Chapter 3

MARINE BIOGEOCHEMISTRY OF RADIONUCLIDES

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Abstract

Radionuclides entering the ocean from runoff, fallout, or deliberate release rapidly become involved in marine biogeochemical cycles. Sources, sinks and transport of radionuclides and analogue elements are discussed with emphasis placed on how these elements interact with marine organisms. Water, food and sediments are the source terms from which marine biota acquire radionuclides. Uptake from water occurs by surface adsorption, absorption across body surfaces, or a combination of both. Radionuclides ingested with food are either assimilated into tissue or excreted. The relative importance of the food and water pathway in uptake varies with the radionuclide and the conditions under which exposure occurs. Evidence suggests that, compared to the water and food pathways, bioavailability of sediment-bound radionuclides is low. Bioaccumulation processes are controlled by many environmental and intrinsic factors including exposure time, physical-chemical form of the radionuclide, salinity, temperature, competitive effects with other elements, organism size, physiology, life cycle and feeding habits. Once accumulated, radionuclides are transported actively by vertical and horizontal movements of organisms and passively by release of biogenic products, e.g., soluble excreta, feces, molts and eggs. Through feeding activities, particles containing radionuclides are "packaged" into larger aggregates which are redistributed upon release. Most radionuclides are not irreversibly bound to such particles but are remineralized as they sink and/or decompose. In the pelagic zones, sinking aggregates can further scavenge particle-reactive elements thus removing them from the surface layers and transporting them to depth. Evidence from both radiotracer experiments and *in situ* sediment trap studies is presented which illustrates the importance of biological scavenging in controlling the distribution of radionuclides in the water column.

3.1. INTRODUCTION

During the last decade, considerable effort has been expended in the field of environmental sciences to better understand biogeochemical cycles in the sea and how they interrelate to control the basic patterns of climate, primary production and contaminant transport to mention only a few. For this reason, scientists from various fields have joined the growing ranks of biogeochemists to try to solve some of the major problems facing mankind today. As a biological oceanographer with a keen interest in the effects of pollution, in this chapter I will emphasize the importance of the biology in the discipline of biogeochemistry. In this respect, I intend to show how marine organisms control the movement and fate of radioactivity and related trace elements and, in general, their overall importance in the cycles of these materials in the sea.

3.2. GENERAL CYCLES

A conceptual diagram which covers most of the biological parameters in marine biogeochemical cycles is shown in Figure 1. Input into the cycle can arise from several sources of which radionuclides are just one example. It is also valid for pesticides and a variety of other contaminants. These materials enter the sea and become involved in various biotic and abiotic cycles. In Figure 1, the principal biological groups in the sea are circled and the arrows indicate how they accumulate and release radioactivity. Also illustrated is how these species pass radioactivity through the food chain and how, during their metabolic activities, radioactivity is lost

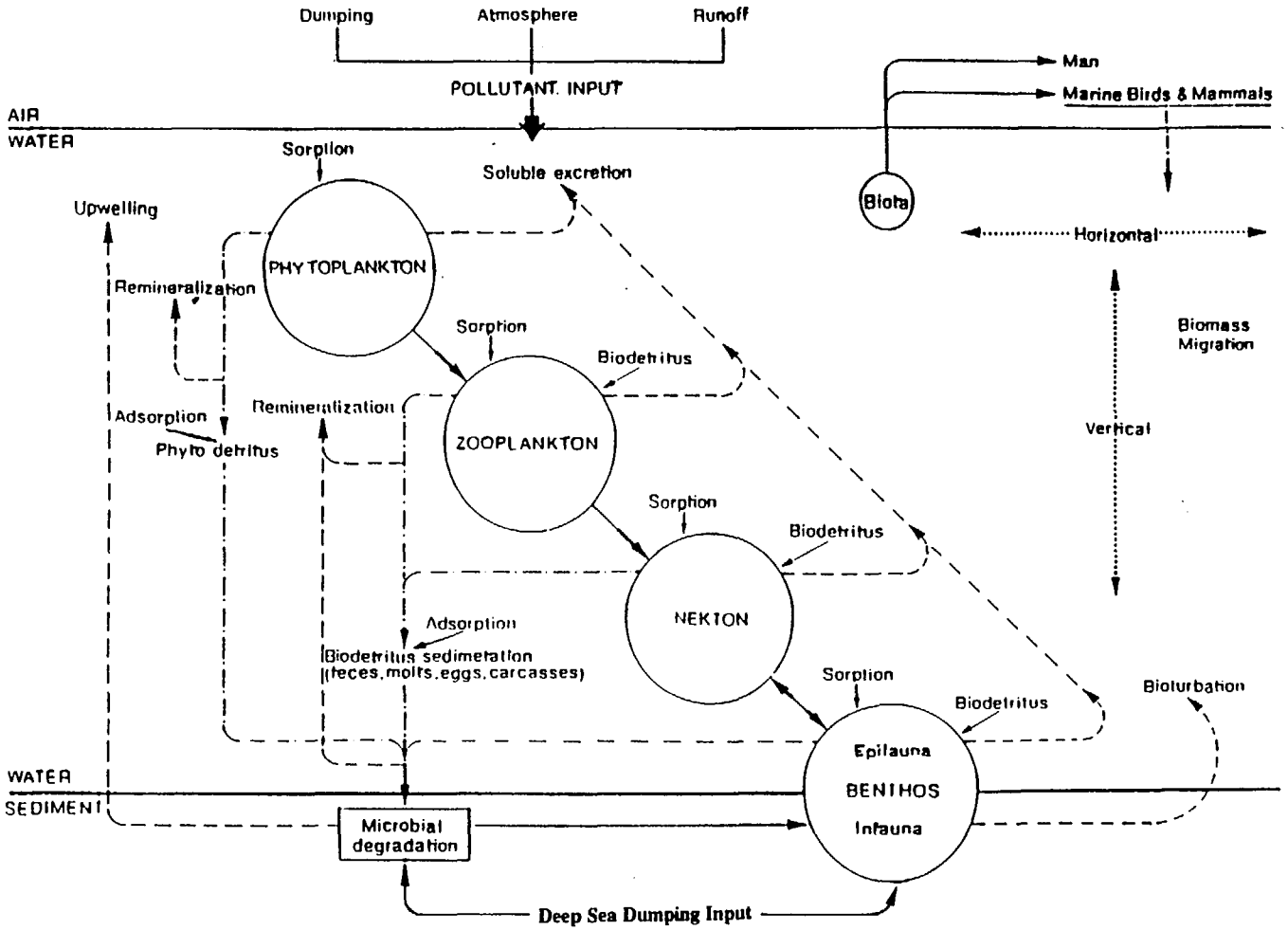

FIG. 1. General marine biogeochemical cycling scheme [1]. Reprinted with permission from Pollutant Transfer and Transport in the Sea, Vol. II, 1982 (ed. G. Kullenberg): Chapt. 1, 'Biological Transfer and Transport Processes' (S.W. Fowler), p.34. Copyright CRC Press, Boca Raton, Florida.

through excretion, is transported vertically and, in some cases, is remineralized. Another important link are the organisms that migrate vertically in the sea, e.g., the zooplankton and nekton. It is noteworthy that most of the biomass in the sea is found in the upper 800 m; relative to the entire sea there is very little biomass below 800 to 1000 m. There are also ways in which contaminants can escape the sea, for example, through the feeding activities of marine birds, mammals and man. The horizontal migration of marine organisms, particularly during their breeding cycles is another manner in which biomass transports radioactivity to different localities in the sea. One example is invertebrate and fish swarming. In this presentation, emphasis will be given principally to the smaller organisms in the sea because they are the most important in terms of their interactions with radioactivity and its eventual transport.

Marine species range in size from sub-micron (picoplankton, bacteria, etc.) to greater than centimetres in length (e.g., macroplankton and micronekton). These latter species are principally the large organisms termed nekton. Most of the information presented here is related to species in the range from a few μms to cms, that is the microplankton, zooplankton, and some of the small nektonic species which make up the bulk of the marine biomass.

In recent studies on marine particle types [2], it has been shown that in the 0.4-1 μm range there are as many as 10^7 particles in the top 50 m. This is an enormous number; however, more than 95% of these particles are non-living organic detritus from marine species. In the lower μm range there are the nanoplankton represented by silico-flagellates, coccolithophores, green flagellates, and many other very small species. Then, there are the slightly larger microplankton which are represented by chain forming phytoplankton such as certain diatoms and dinoflagellates. The main point to note is the wide variety of existing shapes and surface areas that occur in these microscopic species, since this aspect is of prime importance in the initial stages of radionuclide transfer from water to organisms.

Larger organisms, which are termed mesoplankton, ingest the smaller species. Most of the forms described above are autotrophes, i.e. they derive their energy from nutrients and sunlight. However, zooplankton species do not and they must ingest their energy requirements. Some examples are the copepods, which are the key microplanktonic species in the sea, krill which form the main food of many large organisms, and the most common gelatinous forms. These zooplankton also occur in a wide range of shapes, sizes and surface composition. They are in continuous contact with their medium which contains many of the trace elements that are important for their nutrition and growth. Therefore, if one understands the biogeochemistry of selected trace elements, one can very easily derive the biogeochemical cycles of the radionuclides of those elements. In fact, much of our knowledge about marine radioactivity has been gained from studies of the corresponding trace elements and heavy metals in marine species [3].

3.3. BIOACCUMULATION POTENTIAL

Small phytoplankton species accumulate elements and radionuclides principally by adsorption onto their surface followed by absorption through their cell membranes. Even as they accumulate elements, they also release them in element exchange processes and eventually attain what is termed isotopic equilibrium, i.e. they reach a steady state in their element uptake process. Often these species are sampled to assess their ambient element or radionuclide concentrations. In terms of radioactivity monitoring, it is necessary to measure the concentration of the radionuclide in the organism and then perhaps relate it to the corresponding radionuclide concentration in sea water. For this purpose marine radioecologists have derived a relationship which is called the concentration factor (CF). The CF is defined as the ratio of the element/radionuclide in a gram of tissue to the amount in a gram of sea water, and it is used to assess the relative ability of these species to take up elements. Therefore, assuming organisms are in isotopic equilibrium with their environment, estimates of radionuclide concentration in sea water can be derived from known CFs and radionuclide concentrations in the organisms.

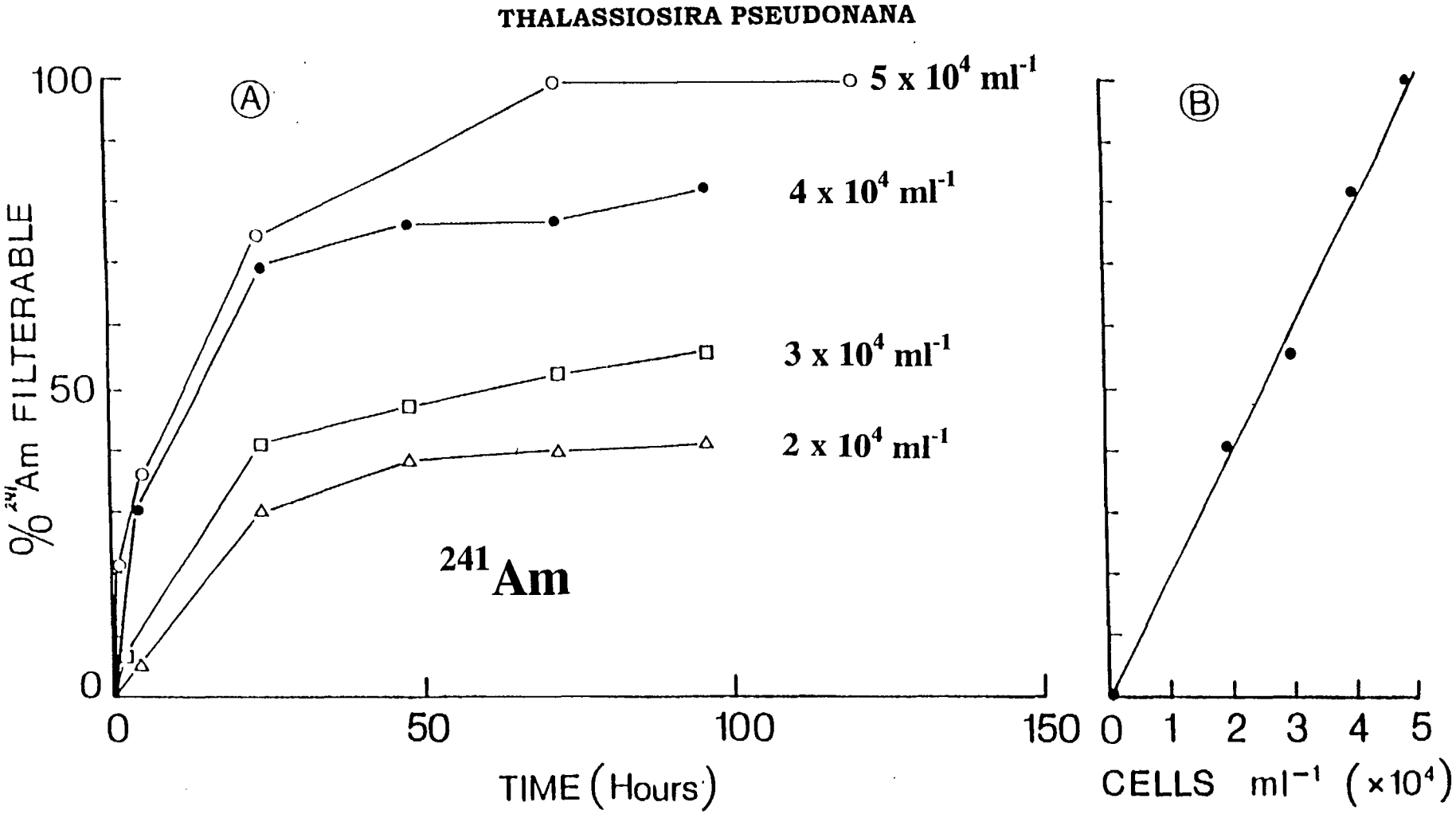

FIG. 2. ^{241}Am uptake by a diatom species (phytoplankton) present at different cell concentrations [6]. Reprinted from Limnology & Oceanography, Vol. 28(3), pp. 432-447, 1983, 'Interactions of marine plankton with transuranic elements. 1. Biokinetics of neptunium, plutonium, americium and californium in phytoplankton' (Fisher, et al.), with kind permission from the American Society of Limnology & Oceanography, KS.

TABLE I. ELEMENT AND RADIONUCLIDE CONCENTRATION FACTORS+ FOR PHYTOPLANKTON AND CRUSTACEAN ZOOPLANKTON [4]

Element or RN	Phytoplankton	Microzooplankton++	Macrozooplankton*
**Hg	5×10^4	1×10^4	2×10^4
**Ni	2×10^3	4×10^3	6×10^2
**Co	1.5×10^3	1×10^4	6×10^3
**Cd	2×10^4	4×10^4	2×10^4
**Cu	1×10^4	5×10^4	1×10^5
**Fe	5×10^5	3×10^5	2×10^5
**Zn	5×10^5	2×10^5	1×10^5
Tc	$\approx 10^0$	10^0-10^1	$\approx 10^0$
$^{239+240}$Pu	9×10^4 - 1×10^5	5×10^3	1×10^2
^{241}Am	2×10^4 - 1×10^5	3×10^3	1×10^3
^{144}Ce	9×10^4	1×10^3	-
^{106}Ru	1×10^5	3×10^3	-
^{238}U	1×10^1	5×10^0	-
^{232}Th	2×10^4	2×10^4	-
^{230}Th	8×10^3	4×10^3	-
^{228}Th	2×10^4	6×10^3	-
^{226}Ra	2×10^3	1×10^2	-
^{210}Po	1×10^4	2×10^4	1×10^4

+Defined as element/g wet animal divided by element/g water
++Mainly copepods
* Euphausiids
**Computed using recent values for element concentration in sea water

There is a very wide range in the degree to which phytoplankton concentrate these radionuclides. Newly revised CFs for phytoplankton range from approximately 1 for Tc (very low affinity for a radionuclide) to greater than 10^6, i.e. a million times over the amount of the element/radionuclide in sea water (Table 1). Phytoplankton, being very small, have a very large surface area to volume ratio. One of the key points in the initial entry of radionuclides into the food chain is at the level of the phytoplankton. Certain elements like Ni, Co, Cu, which are biologically essential, are needed in the enzyme systems of marine organisms. With these elements, high CFs are typical. For others, like Tc, which are conservative in sea water, they do not readily associate with particles and the CF is therefore very low.

With zooplankton which are slightly larger, there is also a wide range of CFs although normally they are not quite as high as in phytoplankton [5]. As in the case of phytoplankton, the Tc concentration factor is very low, elements like Pu and Am are mid-range, and Zn tends to be very highly concentrated by zooplankton (Table 1).

An idea of the time scale involved in the accumulation process can be seen in Figure 2a which depicts the accumulation of ^{241}Am by phytoplankton. In this case the cells are a species of diatom which contains a silica frustule. One important point is that the uptake process is relatively rapid, i.e. it occurs over a period of a day or two rather than weeks or months. Moreover, an equilibrium between uptake and loss in the medium is established very quickly. These experiments were performed at different cell concentrations. If the concentration of the cells per ml is plotted against the amount of Am which is retained on those cells, a clear relationship is found (Fig. 2b). This simply means that the more phytoplankton there are in the water (high biomass), the more radioactivity will be taken up.

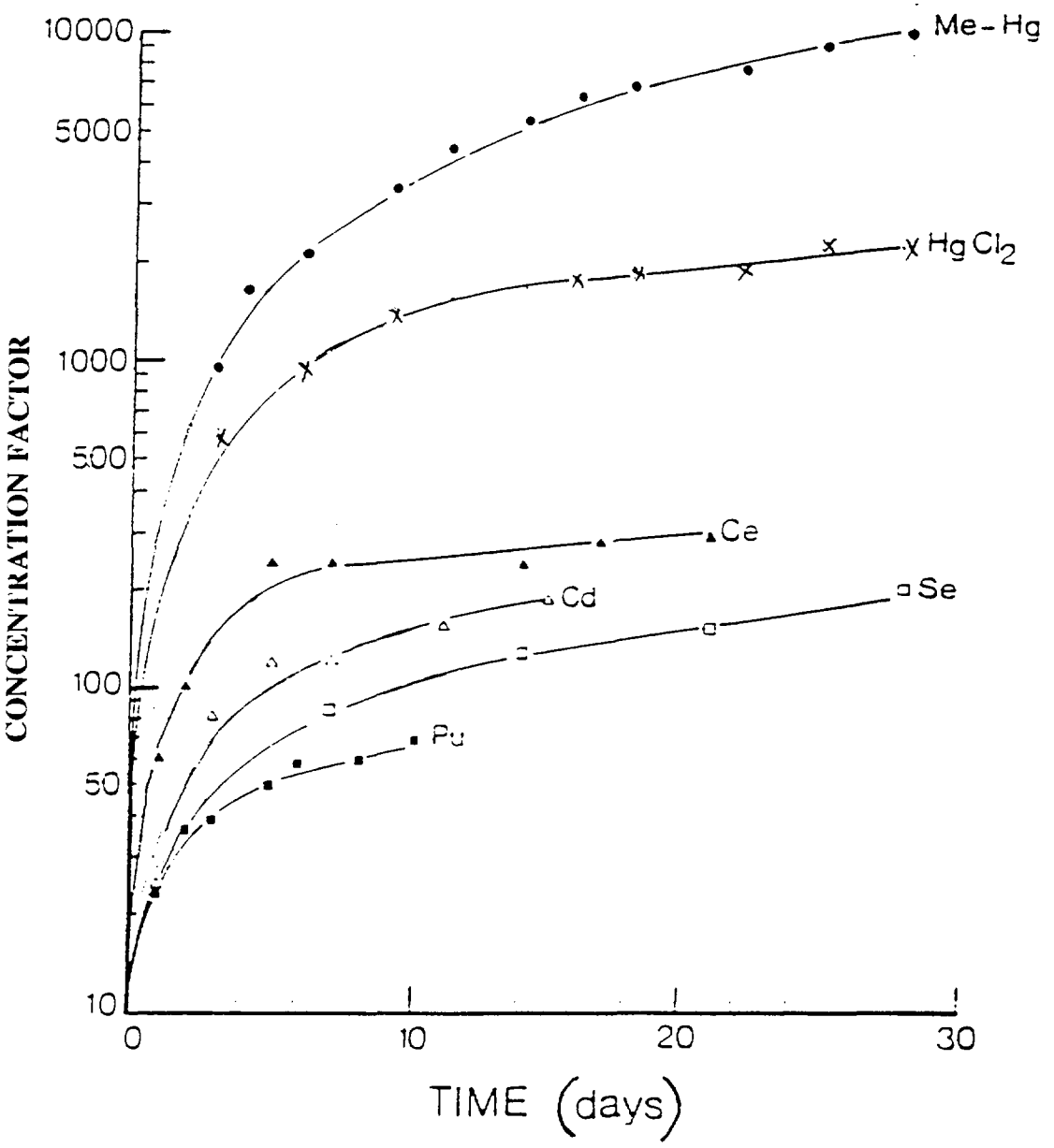

FIG. 3. Accumulation from water of various metals and radionuclides by the euphausiid Meganyctiphanes norvegica [1]. Reprinted with permission from Pollutant Transfer and Transport in the Sea, Vol. II, 1982 (ed. G. Kullenberg): Chapt. 1, 'Biological Transfer and Transport Processes' (S.W. Fowler), p.4. Copyright CRC Press, Boca Raton, Florida.

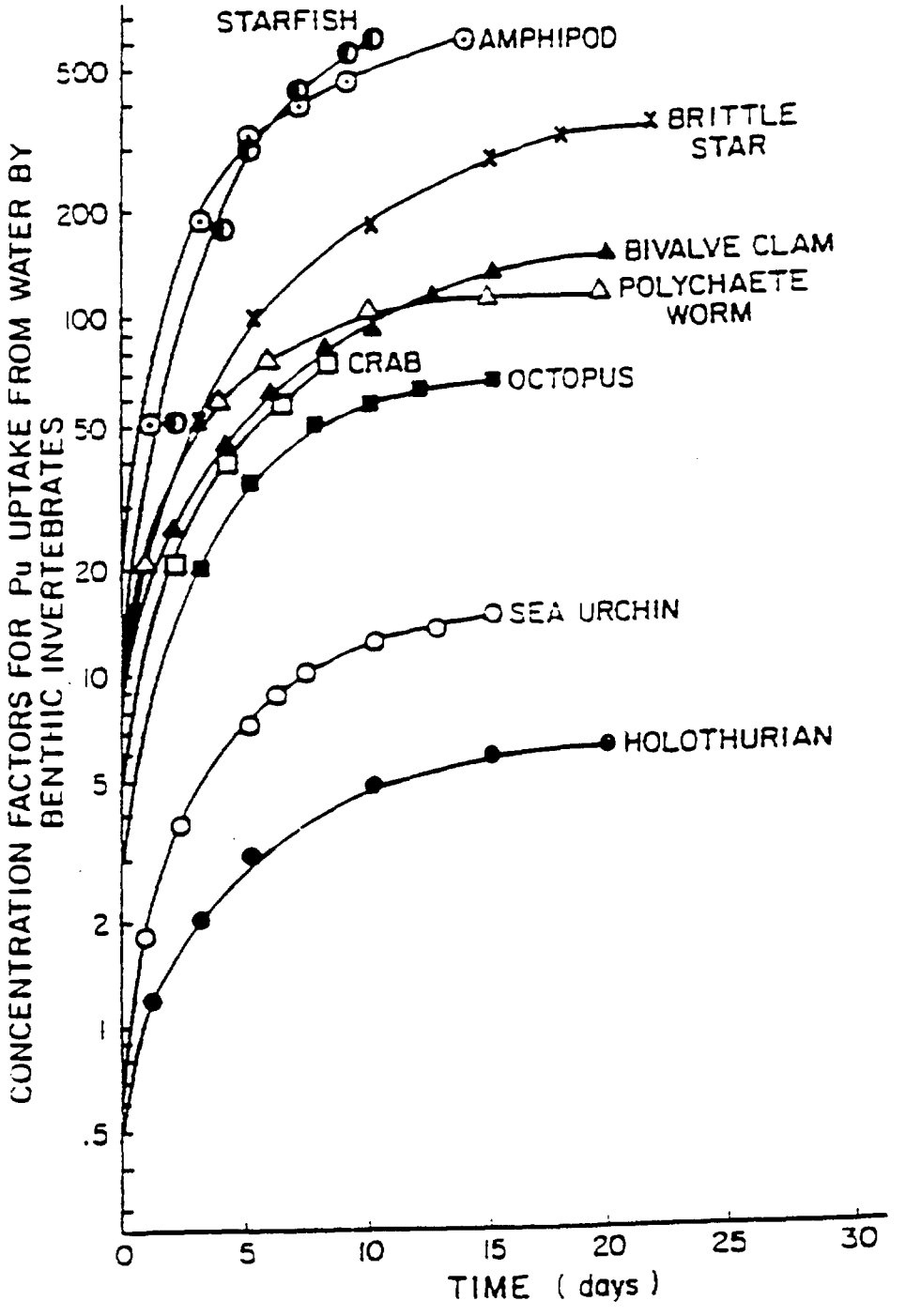

FIG. 4. Experimentally-determined plutonium uptake in various marine invertebrates [after 7].

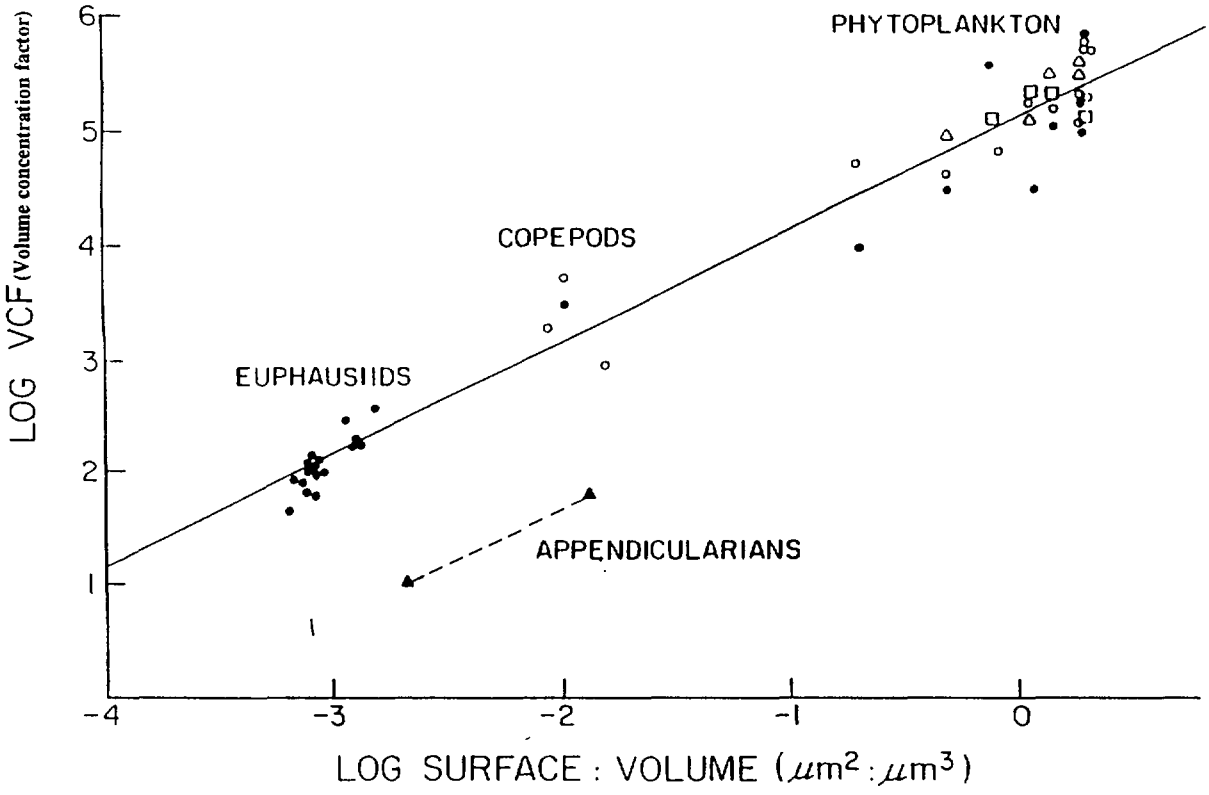

FIG. 5. Correlation of log VCF for transuranic elements with log surface: volume ratio of marine plankton (•) Am, (O) Pu, (Δ) Cf, () Cm, and (Δ) Am in appendicularians [9]. Reprinted from Oceanic Processes in Marine Pollution, Vol. 2, 'Physicochemical Processes and Wastes in the Ocean' (Connor, Burt & Duedall, eds.), Fig. 16.1, p. 199. Copyright from Krieger Publishing Company, P.O. Box 9542, Melbourne, FL32902-9542, USA.

In the case of slightly larger species, for example the krill, there is a large difference in CF depending on the element (Fig. 3). These CF values range from around 50 to 60 for Pu to 10,000 for the metaloid Hg. Hg tends to bind with sulfydryl groups in proteins, so because of this large internal binding pool, enhanced uptake occurs. In the case of elements like plutonium, which are not metabolically essential, they tend to bind to animal surfaces which results in somewhat lower CFs.

The biokinetic behaviour of plutonium in a variety of marine species is shown in Figure 4. There is a wide range of measured CFs and, for an as yet unknown reason, asteroid starfish take up Pu to relatively high levels. This organism appears to be an extremely good bioindicator for Pu in the marine environment. Starfish have mucus on their parapodia which tends to accumulate and firmly bind this transuranium element. They effectively retain Pu, whereas other organisms do not. When the source of Pu is in the water, the radionuclide accumulates on the hard surface of the starfish [8]. If starfish ingest contaminated food, Pu is digested and enters the pyloric caecum which is the liver-like storage tissue found in each arm of the starfish. Nearly 100% of the ingested Pu is found in the caeca where it becomes more or less immobilized [8]. Both uptake from water and from food results in CFs ranging from 10s to 1000s in these species.

To underscore the importance of surface area in the radionuclide uptake process, the relation between transuranic uptake and surface area for numerous types of plankton ranging from microscopic phytoplankton species to small crustaceans such as copepods and krill is shown in Fig. 5. In the case of Am, Pu, Cm, a straight line relationship is found which indicates that uptake in

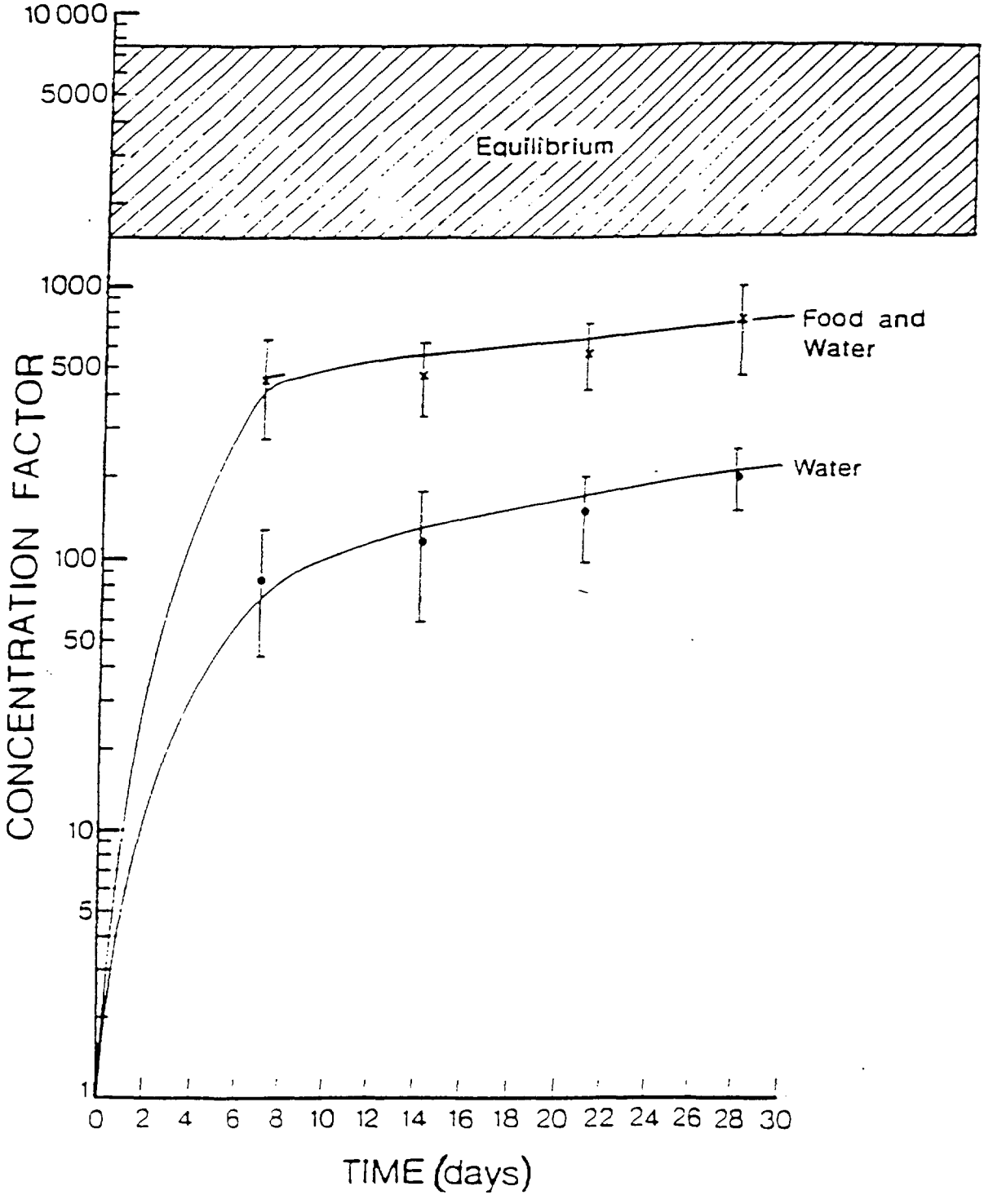

FIG. 6. Accumulation of Se-75 from water and from food and water together by the euphausiid Meganyctiphanes norvegica. Shaded area represents range of equilibrium concentration factors based on stable selenium concentrations in euphausiids and water [1]. Reprinted from Marine Science Communications, Vol. 2, No. 1, (Fig. 1) 1976, 'Selenium Kinetics in Marine Zooplankton' (Fowler, S.W. & Benayoun, G.), pp. 43-67, with kind permission from Marcel Dekker Inc., N.Y.

these species is basically a surface area effect, not a metabolic one. It is noteworthy that appendicularians fall off the regression line. These are gelatinous planktonic forms, with a surface chemical composition that is very different from the siliceous and chitinous surface of diatoms and crustaceans respectively. Thus, in the case of appendicularians, for a given surface/volume ratio there is considerably less uptake than in the other species with hard surfaces.

Another way in which uptake can occur, particularly in large species, is through the food chain. Fig. 6 shows an example with krill. One group of krill was exposed to ^{75}Se in water, and another group was exposed to the same radiolabelled sea water plus radioactive food. Over time, there was quite a significant increase in the CF due to the amount of ^{75}Se which was ingested with food. However, even after approximately one month, these organisms had not reached the derived equilibrium CF. This concentration factor is based on stable Se levels in the organism and in the water. Even by accumulating selenium from both water and food, the combined process is very slow and after a month the organism had not reached isotopic equilibrium. This is one example of food chain transfer which facilitates the organism in reaching a steady state with the element or radionuclide in its surroundings.

Much has been written about contaminant bioaccumulation in higher trophic levels of the food chain. For example, cases abound where following a contamination, the contaminant enters an organism which, in turn, is eaten by another and finally is ingested by the top predator which is often man. Thus, one is tempted to say that the contaminant passes along the food chain and is concentrated with each increasing link in the food chain. In most cases, this is an incorrect assumption. If one follows concentrations of a given element or radionuclide in different organisms of a food chain, a decrease in concentration or concentration factor is actually noted [10]. This effect is the opposite of "biomagnification" in which case the top predator displays the highest concentration. It is commonly believed that there is a biomagnification of Hg at higher levels in the food chain. In some cases this may be true since methyl-Hg tends to accumulate in marine organisms and is not excreted. However, for an element like Pu and many other trace elements, it is exactly the opposite. For radionuclides, the only example that has been reported to date where this may not hold is ^{137}Cs, and particularly in fresh waters [10]. Where this effect has been examined in detail in both fresh water and marine species, there is a slight tendency to increase along the food chain possibly because Cs follows K in its accumulation pattern and both tend to accumulate in the muscle tissue of large predator fish. Furthermore, Cs is accumulated to higher levels in fresh water species than in marine organisms, because in a low salinity environment there are fewer cations to compete with Cs.

3.4. EXCRETION

In concert with element accumulation, there is an opposing process operating to eliminate or excrete elements. An example of radionuclide excretion from phytoplankton is given in Fig. 7. Note that when phytoplankton previously exposed to ^{241}Am are transferred to clean sea water, there is a very rapid elimination of the radionuclide. This initial, rapid loss is usually thought to be due to desorption from the surface of contaminated species. This initial loss is followed by a subsequent, rather gradual elimination process. From the slope of the regression lines a rate can be derived which can be converted to a biological half-life, i.e. the time for half the element to be eliminated from the organism. It is evident from these slow components that loss often proceeds at a less rapid rate than the accumulation phase. The other parameter to note in this example is the effect of the duration of initial exposure. These diatoms were exposed to ^{241}Am for 5 hr and for 72 hr. There was an overall increase in the retention time for the longer exposure, suggesting some form of penetration and stronger binding of radionuclide within the cells which were in contact with the contaminant for a longer period.

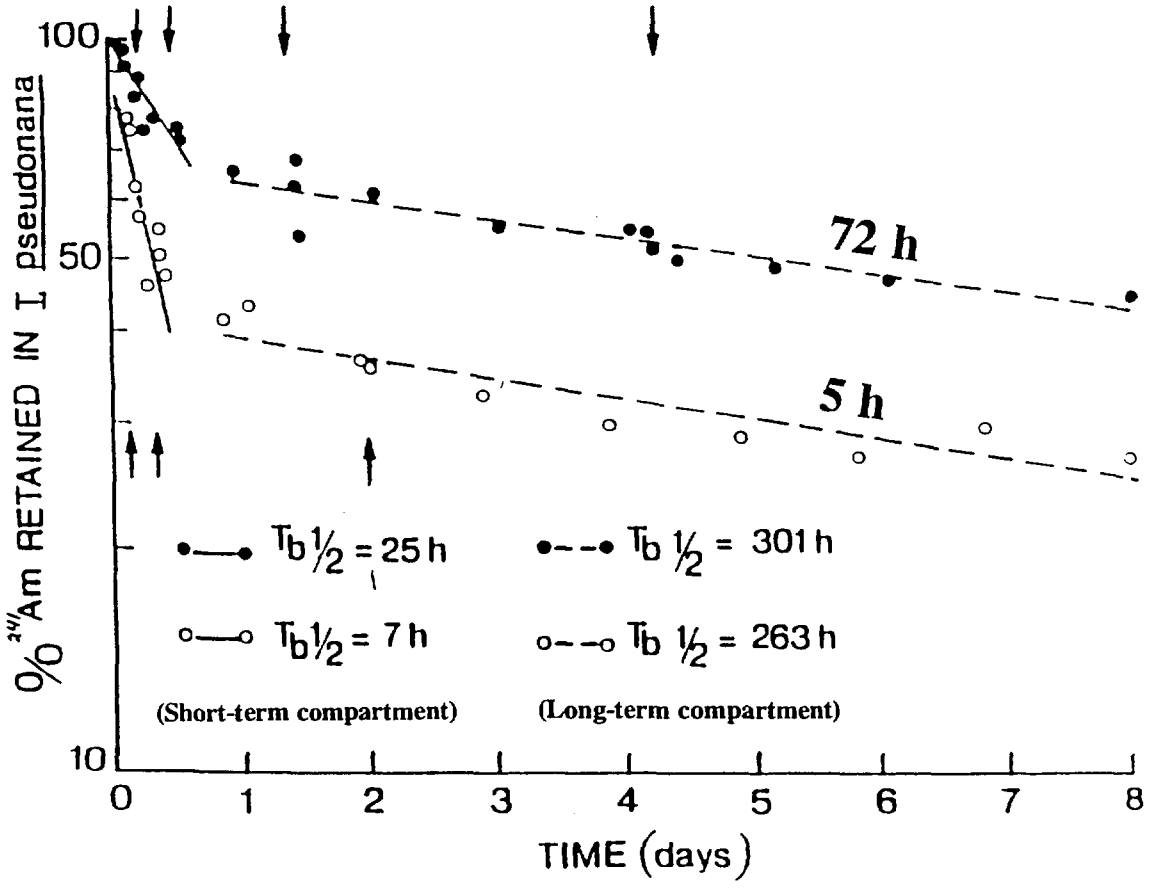

FIG. 7. ^{241}Am excretion by a diatom previously exposed to ^{241}Am for two periods of time (5 hr and 72 hr). $T_{b½}$ (biological half-life) is time necessary to lose one half of the body burden in the long-term and short-term compartments [6]. Reprinted from Limnology & Oceanography, Vol. 28(3), pp. 432-447, 1983, 'Interactions of marine plankton with transuranic elements. 1. Biokinetics of neptunium, plutonium, americium and californium in phytoplankton' (Fisher, et al.), with kind permission from the American Society of Limnology & Oceanography, KS.

Similarly, with heterotrophes there are a variety of loss curves depending upon the radionuclide under study. In the case of krill (Fig. 8), there are two elements which are not biologically essential (Ce and Pu) and consequently, they are lost very rapidly, whereas methyl-Hg and Se (two elements that are involved in protein cycles) are more strongly retained. Heterotrophes display radionuclide loss kinetics which are a combination of soluble excretion and defecation of solid materials. Crustaceans differ from other organisms in that they also lose radionuclides incorporated in their exoskeleton when they molt. Organisms also release eggs, which is another way of excreting radionuclides. Finally, there is simple chemical desorption, i.e. ion exchange from the surface. To discern the relative importance of some of these routes, experiments have been performed where, by use of radiotracers and knowledge of the stable element concentrations, one can calculate and model the percent of a given element that fluxes through the organism. For example, with Zn and Pu in krill, most of the flux takes place through defecation (Table 2). In the case of Se and Hg, both of which are involved in metabolic cycles to some extent, approximately half occurs through defecation and half by soluble excretion via the urine and other products that are released. Note generally that defecation is a very important route for the passage of radionuclides and elements. As will be shown below, this process has a very profound effect on the geochemistry of the sea.

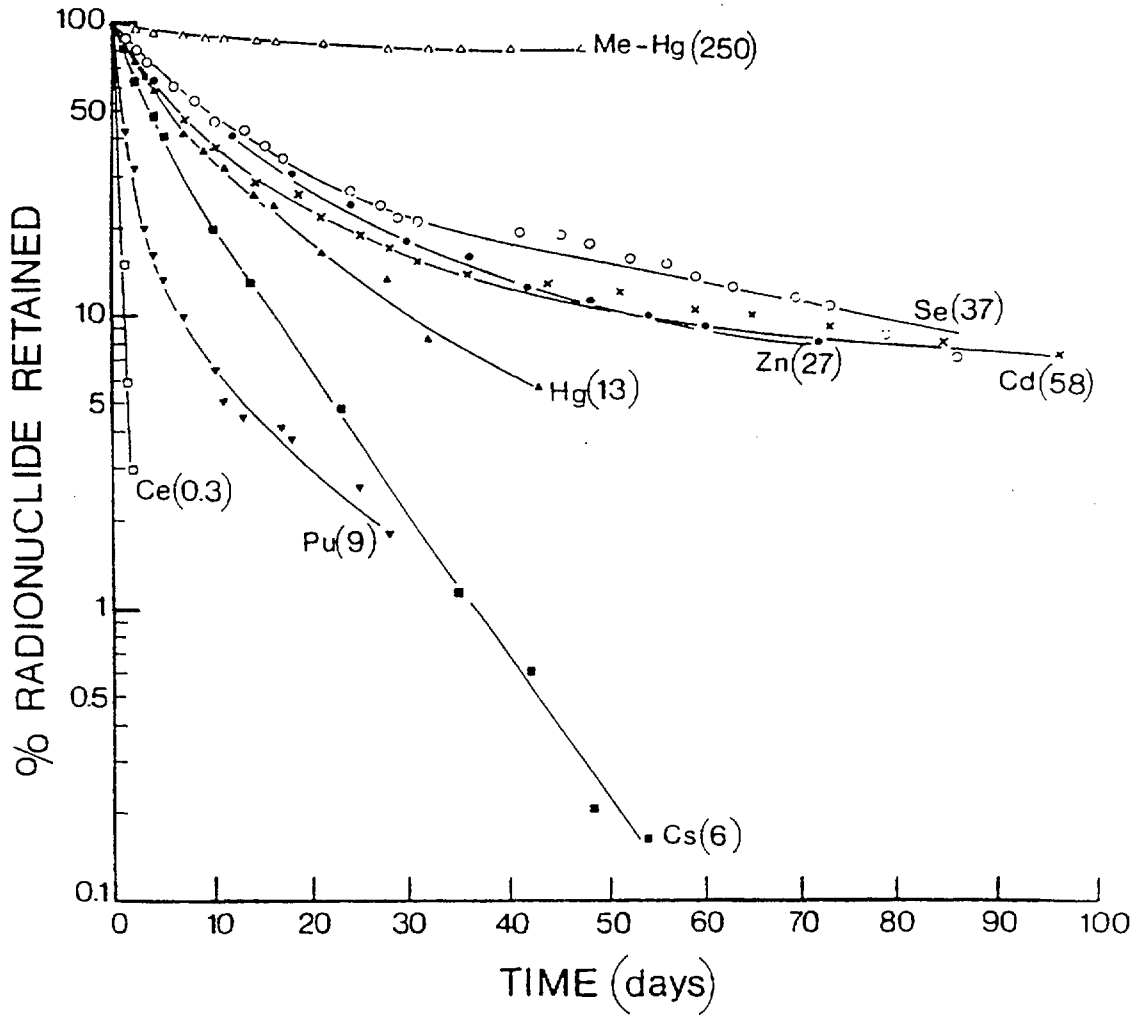

FIG. 8. Long-term excretion of different elements from euphausiids chronically exposed to radiotracers in food and water [1]. Reprinted with permission from Pollutant Transfer and Transport in the Sea, Vol. II, 1982 (ed. G. Kullenberg): Chapt. 1, 'Biological Transfer and Transport Processes' (S.W. Fowler), p.21. Copyright CRC Press, Boca Raton, Florida.

TABLE II. RELATIVE DISTRIBUTION OF ELEMENT EFFLUX[a] THROUGH THE EUPHAUSIID *MEGANYCTIPHANES NORVEGICA* BY MOLTING, DEFECATION, AND SOLUBLE EXCRETION [1]

Element	Molting (%)	Defecation (%)	Soluble excretion (%)
Zn	1.1	92.6	6.3
Cd	3.3	84.5	12.2
Se	2.4	54.4	43.2
Hg (inorg.)	2.5	29.1	68.4
^{239,240}Pu	0.8	98.6	0.6

[a]Calculations based on euphausiids grazing under sufficient food conditions during the non-egg-laying period.

Particulate radionuclide excretion is relatively easy to determine. In practice the fecal pellets can be collected and the amount of elements or radionuclides contained in them are then measured. The same can be done with molts. To follow soluble excretion, the organism must be labelled with radiotracer and loss (corrected for defecation) from the organism's body measured. With these data, complete budgets of element excretion can be derived [11].

3.5. TRANSFER AND TRANSPORT

Some small crustacean species molt very rapidly, on the order of once per week. There is a strong relationship between temperature and crustacean molting, i.e. the warmer the water, the higher the molting frequency. Therefore, one might expect that in warmer, upper layers, for example in tropical areas, there would be a greater molting effect on radionuclide release than would occur in the deep sea where temperatures are typically very cold. At present, there is little information on molting rates in deep sea species.

Zooplankton fecal pellets vary greatly in size and shape. A typical euphausid fecal pellet is several hundred microns in length. Phytoplankton cells are "packaged" within this cylinder which is covered with a thin layer of chitin called the peritrophic membrane. This membrane retards bacterial degradation and keeps the packaged material contained inside. When pellets are released in the water column they sink very rapidly, at speeds ranging between tens and several hundreds of metres per day [12]. Researchers have recently recognized the importance of fecal pellets in biogeochemical cycles and have begun to examine the speed at which they sink in the sea. The sinking speed (m/day) is closely related to pellet density. For large dense pellets that might typically sink at speeds of greater than 100 m d^{-1}, they have the potential to reach great depths in a matter of days. Large shrimp, fish and salps produce such pellets and thus, they are probably very important in packaging surface radioactivity and transporting it to depth by this means.

However, rapid sinking is not the only factor governing the importance of fecal pellets and other biogenic particles in element transport. The concentration of trace elements and radionuclides in these materials are given in Table 3. Included are data for micro-plankton species which form the base of the foodchain, the zooplankton that ingest them, and the fecal pellets and molts that are produced. It is evident from the data that the zooplankton themselves have element and radionuclide concentrations that are much lower than those in their fecal pellets.

The fecal pellets tend to concentrate elements and radionuclides, particularly those that are not metabolically essential (Table 3). In other words, animals strip the nutrients such as carbon and phosphorous from the food source and then package the refractory portion of the food (e.g., silica frustules, exoskeleton and other undigestible tissues). These detrital particles which are released as pellets then form a new aggregate particle which is relatively enriched in certain elements and radionuclides. For example, the data in Table 3 show that euphausiid fecal pellets contain as much as 2.4% Fe. Molts usually contain lower concentrations which are a function of the surface chemistry of the element. For example, for elements like Pb which adsorb to surfaces, nearly 100% of what is associated with the animal is found in the molt. In contrast, other elements like Se and Hg that tend to be bound within the animal show very low percentages in the molt. In summary, the overall enrichment of many elements and radionuclides in fecal material, and biogenic detritus in general, is the main factor which determines their importance in biogeochemical cycles and vertical transport.

Some relationships with fecal pellets have been examined, particularly the CF in a fecal pellet and the residence time of that element in sea water. The residence time is a measure of how long the element resides in sea water before it is removed. Residence time can be calculated if the element concentration in the water (assumed to be in steady state) and the element input or output are known. This has been done for known river inputs of elements to the sea, and the resultant plot of the CF in fecal pellets against the residence time shows a very clear inverse relationship [13].

TABLE III. LEVELS ($\mu g\ g^{-1}$ dry) OF TRACE ELEMENTS, RADIONUCLIDES, AND PCBS IN ZOOPLANKTON AND BIOGENIC PARTICLES [1,4]

Element	Macro-zooplankton	Fecal Pellets	Molts	(%)*	Food (microplankton)	Marine snow
Ag	0.7	2.1	2.9	(31)	0.7	
Cd	0.7	9.6	2.1	(22)	2.1	3.4
Co	0.2	3.5	0.8	(34)	0.9	
Cr	0.9	38	5.3	(48)	4.9	
Cu	48	226	35	(6)	39	10
Fe	64	24000	232	(28)	570	12800
Mn	4	243	12	(21)	18	148
Ni	0.7	20	6.7	(78)	8.1	25
Pb	1	34	22	(≈100)	11	9
Zn	62	950	146	(18)	483	40
Hg	0.3	0.4	0.2	(4)	0.1	
Se	4	7	2	(3)	3	
Ce	0.2	200	1.2	(44)	0.3	
Eu	0.002	0.66	0.008	(26)	0.013	
Cs	0.062	6.0	0.019	(2)	0.08	
Sb	0.071	71	0.8	(87)	0.22	
Sc	0.009	2.8	0.03	(26)	0.13	
Sr	117	78	350	(23)	520	
$^{239+240}Pu$**	0.4	98	4.8	(90)	4.0	19
^{241}Am**		72				10
^{210}Po**	1100	24500	360	(2.5)	3400	
^{210}Pb**		10400				
^{232}Th**	0.35	250	2.6	(57)	17	
^{238}U**	21	520	245	(90)	340	
PCBs	0.62	16	1.4	(17)	4.5	

*Percent total body burden in molt
**pCi/kg dry. 1 pCi = 37 mBq

From this relationship there is a good indication that element scavenging and subsequent removal from the sea is due to biogenic materials like zooplankton fecal pellets.

3.6. ELEMENT PROFILES

What evidence for the importance of these biological processes exists? Marine chemists have classically examined dissolved and particulate profiles of elements in the sea to determine their oceanographic consistency. Figure 9 shows a series of ^{137}Cs profiles in the Mediterranean. Note that ^{137}Cs is always quite high in the surface waters and then decreases with depth. This is termed a conservative element profile because the element concentration follows salinity very closely. In the Mediterranean the ^{137}Cs peaks are prominant in the eastern Mediterranean but decrease going into the Ionian Sea near Greece and again into the Tyrrhenian Sea and off Monaco. The decrease in this peak parallels the decreasing influence of eastern Mediterranean water (of higher salinity and thus higher Cs content) going across the Mediterranean from east to west.

Cadmium behaves somewhat differently and its profile is often called a nutrient-type profile. Cadmium follows phosphate in that it is a minimum at the surface, then displays a shallow maximum before decreasing again at depth. This suggests particle regeneration during sinking,

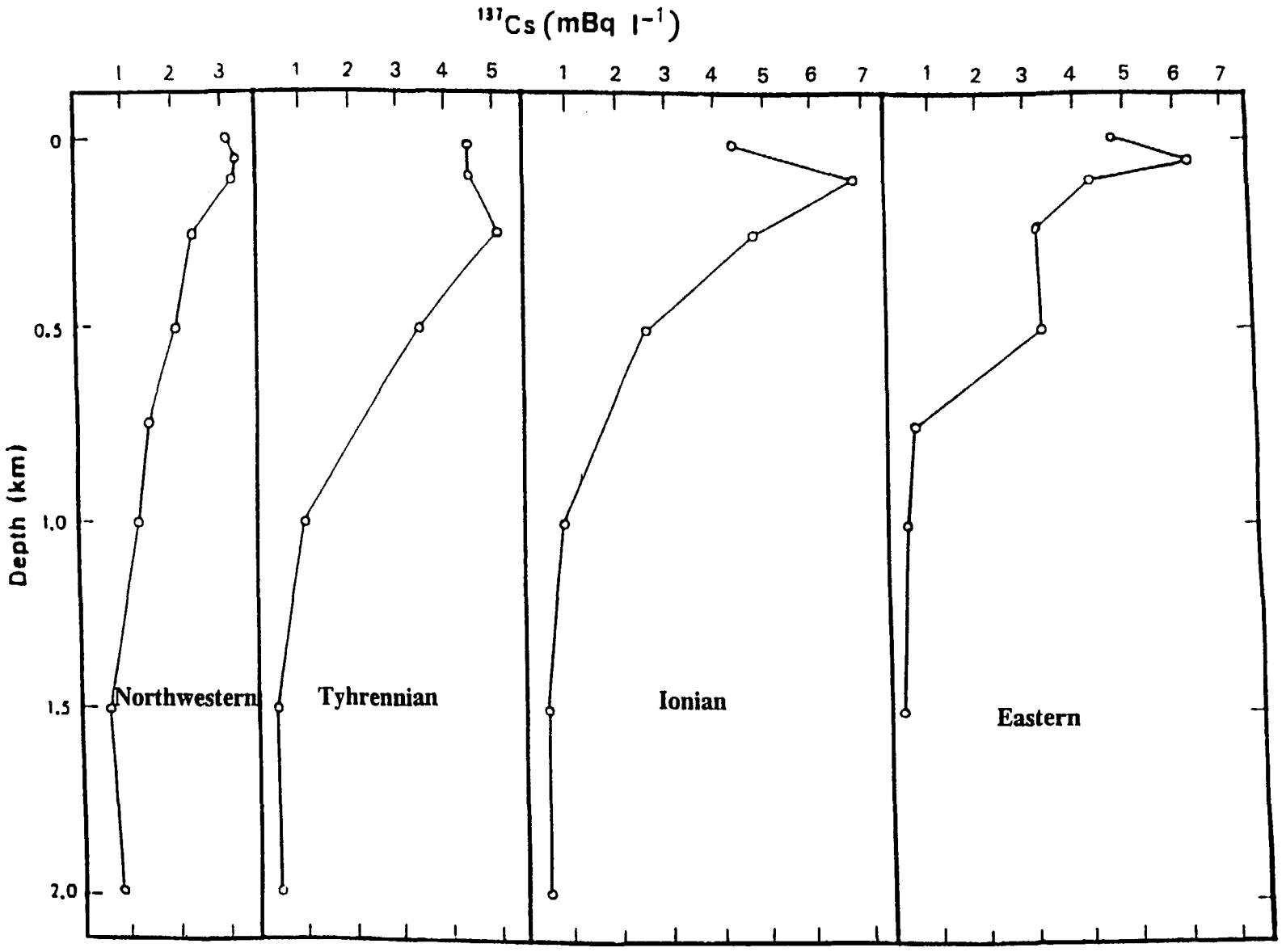

FIG. 9. Dissolved ^{137}Cs profiles in the Mediterranean Sea [after 14].

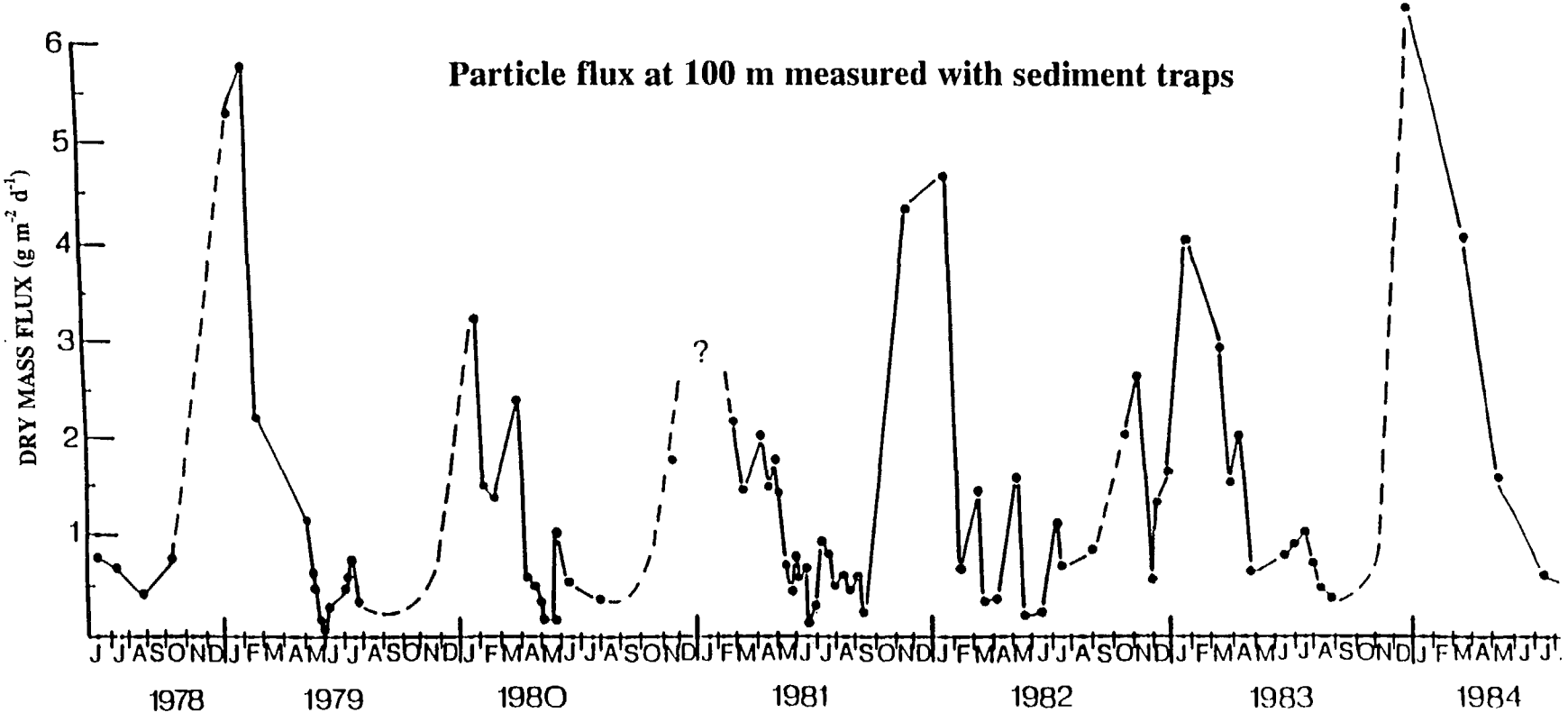

FIG. 10. Temporal change in particle flux through 100 m off the coast of Monaco, NW Mediterranean [17]. Reprinted from Oceanologica Acta, Vol. 14(1): 77-85, 'Seasonal particulate carbon flux in the coastal northwestern Mediterranean Sea, and the role of zooplankton fecal matter' (S.W. Fowler, et al.). Fig. 2, p.79. Copyright with kind permission from Gauthier-Villars Publishers, Paris, France.

much like the pattern for nutrients. This is underscored by the very tight correlation, nearly a 1:1 relationship, between Cd and phosphate profiles noted in the north Pacific [15]. Zinc is similar to silicate and also displays a maximum, but the maximum is found at greater depth than Cd. For this reason, zinc is said to show a deep maximum, nutrient-type profile.

Finally, there is a so-called scavenging type profile. In this type the element displays a surface maximum which then decreases to a minimum in mid water, and then increases again with depth. In the case of Al, this is interpreted as initial atmospheric input through aerosols, scavenging by particles and then remineralization at depth. At the bottom, aluminium diffuses out of the sediments, and creates an Al maximum in near bottom waters. In the case of stable Pb, there is a maximum at the surface, and then as it is scavenged by particles, it decreases very rapidly with depth. However, with Pb no increase near the sediment is seen. The same scavenging trend is also seen for ^{210}Pb, the removal of which is highly correlated with the flux of organic carbon [16].

3.7. VERTICAL FLUX AND SCAVENGING

Chemical oceanographers are now employing sediment traps to study the sinking particles that have been proposed to control the dissolved and particulate element profiles measured in the water column. Sediment traps are simply a series of particle collectors, like rain gauges, which are attached at various depths on a mooring line anchored to the bottom. Particle collections of sinking particles can be made for specific periods of time and then analyzed for their elemental content in order to understand scavenging processes.

Typical information obtained from a sediment trap is shown in Fig. 10. This example is a long time-series data set from coastal waters off Monaco. Each year a definite peak in mass flux is seen during the winter and early spring. This is primarily due to two events: a bloom of phytoplankton at that time of the year, and also increased sediment resuspension in the water during the winter months. During summer, there is a very low particle flux followed in the fall by a secondary peak of flux which may be related to a minor bloom that often occurs at that time in the Northern hemisphere.

Further offshore in the middle of the northwestern Mediterranean, particle flux patterns remain the same but the flux is much lower, i.e. $mg/m^2/d$ instead of $g/m^2/d$. In the open Mediterranean, very large variations in flux over short time scales, e.g. two-week intervals, are also noted [18]. If one examines the flux of these particles over depth, a progression in flux is noted as the water column stabilizes between spring and later summer. For example, during late summer, the particle flux is fairly uniform throughout the entire water column, whereas at other times of the year high flux occurs in the surface waters during periods of high biological productivity, with a much lower flux at depth. The same trend has been seen in other oceans.

The flux of C over depth in $moles/m^2/y$ for the Pacific Ocean is depicted in Fig. 11. There is a sharp drop off with depth which is interpreted to be the zone where most of the C remineralization takes place. As bacteria degrade these particles, a sudden decrease is noted followed thereafter by relatively constant flux with depth. Therefore, if radionuclides are associated with this material, the same process, degradation and radionuclide remineralization, will take place. The first study to show the general relationship between radionuclide flux and biological productivity in overlying waters was carried out in the Sargasso Sea. The time-series measurements showed definite cycles in biogenic particle flux which correlated well with corresponding radionuclide flux [20].

Another radionuclide technique that is commonly being used in marine studies is to examine the disequilibria between ^{238}U and its daughter ^{234}Th in water and particles. When the radionuclide pair is in secular equilibrium, the ratio of Th to U is 1. Samples of sea water down to 100 m are taken and then the Th and the U are measured. A decrease in the Th/U ratio indicates

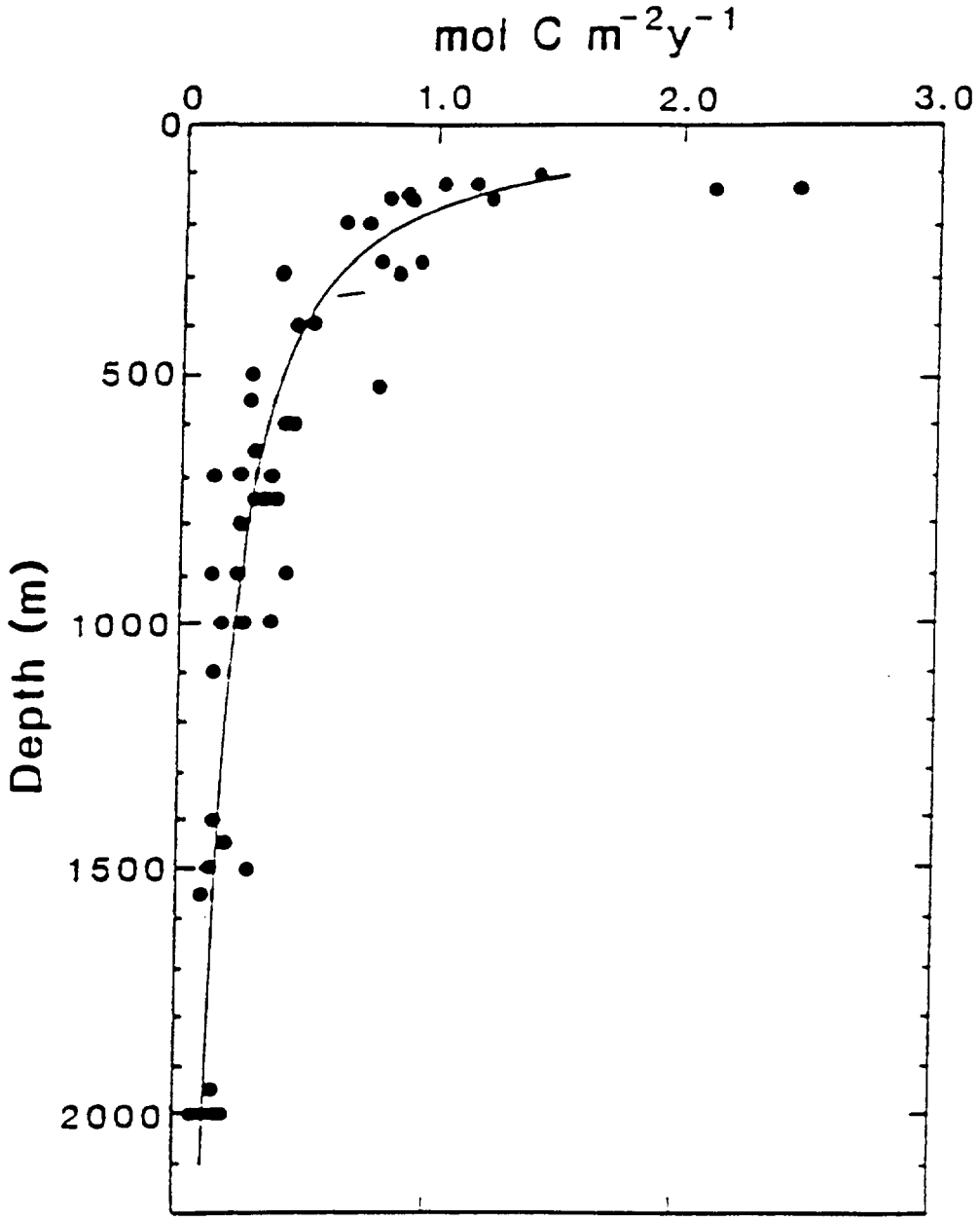

FIG. 11. Open ocean composite fluxes for C using the means of replicates from various stations in the NE Pacific Ocean. $F = 1.53\ (z/100)^{-0.858}$; $r^2 = 0.81$; $n = 48$ [19]. Reprinted from Deep-Sea Research 34(2): pp. 267-285 (1987), 'VERTEX: Carbon cycling in the northeast Pacific' (J.H. Martin, et al.). Copyright 1987, with kind permission from Elsevier Science Ltd., The Boulevard, Langford Lane, Kidlington OX5 1GB, UK.

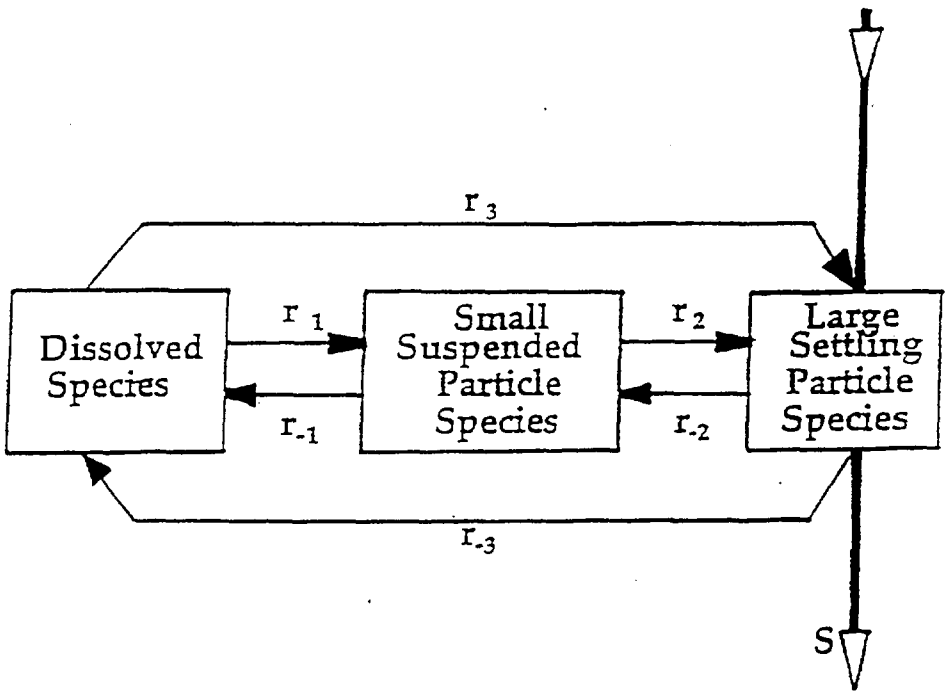

FIG. 12. Scavenging model representing exchanges of dissolved chemical species and different size classes of particulate matter in the oceanic water column. S represents the sinking velocity of the particles; r values are rates for the various transfer processes [22]. Reprinted from SCOR Report Series No. 1, 'Particulate Biogeochemical Processes', Report of SCOR Working Group 71, Sept. 1988, p.9. Copyright 1988, with kind permission from SCOR, Dept. of Earth & Planetary Sciences, The John Hopkins University, Baltimore, MD21218, USA.

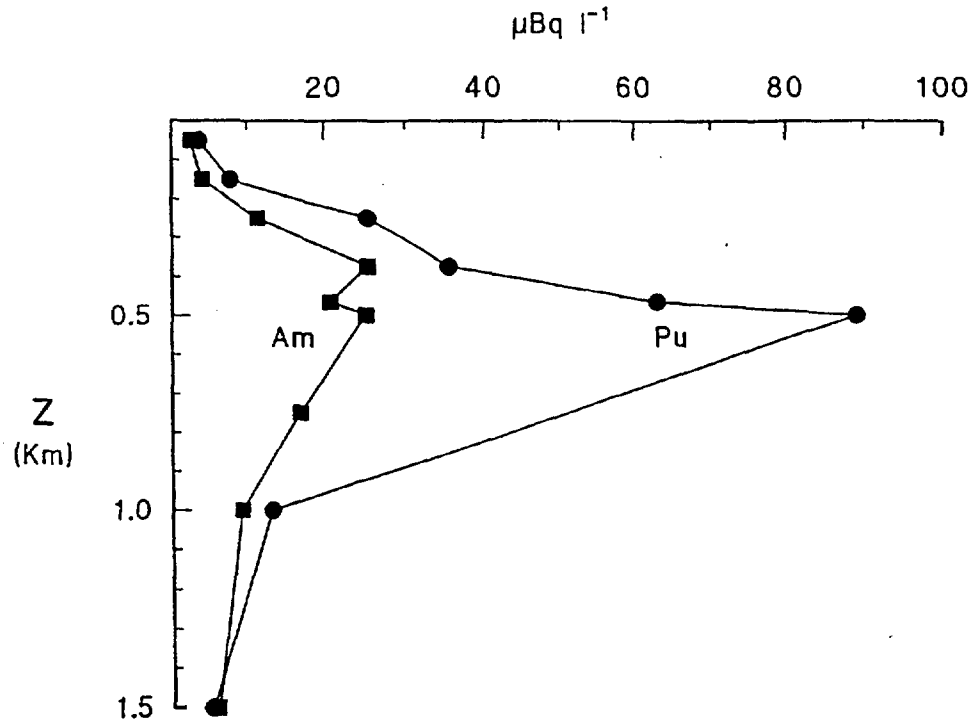

FIG. 13. Dissolved water column profiles of $^{239+240}$Pu and ^{241}Am at the VERTEX IV station (28°N, 155°W) in the NE Pacific Gyre [30].

that Th has been removed from the water relative to U. The cause of this removal is chemical scavenging by the particles in the water, principally the biological particles discussed above. Thorium is a very reactive element and is adsorbed by small particles very rapidly. Uranium is not particle reactive and behaves conservatively; therefore, it remains in the water while the Th scavenged by the particles subsequently sinks with them.

In one study at a location in the north Pacific [21], a very low ratio at about 60 m was found. This depth corresponded exactly to where the chlorophyll maximum was situated. The chlorophyll was a result of the phytoplankton cells which were scavenging and subsequently removing the thorium from the soluble phase. When sediment traps were placed beneath the chlorophyll maximum at 60 m, the sinking particles collected contained high concentrations of ^{234}Th. From the known particulate scavenging rate of ^{234}Th, the residence time for the radionuclide could also be derived. The most important result from their study was that the ^{234}Th scavenging rate was found to closely relate to the rate of C export from the euphotic zone.

To summarize, Fig. 12 depicts current thinking on radionuclide scavenging. The dissolved species is taken up on small particles which, following aggregation into large particles, is removed from the water column by the rapid sinking of the larger aggregates. Aggregation of small particles often occurs through the grazing activity of marine organisms. The resultant large particles in the form of fecal pellets can also accumulate radionuclides and release them at depth [23]. It is essential to remember that although there is a much greater biomass of minute suspended particles than of large particles, it is the latter types which account for most of the mass flux in the sea [24].

3.8. CASE STUDIES

To better understand the basic concepts of radionuclide biogeochemistry outlined above, some case studies are presented which demonstrate the importance of biology in transferring and transporting some of these radionuclides.

During the 1980's, a particle flux programme called VERTEX (Vertical Transport and Exchange Experiment) studied element biogeochemical cycles in various regions of the north Pacific. An examination of results on transuranic flux in the open Pacific gyre, an area of very low production, are particularly interesting. In the Pacific ocean and also in the Atlantic and in some areas of the Mediterranean, there is a sub-surface maximum of Pu and Am concentration [25]. A typical profile of Pu in the Pacific depicting these maxima is shown in Fig. 13. This maximum, particularly for Pu, is found over the entire Pacific Ocean and questions have arisen as to what maintains the quasi-permanence of this feature. It has been hypothesized that sinking particles scavenge plutonium and then release it at the depth of the Pu maximum; i.e. it is controlled by particle interactions in surface waters and at depth.

A series of plankton samples including microplankton, copepods which feed on the microplankton, and fecal pellets produced by these copepods were collected from the surface water at a site in the central north Pacific gyre (Table 4). In general, the copepods had very low Pu and Am concentrations, but their fecal pellets were highly enriched in these transuranics exactly as occurs for other elements. Thus, fecal pellets appear to be good candidates for the packaging and removal of Pu and Am from the water column. The typical Am:Pu ratio in atmospheric fallout is about 0.3, which is not too unlike that measured in copepod zooplankton. However, the data in Table 4 show a ratio of 3.7 in fecal pellets which suggests that fecal pellets were enriched in Am relative to Pu.

At the same time these biological samples were collected, a series of sediment traps were also deployed at the site. Concentrations of Pu and Am in trap material were quite similar to those in the fecal pellets and, in fact, much of the particulate material in these traps consisted of fecal pellets. Taken together, these data indicate that as fecal pellets and other biogenic debris sank from the

TABLE IV. TRANSURANIC CONCENTRATIONS (pCi kg^{-1} dry + 1σ)** AND ACTIVITY RATIOS IN BIOLOGICAL SOURCE MATERIAL AND SAMPLES FROM SEDIMENT TRAPS [26]

Site	Samples		$^{239+240}Pu$	^{238}Pu	^{241}Am	Ratio $^{238}Pu/^{239+240}Pu$	Ratio $^{241}Am/^{239+240}Pu$
Station							
VERTEX IV	Microplankton (>60 μm)		11.4±4.5	2.7±1.4	9.4±2.8	0.24±0.15	0.82±0.41
N. Pacific	Marine snow		18.6±4.8	0.9±0.9	10.4±3.5	0.05±0.05	0.56±0.24
Gyre	Copepods (500-1000 μm)		1.8±0.3	ND	0.8±0.3	-	0.44±0.18
	Copepods (1000-2000 μm)		0.8±0.3	ND	0.6±0.3	-	0.75±0.47
	Copepod fecal pellets		144±110	ND	530±160	-	3.7±3.0
	PIT	150 m	25.1±2.6	1.3±0.5	5.9±1.4	0.052±0.020	0.24±0.06
		250 m	58.1±14.5	3.2±1.6	23.5±5.9	0.055±0.030	0.40±0.10
		500 m	199±30	5.5±3.0	126±11	0.028±0.016	0.63±0.09
		1000 m	405±35	11.9±5.4	271±22	0.029±0.014	0.67±0.06
		1500 m	301±45	8.9±4.4	273±24	0.030±0.015	0.91±0.14
	Bottom sediment Top 1 cm (X_n=4)		1.9±0.3		0.6±0.1		0.3

**1 pCi = 10^{-12}Ci = 37 mBq

ND: Not detectable

TABLE V. CONCENTRATIONS (Bq g^{-1} dry) OF PROMINENT CHERNOBYL FALLOUT RADIONUCLIDES IN LARGE PARTICLES COLLECTED AT 200 m WITH SEDIMENT TRAP MOORED IN 2200 m WATER DEPTH OFF THE COAST OF CORSICA DURING APRIL-MAY 1986 [28]

Sample No.:	1	2	3	4	5	6
Radionuclide	13-20 Apr.	20-26 Apr.	26 Apr.-2 May (Bq g^{-1} dry)	2-8 May	8-15 May	15-21 May
^{95}Zr				≤1.0	24.5±1.4	≤1.0
^{95}Nb				≤1.0	31.8±1.1	≤1.0
^{103}Ru				3.7±0.2	23.6±1.0	14.0±0.4
^{106}Ru				1.1±0.5	5.4±1.8	3.5±0.7
^{134}Cs				0.41±0.05	2.1±0.2	1.9±0.1
^{137}Cs	<0.05	<0.05	0.15±0.08	0.85±0.08	3.8±0.3	4.0±0.1
^{141}Ce	<0.5	<0.5	<0.5	1.3±0.7	12.6±0.6	1.1±0.5
^{144}Ce	<0.5	<0.5	<0.5	<0.5	13.6±0.7	<0.5
$^{239+240}Pu$*	5.43	2.00	3.00	3.22	9.70	4.71
^{241}Am*	0.87	0.68	1.51	1.05	3.63	2.83

*Bq kg^{-1}

surface to 1500 m, they scavenged and accumulated more Pu and Am. However, Am was being scavenged by particles more rapidly than Pu which eventually results in the elevated Am/Pu ratios that have been observed in trap samples [27]. There are few data on transuranic fractionation in particles below 1500 m, however, it is likely that particle remineralization results in a release of Pu and Am at depth. The release of Am and Pu from fecal pellets has been well-documented through laboratory radiotracer experiments [1,23]. One conclusion that can be drawn from this type of study is that in a nuclear waste disposal situation such as has occurred in the Irish Sea and elsewhere, biogenic particles are probably more effective in scavenging and removing Am from the water column than Pu.

One of the more instructive case studies involves the vertical transport of radionuclides following the Chernobyl accident on 26 April 1986. A radioactive plume from the damaged plant drifted across Italy and over the Ligurian Sea where a time-series sediment trap had been deployed in mid-April at a depth of 200 m. Radioactivity was first detected in air samples from Monaco on 30 April, 4 days after the accident occurred. A peak in aerosol activity was measured on 3 May followed by rain on 4-5 May which brought almost all of the fallout down as a single pulse during that period. The sediment traps were set to sample six intervals of 6.25 d each. In the first 3 sediment trap samples (13 April through 2 May), no radioactivity was detectable in the particles. The sample collected between the 2 and 8 May contained some measurable amounts of radioactivity (Table 5). Then, between 8-15 May, a maximum pulse of radioactivity was measured in the trap. This indicated that it took about 7 days for the radioactivity entering the surface to reach 200 m. Following this period, much lower concentrations were evident, particularly for elements like Ce and Nb which are particle reactive and were removed from the water column very rapidly. This observation further highlighted the "pulsed" nature of the fallout which was also transported downward as a pulse in this region of the Mediterranean [28]. To learn to what degree the biology was involved in this process, samples of plankton and their fecal pellets were collected using a zooplankton incubating device [29]. Briefly, the collector consists of a chamber suspended in filtered sea water which allows separating zooplankton from their excreta by sedimentation and seiving through a fine mesh screen. On 6 May, just after the radioactivity reached the surface waters, the zooplankton residing in these waters were sampled. The freshly produced fecal pellets contained similar radionuclide concentrations and ratios to those found in the sediment trap samples

TABLE VI. CONCENTRATIONS (Bq g^{-1} dry) OF CHERNOBYL FALLOUT RADIONUCLIDES IN ZOOPLANKTON AND THEIR FAECAL PELLETS COLLECTED ON 6 MAY 1986 AT THE TRAP SITE OFF THE COAST OF CORSICA [28]

Radionuclides	Zooplankton+	Faecal pellets
^{95}Zr	ND	1.4±0.8
^{95}Nb	0.012±0.003	ND
^{103}Ru	0.28±0.06	16.0±1.9
^{106}Ru	0.07±0.04	5.8±2.9
^{134}Cs	0.022±0.006	3.4±0.6
^{137}Cs	0.034±0.007	6.3±1.0
^{141}Ce	0.02±0.01	0.9±0.4
^{144}Ce	0.10±0.05	2.5±1.3
$^{239+240}Pu$*	0.016	7.4
^{241}Am*	0.004	0.63

+Zooplankton were almost exclusively adult copepods of the species *Centropages typicus* (97%), *Acartia clausii* (1%) and *Clausocalanus* sp. (1%)
*Bq kg^{-1}

TABLE VII. SELECTED RADIONUCLIDE ACTIVITY RATIOS OF CHERNOBYL-DERIVED RADIONUCLIDES IN DIFFERENT SAMPLES FROM THE NORTHWESTERN MEDITERRANEAN [28]

Sample	Collection Date	Activity Ratio			
		$^{137}Cs/^{141}Ce$	$^{103}Ru/^{141}Ce$	$^{103}Ru/^{137}Cs$	$^{241}Am/^{239+240}Pu$
Air	3 May	58	110	1.9	0.13
Unfiltered sea water	7 May	11	8.0	0.73	-
Zooplankton	6 May	1.7	14	8.2	0.22
Fresh faecal pellets	6 May	7.0	18	2.5	0.09
Trapped particles at 200 m	11 May	0.27	1.8	6.8	0.37

Air filter and sea water samples from coastal station at Monaco approximately 90 nautical miles northwest of trap site. The short half-lived radionuclides ^{103}Ru and ^{141}Ce have been corrected to 6 May.

TABLE VIII. VERTICAL FLUXES OF CHERNOBYL-DERIVED RADIONUCLIDES THROUGH 200 m AS DETERMINED BY PARTICLE INTERCEPTOR TRAP MEASUREMENTS IN THE NORTHWESTERN MEDITERRANEAN DURING APRIL-MAY 1986 [28]

Date	Mass flux ($mg\ m^{-2}d^{-1}$)	Radionuclide ^{95}Zr	^{95}Nb	^{103}Ru	^{106}Ru ($Bq\ m^{-2}d^{-1}$)	^{134}Cs	^{137}Cs	^{141}Ce	^{144}Ce	$^{239+240}Pu$*	^{241}Am*
13-20 April	213.7				Pre-Chernobyl					1.16	0.19
20-26 April	111.5				Pre-Chernobyl					0.22	0.076
26 April-2 May	63.9	-	-	-	-	-	0.0096	-	-	0.19	0.096
2-8 May	65.5	-	-	0.24	0.072	0.072	0.0557	0.085	-	0.21	0.069
8-15 May	53.6	1.31	1.70	1.26	0.29	0.11	0.2035	0.68	0.73	0.52	0.19
15-21 May	57.6	-	-	0.81	0.20	0.11	0.2305	0.063	-	0.27	0.16

*$mBq\ m^{-2}d^{-1}$

TABLE IX. CHERNOBYL RADIONUCLIDES (Bq/g dry) IN SINKING PARTICLES (50-250 m) AND BOTTOM SEDIMENTS OFF LA SPEZIA, ITALY, JULY 1986 [30]

9-18 July 25 miles off La Spezia	Dry/Wet Weight Ratio	^{103}Ru	^{137}Cs	^{134}Cs	$^{103}Ru/^{137}Cs$
50 m	0.1085	0.74+0.11	1.37+0.14	0.337+0.067	
150 m	0.1027	0.82+0.10	1.13+0.13	0.51+0.18	0.73
250 m	0.09544	2.14+0.19	0.64+0.15	0.37+0.10	3.34
470 m	(Surface sediment)	ND	ND	ND	

ND: Not detected

(Table 6). This strongly suggested that copepod grazing and the resulting fecal pellets were the prime mechanism that was moving radioactivity down in the water column. From knowledge of depth and collection times, a rough estimate of the sinking speed (30m/d) was made. Microscopic examination of the trap samples verified that copepod fecal pellets made up a substantial portion of the particulate material collected at 200 m. These field observations corroborated many of the vertical transport models that had been derived primarily from laboratory studies.

The ratios of radionuclides in trap material and other samples such as air, sea water, plankton and pellets were also instructive. For example, Cs is conservative in sea water in that it does not react to a great extent with particles. However, Ce in the oxidized form is very particle reactive. It can be seen in Table 7 that the Cs/Ce ratio decreased more or less in regular fashion going from air to sea water to plankton and finally to the material trapped at 200 m. The rather big drop in the ratio suggested that Ce was scavenged by plankton and particles much more than Cs. The net effect was the low Cs/Ce ratio found in the smallest particles.

From the radionuclide flux data shown in Table 8, it is possible to calculate a radionuclide inventory passing 200 m depth. For example, during the interval 26 April - 21 May, approximately 3.1 Bq ^{137}Cs m^{-2} passed through the 200 m layer. Comparing this to the 1383 Bq ^{137}Cs m^{-2} which was deposited as wet and dry fallout in the region through the end of May, it can be shown that only a very small fraction (0.2%) of the deposited ^{137}Cs was transported below 200 m by that time. In the case of Pu and Ce, comparable fractions were 75 and 50%, respectively, which underscores the greater reactivity of these radionuclides with sinking particles and hence their shorter residence time in sea water.

Some puzzles of Chernobyl-derived radionuclide behaviour still remain to be solved. In a similar flux study carried out during July 1986 in the Ligurian Sea near La Spezia, Italy, sediment traps were moored at 50, 150 and 250 m in a water column approximately 500 m deep. Following the 9-day deployment, sediment trap samples and the surface bottom sediment carefully removed from a box core were analyzed by gamma spectrometry. The sinking particles from all three depths showed strong signals of the Chernobyl fallout; however, interestingly, no Chernobyl-derived radioactivity was detected in the surface sediments at that site (Table 9). Thus, in spite of strong evidence that the radionuclides were being transported vertically at least to 250 m by sinking particles, radioactivity had apparently not yet reached the bottom at this site 2.5 months after it entered the surface waters. If these observations at La Spezia are confirmed, they imply that significant recycling of radionuclides was occurring in the upper water column of the Mediterranean. Unfortunately, at present, there are no other published data on Chernobyl-derived radionuclides in deep Mediterranean sediments with which to verify or refute the findings from the La Spezia study.

3.9. TRANSFER FROM SEDIMENTS

In any event, given sufficient time, biogeochemical processes will eventually result in the transport of radionuclides to the sediments. Because sediments tend to be the ultimate repository for many radionuclides, questions often arise regarding the possibility of the bioaccumulation of sediment-bound radionuclides by benthic species and their eventual transfer back to man. Over the years, many data have been gathered from areas contaminated by fallout as well as those directly exposed to radionuclide releases from the nuclear industry or accidents. While the scope of this presentation does not permit an in-depth review of this subject, it has been shown that the transfer of sediment-bound radionuclides to the fauna associated with them is generally very low [31,32]. As one example, laboratory-derived, sediment-organism transfer factors for several radionuclides are listed in Table 10. Transfer factors are defined here as the amount of radionuclide per gram organism divided by the amount of radionuclide per gram sediment. Values ranging between 0.0005 and 0.2 attest to the fact that, in most cases, very little of the sediment-bound radionuclide

TABLE X. CONCENTRATION FACTORS FOR THE UPTAKE OF RADIONUCLIDES FROM SEDIMENTS BY BENTHIC WORKS [1]

Species	Days of Exposure	$^{239,240}Pu$	^{241}Am	^{55}Fe	^{95}Zr - ^{95}Nb	^{137}Cs	^{106}Ru	^{60}Co
Polychaete								
Nereis diversicolor	40	0.0014	0.0005					
	(15)	(200)						
Nereis diversicolor	88			0.019				
Nereis japonica	11				0.01	0.2	0.006	0.06
	(11)				(4)	(6)	(6)	(6)

Numbers in parentheses are concentration factors based on uptake from water.

is taken up by benthic fauna. In fact, field studies such as those carried out at the plutonium contaminated site off Thule, Greenland, bear out the laboratory results [33].

Generally speaking, the higher the distribution coefficient (Kd) of the radionuclide between sediment and water, the lower the degree of uptake by organisms living in the sediments. On the other hand, as the corresponding concentration factor data in Table 10 show, when the source term for the radionuclide is water, the relative bioaccumulation by benthic species is one to four orders of magnitude greater than from contaminated sediments. Given this fact, it may be that the measured uptake from bulk sediments is really a function of bioaccumulation of the radionuclide associated with the interstitial waters in sediments. This subject has been an active area of radioecological research in recent years.

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