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LABORATORY EXPERIMENTS ON THE TRANSFER DYNAMICS

OF PLUTONIUM FROM MARINE SEDIMENTS TO SEA

WATER AND TO MARINE ORGANISMS¹

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ABSTRACT. The leachability of $^{239}(240)\text{Pu}$ from a fine contaminated calcareous sediment to aerated open sea water and to anoxic sea water was measured. The distribution coefficient for $^{239}(240)\text{Pu}$ from sediment to sea water was 6.1×10^{-5} for aerated water and 2.6×10^{-6} for anoxic water.

Experiments on the uptake of $^{239}(240)\text{Pu}$ by the clams Donax denticulatus, and Lucina pectinata, were done in aquaria containing kilogram quantities of sediment from the Bravo Crater at Bikini Atoll. The concentration factor for $^{239}(240)\text{Pu}$ by the soft parts of these clams was about 200. All the plutonium taken up in the soft parts was associated with the gill, mantle and siphon. No plutonium was detected in the adductor muscles or hepatopancreas. The smooth surfaces of the shells of the Donax did not show any detectable plutonium, but the rough shell surfaces of the Lucina concentrated plutonium by a factor of 1.10×10^4 over that in the sea water.

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Marine periphyton cultured on glass plates in an aquarium concentrated $^{239}(240)\text{Pu}$ by a factor of about 7×10^3 over that in the sea water.

Key words: Plutonium; leaching; sea water; sediment; uptake; clams; periphyton.

INTRODUCTION

The transuranic elements, plutonium (Pu) and americium (Am), are potentially among the most hazardous materials known to man (Langham, 1972). Increasing usage of these elements as predicted by Seaborg (1970) for both defense and peaceful applications will enhance the potential for environmental contamination from accidental release. It is expected that at least 10,000 metric tons of plutonium will be recycled annually by the year 2000 (U.S. Atomic Energy Commission, 1972).

Several accidents have occurred in the past involving plutonium (Noshkin, 1972). In addition to these accidents, higher than background levels $^{239}(240)\text{Pu}$, ^{241}Am , and ^{242}Cm have been detected in aquatic samples near fuel reprocessing facilities. The release of transuranium elements into the environment from accidents, reactor fuel processing, weapons testing and, in the future, by the operation of fast breeder reactors provide the basis for a need to understand the behavior of plutonium and the other transuranics in the natural environment. The knowledge gained should enable the orderly development of methods for radiation protection (Rogers and Henderson, 1971). Investigations on the environmental fate of the transuranium elements are more urgently needed now than before since the results may serve as a realistic guide for the safe development of increased nuclear power generation which is necessary to the United States for regaining and maintaining energy self-sufficiency (Rogers and Henderson, 1971;

Straugh, 1971; Ray, 1973).

Although several papers have been published on the physical, chemical, and biological properties of plutonium, little is understood about its biogeochemistry and radioecology, especially in the marine environment. With the release of plutonium into marine waters, data on the distribution coefficients and transfer rates of plutonium through the marine environment and the contained ecosystems would be an important factor for evaluating or predicting possible contamination of marine food sources important to humans and other animals.

For these reasons, a study of the transfer dynamics of plutonium from a contaminated calcareous sediment to sea water and to marine organisms under laboratory conditions was started.

The first phase of the investigation involved the measurement, under controlled laboratory conditions, of the leaching of plutonium from contaminated calcareous sediments (from the Bikini Atoll) to aerated open sea water and to anoxic sea water collected at or near Puerto Rico.

The second phase of the investigations was concerned with the measurement of uptake rates of the transuranium nuclides by tropical marine organisms under laboratory experiments.

These investigations may be easily done in Puerto Rico because many species of marine organisms similar to those found at Bikini Atoll are available in the marine waters of Puerto Rico and the analytical facilities are nearby.

EXPERIMENTAL

Radiochemical Procedures

Calibrated solutions of ^{242}Pu and ^{243}Am , were used for the simultaneous evaluation of recovery and counting efficiency in the isotopic dilution and alpha spectrometric determination of $^{239}(240)\text{Pu}$ and ^{241}Am .

For sediment and biological samples, known amounts of ^{242}Pu and ^{243}Am spikes were added and the samples dissolved by wet ashing with concentrated hydrochloric, nitric acid and hydrogen peroxide or with perchloric and hydrofluoric acids. The solution was evaporated to dryness and the residue dissolved in 4N. nitric acid containing a few drops of 50% hydrogen peroxide. Plutonium was separated and purified by extraction with the liquid ion exchanger triisooctylamine (Butler, 1965) and by subsequent extraction with thenoyltrifluoroacetone from an aluminum nitrate/nitric acid solution (Everett et al., 1949).

Americium was separated by a series of coprecipitation steps (Bokowski, 1965) with lanthanum hydroxide, bismuth phosphate and lanthanum fluoride and by selective adsorption on and elution off of AGI-X8 anion (Bojanowski and Livingston, 1972) and Dowex 50W-X8 cation (Chetham-Strode, Jr., 1956) resin columns.

After obtaining the radiochemically pure Pu and Am fractions, a lanthanum fluoride coprecipitation technique (Mo and Lowman, 1975) was used to prepare the alpha counting sources on membrane filters for quantitative alpha spectrometric determination of $^{239}(240)\text{Pu}$, ^{238}Pu , and ^{241}Am with silicon surface barrier detectors. Electroplating

was not used.

For the analysis of ^{239}Pu in sea water samples, ^{242}Pu spike was added to the sea water sample, and the solution adjusted to 1N hydrochloric acid. Plutonium was then coprecipitated on ferrous hydroxide by the addition of sodium bisulphite, ferric chloride and an excess of concentrated ammonium hydroxide solution. The ferrous hydroxide precipitate was then dissolved in 4N. nitric acid. The remainder of the procedure was the same as for the sediment and biological samples.

Counting Equipment and Technique

The samples were counted in an alpha spectrometer system consisting of silicon surface barrier detectors, each in a vacuum chamber connected to a liquid nitrogen trap and a mechanical vacuum pump. The detectors were connected to an analyzer with 8K of memory through the appropriate preamplifiers, amplifiers and mixer-routers. Standard procedures for alpha spectrometry with silicon surface barrier detectors were followed.

RESULTS AND DISCUSSION

Assay of Pu and Am Isotopes in Sediment and Plankton

Typical alpha spectra of the Pu and Am fractions separated from, (a) the Tewa (Nuclear Test) Crater sediment, and (b) a Bikini plankton samples are shown in Figure 1. Although the samples were precipitated onto membrane filters instead of being electroplated the resolution of the α peak of ^{242}Pu , $^{239(240)}\text{Pu}$, ^{238}Pu and the ^{243}Am , ^{241}Am is good enough in every case to allow a quantitative determination of the

Figure 1. Alpha spectra of the Pu and Am fractions separated from (a) the Tewa Crater sediment and (b) a Bikini plankton sample.

isotopes $^{239}(240)\text{Pu}$, ^{238}Pu and ^{241}Am . The results of the assay are shown in Table 1. Better agreement is obtained among the results of the duplicate analysis for $^{239}(240)\text{Pu}$ in the sample aliquots after the sediment was ball milled for 10 days. This would be expected if the $^{239}(240)\text{Pu}$ were present as particulate contamination in the sediment particles.

The alpha spectrum of the purified Pu fraction from a liter of Caribbean sea water is shown in Figure 2. This sample should not contain detectable amounts of $^{239}(240)\text{Pu}$ and the only alpha peak in the spectrum was that due to ^{242}Pu tracer that was added to determine chemical yield.

Leaching Studies

100 grams of the ball milled Tewa sediment were shaken with 10 liters of aerated Caribbean sea water which had been filtered through a membrane filter (pore size, 0.45μ) into a polyethylene carboy. The carboy was fastened to a sieve shaking machine and the top of the carboy was fitted with an 8.73 cm OD polyvinyl chloride (PVC) tube about 60 cm long. The PVC tube was covered with a large mesh number nylon screen to permit exchange of air between the sediment suspension and the atmosphere and also served to arrest any vertical component of sea water spray during periods of shaking. One liter aliquots of supernatant water were removed at time intervals of 1, 3, 5, 7, 9, 18, 24, 30, 48, 80 and 122 hrs, filtered through 0.45μ pore size, membrane filter and the leachates analyzed for

Table 1. Assay of Pu and Am isotopes in sediment and plankton samples

Sample Form	Wt.	$^{239}(240)\text{Pu}$	^{238}Pu	^{241}Am	$^{238}\text{Pu}/^{239}(240)\text{Pu}$
Aliquot No.	(g)	(dpm/g)	(dpm/g)	(dpm/g)	(Ratio)
Tewa sediment					
Not ball milled					
1.	0.5000	142 ± 8	11.4 ± 1.0	----	0.080
2.	0.5000	93.8 ± 5.9	7.80 ± 0.65	56.2 ± 4.4	0.080
3.	0.2180	163 ± 5	15.2 ± 0.85	81.2 ± 4.2	0.094
Tewa sediment					
Ball milled					
1.	0.3000	152 ± 5	13.0 ± 0.6	71.9 ± 4.4	0.085
2.	0.3000	214 ± 12	16.0 ± 1.4	109 ± 4.0	0.075
3.	0.3006	149 ± 9	11.0 ± 1.1	79.3 ± 3.7	0.074
4.	0.3069	162 ± 8	13.0 ± 1.1	45.1 ± 3.1	0.075
5.	0.3181	170 ± 9	12.1 ± 1.0	----	0.071
6.	0.3592	190 ± 11	15.1 ± 1.3	----	0.079
Bikini plankton					
Dry ash	0.2814	126 ± 5	1.69 ± 0.26	39.5 ± 3.3	0.020

Figure 2. Alpha spectrum of the Pu fraction separated from 1005 grams of Caribbean sea water.

plutonium. In order to keep the relative amounts of sediment to sea water constant, a liter of filtered uncontaminated sea water was added each time after a liter of the contaminated sea water was withdrawn. At each time of sampling the pH of the filtrate or leachate solution was measured and the content of $^{239}(240)\text{Pu}$ determined. The results are given in Table 2 and Figure 3.

Most of the plutonium that could be leached from the sediment into the sea water reached an almost constant maximum concentration of about 4 dpm/liter after about 1 or 2 hrs of shaking with the contaminated sediment.

The pH of the sea water remained constant at about 8.1 during the duration of the experiment which lasted 122 hours. The distribution coefficient of the $^{239}(240)\text{Pu}$ between the water and sediment was calculated to be 6.1×10^{-5} (ratio water/sediment).

The experiment was repeated with anoxic sea water in a closed system. The distribution coefficient of $^{239}(240)\text{Pu}$ between the anoxic sea water and sediment was 2.6×10^{-6} (ratio, water/sediment).

Thus under the conditions of the above experiments plutonium was an order of magnitude more soluble in aerated sea water than in anoxic sea water.

Uptake of Plutonium by Marine Clams

The solubility of plutonium in sea water is quite small (Hersh and Linquen, 1964) and plutonium introduced into estuarine and other marine areas would rapidly become associated with suspended particles

Table 2. Leaching $^{239}(240)\text{Pu}$ from "Tewa Crater"
sediment by sea water

Time of agitation (hours)	pH of leachate	$^{239}(240)\text{Pu}$ Alpha dpm/1. leachate
1	8.1	4.4 ± 0.4
2	8.1	4.7 ± 0.2
3	7.9	4.0 ± 0.3
5	8.0	3.7 ± 0.2
7	8.0	4.1 ± 0.4
9	8.1	4.4 ± 0.4
18	8.1	4.1 ± 0.4
24	8.1	3.7 ± 0.2
30	8.0	4.0 ± 0.2
48	8.1	3.6 ± 0.3
80	8.1	3.7 ± 0.2
122	8.1	3.2 ± 0.2

Figure 3. Leached $^{239}(240)\text{Pu}$ from Tewa sediment into aerated
Caribbean sea water.

LEACHING OF $^{239}(240)\text{Pu}$ FROM TEWA SEDIMENT BY
CARIBBEAN SEAWATER

and would be subject to normal sedimentary processes. In both fresh water and marine environments the sediments constitute the principal repository for plutonium (Amphlett, 1961; Welander et al, 1966). Plutonium, once deposited in the sediments is removed slowly by the water. This is confirmed by the leaching experiments reported in the present paper.

Benthic organisms have been reported by several investigators to concentrate plutonium in the soft and hard parts to levels higher than those in the water. Pillai et al (1964) reported that the Mussel Mytilus californianus concentrated ^{239}Pu 230 to 290 times the amounts present in the water, and Ward (1966) observed concentration factors of 200-300 in the viscera and 1.5 to 2.5 in the muscle of the lobster Homarus vulgaris. Wong et al (1970) reported a concentration factor of 380 in the Atlantic soft shell clam (Mya arenaria). Noshkin et al (1971) reported concentration factors of 300 in the soft parts of the blue mussel and Aarkrog (1971) a concentration factor of 1800 in a marine crustacean. However, all of these organisms live in or on the surface of the sediments, are detritus feeders or scavengers and probably are mainly exposed to sources of plutonium in the sediments and not in the water.

Weiss et al (1956) and Gong et al (1957) reported that clams are good biological indicators of radionuclide contamination of marine waters. Their conclusion was based upon the ability of these animals to concentrate radionuclides introduced into marine waters in fallout.

The concentrating ability of the clam for most radionuclides probably derives from its mechanism of feeding which involves the filtration of particles from large volumes of water. It is estimated that in one year a 7 cm clam filters about 2.3×10^4 liters of water.

In view of the ability of bivalve marine molluscs to concentrate many elements to levels exceeding those found in the water, the following experiments were done with two marine clams with different feeding habits.

Donax denticulatus, Linne (Abbott, 1974) is a select deposit feeder which lives in the surf zone of sandy beaches near the outflows of rivers in Puerto Rico. The clams are from 2.5 to 3 cm in length and weigh 2.5 to 3 grams. The weight ratio of soft parts to shell is about 1:3. These small clams inhabit well oxygenated zones of the marine environment.

The aquarium for the experiment contained 20 liters of continuously aerated sea water with about a kilogram of Bravo Crater sediment forming a 2.5 to 3 cm thick deposit at the bottom. The sediment was assayed for ^{239}Pu content and found to contain about 124 dpm of ^{239}Pu per gram. Thus, a total of 1.2×10^5 dpm of ^{239}Pu was present in the aquarium sediment. The aquarium water had a constant specific activity of 1.09 dpm ^{239}Pu /liter at equilibrium. 100 clams were placed in the aquarium and were fed on phytoplankton every two days. The clams started pumping actively after a few hours. Ten of the clams were removed from the aquarium at time intervals of 13, 28, 63, and 88 hours. After thoroughly cleaning the sediment particles that stick to the outside of each clam

with a spray of sea water from a wash bottle they were placed inside a beaker containing about 1 liter of uncontaminated sea water for 24 hours. They were then taken out, rinsed with sea water and the soft parts separated from the shells and the wet weight of the combined soft parts determined. Both the soft parts and the shells were dried overnight at 110°C in a drying oven and ashed at 550°C in a muffle furnace for about 16 hours. The dried ash was assayed for ^{239}Pu content in each case after addition of Pu-242 spike and after complete dissolution of the ash.

The results are shown in Figure 4. At the end of 90 hours the soft parts of the clams had concentrated $^{239}(240)\text{Pu}$ by a factor of 190 over the $^{239}(240)\text{Pu}$ concentration of sea water. Complete equilibrium was not reached so that the equilibrium concentration factor, based on the activity in the water, would exceed 190. However, these clams were living in the sediment and probably derived most of the $^{239}(240)\text{Pu}$ from the sediment, not the water. Based upon the plutonium activity in the sediment and in the clam soft parts, the clams discriminated against the uptake of $^{239}(240)\text{Pu}$ by a factor of 590.

Samples of Donax shells from clams taken at each collection time were etched with 1 N hydrochloric acid for a fixed time of 6 seconds. The resulting solution was assayed for $^{239}(240)\text{Pu}$ by alpha spectrometry and calcium by atomic absorption spectrometry. The results indicated that the outside surface of the shells of these animals did not take up $^{239}(240)\text{Pu}$ during the period of the experiment.

Figure 4. Pu-(239+240) uptake by the soft parts of the clam (Donax denticulatus, Linne).

$^{239}(^{240})\text{Pu}$ UPTAKE BY CLAM (DONAX DENTICULATUS, LINNE)

A separate experiment was done in which 15 clams were placed in a plastic basket hung in the water above the sediment for 5 days. For this length of exposure the clams concentrated $^{239}(240)\text{Pu}$ by a factor of 40 over that in the water, thus confirming the view that most of the plutonium accumulated in the soft parts of the clams was derived from the sediment and not the water.

A similar uptake experiment was repeated with another species of clam, Lucina pectinata, Gmelin (Abbott, 1974). Lucina are larger in size than Donax and live in and under anoxic mud in mangrove swamps. Lucina samples collected for these experiments varied from 7-8 cm in diameter and weighed 70-80 grams. The weight ratio of soft parts to shell was about 1:3.

Twenty-two clams were placed in an aquarium containing a kilogram of Bravo Crater sediment and 20 liters of continuously aerated sea water. The clams were fed phytoplankton every two days. Two clams were removed from the aquarium at time intervals of 1, 2, 4, 8, 14, and 28 days. These clams were subjected to the same cleaning procedures as those described above for Donax. The Lucina samples were dissected into the adductor muscle (3% of total weight), the hepatopancreas (10% of total weight), a combined sample of gill, mantle, and siphon (12% of total weight), and the shell (75% of total weight). The soft parts of the clam were assayed for ^{239}Pu following the same procedures as for the Donax clams. The results are shown in the Table 3 and Figure 5.

Table 3. Uptake of $^{239}(240)\text{Pu}$ by the clam (Lucina pectinata, Gmelin)

Exposure Time (Days)	Alpha -dpm/kg. wet wt.			
	Gill, mantle siphon	Adductor muscle	Hepatopancreas	Shell
1	162 $\pm$ 51	N.F.	N.F.	3310 $\pm$ 332
2	163 $\pm$ 19	N.F.	N.F.	6908 $\pm$ 521
4	210 $\pm$ 24	N.F.	N.F.	11033 $\pm$ 1103
8	184 $\pm$ 12	N.F.	N.F.	11013 $\pm$ 743
14	123 $\pm$ 16	N.F.	N.F.	10036 $\pm$ 782
28	243 $\pm$ 16	N.F.	N.F.	12251 $\pm$ 1043

N.F.: Not Found

Figure 5. Plutonium-239(240) uptake by the clam (Lucina pectinata,
Gmelin).

$^{239} (^{240})\text{Pu}$ UPTAKE BY CLAM (LUCINA PECTINATA, GMELIN)

No plutonium was found in the adductor muscle or hepatopancreas samples. This finding is consistent with the results of Noshkin (1972) who reported that the adductor muscle of the scallop contained levels of plutonium about two orders of magnitude lower than the rest of the body and that the plutonium content of all of the fish muscle samples was so low that it was almost undetectable. The plutonium uptake data by lobster obtained by Ward (1966) also indicated that the muscle of the lobster contained insignificant amounts of Pu compared to the rest of the body. Reichle and coworkers (1970) reported that the lanthanides and actinides are poorly absorbed (less than 1%) through the gastrointestinal walls of most animals. Thus, it appears that plutonium and other actinides pass through biological membranes with very low efficiency. The combined sample of gill, mantle, siphon and foot indicated levels of $^{239}(240)\text{Pu}$ greater than the amounts found in the aquarium water. This might be expected since the gills, the mantle and the siphon were directly exposed to the contaminated sediments. The $^{239}(240)\text{Pu}$ distribution among the different organs of this group was determined on clams that had lived in the aquarium for two months. The ratio of the $^{239}(240)\text{Pu}$ specific activity for the mantle, siphon and gills was 3.5/1.5/1 respectively.

Uptake equilibrium was established in the combined sample of gills, mantle, siphon and in the shell after about 5 days of exposure (Figure 5).

A concentration factor of 193 was observed for the combined sample of gill, mantle, and siphon for $^{239}(240)\text{Pu}$ uptake, based on the amounts of Pu in the water. This value for the Lucina is consistent with the value of 193 observed in the soft parts of the Donax clams. However, in Lucina as in Donax the source of plutonium found in the soft parts probably was the sediments in which the clam lived and not the overlying water. The fact that $^{239}(240)\text{Pu}$ was not found in the muscle or hepatopancreas, neither of which were directly exposed to the sediments reinforces this view. In the case of the gill, mantle and siphon most of the plutonium probably was adsorbed to the surfaces of the organs. Based upon the amounts of plutonium observed in (on) the gill, mantle and siphon and the amounts which were present in the sediments these clams also discriminated against the uptake of $^{239}(240)\text{Pu}$ by a factor of at least 590. Lucina pectinata placed in plastic baskets hung in the aquarium water without touching the sediment for 5 days did not take up detectable amounts of $^{239}(240)\text{Pu}$. This suggests that the Lucina were more efficient than the Donax in removing excess sediments from the surfaces of the soft parts.

Based on the amounts of $^{239}(240)\text{Pu}$ in the water, a concentration factor of about 1.1×10^4 was calculated for the surfaces of the shells of these clams after 5 days. Most of the plutonium activity associated with the clams was, therefore, on the outside surface of the shells. The shells of Lucina were in direct contact with the sediments, however, and based upon the activity in the sediments and in (on) the outside

surfaces of the shells of this clam, the plutonium was not concentrated by the surface of the shell. The ratio of $^{239}(240)\text{Pu}$ per gram of shell surface to that per gram of sediment was 0.09. In all of the clam samples in which plutonium was detected the $^{238}\text{Pu}/^{239}(240)\text{Pu}$ was 0.08. The same ratio was observed in the sediment (Table 1).

Uptake of Plutonium by Marine Periphyton

Marine periphyton was raised on six glass plates (25 cm x 35 cm) each of which two thirds was immersed in 60 liters of sea water in a 100-liter glass aquarium. The aquarium also contained 3 kilograms of Bravo Crater sediment which formed a deposit of about 2 cm at the bottom. The sediment contained 3.72×10^5 dis/min of $^{239}(240)\text{Pu}$ (124 d/m/g). The sea water had a specific activity of 1.1 dis/min of $^{239}(240)\text{Pu}$ /liter at equilibrium. One, 2 and 3 glass plates were removed from the aquarium at times of 1 month, 2.5 months and 3.5 months respectively, allowed to drain thoroughly and the periphyton removed with a stainless steel razor blade. In each case, a portion of the sample subjected to neutron activation analysis indicated that one third of the wet weight of the sample was due to sodium chloride. These results were used to make a "salt" correction for the wet weight of the remaining portions of the periphyton samples. The samples were assayed for $^{239}(240)\text{Pu}$ by using the methods described before. The results are shown in Table 4. At the end of 3.5 months the periphyton had concentrated $^{239}(240)\text{Pu}$ by a factor of 7.1×10^3 over the $^{239}(240)\text{Pu}$ concentration of the sea water. This value approached the value of

Table 4. Uptake of $^{239}(240)\text{Pu}$ by marine periphyton

Time (months)	$^{239}(240)\text{Pu}(\text{dis/min/kg})$		Concentration factor
	Wet periphyton	Water	
1	8200 ± 900	1.09 ± 0.07	7.5×10^3
2.5	5400 ± 800	1.09 ± 0.07	5.0×10^3
3.5	7800 ± 400	1.09 ± 0.07	7.1×10^3

1.2×10^4 obtained for the concentration factor of $^{239}(^{240})\text{Pu}$ by the algal plankton, Chlorococcum and reported elsewhere by Ho and Lowman, (1975).

CONCLUSION

The ^{239}Pu now present in Bikini Lagoon was produced by nuclear weapons fired on or near the coral reefs. Thus, large amounts of calcareous material and varying amounts of sea water were vaporized and mixed with the fireball at the times of detonation. Under the conditions of high temperatures and pressures which exist in the plasma fireball, the vaporized elements would exist as stripped nuclei and free electrons and would be subject to violent mixing. Upon reduction of temperature with time the orbital electrons would be recapture by the plasma nuclei and at about 4000°C to 5000°C the "refractory" elements with boiling points greater than 3000°C , including plutonium, would condense. Calcium, which may be classified as a "volatile" element would condense later on the condensation nuclei provided by the "refractory" and "intermediate" elements. Thus, the condensed plutonium oxide would be distributed throughout the sediment particles and not on their surfaces, a factor which would influence the availability of the plutonium to periphyton and sea water.

In the present experiments extremely small amounts of ^{239}Pu were leached from the calcareous sediments. The observed distribution coefficient of 6.1×10^{-5} (water/sediment) for aerated water is in agreement with the observations of other investigators on the solubility of plutonium in natural waters (Hersh and Lingren, 1964).

The solubility of plutonium at the sediment-sea water interface

is dependent upon pH, Eh, ionic strength and complexing ions including carbonate and bicarbonate, organic chelators and living organisms. (Gel'man and Moskvina, 1958; Andelman and Rozzell, 1970). Plutonium normally exists in four oxidation states in water (Pu (III), Pu (IV), Pu (V) and Pu (VI)) and the fate of soluble plutonium in the sea depends upon the interactions of these factors with the chemical-physical form of plutonium introduced into the marine environment. The actinide metals are similar to the lanthanides in being highly electropositive although the actinide ions tend to form stronger complex ions than do the corresponding lanthanides (Seaborg, 1958). Plutonium, like the rare earths thorium and zirconium, hydrolyzes in sea water to form insoluble oxides and hydroxides as well as sequestered ions. Polymerized plutonium may form a positive colloid and become adsorbed onto a variety of surfaces. Adsorption of plutonium increases with decreased acidity and the element is strongly adsorbed onto surfaces that become negatively charged in water. The ability of plutonium to attach to these surfaces is also influenced by the size of the colloidal particles and increases with decreased particle size (Starik, 1959).

The ability of dissolved or colloidal plutonium to adsorb onto inorganic and organic surfaces strongly influences its incorporation, transfer and deposition in marine food webs or ecosystems. The strong adsorptive characteristics of the element enhances its association with small organisms, however, the same characteristic

strongly reduces its ability to pass through biological surfaces, including membranes. Because of this, plutonium which is passed up through food webs, is subject to strong discrimination factors with increased trophic level.

Periphyton and marine phytoplankton are composed of small organisms and the concentration factors for plutonium from sea water of 7.1×10^3 for periphyton and 1.2×10^4 for phytoplankton reported here are among the highest concentration factors which have been found for plutonium in marine organisms. In contrast, extremely low amounts of the radioelement were able to enter the internal organs of the two species of clams studied in the present work although the clams were living in highly contaminated sediment.

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