Paper

A MILLENNIUM PERSPECTIVE ON THE CONTRIBUTION OF GLOBAL FALLOUT RADIONUCLIDES TO OCEAN SCIENCE

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Abstract—**Five decades ago, radionuclides began to enter the ocean from the fallout from atmospheric nuclear weapons tests. The start of the 21st century is an appropriate vantage point in time to reflect on the fate of this unique suite of manmade radionuclides—of which more than two-thirds arrived at the surface of the oceans of the planet. During these five decades much has been learned of the behavior and fate of these radionuclides and, through their use as unique tracers, of how they have contributed to the growth of basic knowledge of complex oceanic physical and biogeochemical processes. Some of the highlights of the ways in which fallout radionuclides have given new insights into these processes are reviewed in the historical context of technological and basic ocean science developments over this period. The review addresses major processes involved, such as physical dispersion and mixing, particle association and transport of reactive nuclides, biological interactions, and mixing and burial within ocean sediments. These processes occur over a range of scales ranging from local to global. Finally, an account is given of the present spatial distribution within the oceans of the various components of the fallout radionuclide suite.**
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INTRODUCTION

As discussed in detail elsewhere in this issue, the fallout from the atmospheric nuclear weapon's tests in the mid-20th century has by now largely been transferred from the atmosphere to the surface of Earth. Since about 70% of the surface of the planet is ocean, much of this mixture of artificial radionuclides has ended up there. Despite the fact that most atmospheric nuclear weapons tests took place in the Northern Hemisphere, it can be shown from the proportions of land and ocean in each hemisphere, and from the latitudinal fallout patterns over time, that over two-thirds of the fallout entered the ocean. Following entry to the ocean, the behavior of these radio-elements was determined by their physical and chemical properties and their fate by oceanic physical and biogeochemical processes. As in the terrestrial environment, the post-delivery behavior of these fallout radionuclides provided a unique opportunity to gain insights into the nature of the processes controlling their fate. The great depth and dynamic nature of the oceans have resulted in substantially greater dispersion and dilution than on land. This review is intended to trace the key events and trends of studies during the second half of the 20th century associated with the entry of global fallout to the world oceans. Initially these studies were driven by the need to assess the impact and fate of fallout radioactivity on marine life and associated transfer to man through marine exposure pathways. Subsequently, fallout radionuclides were widely used as oceanic process tracers, due to their unique time-marker labeling of sea water, marine organisms, and sediments according to their individual chemical properties. In this sense they contributed in no small way to the broad growth of knowledge of the intricate physical, chemical, biological, and sedimentological processes which occur in the oceans. So this review will also try to present a broad account of the major research areas where fallout radionuclide studies have contributed to advances in ocean science—with some illustrative examples where appropriate.

CHARACTERISTICS OF FALLOUT INPUT TO THE OCEANS AND THE STUDY TIME FRAME

The course of ocean fallout studies, by the large body of international scientists and institutions involved, followed a series of phases and trends. They were based on a common set of physical and chemical characteristics and were accompanied by a variety of perturbations from other sources of artificial radionuclides which impacted the oceans. An account of these provides the context in which the whole range of ocean fallout research was framed.

The primary time-frame of ocean fallout studies was set by the history of nuclear weapons testing and the resultant transfer to the atmosphere of debris which was delivered to land and sea at rates documented elsewhere. The main periods of delivery of fallout to the oceans were the years following the two intense series of

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atmospheric tests in the mid-1950's and early 1960's. The scale of testing by countries that continued atmospheric tests after the 1962 Test Ban Treaty was much diminished and represented a relatively small continued input. The majority of the fallout produced in atmospheric tests was injected into the stratosphere and subsequently returned to Earth's surface as global fallout. Some tests, especially in the earlier test series in the 1950's time frame, were conducted at or near ground level and produced tropospheric fallout with a regional impact. The oceanic impact of ground level tests was significant from testing at oceanic locations such as those on Pacific islands—especially the United States test series at the Pacific Test Site in the Marshall Islands. So this regional impact was largely confined to the Western North Pacific. Minor traces of tropospheric fallout from the U.S. Nevada Test Site were identified in the Atlantic Ocean and Mediterranean Sea (Buesseler and Sholkovitz 1987) but were not quantitatively very significant.

The pattern of global fallout delivery to the world oceans was anticipated to be very similar to that recorded on land networks of sampling stations. This would follow the now classic depositional pattern with maxima at mid-latitudes, minima towards the equator and the poles and with much larger input rates in the Northern Hemisphere. In fact, the patterns of surface sea water concentrations observed during peak fallout years [summarized by latitude and time in the comprehensive RIME report (Volchok et al. 1971)] proved rather complex due to seasonal fluctuations in delivery, ocean mixing, and nonstandard observational networks. Sometimes the overland depositional pattern was observed, e.g., in the mean ^{90}Sr surface values reported in the North Atlantic in 1960 and 1961, but often the data were difficult to interpret. A more convincing demonstration of oceanic distributions came more than a decade later, and relied on water column inventory patterns of ^{137}Cs (Kupferman et al. 1979; Livingston and Jenkins 1982). The interesting point about these inventory patterns was that not only did they show a latitudinal pattern similar to overland fallout patterns, but that these patterns still were preserved substantially later than the major inputs—despite the dynamic nature of the ocean, with the active gyral circulation of surface and sub-surface currents.

A further general comment needs to be made concerning physico-chemical properties of global fallout entering the ocean and which explains subsequent observations of its fate and movements. The basic hypothesis, which has been developed over the history of studies of fallout in the oceans, is based on the premise that most of the fallout quickly behaves essentially as in solution, in ways controlled by the elemental chemical characteristics of each radionuclide. There is evidence that, while this is mostly the case, a relatively small fraction of fallout debris can be in the form of rather refractory particles, which do not fully solubilize and whose fate is more determined by oceanic particle processes and can end up rather quickly in ocean sediments.

As a result of this behavioral setting premise above, it has been common practice (Livingston and Jenkins 1982) to classify the oceanic behavior of fallout radionuclides as broadly "conservative" or "nonconservative." The conservative group includes radionuclides having a very small affinity for particle association and whose behavior consequently is controlled by the physics of ocean circulation and mixing. Examples of these include ^{3}H, ^{90}Sr, ^{85}Kr, and (in most situations) ^{137}Cs. The nonconservative group includes radionuclides with a range of strengths of particle association and whose ultimate behavior is linked to these affinities. Examples of these include ^{55}Fe, ^{147}Pm, plutonium and americium isotopes, etc. It is important to note that, despite this classification, the time scales over which such behavioral differences may be observed are also important to keep in mind. For example, it is well established that the association of nonconservative radionuclides with plankton blooms provides a mechanism by which reactive fallout nuclides can be removed from surface sea water and released through remineralization processes below the euphotic zone. This can occur over seasonal time scales and produces separation of nonconservative and conservative fallout nuclides. But as even reactive radionuclides can have oceanic residence times of hundreds to thousands of years, physical circulation processes can dominate in the short term—at least in the open ocean which received the major part of the fallout input by far. Different considerations apply in productive coastal shelf areas, and in estuaries, where particle association processes can exert substantial controls on reactive radionuclides over much shorter time scales.

HISTORICAL PERSPECTIVES ON OCEAN FALLOUT STUDIES

Inevitably, studies of the fate of fallout from nuclear weapon's tests and its use as a tool for ocean science have been limited by the availability and state of development of analytical measurements and sampling technologies. They have also been limited by the contemporary level of the state of knowledge of the oceans and its processes. Ocean fallout studies span a half century, which has been marked by a staggering growth of scientific and technical knowledge. The period began on the nuclear side at a time of slide rules, gas filled detectors and the first generation of radiochemical techniques. It ended in an age of computers, sophisticated semi-conductor detectors, advanced radiochemistry, and powerful mass spectrometric techniques. On the ocean side it began in the era of the reversing Nansen bottle and bathythermograph and ended in the age of satellites, robotic vehicles, and the sophisticated sampling technologies of integrated, international big ocean science. It is an accident of history that the early days of ocean fallout studies were constrained by the available technologies. The wide range of shorter lived radio-tracers received scant study and the benefits of growth in technology has only seen application to the suite of medium to long half-life radionuclides.

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The nature of ocean fallout studies falls into several phases, defined by technical development and political trends. For example, the earliest period was one of evolving nuclear weapons confrontation, priority on weapon's development, and an atmosphere of secrecy. Gradually concerns over radiation risks lead to an increase in health and environment studies. As these did not uncover critical problems, support for fallout studies began to decline—in parallel with the increased potential through developments in ocean science and technology. Technical developments also exerted influence on the direction of these studies. The early days were characterized by complex radiochemical analyses and relatively simple radiation detectors, which were frequently limited in energy discrimination properties. The 1960's and 1970's saw a massive growth in solid state detector technology—in parallel with an increase in sophistication of the electronics required to handle the detector output. First sodium iodide, NaI(Tl), detectors and then germanium, Ge(Li), detectors brought new gamma counting capabilities to the field—albeit with somewhat lower sensitivities of radionuclide detection. In the area of alpha emitting radionuclide detection, the arrival of surface barrier silicon detectors permitted an increase in the use of studies of this class of radionuclides—which included the important group of transuranic elements (plutonium, americium, etc.) This development was an important event in ocean fallout studies—and no doubt in other fields—for another reason. Studies of ocean processes which could be traced by reactive radionuclides mostly used beta or gamma emitting radionuclides with half-lives of relatively short (5 y or less) duration. In contrast, the transuranic group of radio-elements are often very long lived and so became very convenient replacements for the rapidly disappearing shorter lived group.

These political and technical considerations apply regardless of the study scale involved, e.g., small scale near shore studies or larger scale open ocean studies. However, other factors marked the course of the larger scale studies. These included the way in which ocean science was changing over the years. In the period of the first two decades, roughly 1950 to 1970, ocean studies were planned and carried out largely at the national and institutional level, with limited coordination and generally single ship projects. Looking back, it seems that was ended and summarized by the publication in 1971 of a prestigious report by the U.S. National Academy of Sciences—the so-called RIME (Radioactivity in the Marine Environment) report (Seymour 1971). It essentially pulled together in a comprehensive way the full suite of marine fallout studies completed as of then, against an account of the relevant features of nuclear weapon's testing.

At about the same time as the publication of the RIME report, a new approach to ocean science began—the so-called era of big ocean science. At first at the national level, teams of scientists began to plan projects on a geographical scale previously not attempted. In part, this was a beginning of a recognition that an integrated suite of measurements covering major ocean areas in a short period produced a much better global description of ocean properties or processes. For ocean fallout studies, the GEOSECS (Geochemical Ocean Sections Study) project (Craig and Turekian 1976) was a milestone in marine chemistry and was a major event in the history of ocean fallout studies and their application as tracers of ocean physical and biogeochemical processes. This project spanned the decade of the 1970's with the objective of obtaining a global inventory of chemical components in the world oceans with special emphasis on the deep waters. Intensive sampling was carried out in the Atlantic in 1972–1973, the Pacific in 1973–1974, and the Indian Ocean in 1977–1978. The project culminated in the publication of the full data set of measurements and a series of illustrative atlases of sections of the various parameters in the ocean basin's sampled. The GEOSECS project produced a comprehensive data set for the fallout radionuclides ^{137}Cs, ^{90}Sr, ^{239,240}Pu (and limited ^{241}Am) for the Atlantic and Pacific Oceans which permitted very useful inventory estimates for each ocean to compare with input estimates (Bowen et al. 1974). It also saw the collection of even more detailed data sets of the distributions of ^{3}H and ^{14}C in all three oceans. This led to the specific use of these conservative tracer radionuclides by the physical oceanographic community. As will be elaborated on later, this application of fallout radionuclides has to be high on the list of their contributions to ocean science knowledge.

The last 20 years have seen a decrease in the scale of effort applied to studies of fallout in the oceans. This is, no doubt, in part due to the reductions in research funding as fallout levels in the ocean declined, both by radioactive decay and by dilution through mixing and dispersion. Other new sources of radioactivity impacted the ocean in this period, as noted in the following section, and their study and assessment had to be made against the pre-existing fallout signal. However, the techniques and knowledge developed in the ocean fallout study era were quickly turned to the new sources and greatly facilitated observations of their fate and impact. It is also probably true to observe that measurements of fallout radionuclides in the oceans in recent years were on applications in which the fallout signal had been shown to be a proven ocean research tool; these types of measurements found their place amongst the suite of techniques available for addressing emerging questions or problems. Many of the examples of ocean fallout applications noted later would typify this recognition.

PERTURBATIONS OF THE OCEAN FALLOUT SIGNAL

Although the introduction to the ocean of fallout radionuclides from atmospheric nuclear weapons tests by far outweighs the contributions from other sources of artificial radioactivity from accidents and planned releases (Table 1), on local to regional scales, these

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Table 1. Principal sources of anthropogenic radionuclides in the oceans (PBq)[a,b]

		Global fallout		Discharges		Chernobyl accident	
Radionuclide	Total	Ocean input	Present ocean inventory	Sellafield	La Hague	Total	Input to oceans
^{3}H	186,000	112,693	13,300	39	87		
^{90}Sr	622	377	151	6	1.1	10	negligible
^{137}Cs	948	603	251	41	1.0	85	16
^{239,240}Pu	10.9	6.6	6.6	0.6	0.1	0.06	negligible

[a] Based on data from Guegueniat et al. (1996), Mitchell et al. (1996), and IAEA (2001, 2002a,b).
[b] River run-off may increase the ^{90}Sr and ^{137}Cs inventories by about 6% and 1%, respectively.

perturbations were often substantial but the underlying fallout signal needed to be known to evaluate and detect the new source impacts and to define their dispersion pathways. The largest of these perturbations primarily affected the oceans and seas around Europe. These came from the discharges of nuclear fuel reprocessing wastes to the Irish Sea [from the British Nuclear Fuel Laboratory (BNFL) at Sellafield] and the English channel (from the COGEMA Plant at Cap de la Hague in northwest France) and from fallout into European Seas from the 1986 Chernobyl nuclear power plant accident. The nuclear fuel reprocessing discharges have been widely described (IAEA 1985a; Guegueniat et al. 1996; Mitchell et al. 1996) and studied and are beyond the scope of this review. It is sufficient to note here that they have been ongoing since 1952 (United Kingdom) and 1965 (France) and tended to be maximal in the mid-1970's. Their impacts were not only local—especially in the Irish Sea—but regional. Due to the rapid northward flushing of the European shelf seas, in the scale of a few years most of the soluble radionuclides discharged were transported into the Arctic Ocean and adjacent seas (Kershaw and Baxter 1995). There they represent a substantial new tracer signal following on the heels of the global fallout signal. Over longer time scales, these two sources have labeled the source waters forming the precursors of the cold waters sinking and moving southward to form North Atlantic Deep Water. The distinctive time and composition signatures of the fallout and nuclear fuel reprocessing signals can be expected to be of great value in future studies of the rates and routes of ventilation of the deep Atlantic and beyond from the Arctic. Given the relevance of this process to studies of the linkages between oceans and climate, these are likely to be exploited as useful tracer tools for many decades and centuries to come. A similar set of considerations apply to the perturbation from Chernobyl fallout. This was primarily by ^{137}Cs (after decay of the short-lived ^{134}Cs) and impacted the Baltic, Black and Mediterranean Seas and the European and Arctic shelves and enclosed seas. Again the combination of the two tracer ^{137}Cs signals, from global fallout and from Chernobyl, have been and will continue to be used as ocean tracers in these regions.

There is another class of additional nuclear sources which have entered the ocean but which are of minor and localized input compared with those described above. They include leakages from oceanic nuclear weapons test sites (Povinec et al. 1999; Robinson and Noshkin 1999), from the few small underwater nuclear weapons tests conducted in the oceans (Joseph et al. 1971; Smith et al. 1995), and from the dumping of nuclear wastes in both surface and deep ocean (IAEA 1991; NEA 1996). Included in this latter group are the relatively large radionuclide activities inside the fueled reactors disposed of in shallow waters of the Kara Sea by the former Soviet Union (Sjoblom et al. 1999). No releases of any significance have been observed at these sites (Osvath et al. 1999) and the assessment of any potential release has been evaluated as unlikely to present any significant radiological risk (Osvath et al. 1999; Sjoblom et al. 1999). In the category of accidental inputs there have been a number of nuclear submarines lost in both the deep (IAEA 2002a) and shallow ocean (the Russian nuclear submarine "Kursk" which sank on the Barents Sea's shelf in 2000), the loss of radioactive sources (Naoaki 1993), and the loss of nuclear weapons (Aarkrog et al. 1984). No releases of consequence have been noted from the former and the plutonium contamination from the latter has been found to be very localized and of minor radio-ecological risk.

Finally, there was one accident involving a nuclear source which contributed to global fallout. This was a ^{238}Pu source from a satellite (SNAP-9A) that burned up over the Indian Ocean in 1964 (Hardy et al. 1973). The fallout from this source was distributed inversely to global fallout, i.e., a majority in the Southern Hemisphere and a minority in the Northern Hemisphere. Again, this provided an additional tool to be used in conjunction with nuclear weapons testing fallout as it had a distinctive isotopic (e.g., the ^{238}Pu:^{239,240}Pu ratio) and temporal signature, which has been used as a time-marker and has potential for future use, especially in the Southern Hemisphere.

OCEAN DEPOSITION OF NUCLEAR FALLOUT

In the years during and after the intense period of atmospheric nuclear weapons tests, great effort was expended into monitoring and quantifying atmospheric fallout levels and consequent delivery to the surface of Earth—as reviewed elsewhere in this issue. Although these programs were by no means easy or without problems and uncertainties from overland observation sites, the situation was much more difficult with respect to oceanic deposition. The enormous area of the oceans

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and the lack of fixed observational systems meant that, in contrast to the overland situation, there were very few systematic observations of fallout arrival and delivery to the surface ocean. The nearest suite of systematic over-ocean measurements were made from vessels occupying permanent oceanic sites for military or meteorological observations. These were limited in terms of coverage but did provide some information to compare with land deposition. In the United States, four fallout collection platforms on weather ships were operated in the North Atlantic for several years (HASL 1968) and on other vessels in the North Pacific (data reported separately) (Hammond et al. 1966; Volchok 1967; Volchok and Kleinman 1968).

Given the limited and patchy nature of the oceanic deposition observational coverage, it is hardly surprising that many uncertainties and controversies surrounded the period of major fallout entry to the oceans. For example, in a review of ocean fallout in 1971 (Volchok et al. 1971) it was noted that the pattern of seasonal deposition, with a pronounced maximum each spring, was not found generally in over-ocean measurements and that even other means of deposition than precipitation could deliver fallout to the ocean. Subsequently, it was pointed out (Kupferman et al. 1979) that even precipitation has been historically difficult to measure at sea—in addition to further uncertainties in the measurement of the fallout.

At the time of the RIME report, there was another controversy over the question of whether soluble fallout radionuclides (^{90}Sr, ^{137}Cs) were being mixed into the deep ocean, as some measurements indicated (discussed in Volchok et al. 1971). This was a major issue in establishing oceanic fallout inventories, as small concentrations averaged over large deep water volumes represented correspondingly large inventories. In time (Kupferman et al. 1979), this was seen to have been a deep water blank problem and that there appeared to be a reasonable balance between ^{137}Cs and ^{90}Sr inventories in the North Atlantic when compared with the best estimates of delivery overland (on an area adjusted basis).

The depositional situation in the Pacific turned out to be considerably more complex than in the Atlantic. Although there were scattered references (mostly non-United States) to very high surface ^{90}Sr concentrations in the Western Pacific in 1957 to 1959 (in RIME Report Table 2, Volchok et al. 1971), this began to be recognized more fully in the years following the Pacific GEOSECS expedition. Both the GEOSECS datasets (Bowen et al. 1980) and subsequent Japanese measurements (Nagaya and Nakamura 1984, 1987), as well as previous studies by Noshkin (1980) and Noshkin and Wong (1980) revealed substantial excesses of measured inventories of ^{137}Cs, ^{90}Sr, and ^{239,240}Pu at many locations in the Western North Pacific compared with estimated delivery from global fallout. It has become clear that the 68 near surface nuclear tests (Robinson and Noshkin 1999) conducted at Bikini and Enewetak produced not only local fallout but regionally deposited tropospheric fallout, which led to widespread deposition to the western North Pacific on a scale similar to that from global fallout. Its presence has been detected not only by the substantial increments to fallout nuclide inventories and concentrations but also by the distinctive enriched ^{240}Pu/^{239}Pu signature in Pacific seawater and sediments over a wide area (Buesseler 1997; IAEA 2002b). The Western Pacific Ocean is, therefore, unique in the history of atmospheric nuclear weapons test fallout in that it is the one area of the globe where fallout was substantially enhanced over the patterns globally observed.

USES OF FALLOUT RADIONUCLIDES AS TRACERS OF OCEAN CURRENTS AND MIXING

As indicated earlier, several of the radionuclides that entered the oceans as fallout from atmospheric nuclear weapons tests had chemical properties which resulted in their behavior as truly soluble elements, moved by and tracing the physical dispersive and mixing processes in the oceans. This behavior resulted in their dilution and dispersion in response to physical forcing such as wind and thermal factors or by major gyral circulation driven by Earth's rotation. Through these processes, this class of nuclides has been rapidly mixed down the main thermocline in the oceans and moved in association with both surface and sub-surface advective processes. This has led to their arrival also at oceanic mid-depths, e.g., in the Mediterranean outflow tongue spreading to depths greater than 1,000 m in the North Atlantic or to ocean deep water, for example in the cooling and sinking of Arctic waters into the North Atlantic Deep Boundary Current which travels south at the bottom of the North American continental slope.

A large number of studies of fallout in the ocean have contributed either directly or indirectly to this body of knowledge on the rates and routes of physical mixing. While a review of this field is far beyond the scope of the present overview of fallout in the oceans, it is appropriate to classify such studies into two broad groups.

Firstly, studies of the evolving three dimensional distribution of soluble fallout tracers provides a basic set of reference data against which the contrasting behavior of particle active tracers can be compared. For example, the distributions over depth and time of ^{90}Sr and ^{137}Cs have evolved in a tightly coupled way in the open ocean, at least, and can be taken as tracing their dispersion from the ocean surface by physical processes. In the open ocean, their relative ratio, ^{137}Cs to ^{90}Sr, has been shown to remain very constant, at a ratio, when adjusted for radioactive decay, equal to that of their fission yields (Bowen et al. 1974). Tracers that have been shown to deviate from the patterns traced by ^{90}Sr and ^{137}Cs, e.g., ^{147}Pm, plutonium isotopes, and ^{241}Am (Livingston and Jenkins 1982), are classified as particle-active or nonconservative tracers. The degree of deviation is a function of strength of their particle association properties, the elapsed time since fallout arrival, and the intensity of the

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particle transport processes. These latter are typically primarily biologically driven, strongest in oceanic regions of high biological productivity and lowest in the so-called oligotrophic or biologically poor regions—such as in the central ocean gyres. By way of example, we can cite a recent and striking study by Nozaki et al. (1998) of the distribution of these radionuclides in the water columns of and above two deep ocean trenches south of Japan. In this study, the distributions of ^{90}Sr and ^{137}Cs decline to essentially undetectable levels somewhat below 1,000 m, i.e., in the main thermocline. In contrast, ^{239,240}Pu continues to be found in two trenches at significant and little varying levels down to depths of 7,585 m and 9,750 m, respectively. In this relatively high ocean productivity area, the major fraction of the ^{239,240}Pu inventory is found in the deep water below the depths containing the ^{90}Sr and ^{137}Cs inventories.

The second broad group of studies are those which were directed specifically to illustrate or quantify specific features of physical mixing. Early examples include studies by Broecker (1966) and Rooth and Ostlund (1972). However, the direction of real growth of these studies came in the growth of the use of ^{3}H in the large national/international ocean projects ushered in with the GEOSECS project like Transient Tracers in the Oceans in 1981 to 1983 and South Atlantic Ventilation Experiment in 1988 to 1989 in the Atlantic Ocean. This could be characterized as the start of recognition by the physical oceanographic community of the potential and power of the use of anthropogenic ocean tracers to be used in conjunction with standard physical property measurements. In addition to fallout ^{3}H (and its radiogenic progeny ^{3}He), chlorofluorocarbons (CFCs) started to be used in parallel applications as physical tracers. CFCs have had some similarities in atmospheric input function as ^{3}H (see review in Schlosser et al. 1999). When used together, these tracers have seen the greatest level of adoption for physical tracer use. Both have the advantage that sampling and analysis developed rapidly, permitting the collection of quite high resolution two- and three-dimensional data sets which found ready assimilation to the physical oceanographic modeling community. As an example of such high resolution data sets, Schlosser et al. (1999) has described the tritium/helium data sets being developed in the framework of the World Ocean Circulation Experiment (WOCE). He gives as an example, the P17 north-south line at about 135°W in the Pacific. It is a beautiful snapshot of the near century end distribution of this tracer in the Eastern Pacific Ocean and illustrates the application of this fallout tracer in a most impressive way. The WOCE program is strongly connected to ocean/climate studies so the application of ^{3}H (^{3}He) as part of this program demonstrates one of the most striking contributions of fallout to advances in marine science.

The ^{3}H (^{3}He) technique has also contributed very specifically and precisely to particular aspects of the study of ocean circulation, viz., the "age" of given water mass. This age, the so called tritium/^{3}He age, is a measure of time that has elapsed since the last equilibrium of a given water mass with the atmosphere (Jenkins and Clarke 1976). It rests on the resetting of this tritium/^{3}He clock when a surface mass degasses any helium and the clock begins to record ^{3}He ingrowth when the body water becomes isolated from the surface by convective cooling and sinking, for example. This work was pioneered by W.J. Jenkins and described generally in Jenkins (1988) and Fine (1995). A classic application of the method to gain insights into ventilation of the deep western North Atlantic was reported by Doney and Jenkins (1988) and is but one of many examples of this technique which could be cited.

USES OF FALLOUT RADIONUCLIDES AS TRACERS OF PARTICLE ASSOCIATION, TRANSPORT AND FATE

Water column studies

Although the early period of ocean fallout studies included a significant number of studies of such particle-active radionuclides as ^{95}Zr-^{95}Nb, ^{103}Ru, ^{106}Ru, ^{141}Ce, ^{144}Ce, and ^{147}Pm, the summary of such studies in the RIME report (Seymour 1971) indicates that much more emphasis had been directed to studies of the less particle-active, or soluble, radionuclides. Indeed, very little progress was made in their use to study particle processes beyond the confirmation of some vertical separation in the ocean of this class of radionuclides from the soluble elements which accompanied them as they entered the ocean. In fact, some early theories (Bowen and Sugihara 1958, 1965) suggesting that this class of radionuclides would primarily be associated with particulate matter were subsequently revised in the light of the next generation of studies. So, when technical developments permitted the measurements of the long-lived alpha emitting nuclides of the transuranic element group, progress in this type of study began to gather pace. An important partner in these studies came through the parallel studies of the particle-active radionuclides of the naturally occurring family—such as the isotopes of thorium, protactinium, and lead.

Two distinctively different branches of fallout particle association study can be distinguished as proceeding somewhat independently. The first developed earlier and comprised the suite of fallout sediment studies in both the shallow, near-shore, and deep, open ocean. In these, the comparison of the total fraction of, e.g., ^{239,240}Pu in sediments relative to that which had entered the ocean at the location of study was determined. This provided an index of the particle-association and removal process as a function of location and oceanographic setting. The internal uses of the in-sediment distributions for either dating or bioturbation will be mentioned separately. The early picture that emerged from such studies (Noshkin and Bowen 1973; Livingston and Bowen 1979) was one in which quite intensive removal of plutonium was recorded in shallow, coastal settings with substantially smaller fractions of fallout plutonium delivery being

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found in deep ocean sediments. In fact, the many studies of plutonium distribution in open ocean water columns (Bowen et al. 1980) confirmed that most open ocean fallout plutonium was to be found within the ocean water column. The keys to understanding the totality of ocean processes on the cycling of particle-active elements or radionuclides came from the other branch of fallout particle-association studies, viz., through studies of their association with both suspended and sinking particles in oceanic water columns.

The suspended particle populations were studied following their separation by filtration by various techniques, either on bulk water filtered on board ship or by in situ filtration techniques (Livingston and Cochran 1987; Sachs et al. 1989), which permitted the filtration of the large volumes usually needed to measure the small proportion of the fallout plutonium (or other particle active fallout or naturally occurring radionuclide) associated with this particle class. The second class of particles, generally large and rapidly sinking, were collected in so called sediment traps, basically open containers into which the sinking particles could be collected. This area of study developed rapidly as sophisticated systems were designed and deployed. They were sometimes free floating but frequently moored systems. Left in the oceans for months and years, they provided the means to sample the sinking population of large particles over time increments set in advance. Sometimes traps were deployed at several depths of the water column allowing the measurement of the sinking particle flux as a function of depth. These traps provided a record of the seasonal and inter-annual changes in flux when deployed over several years. The longest running deployment of such traps in the Sargasso Sea (Deuser 1986) pioneered these studies of particle flux change over time. These studies and many others (Cochran et al. 1993) have broadened our knowledge of ocean particle dynamics and their link with the ocean carbon cycle. The incorporation of fallout radionuclides, together with natural series radionuclides, in these studies has brought about significant advances in knowledge in general about the ocean carbon cycle and a new model framework for the ocean cycling of particle-active elements and radionuclides (Bacon et al. 1985; Cochran et al. 1993). This framework now makes it possible to have a better integrated understanding of oceanic behavior of these radionuclides and of their present distribution in the ocean. In fact, the discussion is now only really applicable to plutonium (and ^{241}Am from ^{241}Pu decay) as most other particle reactive fallout radionuclides are relatively short lived.

Although plutonium is a particle reactive element, its oceanic presence has been shown to be to a large extent in the more soluble, oxidized ^{VI}Pu state (Nelson et al. 1984). This means that it is less particle active than, for example, ^{241}Am or lanthanide radioelements or thorium isotopes. Nevertheless, its behavior has shown strong particle process influence. Basically the primary process acting on plutonium or other particle-active species is the association with particles of biogenic origin. This means that in oceanic areas of high surface biological productivity, intense uptake and sub-surface transfer is a major vector of transport. This is especially active in coastal areas, where riverine and resuspended particles can also contribute, or in areas of upwelling, where fresh nutrient supply fuels the biological production processes.

The nature of this process is very different when one compares shallow coastal areas, the margin areas of the major ocean basins, and the deep ocean basins. In shelf areas, high removal rates keep particle-active nuclide concentrations low in seawater and explain their rapid accumulation in areas of fine-grained sediment deposition. This is also true on continental slopes, which are not only themselves productive, but also represent particle sinks for material formed on adjacent shelves (Carpenter et al. 1987). In open ocean areas, as noted earlier, particle production in the surface ocean is primarily dependent on the productivity in surface waters. This varies both seasonally and regionally. The large central ocean gyres are mostly oligotrophic (low productivity), whereas upwelling areas, and areas which receive subsurface nutrient supply through winter mixing, show higher productivity with consequent biological particle formation and vertical transport of reactive elements and radionuclides. A striking example is the North Atlantic spring bloom—which spreads across the northern North Atlantic producing a huge vertical flux of material which reaches the ocean floor in a matter of a few days. Much of the sinking material is, however, oxidized, or remineralized, during descent through the water column. It is this process that is the core of the ocean cycle—critical to the oceanic removal and cycling of the critical greenhouse gas, carbon dioxide. This major process was the focus of a recent major international ocean science program, the Joint Global Ocean Flux Study (JGOFS) (Hanson et al. 1999). JGOFS included many uses of radionuclides as study tools of carbon transport processes. Although many of these used the reactive nuclides from the naturally occurring uranium or thorium series, many of the techniques used were developed with earlier support of studies of reactive fallout radionuclides.

Sediment studies

Oceanic fallout studies in marine sediments have made substantial contributions to the growth of knowledge of marine sedimentology. Again, this is an area which has seen a very large number of studies. For the purpose of this review it is possible only to touch on the broad areas of study with some illustrative references. There are essentially two areas: (a) sediment dating and (b) sediment mixing (bioturbation). In both areas most fallout radionuclide studies have included ^{137}Cs and plutonium isotopes. But in addition they have benefited from inclusion of naturally occurring ^{210}Pb—especially

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the unsupported ^{210}Pb which was introduced from the atmosphere. This latter approach has been the basis of a well established sediment dating methodology—^{210}Pb method. Smith points out (Smith 2001) that it can only be used in sediments where sediment deposition dominates sediment mixing and it is essential that the two processes be kept in mind. The areas where deposition dominates can be certain estuarine environments or basins where accumulation is very rapid. For example, the sediments in basins off Southern California accumulate rapidly, are not mixed, are anoxic, and are deposited in annual layers (varves) (Koide et al. 1975). These sediments have been dated by several methods and recorded very well the fallout isotope signature in the flux of biogenic materials accumulating there. For example, the high ^{238}Pu signal from the SNAP-9A satellite burn up appears at a mid-60's time horizon. The very high fallout plutonium inventories in these sediments have been explained by the upwelling of offshore plutonium rich subsurface water causing high biological productivity and plutonium fluxes (Sholkovitz 1983). In contrast, most marine shallow sediments in shelf or slope environments are dominated by active biological mixing at or near the sediment/water interface. Classic studies of this process of bioturbation were made in shallow sediments of Long Island Sound (Benninger et al. 1979; Benninger and Krishnaswami 1981) and both natural series radionuclides and fallout plutonium were used to trace the dynamics of the mixing processes. Shelf and slope (Carpenter et al. 1987) studies have been used to quantify the rates and intensity of biological mixing. This has led to the development of models of in-sediment processes in which both mixing and accumulation are taken into account (Smith et al. 1995).

In the deep ocean, sediments accumulate very slowly and fallout radionuclide measurements have been used to trace the rates of biological mixing by benthic fauna. Even though bioturbation is much less active than in shelf and slope sediments, fallout isotopes have been mixed to substantial depths, e.g., abyssal sediments in the western North Pacific showed fallout isotopes mixed frequently and significantly deeper than 10 cm from the interface (Livingston 1986). This situation has been observed in several ocean basins (Cochran 1985; Smith et al. 1986; Cochran et al. 1987) and has demonstrated conclusively that biological mixing in deep ocean sediments can be quite significant and must be kept in mind in all types of deep ocean sediment studies in the present day or in paleo-sedimentological studies. For example, the distribution of both bomb ^{14}C and pre-bomb (cosmogenic) ^{14}C in abyssal red clay sediments became more understandable through parallel studies of fallout isotopes (Druffel et al. 1984). Finally, the knowledge derived from deep ocean sediment studies of fallout radionuclides was an essential prerequisite baseline against which all assessments were made of the dumping of low level radioactive waste in the deep ocean (NEA 1996).

FALLOUT RADIONUCLIDES AND BIOLOGICAL OCEANOGRAPHY

Initially the focus of attention of the uptake of fallout radionuclides by marine organisms was the possible radiation dose to humans from seafood consumption. This has been consistently shown to be very small, orders of magnitude less than that derived from the naturally occurring radionuclides in seafood (Aarkrog et al. 1997).

In the framework of the Coordinated Research Programme "Sources of radioactivity in the marine environment and their relative contributions to overall dose assessment from marine radioactivity (MARDOS)," IAEA Marine Environment Laboratory (MEL) coordinated a marine radioactivity study with the aim of comparing radiation doses delivered to the human population through ingestion of anthropogenic ^{137}Cs and natural ^{210}Po in sea food. These two radionuclides were chosen, as they are the most representative of each of the two classes of marine radioactivity on a global scale. ^{137}Cs is the most abundant anthropogenic radionuclide in the marine environment and ^{210}Po is the main contributor to doses from natural radionuclides by ingestion of seafood. Concentrations of ^{137}Cs and ^{210}Po in seawater and biota (fish and shellfish) have been estimated for the FAO fishing areas on the basis of measurements carried out in recent years. While ^{210}Po is uniformly distributed in seawater at a concentration of about 1 mBq L^{-1}, the ^{137}Cs concentrations in the world's oceans and seas differ considerably (Fig. 1). Collective doses were calculated for each FAO area using radioactivity data for water and biota. A reasonably good agreement was found between the results calculated by both methods. The collective effective dose commitment for ^{137}Cs in marine food estimated for the year 2000 is about 120 person Sv with an estimated uncertainty of 50%. The corresponding dose from ^{210}Po is about 30,000 person-Sv per year with an estimated uncertainty within a factor of five. The average annual individual dose estimated for the world population from the seafood consumption is about 0.03 μSv from ^{137}Cs and 9 μSv from ^{210}Po. The annual dose from ^{137}Cs for a hypothetical critical group living on the northeast Atlantic coast and consuming 100 kg of fish and 10 kg of shellfish per year would be 3 μSv, while the contribution from ^{210}Po would be 160 μSv. These values are well below the value of 1 mSv accepted for the public. The results confirm that the dominant contribution to doses comes from natural ^{210}Po in fish and shellfish and that the contribution of anthropogenic ^{137}Cs is generally negligible (100 to 1,000 times lower).

However, in the course of studies of the biological uptake of fallout much valuable information on marine biological processes and their interaction with both fallout radionuclides and their elemental analogues was obtained. In terms of the extent of their uptake, over the years, various reports have summarized the experimental and field observations that have led to refinements of the relevant parameters, concentration factors, for at least the major marine organisms categories and, where available,

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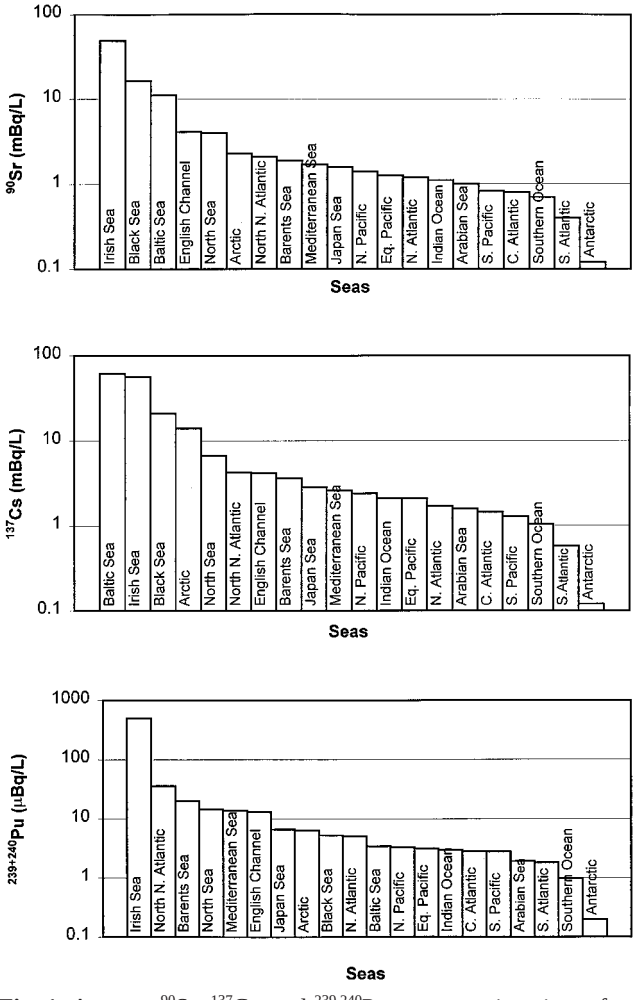

Fig. 1. Average ^{90}Sr, ^{137}Cs, and ^{239,240}Pu concentrations in surface waters of the world oceans and seas adjusted to the year 2000.

the component tissues and organs [Gomez et al. 1985; IAEA 1985b (revised and updated in IAEA 2001)]. It has been found that the concentration factors (CFs) differ substantially for different radionuclides (elements) as well as for different marine organisms. For example for plutonium the CFs as low as 0.5 were found for bottom fish, about 50 for octopus and about 100,000 for phytoplankton (Fowler 1983). The variety and volume of studies on which these summaries were based is both broad and massive. It has led to an exponential increase in the knowledge base of the transfer of radionuclides and critical elements, such as toxic metals, to marine organisms. This has been especially useful in understanding and assessment of the impact of both nuclear and non-nuclear marine pollution in coastal areas—which are impacted by land based contaminants and which are used extensively in terms of their marine food resource.

One development of such studies was the concept of using marine organisms as biological indicators. The concept is that they reflect the marine environment in which they live in a way that has been understood by the studies of their interaction with pollutants—both nuclear and non-nuclear. So by regular collection and analyses, a picture can be developed of the time-varying pollutant record in specific areas and how they compare from place to place. Perhaps the best known example of this was the program called Mussel Watch (Goldberg et al. 1978). Originally an United States program, this developed into a global program in which the blue mussel, *Mytilus Edulis*, was used to monitor contaminants in its locality. Although this approach has been used to monitor coastal environments with respect to metals, organics, and radionuclides, the early demonstration of the technique benefited from the example of fallout monitoring. An interesting aspect of the U.S. Mussel Watch data on fallout plutonium was the finding of much higher levels of plutonium in West Coast mussels relative to those on the East Coast. In hindsight, it became clear that the West Coast mussels were in fact reflecting the influx to Californian coastal waters of plutonium rich sub-surface waters that were being upwelled on to the shelf (Sholkovitz 1983).

Fallout radionuclides have been used in several other marine biological areas and have contributed to the growth of basic scientific knowledge there. Some radionuclides (^{90}Sr and plutonium) accumulate in the coral layers created annually and reflect seawater levels of each radionuclide. So, for example, in the dating of corals, coral rings are well established dating tools and fallout radionuclides have been used in such studies. They have on the one hand been able to confirm the reliability of the annual ring formation dating, and, in addition, have recorded the time-history of fallout levels in the waters in which they grew (Purdy et al. 1989).

GLOBAL FALLOUT IN THE OCEAN—50 YEARS LATER

As a way of concluding this review of 50 years of the uses of fallout in ocean science, some brief observations can be made on the state of the fate of this suite of radionuclides at the start of the 21st century. The passage of several decades since their ocean arrival has led to their dispersal as described previously. To close this review, an account is presented of an ongoing IAEA project being conducted by the Monaco Laboratory [Marine Environment Laboratory (MEL)] to capture the present global picture of fate of fallout in the world's oceans.

One of the most recent and the most comprehensive open ocean studies of anthropogenic radionuclides in the water column has been the IAEA-MEL research program on Worldwide Marine Radioactivity Studies (WOMARS). The primary objective was to develop an understanding of the present open ocean distribution of radionuclides in the water column and sediment and to contribute to scientific knowledge of the processes, which affect radionuclide behavior in the marine environment. Also, this will enable easy identification of any contributions over established background levels due to additional nuclear discharges or accidents (IAEA 2002a).

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Three anthropogenic radionuclides (^{90}Sr, ^{137}Cs, and ^{239,240}Pu) were chosen as the most important and radiologically typical of each class of marine radioactivity. They are the most abundant anthropogenic radionuclides present in the marine environment and they may lead to the highest radiation doses to humans and marine biota.

For the purposes of the WOMARS project, the world ocean was divided into latitudinal belts for which average radionuclide concentrations were estimated. ^{90}Sr, ^{137}Cs, and ^{239,240}Pu concentrations in surface waters were found to vary considerably, still preserving the latitudinal dependence observed in the sixties, with lowest values in the southern and highest values in the northern latitudes. Fig. 1 shows averaged ^{90}Sr, ^{137}Cs, and ^{239,240}Pu concentrations in surface waters of the world ocean adjusted to the year 2000. The results confirm that the main source of these radionuclides in the marine environment is still global fallout. Higher radionuclide concentrations observed in the northeast Atlantic Ocean (the Irish and North Seas) and the Arctic are due to the release and transport of the radionuclides from Sellafield and La Hague reprocessing plants. The Baltic, Black, and Mediterranean Seas were the main reservoirs for radionuclides released after the Chernobyl accident. The ^{90}Sr distribution in surface waters shows a similar pattern to that of ^{137}Cs, confirming the average global fallout ^{137}Cs/^{90}Sr ratio of 1.6 ± 0.1, with the exception of seas affected by discharges and the Chernobyl accident.

Further, time trends in radionuclide concentrations were studied and mean residence times of radionuclides in surface waters of the world ocean were estimated. The results confirm a similar residence time for ^{90}Sr and ^{137}Cs in surface waters of 27 ± 2 y. A lower value of 13 ± 1 y has been obtained for ^{239,240}Pu.

Entire water column radionuclide time series were developed in the same way as the surface water time series for a few stations in the Pacific and Atlantic Oceans. By re-visiting some of the GEOSECS stations, it was possible to evaluate water column radionuclide data over time. As an example, Fig. 2a and b show ^{137}Cs and ^{239,240}Pu profiles for the northwest Pacific Ocean obtained from the GEOSECS expedition (Bowen et al. 1980), from the Hakuho Maru expedition (Nagaya and Nakamura 1984), and from the IAEA '97 Pacific Ocean Expedition (Livingston et al. 2001). Both the ^{137}Cs and ^{239,240}Pu profiles show typical behavior for these radionuclides in the water column. Twenty-four years later, the sub-surface ^{239,240}Pu maximum had become much smaller

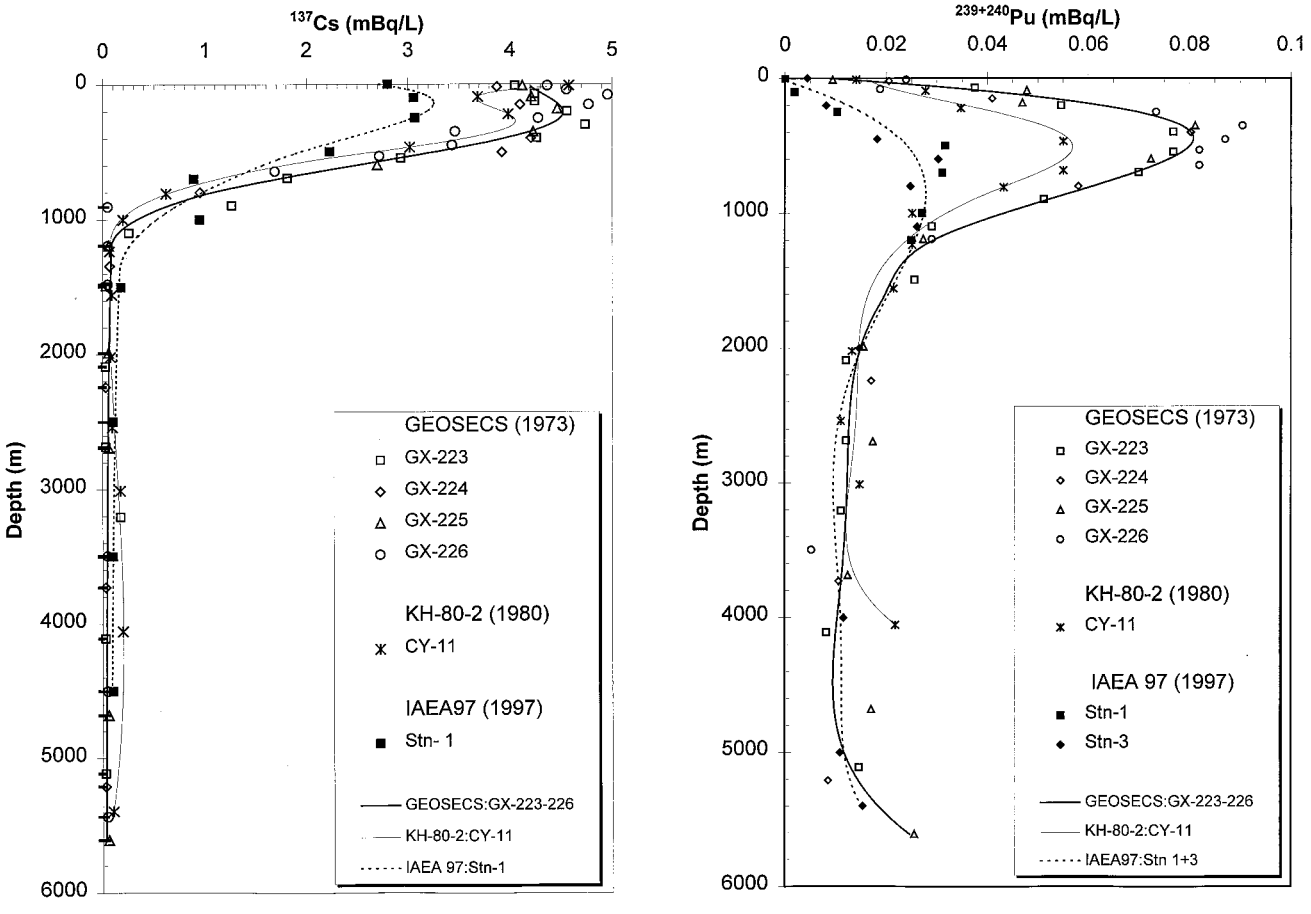

Fig. 2. ^{137}Cs (a) and ^{239,240}Pu (b) profiles in water column of the GEOSECS (1973), Hakuho Maru (1980), and IAEA (1997) stations in the central northwest Pacific Ocean.

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and less pronounced (a decrease by about a factor of four) and had moved to deeper layers (from 450 to 850 m). The decline in the ^{137}Cs maximum is clearly seen as well (the data were decay corrected to 1 January 1997), although the decrease 24 y later is less than a factor of two. This observation would emphasize that the observed changes in plutonium over time could not only be caused by the association of plutonium with sinking particles, but also could be due to physical circulation in the upper water column which has brought water masses bearing significantly lower levels of fallout radionuclides advectively into the region.

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This paper is dedicated to the life and inspiration of V. T. Bowen, Woods Hole Oceanographic Institution, Woods Hole, MA.

REFERENCES

Aarkrog A, Dahlgaard H, Nilsson K, Holm E. Studies of plutonium and americium at Thule, Greenland. Health Phys 46:29–44; 1984.

Aarkrog A, Baxter MS, Bettencourt AO, Bojanowski R, Bologa A, Charmasson S, Cunha I, Delfanti R, Duran E, Holm E, Jeffree R, Livingston HD, Mahapanyawong S, Nies H, Osvath I, Pingyu L, Povinec PP, Sanchez A, Smith JN, Swift D. A comparison of doses from 137Cs and 210Po in marine food: A major international study. J Environ Radioact 34:69–90; 1997.

Bacon MP, Huh, C-A, Fleer AP, Deuser WG. Seasonality in the flux of natural radionuclides and plutonium in the deep Sargasso Sea. Deep-Sea Res 32:273–286; 1985.

Benninger LK, Krishnaswami S. Sedimentary processes in the inner New York Bight: evidence from excess 210Pb and 239,240Pu. Earth Planet Sci Lett 53:158–174; 1981.

Benninger LK, Aller RC, Cochran JK, Turekian KK. Effects of biological sediment mixing on the 210Pb chronology and trace-metal distribution in a Long Island Sound sediment core. Earth Planet Sci Lett 43:241–259; 1979.

Bowen VT, Sugihara TT. Marine geochemical studies with fallout radioisotopes. In: Proceedings of the second U.N. International Conference on the Peaceful Uses of Atomic Energy, Geneva, 18. New York: United Nations; 1958: 434–438.

Bowen VT, Sugihara TT. Oceanographic implications of radioactive fallout distributions in the Atlantic Ocean: from 20° N to 25° S, from 1957 to 1961. J Mar Res 23:123–146; 1965.

Bowen VT, Noshkin VE, Livingston HD, Wong KM. Cesium 137 to strontium 90 ratios in the Atlantic Ocean 1966 through 1972. Limnology Oceanography 91:670–681; 1974.

Bowen VT, Noshkin VE, Livingston HD, Volchok HL. Fallout radionuclides in the Pacific Ocean; vertical and horizontal distributions, largely from GEOSECS stations. Earth Planet Sci Lett 49:411–434; 1980.

Broecker WS. Radioisotopes and the rate of mixing across the main thermocline of the ocean. J Geophys Res 71:5827–5836; 1966.

Buesseler KO. The isotopic signature of fallout plutonium in the North Pacific. J Environ Radioactivity 36:69–83; 1997.

Buesseler KO, Sholkovitz ER. The geochemistry of fallout plutonium in the North Atlantic: II. 240Pu/239Pu ratios and their significance. Geochim. et Cosmochim Acta 51:2623–2637; 1987.

Carpenter R, Beasley TM, Zahnle D, Somayajulu BLK. Cycling of fallout (Pu, 241Am, 137Cs) and natural (U, Th, 210Pb) radionuclides in Washington continental slope sediments. Geochim. et Cosmochim Acta 51:1897–1921; 1987.

Cochran JK. Particle mixing rates in sediments of the eastern equatorial Pacific: Evidence from 210Pb, 239,240Pu and 137Cs distributions at MANOP sites. Geochim. et Cosmochim Acta 49:1195–1210; 1985.

Cochran JK, Livingston HD, Hirschberg DJ, Surprenant LD. Natural and anthropogenic radionuclide distributions in the northwest Atlantic Ocean. Earth Planet Lett 84:135–152; 1987.

Cochran JK, Buesseler KO, Bacon MP, Livingston HD. Thorium isotopes as indicators of particle dynamics in the upper ocean: results from the JGOFS North Atlantic Bloom Experiment. Deep-Sea Res I 40:1569–1595; 1993.

Craig H, Turekian KK. The GEOSECS program: 1973–1976 Earth Planet. Sci Lett 32:217–219; 1976.

Deuser WG. Seasonal and interannual variations in deep-water particle fluxes in the Sargasso Sea and their relation to surface hydrography. Deep-Sea Res 33:225–246; 1986.

Doney SC, Jenkins WJ. Ventilation of the deep western boundary current and abyssal western north Atlantic: estimates from tritium and 3He distributions. J Phys Oceanogr 24:947–965; 1988.

Druffel ERM, Williams PM, Livingston HD, Koide M. Variability of natural and bomb-produced radionuclide distributions in abyssal red clay sediments. Earth Planet Sci Lett 71:205–214; 1984.

Fine RA. Tracers, time scales, and the thermocline circulation: the lower limb in the North Atlantic Ocean. Rev Geophys (Suppl):1353–1365; 1995.

Fowler SW. Radioecological aspects of deep-sea radioactive waste disposal. In: Atti del 3e Convegno Nazionale di Radioecologia. Bologna, Italy: Associazioine Italiana di Protezione contro le Radiazioni; 1983: 236–258.

Goldberg ED, Bowen VT, Farrington JW, Harvey G, Martin JH, Parker PL, Risebrough RW, Robertson W, Schneider E, Gamble E. The mussel watch. Environmental Conservation 5:101–125; 1978.

Gomez LS, Marietta MG, Jackson DW. Compilation of selected marine radioecological data for the U.S. Subseabed Program: Summaries of available radioecological concentration factors and biological half-lives. Albuquerque, NM: Sandia National Laboratories; Report SAND84-2087; 1985.

Guegueniat P, Hermann J, Kershaw P, Bailly du Bois P, Baron Y. Artificial radioactivity in the English Channel and the North Sea. In: Radionuclides in the oceans, inputs and inventories. Paris: Les Editions de Physique; 1996: 121–154.

Hammond RR, Shirasawa TH, Minard DR. Fallout in the North Pacific Ocean. Washington, DC: US Atomic Energy Commission; USAEC Fallout Program Quarterly Summary Report No. HASL-171:357; 1966.

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Hanson RB, Ducklow HW, Field JG. The changing ocean carbon cycle, a midterm synthesis of Joint Global Ocean Flux Study. International Geosphere-Biosphere Programme Book Series (5). Cambridge: Cambridge University Press; 1999.

Hardy EP, Krey PW, Volchok HL. Global inventory and distribution of fallout plutonium. Nature 241:444–445; 1973.

Health and Safety Laboratory. Radiostrontium deposition at Atlantic Ocean weather stations. Washington, DC: US Atomic Energy Commission; Fallout Program Quarterly Summary Report; HASL-197:B:49; 1968.

International Atomic Energy Agency. Sediment Kds and concentration factors for radionuclides in the Marine Environment. Vienna: IAEA; IAEA Technical Report Series No 247:73; 1985a.

International Atomic Energy Agency. Behaviour of radionuclides released into coastal waters. Vienna: IAEA; IAEA TECDOC 329:1–183; 1985b.

International Atomic Energy Agency. Inventory of radioactive material entering the marine environment: Sea disposal of radioactive waste. Vienna: IAEA; IAEA TECDOC No 588; 1991.

International Atomic Energy Agency. Sediment Kds and concentration factors for radionuclides in the marine environment. Vienna: IAEA; IAEA Technical Report Series No 247:73; 2001.

International Atomic Energy Agency. Inventory of accidents and losses at sea involving radioactive material. Vienna: IAEA; IAEA TECDOC; 2002a.

International Atomic Energy Agency. Worldwide marine radioactivity studies (WOMARS). Vienna: IAEA; IAEA TECDOC; 2002b.

Jenkins WJ. The use of anthropogenic tritium and helium-3 to study subtropical gyre ventilation and circulation. Philos Trans R Soc Lond A325:43–61; 1988.

Jenkins WJ, Clarke WB. The distribution of 3He in the western Atlantic Ocean. Deep-Sea Res 23:481–494; 1976.

Joseph AB, Gustafson PF, Russell IR, Schuert EA, Volchok HL, Tamplin A. Sources of radioactivity and their characteristics. In: Radioactivity in the marine environment. Washington, DC: National Academy Press; 1971: 6–41.

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

Koide M, Griffin JJ, Goldberg ED. Records of plutonium fallout in marine and terrestrial samples. J Geophys Res 80:4153–4162; 1975.

Kupferman SL, Livingston HD, Bowen VT. A mass balance for 137Cs and 90Sr in the North Atlantic Ocean. J Mar Res 37:157–199; 1979.

Livingston HD. Fallout plutonium in western North Pacific sediments. Vienna: IAEA; IAEA-TECDOC-368:27–34; 1986.

Livingston HD, Bowen VT. Pu and 137Cs in coastal sediments. Earth Plan Sci Lett 43:29–45; 1979.

Livingston HD, Cochran JK. Determination of transuranics and thorium isotopes in ocean water: in solution and in filterable particles. J Radioanalytical Nucl Chem Articles 115:299–308; 1987.

Livingston HD, Jenkins WJ. Radioactive tracers in the sea. In: The future of oceanography. New York: Springer-Verlag; 1982: 163–191.

Livingston HD, Povinec PP, Ito T, Togawa O. The behaviour of plutonium in the Pacific Ocean. In: Kudo A, ed. Plutonium in the environment, Vol. 1, Radioactivity in the environment. New York: Elsevier Press; 2001.

Mitchell PI, Kershaw PJ, Leon Vintro L. Radioactivity in the Irish Sea: Past practices, present status and future perspectives. In: Radionuclides in the oceans, inputs and inventories. Paris: Les Editions de Physique; 1996: 155–175.

Nagaya Y, Nakamura K. 239,240Pu, 137Cs and 90Sr in the central Pacific. J Oceanogr Soc Japan 40:416–424; 1984.

Nagaya Y, Nakamura K. Artificial radionuclides in the western Northwest Pacific (II): 137Cs and 239,240Pu inventories in water and sediment columns observed from 1980 to 1986. J Oceanogr Soc Japan 43:345–355; 1987.

Naoaki V. Russia won't stop sea dumping but supplies details to Tokyo. Nucleonics Week (May) 20:17–18; 1993.

Nelson DM, Carey AE, Bowen VT. Plutonium oxidation state distributions in the Pacific Ocean during 1980–81. Earth Planet Sci Lett 68:422–430; 1984.

Noshkin VE. Transuranium radionuclides in components of the benthic environment of Enewetak Atoll. In: Transuranic Elements in the Environment. Washington, DC: US DOE; DOE/TIC-22800:578; 1980.

Noshkin VE, Bowen VT. Concentrations and distributions of long-lived fallout radionuclides in open ocean sediments. In: Radioactive contamination of the marine environment. Vienna: IAEA; IAEA Symp. SM-158/45:671–686; 1973.

Noshkin VE, Wong KM. Plutonium in the North Equatorial Pacific. In: Processes determining the input behavior and fate of radionuclides and trace elements in continental shelf environments. Conf-790382:11; 1980.

Nozaki Y, Yamada M, Nakanishi T, Nagaya Y, Nakamura K, Shitashima K, Tsubota H. The distribution of radionuclides and some trace metals in the water columns of the Japan and Bonin trenches. Oceanologica Acta 21:469–484; 1998.

Nuclear Energy Agency. Co-ordinated Research and Environmental Surveillance Programme related to sea disposal of radioactive waste. Paris: Organisation for Economic Co-operation and Development; CRESP Final Report 1981–1995; 1996.

Osvath I, Povinec PP, Baxter MS. Kara Sea radioactivity assessment. Sci Total Environ 237/238:167–169; 1999.

Povinec PP, Woodhead D, Blowers P, Bonfield R, Cooper M, Chen Q, Dahlgaard H, Dovlete C, Fox V, Froehlich K, Gastaud J, Groning, M, Hamilton T, Ikeuchi Y, Kanisch G, Kruger A, Liong Wee Kwong L, Matthews M, Morgenstern U, Mulsow S, Pettersson H, Smedley P, Taylor B, Taylor C, Tinker R. Marine radioactivity assessment of Mururoa and Fangataufa atolls. Sci Total Environ 237/238:249–267; 1999.

Purdy CB, Druffel ERM, Livingston HD. Anomalous levels of 90Sr and 239,240Pu in Florida corals: Evidence of coastal processes. Geochim. et Cosmochim Acta 53:1401–1410; 1989.

Robinson WL, Noshkin VE. Radionuclide characterization and associated dose from long-lived radionuclides in close-in fallout delivered to the marine environment at Bikini and Enewetak Atolls. Sci Total Environ 237/238:311–327; 1999.

Rooth CG, Ostlund HG. Penetration of tritium into the Atlantic thermocline. Deep Sea Res 19:481–492; 1972.

Sachs PL, Hammar TR, Bacon MP. A large-volume, deep-sea submersible pumping system. Woods Hole, MA: Woods Hole Oceanographic Institution; Technical Report WHOI-89-55; 1989.

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Schlosser P, Bayer R, Bonisch G, Cooper LW, Ekwurzel B, Jenkins WJ, Khatiwala S, Pfirman S, Smethie WM. Pathway and mean residence times of dissolved pollutants in the ocean derived from transient tracers and stable isotopes. Sci Total Environ 237/238:15–30; 1999.

Seymour AH. Radioactivity in the marine environment. Washington, DC: National Academy Press; 1971.

Sholkovitz ER. The geochemistry of plutonium in fresh and marine water environments. Earth-Science Reviews 19:95–161; 1983.

Sjoblom K-L, Salo A, Bewers JM, Cooper J, Dyer RS, Lynn NM, Mount ME, Povinec PP, Sazykina TG, Schwarz J, Scott EM, Sivintsev YV, Tanner JE, Warden JM, Woodhead D. International Arctic Seas Assessment Project. Sci Total Environ 237/238:153–166; 1999.

Smith JN. Why should we believe 210Pb sediment geochronologies? Editorial. J Environ Radioact 55:121–123; 2001.

Smith JN, Boudreau BP, Noshkin VE. Plutonium and 210Pb distributions in northeast Atlantic sediments: subsurface anomalies caused by non-local mixing. Earth Planet Sci Lett 81:15–28; 1986.

Smith JN, Ellis KM, Naes K, Dahle S, Matishov D. Sedimentation and mixing rates of radionuclides in Barents Sea sediments off Novaya Zemlya. Deep Sea Res II 42:1471–1493; 1995.

Volchok HL. 90Sr deposition in the Atlantic Ocean. Washington, DC: US Atomic Energy Commission; USAEC Health and Safety Laboratory, Fallout Program Quarterly Summary Rep. HASL-181:I-25–I-30; 1967.

Volchok HL, Kleinman MT. 90Sr deposition in the Atlantic Ocean. II. Washington, DC: US Atomic Energy Commission; USAEC Health and Safety Laboratory, Fallout Program Quarterly Summary Rep. HASL-197:1227–1232; 1968.

Volchok HL, Bowen VT, Folsom TR, Broecker WS, Schuert EA, Bien GS. Oceanic distributions of radionuclides from nuclear explosions. In: Radioactivity in the Marine Environment. Washington, DC: National Academy Press; 1971: 42–89.

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