

Report Rapport



Atomic Energy
Control Board

Commission de contrôle
de l'énergie atomique

INFO--0324

CA9200011

TRANSFER OF SUSPENDED PARTICLES
FROM LIQUID EFFLUENTS OF
NUCLEAR GENERATING STATIONS
THROUGH THE ENVIRONMENT

by

Canada



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July 1989

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ABSTRACT

Due to the complexity of the environmental transfer of suspended particles in aquatic systems, the available literature usually deals with specific pathways and mechanisms of the transfer process. This paper attempts to give a brief overview of the entire transfer process. Potential routes of transfer in both the marine and freshwater environments are examined, and tentative conclusions presented.

This work was performed while the author was employed by AECS under the McMaster University cooperative program.

RÉSUMÉ

À cause de la complexité du transfert environnemental des particules suspendues dans les systèmes aquatiques, la bibliographie disponible traite habituellement de voies et de mécanismes spécifiques du processus de transfert. Ce document tente de survoler le transfert entier. Les voies potentielles de transfert dans les milieux marins et d'eaux douces sont examinées, et des conclusions tentatives y sont présentées.

Ce travail a été complété lorsque l'auteur travailla en tant qu'étudiant à la CCEA dans le cadre du programme coopératif de l'Université McMaster.

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Introduction

Radionuclide releases to the environment from Canadian nuclear generating stations are regulated by the Atomic Energy Control Board (AECB). To ensure that the risk to the general public is kept as low as reasonably achievable, the AECB must have an adequate understanding of the transfer pathways, and ultimate repositories of the radionuclide.

The following literature study was conducted to determine the modes of suspended particle transfer from liquid effluents released by nuclear generating stations through the environment. The majority of studies reviewed dealt with sources (fuel reprocessing, atomic bomb testing, etc.) that introduce more significant quantities of radionuclides into the environment than do nuclear generating stations. However, these sources release the same radionuclides as do nuclear generating stations, and their environmental behaviour can thus be used to infer the behaviour of radionuclides present in liquid effluents of nuclear generating stations.

Considerable quantities of transuranic radionuclides have been discharged directly into the coastal zone as a result of nuclear reprocessing activities (Fowler et al., 1983). Amongst long-lived nuclides, Pu-239 and Pu-240 (half-lives 2.4×10^4 and 6.6×10^3 years respectively) are interesting due to their relative abundance in wastes arising from such processes, as well as to their radiotoxicities. Also, Pu-241 is of interest because it is the parent of Am-241 ($T_{1/2} = 431$ years) which decays to Np-237 ($T_{1/2} = 2.2 \times 10^6$ years) (Aston and Fowler, 1984a). In addition, Tc-99 enters the environment as a result of nuclear fuel reprocessing, nuclear power production, nuclear waste storage, nuclear weapons testing and pharmaceutical use (Fowler et al., 1981).

Ocean basins are the ultimate sinks for low-level radioactive wastes released under controlled conditions from a variety of land-based facilities. Radioactive effluents are introduced either directly to coastal seawater or to river water in which they undergo mixing and dilution before being discharged into the sea. Regardless of their way of introduction, radioactive materials are transported away from the zone of discharge and dispersed into receiving waters by currents and turbulent diffusion (Ballestra et al., 1984). Although discharges are released to both freshwater and seawater environments, the majority of the available literature deals with the marine environment and consequently is the focal point of this review.

Chapter 1 - Transport Characteristics

1.1 Profiles with Depth

Most studies have identified sediments as the eventual repository for Pu-239, Pu-240 and Am-241 in marine waters, and sedimentation of particulate materials has been proposed as the primary means by which surface-introduced transuranic elements are transported to sediments. Experiments conducted during 1980 in the North East Pacific noted a strong sub-surface Pu (Pu-239 and Pu-240) concentration maximum at 250 m (Fowler et al., 1983), whereas using more closely-spaced sampling in 1974, Bowen et al., (1980), located a sub-surface Pu maximum at 465 m over a widespread area. Bowen et al., (1980) also reported that between 1974 and 1978, the depth of the Pu maximum had, in fact, moved downward to 515 m. The different profiles suggest that the depth of the sub-surface Pu maximum may not be the same everywhere in the Pacific but may change, particularly near the coast where lateral advection becomes important. As with Pu, a smaller but widespread Am-241 maximum appeared to exist between 250 and 750 m depth. Bowen et al., (1980) also observed an Am-241 sub-surface maximum in the Pacific but at a slightly greater depth (750 m) than that of Pu.

In Mediterranean studies, transuranic elements were vertically transported faster compared with Cs-137 in the upper few hundred metres, and vertical transport of Pu in the Mediterranean is rather slow compared with the estimated sinking rate of Pu in the North Atlantic. The vertical distribution of Am-241 is more complicated than that for Pu isotopes. Nevertheless, concentrations of Am-241 below 500 m, contrary to those of Pu, tend to be higher than those above 250 m. This vertical distribution of Am-241 is considered to reflect its complex delivery characteristics; in addition to the supply at the surface (effluent or fallout) followed by vertical transport through the water column or lateral advective transport, Am-241 is produced in situ from the decay of Pu-241 (half-life 14.9 years). Thus, an understanding of Am-241 concentration below 500 m, relative to upper layers, must take into account both transport and in situ build up (Fowler et al., 1983).

In Mediterranean water columns, the vertical distribution of transuranic nuclides is generally characterized by the following features:

- 1) the subsurface concentration maxima of Pu appear around 250 m depth,
- 2) the substrate concentration maxima appear below 500 m in depth, and
- 3) changes with time of these vertical distributions are likely to be slow.

These characteristic vertical distribution patterns of Pu and Am-241 are postulated to be caused by the difference in their affinity to various sorts of particles sinking through the Mediterranean water column. The subsurface concentration maximum of Pu has also been observed in the Pacific but at much greater depths, between 400-700 m.

The fraction of Pu involved in the subsurface maximum in the Pacific behaves as a "soluble" tracer, being moved largely by water mixing processes with more horizontal than vertical components. Vertical displacement of the Pu maximum in the Mediterranean appears to be very slow - the depth difference of the Pu maximum between the Mediterranean and the Pacific, ~ 250 m versus ~ 500 m below the surface, represents the difference in their sedimentation regimes. Similarly, the variation of the depth and concentrations of the Am-241 maxima are likely to depend strongly on sedimentation regimes and on prevailing hydrographical conditions (Fukai et al., 1983).

1.2 Mechanisms Varying the Concentration of Transuranic Nuclides with Depth

The following describes the various mechanisms responsible for the concentration variations of transuranic elements with depth as presented in the preceding discussion. In studies of the N.E. Pacific, it appears that Pu is preferentially scavenged by particulate matter from surface waters, a hypothesis supported by the results from filtered particle samples that indicate that approximately 32% of the Pu and 21% of Am-241 is associated with particulate matter in surface waters (Table 1). The Pu fraction associated with suspended particles is substantially higher in the N.E. Pacific study than has been observed either in the North Pacific (18%) or the Mediterranean (~ 4% - 5%). This increase in the Pu fraction associated with suspended particles appears to be due to the high particulate load (~ 3.5 mg dry l^{-1}) associated with phytoplankton "bloom" (i.e., peak of growth) which was observed in the surface waters at the time of sampling.

Laboratory studies (Fisher et al., 1983b) have shown that phytoplankton can effectively accumulate transuranic elements, reaching concentration factors of $\sim 10^5$ for both Pu and Am-241 (Fowler et al., 1983). Considerable insight into transuranic element behaviour in the water column has been gained by examination of the activity ratios of the various radionuclides in suspended particles. The data indicate that, except in surface waters, Am-241:Pu-239 and Pu-240 ratios in unfiltered seawater tend to increase between 250 and 1500 m, although not in any regular fashion. The increase in the ratio observed with depth further supports the contention that Am-241 is moving downward in the water column more rapidly than Pu-239 and Pu-240.

There is convincing evidence for fractionation of transuranic elements between the aqueous and particulate phase from the analyses of Am-241 and Pu in larger, fast sinking particles. To explain the reason for fractionation between Pu and Am-241 in the Mediterranean water column, it has been postulated that the inorganic component of the Mediterranean particle population is much higher than in the Atlantic, and that these inorganic particles preferentially retain Am-241 over Pu, leading to greater downward transport of Am-241 (Holm et al., 1980). The relatively high Am-241:Pu ratios (maximum 2.8) in the N.E. Pacific experiment support the hypothesis of an overall enrichment of Am-241 over Pu in oceanic particles. From the data, there is a strong suggestion that there is a net accumulation of Pu and Am-241 on the particles with depth (Fowler et al., 1983).

1.3 Significance of Zooplankton Pellets in Transport Processes

The bulk of particulate matter collected in sediment traps in the upper few hundred metres of the water column consist of zooplankton pellets. Experiments in the N.E. Pacific suggest that freshly produced zooplankton fecal pellets, already richer in Am-241 and Pu than the organisms' food (principally phytoplankton), can scavenge or accumulate more of the transuranic elements (Am-241 to a greater extent than Pu) as they sink through the water column and/or are repackaged by other foraging animals during their descent. The trend of increasing transuranic concentration in large particles with depth suggests active scavenging of these elements from the aqueous phase.

Reactive transuranic elements are known to be strongly sorbed by clay particles which are readily filtered by zooplankton and incorporated into fecal pellets. Thus, such sinking biogenic aggregates with their incorporated transuranic element load could feasibly scavenge additional clay particles during their descent and rapidly reach depths of up to 3200 m in less than 60 days (Deuser et al., 1981). From sedimentation studies in the Atlantic and Pacific, it has been found that 60% to 90% of the total flux in the deep ocean is biogenic (i.e., produced by living organisms). While it appears from visual observations that only 2 to 3% of the total flux is provided by intact fecal pellets, it is becoming increasingly evident that large biogenic aggregates such as fecal pellets are responsible for transporting even the very fine particles to depth in the open ocean, either by concentrating the particles or by scavenging them during descent (Fowler et al., 1983).

The relatively rapid vertical transport of Am-241 plus the in situ buildup of Am-241 from Pu-241 results in an increase of the Am-241/Pu-239 and Pu-240 activity ratios in the deeper water layer. In addition, advective transport may change the ratios at certain depths. All these factors complicate the interpretation of the vertical variation of Am-241/Pu-239 and Pu-240 ratios (Fukai et al., 1979).

Noskin and Bowen (1973) proposed a model which associates Pu with a mixed population of particles, sinking at rates of 70 - 392 m a⁻¹ and suggested that algae and, perhaps, the main phytoplanktonic biomass may have an important role in determining the chemical and physical forms of Pu predominant in the ocean. It is known that for several elements, zooplankton metabolism may be an important biological factor in the removal of the elements from the surface layer of the ocean to depth. As mentioned previously, zooplankton fecal pellet deposition is thought to be an important vector in the vertical oceanic transport of elements.

Studies of a common zooplankton species, the euphausiid Meganycitipanes norvegica, indicates that 99% of the Pu taken up by M. norvegica is excreted in fecal pellets. Euphausiid pellets sink at rates ranging from 126 to 862 metres per day, and they decompose relatively slowly; it seems clear that they have the potential to reach the sea bottom in most areas. If

M. norvegica data can be considered as typical of the marine zooplankton biomass as a whole, a rough estimate of the removal time of Pu from the upper mixed layer of the ocean by zooplankton fecal pellets alone can be made. Estimates of the removal time of Pu from the upper mixed layer due to zooplankton fecal pellets alone is of the same order as for estimates of removal from the upper mixed layer by all routes (3.6 a versus 1 a). This suggests that fecal pellet removal has a significant role in the removal of Pu from the surface layers of the sea (Cherry et al., 1977).

There exists some conflict as to whether water movement or fecal pellet sinking is the major mode of vertical nuclide transport. A simplified scenario for the vertical transport of Pu and Am-241 through the Mediterranean has been postulated. In the upper layer, near-surface fractions of Pu and Am-241 are preferentially associated with particles from biogenic and terrigenous origin respectively. These particles sink under gravitational fields at various rates, depending on size and density. The fractions of Am-241 transported downwards are larger than Pu, since biological productivity in the open Mediterranean is generally low, hence the population of terrigenous particles exceed those of biogenic particles in most areas. At about 250 m depth, biogenic particles bearing, preferentially Pu, release Pu as well as Am-241 due probably to their dissolution, while the Am-bearing particles continue to sink further, accreting the released and ingrown Am-241 with them. Although the sinking rates of these particles are generally slow, fast-sinking particles reach the bottom, enriched substantially with Am-241 over Pu-239 and Pu-240. Below 500 m depth, the Am-241 concentration is maintained either by short distance association/release cycle of Am-241 with biogenic particles and slow rates of sinking of the main population of the Am-241 bearing particles (Fukai et al., 1983).

Mediterranean studies, (Linsalata et al., 1986), have found a systematic variation of Cs-137 concentration which suggests that the vertical distribution of Cs-137 is closely related to vertical water mass movements. The computed inventories confirm the assumption that Cs-137 in the Mediterranean remains in the water column long after its introduction, and its behaviour is chiefly influenced by water mass movements (Fukai et al., 1980).

In contrast, evidence has been presented (Karl and Knayor, 1983) that indicates that among other parameters, an increase in fecal pellet flux between 500 and 1100 m is a result of in situ rather than advective processes. This leads to the belief that large particle sinking is normally the predominant process controlling the vertical transport of Pu and Am-241 in the upper 750 m (Fowler et al., 1983).

Chapter 2 - Physical and Chemical Considerations

2.1 Effect of pH Levels

Turning to the behaviour of radionuclides in the freshwater environment, there are notable differences with the behaviour exhibited in the marine environment. Americium-241 in freshwater is unlikely to form particles to an appreciable extent at $\text{pH} \leq 8$. Murray and Fukai (1975) noted that the addition of Am-241 to the Var River (Nice, France) water at pH 8.0, which had been filtered through 0.1 or 0.45 μm Millipore filters, resulted in 64% (1.2 μm) to 100% (0.1 μm) particle formation of Am-241 within 10 hours; similar findings were reported for Pu-237 by the same authors. Theils (1983) observed that the Am-241 particle formation ($> 0.45 \mu\text{m}$) after 1 day was on the order of 30% for most lake water tested, but as high as around 70% at pH 8; similar findings for other freshwater systems have also been reported. Extrapolating from these laboratory studies to the field, one would expect that nearly all of the Am-241 or Pu in natural waters (at $\text{pH} \geq \sim 6$) would form particles $> 0.45 \mu\text{m}$. However, there is now some information for fractionation of Am-241 and Pu in various bodies of fresh and saltwater which indicate that the greatest fraction is usually dissolved (i.e., $< 0.45 \mu\text{m}$), particularly in low particulate waters. In Lake Michigan, typically only about 10% of Pu-239 and Pu-240 in the water column is present as particles $> 0.45 \mu\text{m}$, and there is minimal association with the colloidal fraction $> 0.003 \mu\text{m}$ and < 0.45 . Although, substantial variation exists in different regions, probably due to variation in the load of suspended particulate matter in the various water columns (Fisher 1985).

Examining the effect of pH more closely, the laboratory experiments with Pu-237 have shown that the particulate formation of hexavalent Pu in seawater (30% at pH 8) is much less than that of either quadrivalent Pu or trivalent Am-241 ($> 90\%$ at pH 8). Results suggest that a major portion of "soluble" Pu in seawater occurs in hexavalent form, while Am-241 is in trivalent form. Thus, the difference in oxidation state substantially influences the fractionation between Pu and Am-241 in the seawater medium (Holm et al., 1980).

As stated above, Murray and Fukai (1975) and Thiels (1983) identified pH as being an important determinant in Am-241 particulate formation in river and lake water. Rai et al., (1981) found that solubility of Am-241 in solution decreased approximately tenfold with each unit increase in pH. On the basis of thermodynamic data, they further argue that the likely form for aqueous Am is $\text{Am}(\text{OH})_2^{-1}$ above pH 5. Murray and Fukai (1975) reported that 97% of Am "formed" particles in filtered water, pH 8, after 1 day (Fisher 1985). Also, it is stated that Pu in seawater exists as its higher oxidation state in the dissolved fraction and as its lower states in the particulate form (Aston et al., 1984a).

There is some disagreement as to the degree to which Am is associated with particles. For example, Fisher (1985) reports that at pH ~ 8, at 24h, about 5% of the total water column Am was bound to particles > 1 μm and that there were small differences in Am content between 0.2 and 1 μm particles. In contrast, Thiels (1983) reported that at pH 8, ~ 30% (but as high as 70%) of the Am was bound to particles > 0.45 μm after 1 day. Additionally, naturally occurring dissolved organic matter (DOM) in freshwater appears to complex Am and Pu in some lakes (Nelson et al., 1980; Orlandini, 1981) and in laboratory experiments (Bertha and Choppin, 1978; Vangenechten et al., 1984), while in seawater there is no apparent complexation of these elements by DOM (Fisher et al., 1983a). The differences may be attributable to the lower levels of competing ions in freshwater systems or possibly to quantitative and qualitative differences in the DOM normally encountered in freshwater and marine environments (Fisher, 1985).

2.2 Sedimentary Trapping Mechanisms

Experiments in the N.E. Mediterranean reveal higher seawater concentrations of Pu-239 and Pu-240 taken from bottom layers (50 m depth), as compared with the corresponding surface water stations (5 m). It is suggested that suspended material containing high Pu-239 and Pu-240 concentrations, brought by resuspension of bottom sediments and/or by land runoffs, contribute significantly to the elevated Pu-239 and Pu-240 concentrations noted in the

bottom waters. Nevertheless, it is reasonable to assume that these higher concentrations in the bottom waters indicate an effective trapping mechanism for Pu-239 and Pu-240, supplied by runoff into the sea, operating in the near shore bottom layers (Fukai and Murray, 1978).

Effective trapping is also evident from analysis of particulate distribution in rivers and estuaries. A comparison of the Pu-239 and Pu-240 concentration in river water with that in estuary water revealed less Pu-239 and Pu-240 than did unfiltered mid-river water, with the concentrations of Sr-90 and Cs-137 remaining constant while the water flowed from the river into the sea. These observations suggest that the supply of Pu-239 and Pu-240 through rivers or from direct land runoff into the sea has little influence on the offshore inventory of Pu-239 and Pu-240 due to effective trapping of Pu-239 and Pu-240 entering estuarine and near-shore areas (Fukai and Murray, 1978).

Measurements of Cs-137, Pu-238, Pu-239 and Pu-240 and Am-241 were carried out on river water, as well as its suspended matter collected seasonally in 1977 from two Mediterranean rivers, the Var and the Rhone. The Var and the Rhone are considered to represent, respectively, Mediterranean rivers not affected by reactor effluent releases and those receiving radioactive effluents. Essentially, identical average Pu-238/Pu-239 and Pu-240 (0.26) and Am-241/Pu-239 and Pu-240 (~ 0.35) activity ratios were obtained for both rivers. These ratios were not significantly different statistically, although the concentration levels between the two rivers are different by a factor of approximately 10 (Fukai et al., 1981).

In both rivers, the percentages for the fraction of matter in suspension exceeded 65% for all nuclides studied, and especially for Pu-239 and Pu-240, they exceeded 90%. In summary, on arrival at the estuary, suspended matter is quickly deposited to the bottom without allowing much time for interaction with seawater. Thus, the major fraction of these radionuclides carried by rivers does not affect the distribution of the radionuclides in seawater beyond the vicinity of the estuarine areas (Fukai et al., 1981).

On the basis of literature for freshwater and estuarine sediments, organic matter (humics), which is chiefly of terrestrial origin, may play a much more important role than might be expected on the basis of its fractional content

in sediments. In view of the diversity of chemical properties of radionuclides, it would be expected that their behaviour in sediments would be equally diverse. There is, however, substantial evidence showing that most radionuclides and trace metals are strongly associated with suspended particulate matter and bottom sediments and that the primary pathway of contamination of sediments is by way of sedimentary metal-contaminated particles. This phenomenon is, moreover, not limited to marine systems but applies to freshwater bodies as well. In estuarine systems, the solid-liquid distribution behaviour of radionuclides appears to be essentially governed by organic matter and hydrous oxides, the relative effect of these two sinks being dependent on phase component concentrations and redox conditions (Cremers and Herrion, 1985).

2.3 Adsorption and Desorption Processes

Americium, Pu and other transuranic elements (e.g., Cm, Cf) are very reactive for surfaces in aquatic or marine environments. K_d^* values on the order of $10^4 - 10^5$ have been reported for sediments in freshwater and around 10^5 for suspended matter in rivers. Given these high affinities for suspended matter and the high number of particles normally encountered in rivers, Simpson et al., (1980) estimate that most river-borne Pu would be associated with particles, although the degree can vary seasonally, as in the Hudson River.**

The finding of relatively low transuranic particle association in the Great Lakes (Wahlgren and Mawholl, 1975) suggests that the high particle association in rivers is attributable to adsorption to suspended matter rather than particle formation of the elements. As in marine systems, the extent of particle association of Am-241 in freshwater bodies should primarily be a function of the number of suspended particles, and should, therefore, be low in particulate (e.g., many oligotrophic systems) or filtered water. In acidic water, most Am-241 would likely be in the "dissolved" phase (Fisher, 1985).

*The distribution of radionuclides (K_d) between pore waters and settled sediments.

**Desorption of radiocesium during salinity intrusions (due to dry conditions) has been reported in brackish water areas of the Hudson River Estuary and is a major mechanism for removing radiocesium (Linsalata et al., 1986).

Concerning the adsorption of long-lived radionuclides onto sediments, it has been found that transuranic elements Pu and Am are found to rapidly and effectively adsorb onto the deep-sea and near-shore sediments. Rates of adsorption are, for these elements, so rapid that equilibrium K_d values are frequently obtained in a matter of tens of hours to a few days. In contrast to the transuranics, Tc (valences IV or VII) is very poorly adsorbed to deep sea sediments and has the potential to be lost rapidly to overlying waters. Furthermore, Tc bioavailability from contaminated sediments to benthic fauna, e.g., clams, appears to be low, with most Tc taken up into the shell and subsequently lost at very slow rates. This observation suggests that following acute contamination, the animals living in the deep-sea environment would still retain Tc in their tissues long after the radionuclide has disappeared from the sediment.

Experimental results suggest that Pu and Am are immobile in both oxic and anoxic sediments, while it should be noted that there is contradictory evidence that Pu may be released from sediments which are exposed to regular seasonal fluctuation in redox conditions. Plutonium adsorption to sediments as expressed by K_d values is about two orders of magnitude higher than those for Tc (Aston and Fowler, 1984b). In summary, experiments examining the desorption of radionuclides upon introduction into uncontaminated seawater indicate that the transuranic nuclides are not easily desorbed. However, unlike transuranics, Tc is easily desorbed from sediments (Aston, 1983).

Previous determinations of the distribution coefficient (K_d) of Pu on marine sediments indicated that Pu was bound to sediments to a degree intermediate between that of Sr-90 and Cs-137, i.e., having a distribution coefficient between 10^2 and 10^3 . Although the data suggests, when applicable to Pu-239 and Pu-240 in contaminated areas, that the Pu will be relatively strongly adsorbed to many marine sediments, the question whether Pu, once bound by sediment, cannot be released at the long run is difficult to answer (Duursma and Parsi, 1976).

It is clearly recognized that a great portion of nuclides released into coastal sea is adsorbed into marine sediment with the subsequent possibility of radioactive contamination of marine organisms and then transfer to man.

Studies indicate that the influence of seawater is larger than that of sediments (10^4 times larger for Mn-54, 10^3 times larger for Fe-59, Co-60, Zr-95, Nb-95, Ru-106-Rh-106 and Ce-144-Pr-144, 10^2 times larger for Zr-65, and $10 \cdot 10^2$ times larger for ^{115}mCd and Cs-137). Consequently, seawater can be considered as the main route and sediments as the secondary route in the case of quantitative comparison of the effect on the accumulation of radionuclides by marine organisms and in the transfer of radionuclides to human beings. It is well known that marine sediments accumulate radionuclides, but the relative importance of this phenomenon is still not clear for the overall transfer process of radionuclides from nuclear facilities to human beings via marine products. Seawater appears to give organisms approximately 10^4 times higher radioactivity than sediments in the accumulation of Mn-54 by organisms and $10 \cdot 10^2$ times higher radioactivity of Cs-137 when radioactivity in sediments is the same level as in seawater. The possibility of contamination of organisms by sediments cannot be neglected; however, in general, seawater affects the accumulation of radionuclides more than sediments. Figure 1 illustrates the transfer of radionuclides to human beings via marine organisms; the primary route is B and the secondary route is C, although sediments adsorb a great portion of radionuclides released into coastal sea regions (Ueda *et al.*, 1985).

2.4 Accumulation in Marine Biota

Studies indicate that marine plants and animals are able to concentrate radiocobalt (Co-58 and Co-60 being taken up at the same rate). Unicellular and pluricellular marine algae, as well as the higher plant Callitriche obtusangula, accumulate Mn-54 and I-131, the other nuclides (Cs-134, 136, 137, Sb-124) being taken up at a lower rate. Fish can accumulate 10 times more Cs-137 from water than from food, when the concentration factor of the food organism is 1 and the feeding amount is 6% the fish's body weight per day. On the assumption that marine fish prey on organisms which have concentration factors calculated by Thompson *et al.*, (1972), the principal pathway in the accumulation of radionuclides by marine fish in natural sea might be estimated as shown in Table 2. Food rather than water, supplies most of the Zn-65 and

Mn-54 accumulated by fish, while Sr-85, Ru-106 and Rh-106 are estimated to be mainly accumulated from the water. It is considered that the two pathways play almost the same role in the accumulation of Cs-137, Co-60, Ce-144 and Pr-144 (Suzuki et al., 1979).

The finding that particular radionuclides are taken up preferentially by marine organisms should be borne in mind for a meaningful estimation of the turnover rate of elements in the biological portion of biogeochemical cycles (Bonotto et al., 1981).

Conclusion

There is growing evidence, at least in the near shore and continental shelf areas, that certain radionuclides, including Pu and Am, are laterally transported into the region and subsequently removed to underlying sediments, principally through scavenging by inorganic particles (Fukai et al., 1978). It is now well established that the transuranic radionuclides Pu and Am eventually accumulate in sediments once they are introduced into the aquatic environment. Less well known is the extent to which these elements remain biologically available to organisms once they are bound to sediments. Some experiments have shown that there is preferential uptake of Pu over Am by sediment, contrasted to other experiments where the preferential uptake is reversed. In addition, from the data available so far, it has been concluded that, for both weakly bound Tc and the more strongly adsorbed transuranics, direct uptake from sediments is likely to be a quantitatively unimportant route for the transfer of waste radionuclides from contaminated sediments to the food chain and man (Aston and Fowler, 1984b).

Concerning the available literature, it should be noted that little attention has been given to the pathways by which radionuclides are incorporated into biota or to their retention and ultimate fate in the organism (Fowler et al., 1981). Also, as previously mentioned, the majority of the literature is concerned with the marine environment, with little attention given to freshwater pathways.

Canada has nuclear generating stations in Ontario, Québec and New Brunswick. In addition, two research establishments are located at Chalk River, Ontario and Pinawa, Manitoba. As the primary aquatic release point for reactor effluent is the freshwater pathway, with the exception of New Brunswick, a more comprehensive evaluation of freshwater transfer mechanisms would be desirable to analyze more thoroughly the Canadian situation.

Table 1: Concentration of transuranic elements associated with the particulate phase in seawater from the VERTEX I station, California, 25 August to 9 September 1980 (from Fowler et al., 1983)

Sample Depth* (m)	Sample Volume (L)	Pu-239 and 240 (particulate)			Am-241 (particulate)		
		Total fCi	aCi.L ⁻¹	% on Filters	Total fCi	aCi.L ⁻¹	% on Filters
Surface	1686	33 ± 4	20 ± 2	32 ± 7	23 ± 3	14 ± 2	21 ± 4
100	607	5 ± 1	9 ± 2	1.9 ± 0.4	8 ± 2	13 ± 4	10 ± 3
750	378.5	3 ± 2	9 ± 5	3 ± 2	11 ± 3	30 ± 8	10 ± 3

Table 2: Estimation of the principal pathway for the accumulation of radionuclides by marine fish
(from Suzuki et al., 1979)

Radionuclide	Concentration factor of food organism	$\frac{D_{cf} \cdot F_w}{F_f}^*$		Principal pathway
	(A)	Ff (B)	(B) (A)	
Ce-144 and Pr-144	20	60	3	food and water
Ru-106/Rh-106	3	60	2 x 10	water
Sr-85	2	80	4 x 10	water
Cs-137	40	10	2.5 x 10 ⁻¹	food and water
Mn-54	550	50	9 x 10 ⁻²	food
Zn-65	2000	40	2 x 10 ⁻²	food
Co-60	100	50	5 x 10 ⁻¹	food and water

* where D_{cf} = concentration factor of food, F_w = total body burden of each radionuclide via water and F_f = total body burden of each radionuclide via food

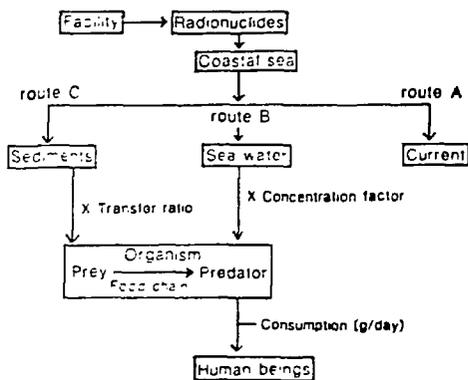


Fig. 1 Flux of radionuclides released into coastal sea.
(from Ueda et al.)

References

- Aston, S.R., (1983), Adsorption and Geochemical Partitioning of Long-Lived Radionuclides in Dumpsite Sediments. Interim Oceanographic Description of the NEA Dumpsite for the Disposal of Low-Level Radioactive Waste, NEA/OECD, Paris, pp. 97-100.
- Aston, S.R. and S.W. Fowler, (1984a). Experimental Studies on the Bioaccumulation of Plutonium from Sea Water and a Deep-sea Sediment by Clams and Polychaetes. J. Environ. Radioactivity, 1, pp. 67-78.
- Aston, S.R., and S.W. Fowler, (1984b). Experimental Studies on the Behaviour of Long-Lived Radionuclides in Relation to Deep-Ocean Disposal of Nuclear Waste. Radioactive Waste Management, Vol. 5, IAEA, Vienna, pp. 339-354.
- Ballestra, S., R. Bojanowski, R. Fukai and D. Vas (1984). Behaviour of Selected Radionuclides in the Northwestern Mediterranean Basin Influenced by River Discharge. International Symposium on the Behaviour of Long-Lived Radionuclides in the Marine Environment, (A. Cigna, C. Myttenaere, eds.), EUR 9214, CEC, Luxembourg, pp. 215-232.
- Beasely, T.M. and S.W. Fowler, (1976). Plutonium and Americium: Uptake from Contaminated Sediments by Polychaete Nereis Diversicolor. Marine Biology, 38, pp. 95-100.
- Bonotto, S., G. Carraro, S. Strack, A. Luttke, J. Colard, G. Koch and R. Kirchmam, (1981), Ten Years of Investigation on Radioactive Contamination of the Marine Environment. Impacts of Radionuclide Releases into the Marine Environment, IAEA, Vienna.
- Bertha, E.L. and G.R. Choppin, (1978). Interaction of Humic and Fulvic Acids with Eu (III) and Am (III), J. Inorg. Nucl. Chem. 40, pp. 655-658.
- Bowen, V.T., V.E. Noshkin, H.D. Livingston, H.L. Volochock, (1980). Fallout Radionuclides in the Pacific Ocean: Vertical and Horizontal Distributions. Earth and Planetary Science Letters, 49, pp. 411-434.
- Cherry, R.D., S.W. Fowler, M. Heyraud and J.J.W. Higgs, (1977). Rapid Removal of Plutonium from the Oceanic Surface Layer by Zooplankton Faecal Pellets. Nature, Vol. 266, pp. 623-624.
- Cremers, A. and P.N. Henrion, (1985). Radionuclide Partitioning in Sediments: Theory and Practice, in Seminar on Behaviour of Radionuclides in Estuaries, Commission of the European Communities, 24 pages.
- Dueser, W.G., E.H. Ross and R.F. Anderson (1981). Seasonality in the Supply of Sediment to the Deep Sargasso Sea and Implications for the Rapid Transport of Matter to the Deep Ocean. Deep Sea Research, 28, pp. 495-505.

- Duursma, E. and P. Parsi, (1976). Distribution of Plutonium-237 between Sediment and Sea Water. *Rapp. Comm. int. Mer. Médit.*, 23, 7, pp. 159-160.
- Fisher, N.S., (1985). On the Measurement of Particulate Association of Americium in Freshwater. *Water Res.*, Vol. 19, No. 1, pp. 79-84.
- Fisher, N.S., P. Bjerregaard, L. Huynh-Ngoc and G.R. Harvey, (1983a). Interactions of Marine Plankton with Transuranic Elements; II - Influence of Dissolved Organic Compounds on Americium and Plutonium Accumulation in a Diatom. *Mar. Chem.* 13, pp. 45-56.
- Fisher, N.S., P. Bjerregaard and S.W. Fowler, (1983b). Interactions of Marine Plankton with Transuranic Elements. I - Biokinetics of Neptunium, Plutonium, Americium and Californium in Phytoplankton. *Limnology and Oceanography*, 28, 432-447.
- Fowler, S.W., S. Ballestra, J. LaRosa and R. Fukai, (1983). Vertical Transport of Particulate-Associated Plutonium and Americium in Upper Water Column of the Northeast Pacific. *Deep-Sea Research*, Vol. 30, No. 12A, pp.1221-1233.
- Fowler, S.W., G. Benayoun, P. Parsi, M.W.A. Essa, and E.H. Schulte (1981). Experimental Studies on the Bioavailability of Technetium in Selected Marine Organisms. *In Impacts of Radionuclide Releases into the Marine Environment*, IAEA, Vienna, RITS No. 71, pp. 319-339.
- Fukai, R., E. Holm and S. Ballestra, (1979). A Note on Vertical Distribution of Plutonium and Americium in the Mediterranean Sea, *Oceanologica Acta*, Vol. 2, No. 2, pp. 129-132.
- Fukai, R. and C.N. Murray, (1978). Measurement of Pu 239 + 240 in the Northwestern Mediterranean. *Estuarine and Coastal Marine Science*, 6, 145-151.
- Fukai, R., S. Ballestra, M. Thein and J. Guion, (1981). Input of Transuranic Elements through Rivers into the Mediterranean Sea. *In Impacts of Radionuclide Releases into the Marine Environment*, IAEA, Vienna, pp. 3-14.
- Fukai, R., S. Ballestra and D. Vas, (1980). Distribution of Caesium-137 in the Mediterranean Sea. *In Management of the Environment*, (B. Patel ed.), pp. 353-360.
- Fukai, R., S. Ballestra and D. Vas, (1983). Characteristics of the Vertical Transport of Transuranic Elements through the Mediterranean Water Column. VI^{es} Journées Etud. Pollutions, Cannes, (C.I.E.S.M.), (1983), pp. 95-101.
- Holm, E., S. Ballestra, R. Fukai and T.M. Beasley, (1980). Particulate Plutonium and Americium in Mediterranean Surface Waters. *Oceanologica Acta*, Vol. 3., No. 2, pp. 157-160.

- Karl, D.M., and G.A. Knauer, (1983). Vertical Distribution, Transport and Exchange of Organic Matter in the Northeast Pacific Ocean: Evidence for Multiple Zones of Biological Activity. In Fowler, S.W., S. Balletra, J. LaRosa and R. Fukai (1983). Vertical Transport of Particulate-associated Plutonium and Americium in Upper Water Column of the Northeast Pacific Deep Sea Research, Vol. 30, No. 12A, pp. 1221-1233.
- Linsalata, P., D. Hickman and N. Cohen, (1986). Comparative Pathway Analysis of Radiocesium in the Hudson River Estuary: Environmental Measurements and Regulatory Dose Assessment Models. Health Physics Vol. 51, No. 3, pp. 295-312.
- Murray, C.N. and R. Fukai, (1975). Adsorption-Desorption Characteristics of Plutonium and Americium Sediment Particles in the Estuarine Environment: Studies Using Plutonium-237 and Americium-241. In Impacts of Nuclear Releases into the Aquatic Environment (IAEA, Vienna), pp. 179-192.
- Nelson, D.M. J.O. Karttunen, K.A. Orlandini and R.P. Larsen, (1980). Influence of Dissolved Organic Carbon on the Sorption of Plutonium to Natural Sediments. Argonne National Laboratory Report ANL-80-115, Pt. III, pp. 19-25.
- Noskin, V.W. and V.T. Bowen, (1973). Concentrations and distribution of long-lived fallout radionuclides in open ocean sediment. In Radioactive Contamination of the Marine Environment (IAEA, Vienna), pp. 671-686.
- Orlandi, K.A., (1981). Observations on the Behaviour of Americium in Natural Waters. Argonne National Laboratory Report ANL-81-85, Pt. III, pp. 53-36.
- Rai, D., R.G. Strickert, D.A. Moore and R.J. Seine, (1981). Influence of Americium Solid Phase on Americium Concentrations in Solutions. Geochim. cosmochim, Acta 45, pp. 2257-2265.
- Simpson, H.S., R.M. Trier and C.R. Olsen, (1980). Transport of Plutonium by Rivers. In Transuranic Elements in the Environment (W.C. Hanson ed.), U.S. Department of Energy, pp. 684-690.
- Suzuki, Y., M. Nakahara and R. Nakamura, (1979). Roles of Food and Sea Water in the Accumulation of Radionuclides by Marine Fish. Bulletin of Japanese Society of Scientific Fishers, 45(11), pp. 1409-1416.
- Theils, G.M.R., (1983). The Bio availability of the Transuranic Elements Pu 237 and Am 241 for the Pond Snail, Lymaea Stagnalis L., and their Behaviour in Selected Natural Surface Waters. Commission of the European Communities, Report EUR 8683 EN.
- Thompson, S.E., C.A. Burton, D.J. Quinn, and Y.C. Ng, (1972). UCRL-50564, Rev. (1).

- Ueda, T., R. Nakamura, and Y. Suzuki, (1985). Position of Sediments in the Transfer of Radionuclides Released into Coastal Sea to Human Beings. Bulletin of Japanese of Scientific Fisheries 51(8), pp. 1319-1324.
- Vangenechten, J.H.D., S. Van Puymbroeck, J. Bierkens, G. Van Keer and O. Vanderborcht, (1984). Speciation of Americium-241 in Fresh Water Systems: Effects of Inorganic and Organic Substances. In Environmental Transfer of Radionuclides to Man from Routine Releases of Nuclear Installations, International Atomic Energy Agency, Vienna.
- Wahlgren, M.A. and J.S. Marshall, (1975). The Behaviour of Plutonium and other Long-Lived Radionuclides in Lake Michigan: I. Biological Transport, Seasonal Cycling, and Residence Time in the Water Column. In Impacts of Nuclear Releases into the Aquatic Environment, IAEA, Vienna, pp. 227-243.