having corrected for decay. This implies that water with lower concentrations has been advected into the region at depth. The need to consider a horizontal advective component when interpreting open ocean plutonium profiles has been discussed previously in respect of the NW Atlantic, and the southwards flow of high-plutonium North Atlantic Deep Water at intermediate depths (35-50% contribution) (Cochran *et al.*, 1987). The NW Pacific profiles also featured an increase in ^{239,240}Pu concentrations in bottom waters of about a factor of 2. This latter observation has not been recorded in the NE Pacific. Upper ocean profiles in the immediate vicinity of Bikini Atoll, which lies in the westward-flowing North

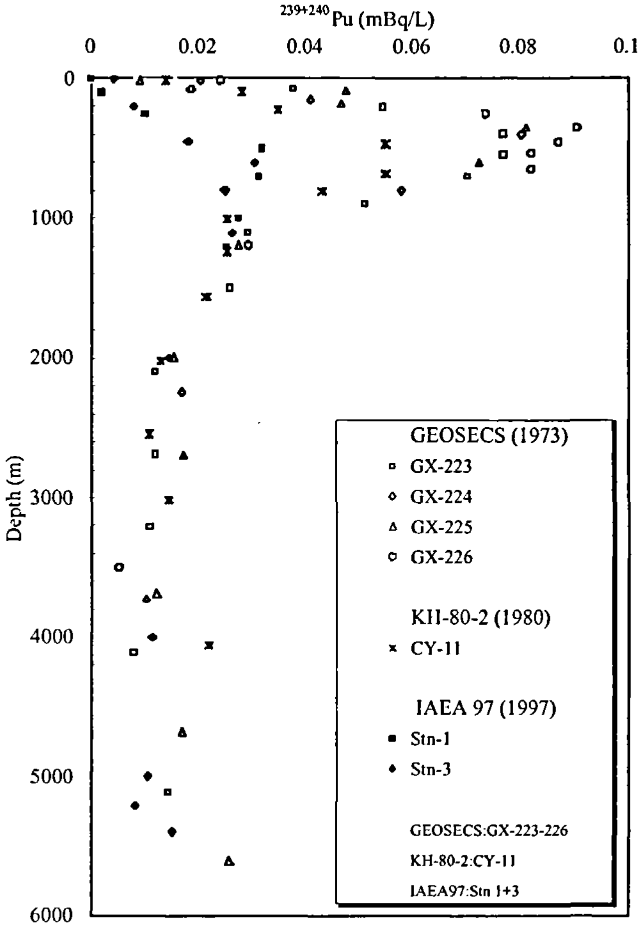


Figure 1

Vertical distribution of ^{239,240}Pu (Bq.m^{-3}) in the NW Pacific, in 1973, 1980 and 1997; from Livingston *et al.*, in press.

Equatorial Current, revealed that ^{239,240}Pu concentrations and inventories did not change significantly between 1973 and 1997. It is suggested that the concentrations were maintained by the continual re-supply of low-productivity water.

It is clear that observations of artificial radionuclide distributions as time-series at specific locations, vertical water-column profiles and spatial surveys can all provide insights into ocean processes on a range of space and time scales. However, it is also apparent that interpretation of the data will be seriously compromised if due account is not taken of the underlying factors controlling the distribution. Some factors may be common to many regions of the world ocean but others may be specific to particular situations. One advantage of using water column measurements is that it may be easier to achieve sample homogeneity compared with sediment sampling.

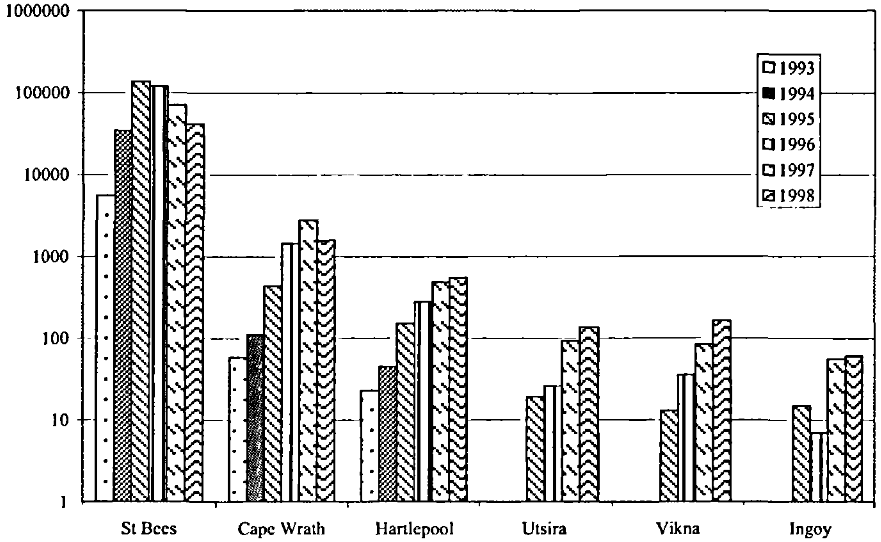
Radiotracers and environmental variability

Water column radiotracer data can be used to identify transport mechanisms/pathways and sometimes to quantify the rate at which transport is taking place. This alone may require a considerable effort. However, to attempt to establish the degree of variability, and systematic change, in such processes presents a formidable challenge. The great advantage of utilising sediment core samples is that they can be dated, often independently, and a single core may encompass the entire period of interest. For water column work it is usually necessary to establish time-series of observations, as well as spatial distributions, and such data sets are often limited to particular locations or regions because of a specific radiological concern (e.g. Sellafield, La Hague, Marshall Islands, French Polynesia) or because of proximity to a research centre (e.g. La Spezia, Italy, annual sampling from 1960 to the present; Papucci *et al.*, 1996). These studies would have been undertaken for a variety of reasons in a variety of circumstances and this may limit their usefulness for environmental change investigations. However, they do provide an excellent basis for designing future programmes.

Fortunately there are examples of where the necessary combination of well-defined radiotracer inputs, oceanography and climate indices occur. The NE Atlantic has received a considerable input of artificial radionuclides as result of nuclear fuel reprocessing activities at Sellafield (UK, discharging into the Irish Sea since 1952) and La Hague (France, discharging into the English Channel since 1966). One of the main purposes of conducting distribution studies throughout the 1960s-1980s was to assess the likely impact of the releases, from the consumption of contaminated fish, by the whole population (i.e. calculating the Collective Dose). However, a number of investigators saw the value in using these freely-available tracers to examine transport pathways, transit times and transfer factors across the shelf and consequently the releases have been traced from the pipelines throughout the NW European Shelf, into the Baltic Sea, across the Nordic Seas (Norwegian, Greenland, Barents) and into the Arctic (see references cited in: Kershaw and Baxter, 1995). Of particular value from Sellafield were ^{134}Cs, ^{137}Cs, ^{90}Sr, ^{99}Tc and ^{129}I although Pu isotopes have also been used. The total quantities discharged from La Hague were much lower but also included characteristic tracers such as ^{125}Sb which has been measured extensively in the English Channel and the North Sea (Guegueniat *et al.*, 1997). In recent years there has been a tendency for discharges of most radionuclides from both sites to decrease substantially. This may have resulted in welcome reductions in the dose to human populations but it has meant that the analytical challenge of making the measurements has increased.

As a consequence of the reductions in direct discharges, the sediments of the Irish Sea, which represent a substantial repository of ^{137}Cs and transuranic elements, have become a significant source of ^{137}Cs and plutonium into the water column. This is in response to the lower water concentrations following flushing by Atlantic Water, characterised by background levels of activity. Despite this mechanism, the outflow from the Baltic Sea, contaminated by Chernobyl, has been the largest source of ^{137}Cs to the North Sea for the past decade. The ^{137}Cs/^{90}Sr ratio due to Chernobyl contamination falls in the same range as that from Sellafield, introducing ambiguity in applying this ratio to transport studies. Fortunately, developments during the 1990s, both analytical and in waste treatment procedures, have provided a further opportunity to utilise reprocessing tracers and compare the results of recent studies with those conducted in earlier years. There has been a steep increase in the quantity of ^{129}I discharged from La Hague, due to increased fuel throughput, and this nuclide can now be detected in small volume samples using the extremely sensitive technique of Accelerator

Mass Spectrometry (AMS) (Smith *et al.*, 1998). Likewise, at Sellafield there has been an increase in the quantity of ^{99}Tc discharged (Figure 2), although this has been in the form of a 'pulse' extending over a number of years, rather than the continuous increase in ^{129}I seen at La Hague. There has also been an increase in ^{129}I discharged from Sellafield (about 20% of the combined release). The development of AMS for ^{99}Tc has been problematic but several groups are now using ICP-MS routinely. Careful separation stages are required to remove interferences, as is the case for more standard radiometric methods.



I Figure 2
Discharge of ^{99}Tc (TBq.yr^{1}) from Sellafield (UK) in to the Irish Sea 1978-1998.

The transport of the initial leading edge of the 1990s ^{99}Tc 'plume' (Figure 3) was much more rapid than had been anticipated, on the basis of the earlier transport time studies, especially within the first 2 years (Figure 4). Seawater samples were augmented with brown seaweed (*Fucus vesiculosus*) collected from around the coast of the

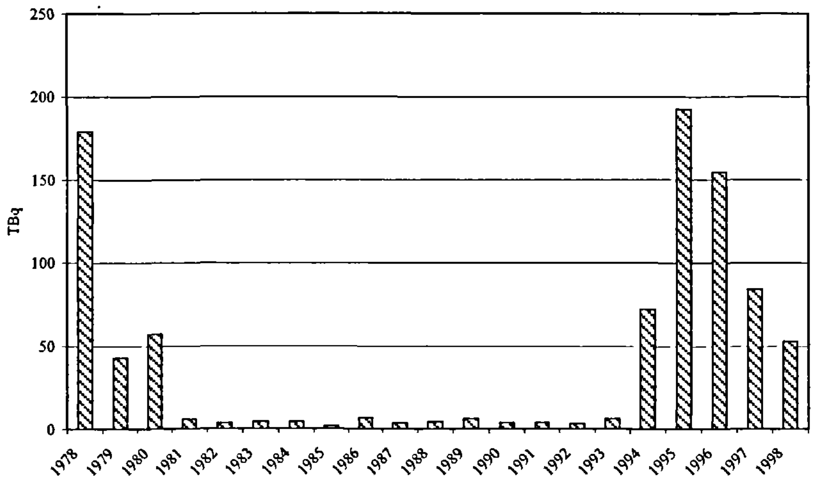
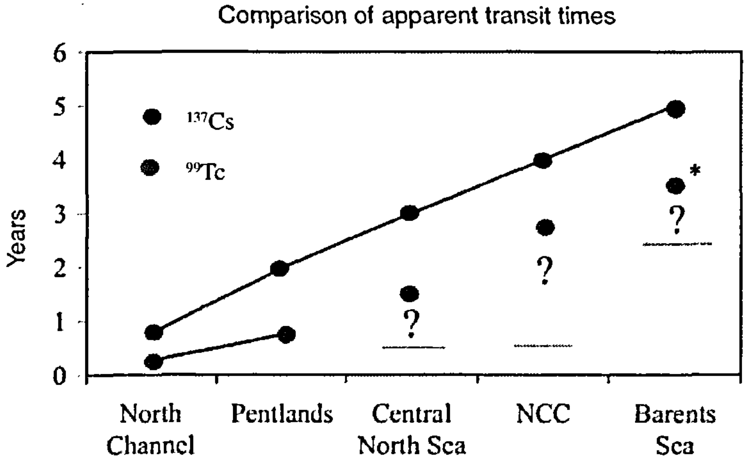


Figure 3
Concentration of ^{99}Tc (Bq.kg^{-1}, dry weight) in the brown seaweed *Fucus vesiculosus* collected from shoreline locations: near Sellafield (St Bees), NW Scotland (Cape Wrath), NE England (Hartlepool), SW Norway (Utsira, ~ 59° N), mid-Norway (Vikna) and northern Norway (Ingoy). The discharged increased in 1994.



* Gordon Christensen pers. comm.

Figure 4
Comparison of apparent transit times from studies using Cs or ^{99}Tc: the North Channel is the northern exit of the Irish Sea, the Pentlands lie to the north of mainland Scotland, the NCC is the Norwegian Coastal Current.

UK and Norway (Heldal *et al.*, in prep) as this has a high concentration factor for ^{99}Tc (~ 1x10^{5}) so provides an effective indicator of the spread of ^{99}Tc contamination. Heldal *et al.* (in prep) suggest that the difference in apparent transport rates is due, in part, to variations in the degree of atmospheric forcing, as indicated by the NAO. The NAO is a simple measure of mean, winter (December-March), surface pressure difference between Iceland (Stykkisholmur) and Lisbon (Portugal). Winters with a high mean pressure difference are described as having a high positive NAO index, and are characterised by increases in the number and intensity of storms, with accompanying higher precipitation, an increase in the flow of the North Atlantic Current and the formation of a shelf-edge jet-like current. There has been an overall increase in both the number of NAO positive years and the value attained by the index. The mid 1990s, when the ^{99}Tc releases increased, were marked by high positive values, in contrast to the lower values of the 1970s and 1980s, when most of the Cs-based transport studies were carried out. There may be other factors which are influencing the derived transit times but there seems to be sufficient consistency to justify further investigation, in particular with the application of mathematical models having adequate spatial and temporal resolution (Nies *et al.*, 1998). The NAO is one of about 12 indices which are routinely monitored, with the results being made freely available via the internet (e.g. National Centers for Environmental Prediction, www.ncep.noaa.gov). There is some evidence that the NAO variation is related to the fluctuations in the ENSO (El Niño Southern Oscillation). Radiotracers may provide a tool to describe the environmental response to these large-scale forcing events, and other climate-related phenomena. For example, it has been suggested that the degree of oligotrophy in the central Pacific is increasing in response to global warming and the consequential strengthening of the thermocline. One way to quantify this mechanism will be to determine whether there is an increase in radionuclide/metal residence times in surface waters, as might be anticipated.

Conclusions

Observations of artificial radionuclide distributions in the water column, in time and space, can provide valuable insights into the presence and rates of ocean processes - on physical scales from local to basin-wide and time-scales of days to decadal - providing something is known about the source term; i.e. the quantity, location and rate of entry into the marine environment, and the chemical behaviour of the radionuclides (i.e. conservative vs. non-conservative). Global stratospheric fallout represents the most significant source on a global scale, although it has had a strong latitudinal dependence and significantly higher deposition in the northern hemisphere (N:S, 3:1), as a result of the history of detonations and resulting yields. The distribution of ^{3}H has been particularly useful in illustrating the extent of ventilation in the North Atlantic. Vertical profiles of $^{239,240}Pu$ in the open ocean tend to exhibit a characteristic sub-surface maximum. This can be attributed partly to biological mediation but it is apparent that in the NW Pacific a component is due to lateral advection – a phenomenon also observed in the North Atlantic. It has been demonstrated, in the NW Pacific, that the magnitude of the sub-surface maxima has decreased and the position in the water column has deepened. Close-in fallout in the Pacific has had a minor impact in the environment around Mururoa and Fangataufa atolls, but near-ground tests in the Marshall Islands have contributed a significant input of plutonium to the NW Pacific. The addition of ^{238}Pu, preferentially into the southern hemisphere, as a result of the SNAP-9A satellite accident represents an under-utilised tracer.

The controlled but variable releases of radionuclides from European reprocessing facilities, combined with the relative proximity of potential sampling sites to centres of population, have provided the best opportunity so far to test whether radiotracers can be used as indicators of environmental variability. A comparison of transport times, from Sellafield to various locations in the North Atlantic and Nordic Seas, based on recent releases of ^{99}Tc (mid 1990s onwards) and older releases of ^{137}Cs (1970s-1980s) has revealed an apparent difference, with the former being faster. It is conjectured that this difference, at least in part, may be due to differences in environmental forcing linked to phenomena which can be characterised by fluctuations in the NAO. To test the validity of this initial conclusion it will be necessary to conduct simulations using sophisticated coupled ocean-ice models, covering the range of years for which Sellafield has been operating.

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