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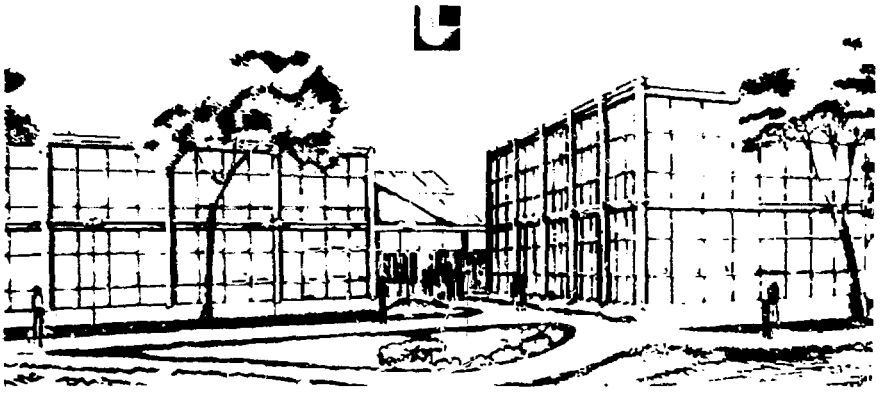

[illegible] MODIFICATION BY [illegible] SEDIMENTARY [illegible] TO [illegible] IN THE MARINE ENVIRONMENT

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ABSTRACT

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INTRODUCTION

Planning for sea-bed deep sea or deposition of radiological wastes (either high or low level) must necessarily account for the various processes, reactions, and rates that influence the fate of specific long-lived radionuclides in the event of their release to the sedimentary environment. Many of the processes leading to release of radionuclides and the migration pathways for these activities from buried and/or disposed containment systems leading back to man have been defined [1, 2, 3] but are still imperfectly understood.

Significant quantities of plutonium isotopes and other transuranics will be among the longer-lived, toxic radionuclides associated with radiological waste materials. It is therefore essential to understand by what processes, if any, and at what rates plutonium, subjected to various chemical, physical, and biological disturbances on the sea floor, migrates back to the oceanic water column.

During the past few years we have been conducting studies at Enewetak and Bikini Atolls to better define the environmental physical, chemical, and biological transport mechanisms and fate of the transuranics and other long-lived radionuclides in the aquatic environments. The radionuclides were introduced to the environments during testing of nuclear devices by the United States at these Pacific Atolls between between 1946 and 1958 and were subsequently deposited to the lagoon sediments in association with settling particulate material. These radionuclides are studied mainly to evaluate their impact on critical processes essential for the establishment and continuity of life at the atolls and partly because we recognize these studies can provide data of some significance related to understanding mobilization and migration of plutonium and other radionuclides from oceanic sedimentary deposits to the water column and back to man. The radiological studies at the atolls are therefore germane to problems related to the disposal of transuranic and other radioactive wastes in the ocean. In this paper we discuss in-situ results related to the partitioning of plutonium between solid sources and sollution in the atoll environment. A great deal of similarity has been found in the aquatic characteristics and behavior of plutonium at Enewetak and Bikini. For example, the rates of plutonium mobilization and atoll residence time are very similar. The results from one atoll have great value in predicting transuranic behavior at other Pacific Atolls and, as we will show, in contrasted marine environments. Results from Enewetak Atoll will be emphasized and supplemented by data from Bikini when it is necessary to clarify the interpretation of data.

Enewetak Atoll and Plutonium Inventory

Enewetak Atoll consists of 39 islands on an elliptical coral reef encompassing a lagoon with an area of 931 km^2. The islands, which were given alphabetic code names during the U.S. occupancy, and several land marks including the locations of craters formed by nuclear tests are identified and shown in Figure 1. The islands, which make a total land area of approximately 6.9 km^2 are situated on a reef 84 km^2, in area. The average depth of the lagoon is 47.4 m; the maximum depth is 60 m.

The U.S. moritorium on testing began on 31 October 1958 and marked the end of all nuclear testing at Enewetak and Bikini Atolls. The fallout history plus other activities during and after the testing period produced a very heterogenous distribution of radionuclides in the lagoon sediments. Today

quantities of long-lived fission products such as ^{137}Cs, ^{90}Sr, ^{155}Eu, and others; activation products such as ^{55}Fe, ^{60}Co, and ^{207}Bi; and transuranics such as ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am persist in the atoll's environment. The largest inventory of plutonium at Eneweta and Bikini is found associated with the lagoon sedimentary components. Analyses of over two hundred core samples collected from Eneweta lagoon in 1971 defined the areal distribution of plutonium in the sediment and showed that approximately 250 curies are associated with the surface 2.5 cm layer and the inventory to a depth of 10 cm in the sediment column is estimated to be three times.

Highest concentrations are associated with the sediments from the northwest quadrant of the lagoon nearby and to east of the islands of Aomoen and Belle (see Figure 1) and several km southwest of Muzen and Kea craters. A second region of relatively high concentrations is in the sediments off the south islands. Most of the plutonium inventory in the surface sediments can be separated roughly into the lesser contaminated map bits by a line extending from the southwest passage to the island of Togo (Murphy), which is south of Yvonne at the eastern reef. The surface $^{239+240}Pu$ concentrations within this area range from 1 to 170 pCi/g dry weight; those outside of this area are less than 1 pCi/g. The average concentration in the lagoon sediment determined from the samples collected during 1971 is 5.2 pCi/g dry weight. Bikini sediment inventories were estimated from substantially fewer data that were available for Eneweta, and future results from Bikini might alter the following estimates. Of the plutonium inventory now resident in the surface 15-cm-thick layer and 47% outside this area, 5% is in the sediment column from 15 to 25 cm and the first 10 cm of sediment is 9.5 pCi/g dry weight of sediment.

For the first nuclear test at Eneweta on 1948, the radioactive fallout debris deposited to the lagoon waters either settled to the bottom, remained as dissolved or particulate species in the water and were eventually discharged to the North Equatorial Pacific by the prevailing exchange of water between the ocean and the lagoon. It has been generally stated that, following introduction to the surface layer of the ocean from any source term, plutonium quickly combines with particles and settles rapidly to the seabed where it remains immobile after deposition. Accepting this argument to be correct, concentrations of $^{239+240}Pu$ and other long-lived radionuclides in the lagoon water column during any year subsequent to 1948 should then only equal the temporal fallout levels in the North equatorial Pacific surface waters.

Concentrations in the Water:

A considerable number of lagoon water samples have been collected and analyzed for $^{239+240}Pu$ by this laboratory since 1972. Several studies are in progress at the present that require data on concentrations in lagoon waters. The number of samples and location sampled during any year will predicate on the requirements of the program. Table I summarizes arithmetic mean $^{239+240}Pu$ concentrations and the range in concentrations detected in filtered water samples collected during different periods from the regions of Eneweta and Bikini indicated. As shown for comparison are concentrations in surface water collected 1-2 miles outside (west and south) Bikini and Eneweta Atolls, surface concentrations in the Equatorial Pacific 25-90 miles directly west of Bikini, concentrations in lagoon water at other Marshall Island Atolls, and levels in the surface North Equatorial Pacific water well away from the Atolls. The range in values in the lagoon shown in Table I indicates there are significant spatial and temporal differences in concentrations

in the water. However, wherever and whenever water was sampled in the lagoon or on the reef, the $^{239+240}Pu$ concentrations in solution and in association with particles greatly exceeded the 0.3-0.5 fCi/l fallout background levels in the Equatorial Pacific surface waters. These results are a direct indication that $^{239+240}Pu$ is mobilized to solution from the solid phases of the environment. Concentrations in the perimeter surface samples and in the surface water at distances west of Bikini show there is a flux of mobilized $^{239+240}Pu$ continuously advecting from the lagoon to the Equatorial Pacific water mass. Small but measurable amounts of $^{239+240}Pu$ are continously mobilized to solution from sources within the atoll, resuspended to the water column for subsequent redistribution both within and outside the atoll, and are concentrated by all lagoon organisms. The term "soluble" plutonium herein refers to the quantity of plutonium in water samples that passes through filters of stated diameters. A considerable number of tests have been run these past years using filters of different pore sizes. We have found that a 1 micron filter, normally used to remove particulates, is as efficient as 0.45- and 0.4-micron filters. We have been unable to identify the species of plutonium in solution but we have identified many of its characteristics. We find, for example, it is present in the lagoon water in more than one valence state; it has solute like characteristics and passes readily through dialysis membranes; less than 6% of the plutonium in solution is found associated with organically bound material; the quantity mobilized to solution on the windward reef is relatively inert and highly complexed when compared to the highly exchangeable species in the lagoon; the species in the lagoon has exchange characteristics similar to fallout levels in the open ocean; dissolved plutonium released on the reef has been traced for considerable distances by a plutonium radionuclide balance that involves the change in the $^{238}Pu:^{239+240}Pu$ ratio in the water; the dissolved plutonium moves in solution apparently without interacting rapidly with sediment deposits during transport; ^{238}Pu behavior is similar to $^{239+240}Pu$, and although we find some similarities between the characteristics of ^{241}Am and $Pu^{239+240}$, there are sufficient differences in properties that set the behavior of the two transuranics apart.

Rates of Plutonium Loss from the Lagoon

A complete description of the biological, physical, and chemical interactions that are potentially capable of mobilizing $^{239+240}Pu$ from the solid sedimentary sources at a coral atoll and of the processes moving $^{239+240}Pu$ to and within the lagoon water mass are beyond the scope of this report. We take a less sophisticated approach in the form of radionuclide budgets to attempt an interpretation at a rather simple level of the rates and processes affecting $^{239+240}Pu$ mobilization. However, by following this procedure we have been able to determine that some processes and mechanisms dominate the mobilization and redistribution of $^{239+240}Pu$ at the atolls; some are insignificant and some appear important but are difficult as yet to test experimentally.

There is a significant quantity of environmental data that now shows that the amount of $^{239+240}Pu$ mobilized to solution in the lagoon has been relatively constant for perhaps as long as the past 13 years. The lagoon water mass, with the dissolved inventory of $^{239+240}Pu$, is constantly replaced by ocean water containing only background levels of $^{239+240}Pu$. In order to maintain a constant, elevated $^{239+240}Pu$ level in the lagoon water, the rate of plutonium removal from the atoll must be balanced by a continuous flux of dissolved plutonium from sources within the atoll. Horizontal and vertical concentration

gradients prevail in the lagoon water mass during any period sampled. To attempt a material balance, it is necessary to assume that a reasonable, average concentration in the lagoon can be derived from the results during those periods, when sufficient samples were obtained from the lagoon for analysis. Results from Enewetak during November 1972, July-August 1974, and April-May 1976 and from Bikini during November 1972 and January-February 1977 show that this is a reasonable assumption. During November 1972 the average concentration of plutonium (soluble plus particulate) in the lagoon water at Enewetak was 32 fCi/l. In 1974 and 1976, the average concentrations of soluble $^{239+240}Pu$ were 24 and 16 fCi/l, respectively, and the particulate concentrations represented 46 and 43%, respectively, of the average total concentrations of $^{239+240}Pu$. Assuming there is, at any time, 44% of the total plutonium in association with the particulate phase, we estimate that in November 1972 10 fCi/l of $^{239+240}Pu$ was in association with suspended particulates and 22 fCi/l was the average soluble concentration. The differences in the soluble concentration determined during these 3 periods is not considered significant. It should be emphasized that the average concentration is based on results each year from a different number of samples. These samples were quite often taken from different lagoon locations. When concentrations were determined in water samples from lagoon stations previously sampled, more than a time or two out of 4 the agreement in measured values was excellent. Therefore, in spite of sampling different water masses during different seasons of different years, the average quantity of $^{239+240}Pu$ in solution, based on results from different numbers of samples, has been reasonably constant, at least since 1972. During 1972 and 1977, the average concentrations of soluble $^{239+240}Pu$ in Bikini lagoon water were 40 and 49 fCi/l, respectively. No significance is attached to these differences and, as at Enewetak, we assume the standing average amount of $^{239+240}Pu$ in the lagoon water mass at anytime has been constant. At Bikini in 1972, the average concentration of $^{239+240}Pu$ associated with lagoon particulates represented 48% of the total concentration in the water. This average is very similar to the mean percentage now measured twice at Enewetak.

With the appropriate dimensions for each lagoon, the average concentrations in solution at Enewetak and Bikini convert to a plutonium standing inventory of 0.9 and 1.9 curies, respectively. The average inventories associated with lagoon suspended particulates are 0.7 and 1.1 curies, respectively. At Enewetak the quantity in solution represents only 0.36% of the $^{239+240}Pu$ inventory in the sediment measured to a depth of 16 cm in the sediment column and 0.075% of the total inventory to a 16 cm depth. At Bikini, the mean soluble inventory of $^{239+240}Pu$ in the lagoon water is 0.40% of the 470 curies estimated in the surface 2.5 cm layer of sediment and 0.09% of the 1470 curies inventory to a 16 cm depth. Particulate inventories in the water column are proportionally smaller fractions of the respective sediment inventories. The amount of $^{239+240}Pu$ mobilized and found at any time in solution at the atolls represents a very small fraction of the inventory in the major atoll reservoirs.

Radiological data from different biological indicators show that the lagoon- water residence times vary between 118-170 days with a reasonable average being 144 days. However, physical circulation data indicate that the lagoon water mass, on the average, is exchanged with the open ocean at a much more rapid rate [4,5]. A resolution of these differences is in progress, but for the present, the slower rate of 144 days will be taken to represent the rate at which the $^{239+240}Pu$ in solution is

exchanged between the lagoon and open ocean. A large fraction of the $^{239+240}Pu$ associated with the lagoon particulates is associated with resuspended sediments. Very little sedimentary material escapes from the lagoon, and resuspended bottom material probably settles out again on the lagoon floor close to its point of origin. Therefore, no plutonium associated with particulate material is assumed lost from the lagoon.

Using the residence time of 144 days and the average soluble inventories of plutonium, 2.8 and 3.2 curies, respectively, are discharged annually to the open ocean from Enewetak and Bikini Atolls. If the inventory to 16 cm in the sediment column (1200 curies) is the reservoir for the remobilized $^{239+240}Pu$ at Enewetak, then the mean life for $^{239+240}Pu$ in this reservoir is 435 years. The mean life is computed from Equation 1, where dN/dt is the annual rate of plutonium depletion, N represents the total sedimentary inventory, corrected, and t is the mean life.

$$\frac{dN}{dt} = \frac{N}{t} \qquad (1)$$

At Bikini, the computed mean life for the $^{239+240}Pu$ in the sedimentary reservoir, to a depth of 16 cm in the lagoon column, is 460 years. At this time the inventory of $^{239+240}Pu$ at any time in these lagoon waters represents a small fraction of the sedimentary inventory if the mobilization processes continue at the same rate. $^{239+240}Pu$ will be depleted from the sedimentary environment in a geological time span that is short compared to the half-life for $^{239+240}Pu$.

Exchange of Plutonium Between Sediment and Seawater

Since $^{239+240}Pu$ has been found in solution at the atoll, there must be some release of $^{239+240}Pu$ from the contaminated sediments to the water phase as the result of continuous or loss by some other means. A number of laboratory and in-situ experiments have been conducted with contaminated sediments from the lagoons to arrive at a value for the distribution coefficient (K_d) for plutonium. Laboratory studies involved measuring the fraction of $^{239+240}Pu$ desorbed from the sediments while field studies involved the collection and analysis of interstitial water and sediment. Different size fractions from different geographical regions and from different depths within the sediment column were used in desorption experiments with uncontaminated seawater. The range in K_d values for the different lagoon sediments was between 0.5×10^5 and 3.6×10^5 with an average K_d value for $^{239+240}Pu$ of 2.3×10^5.

Nelson and Lovett [6], recently studied plutonium distributions in the Irish Sea. They found that the average K_d for Pu (+3 or +4) in seawater was 24.9×10^5 while the average K_d for the oxidized forms of Pu (+5 or +6) in seawater was 0.15×10^5. The method outlined by Nelson and Lovett [6] was followed to separate the reduced from oxidized forms of dissolved $^{239+240}Pu$ in the lagoon seawater at Enewetak and Bikini. Analysis of three lagoon water samples showed that on the average, 92% of the dissolved $^{239+240}Pu$ was in the oxidized (+5 or +6) form while the remaining 8% represented a reduced (+3 or +4) state of dissolved $^{239+240}Pu$. These results demonstrate that different oxidation states of $^{239+240}Pu$ are capable of coexisting in the lagoon water.

Also using the K_d values for the oxidized and reduced states of $^{239+240}Pu$ from Nelson and Lovett [6], and the fractions of oxidized and reduced $^{239+240}Pu$ in seawater at Enewetak, we have been able to correctly predict our average measured K_d value of 2.3×10^5. The value of the distribution coefficient for any element relates to the activity on solids in equilibrium with a quantity in water (K_d = pCi g^{-1}/pCi ml^{-1}). If two species coexist in a seawater/sediment system, then an apparent K_d would fall between the two respective values as shown for the different oxidation states of plutonium by Equations (2) and (3).

$$\frac{\text{pCi g}^{-1}(+3 \text{ or } +4) + \text{pCi g}^{-1}(+5 \text{ or } +6)}{\text{pCi ml}^{-1}(+3 \text{ or } +4) + \text{pCi ml}^{-1}(+5 \text{ or } +6)} = K_d \text{ apparent} \qquad (2)$$

$$\frac{K_{d(+3 \text{ or } +4)} \times \text{pCi ml}^{-1}(+3 \text{ or } +4) + K_d (+5 \text{ or } +6) \times \text{pCi ml}^{-1}(+5 \text{ or } +6)}{\text{pCi ml}^{-1}(+3 \text{ or } +4) + \text{pCi ml}^{-1}(+5 \text{ or } +6)} = K_d \text{ apparent} \qquad (3)$$

Substituting the respective K_d values and the fractions of the oxidized and reduced plutonium in seawater at Enewetak into Equation (3) results in an apparent K_d of 2.1×10^5, which is in excellent agreement with our average measured value. The range in K_d values encountered with sediments from the lagoon may merely reflect the quantity of oxidized and reduced plutonium species capable of disassociation from the particular lagoon sediments tested.

The average concentration of $^{239+240}Pu$ in the surface 10 cm of sediment at Enewetak and Bikini are, as previously given, ... and 6.5 pCi gm^{-1}, respectively. If the surface sediment layer is forcibly mixed with overlying lagoon water, mixing of the interstitial water with dissolved $^{239+240}Pu$ would take place. A fraction of the exchangeable $^{239+240}Pu$ held on the suspended material will also be liberated during each mixing. There are several other identified processes that may cause mixing of the interstitial lagoon fluids and the dissolved $^{239+240}Pu$ with the lagoon water. Assume that a simple chemical balance is continuously maintained between the $^{239+240}Pu$ in the sedimentary sources and the interstitial and interface water. The rate at which the interstitial and interface water mix within the lagoon is balanced by a flux of undersaturated lagoon water which, then rapidly equilibrates with the exposed sediments. This process simulates a huge continuous later extraction in which we allow the $^{239+240}Pu$ retained on the solid sediments to be continuously in equilibrium with the overlying water column, and the desorption mechanism follows the law of mass action applicable to an ion-exchange type equilibrium. Then average concentrations of $^{239+240}Pu$ in the lagoon water at any time should relate to the K_d value and the average sediment concentrations. Computed inventories and average concentrations in the water, using the basic equation relating K_d to concentrations in the water and sediment, are 0.99 curies and 23 fCi/l, respectively, at Enewetak, and 1.3 curies and 41 fCi/l at Bikini. There is general agreement between the average quantities of $^{239+240}Pu$ predicted and those measured in solution during each of the periods of 1972, 1974, 1976, and 1977 at Enewetak and Bikini. For many reasons, it may be argued that this agreement is fortuitous. Nevertheless, the general agreement found between computed and the average concentrations in both lagoons between 1972 and 1977 measured several times supports the contention of a steady state condition and demonstrates the general usefulness of this simple model in predicting long term average concentrations in the lagoon water.

In summary, small quantities of $^{239+240}Pu$ are found to be continuously mobilized to solution from the sedimentary sources at Enewetak and Bikini Atolls. The plutonium is slowly being depleted from the atoll reservoir and is discharged to the surface waters of the North Equatorial Pacific. $^{239+240}Pu$ mobilized to solution has solute-like characteristics, is available for uptake by organisms, and different valence states are capable of coexisting in solution. It will require more than 400 years to mobilize the entire inventory of $^{239+240}Pu$ from these atoll sediments. Although this period of time is long compared to our lifespan, it is small compared to the radiological half-life of plutonium.

The ratio of the average concentration in lagoon water at Bikini and Enewetak is approximately 1.8. This value is nearly identical to the ratio of the mean sediment concentrations of 9.5 and 5.2 pCi gm^{-1}. The computed mean lives for $^{239+240}Pu$ in the sediments of both lagoons are essentially the same.

The average quantity of $^{239+240}Pu$ mobilized to the water column is proportional to the average concentration in the lagoon sediments. Therefore, geological mean lifetimes for plutonium in sediments from any atoll will be similar to the mean life as determined at Enewetak and Bikini, and the average concentration in the overlying water column will be proportional to the respective concentration in the sediment column considered. A simple mass action model can be used to estimate the quantity of $^{239+240}Pu$ capable of dissociation from the sediments to solution.

Concentrations in Equatorial Pacific Waters

During June and July 1978, we participated in a joint oceanographic cruise to regions of the Equatorial Pacific with V.T. Bowen of Woods Hole Oceanographic Institution. Water, sediment, and plankton samples were collected, and several other ancillary experiments were conducted. The following represents a summary of plutonium water-column inventories in samples we collected for analysis. Some of these conclusions may be revised after the results from both laboratories are critically compared.

Inventories of the $^{239+240}Pu$ to different depths for five stations, along with the calculated range, in the North Equatorial Pacific are given in Table II. Stations 2 and 4 are 270 to 300 miles to the east and northeast, respectively, of Bikini Atoll. Station 9 is approximately halfway between Bikini and Enewetak Atolls. Station 11 is 15 miles west of the northwest reef of Enewetak and Station 13 is located about 212 miles northwest of Enewetak. Between the surface and 1000 m depth, there is a slight east to west gradient in the $^{239+240}Pu$ inventory, but the mean values from all stations fall within the average inventory of 1.28 ± 0.10 mCi/km^2. This quantity alone in the upper 1000 m exceeds the estimated global fallout levels delivered to these latitudes [7]. The inventory of plutonium in the 1000 m of water at Stations 9 and 11 are essentially identical to the quantities in the upper 1000 m of water at Stations 2 and 4. Plutonium concentrations from 3 to 10 times higher than fallout levels were previously detected in surface water samples collected west of Bikini and west of Enewetak. An estimated 3 curies of plutonium are remobilized annually from the lagoon sedimentary deposits and exchange with the north equatorieal surface waters. This annual input to the surface layers of the North Equatorial Pacific is so rapidly

diluted and horizontally transported westward that essentially no change in the plutonium inventory is detectable within the upper 1000 m of water west of the atolls when compared to inventories in this layer east of the atolls.

At Stations 2 and 4 the average inventory of $^{239+240}Pu$ between 1000- and 4000-m depths is 1.27 mCi/km^2. At Stations 9, 11, and 13 the plutonium inventory within this depth interval is 1.77 ± 0.26, 3.00 ± 0.38, and 1.79 ± 0.31 mCi/km^2, respectively. These amounts are statistically different from those in the deep water east of the atolls. They show that plutonium may be remobilized to the deeper waters west of these Pacific test sites, possibly from the contaminated sediments previously deposited on the outer slopes of the atolls and surrounding basin. Concentrations in sediments from the region of Station 11 greatly exceed expected fallout concentrations. If the mobilization rate of plutonium from sedimentary deposits outside the atolls does not differ from that within the atolls, the bottom waters of the North Equatorial Pacific must move substantially slower than the surface layers to account for the increased inventories at depth from the atoll source term. Station 13 is 212 miles west of Enewetak, and excess plutonium is still evident in the deep water mass. The areal extent of contaminated bottom water and the fate of the plutonium introduced to the deep ocean are unknown. We can, however, safely assume that plutonium in the deep water, originating from the test series at Enewetak and Bikini, is present at least 212 miles from the atolls' source term and moved with the bottom water to this distance during the last 20-30 years.

References

1. I. Aoyana, M. Yamamoto, and Y. Inoue, "Evaluation of the Radioactive Waste Disposal into the Deep Sea", Health Phys., 33, 227 (1977)

2. C. Machida, A. Ito, A. Matsumoto, S. Sakata, T. R. Nagaraja, and H. Abe, "Developments and Studies for Marine Disposal of Radioactive Wastes", Management of Radioactive Wastes from the Nuclear Fuel Cycle, Vol. II, IAEA, Vienna, 333 (1976).

3. D. R. Anderson, W. P. Bishop, V. T. Bowen, J. P. Bennett, W. N. Caudle, R. J. Detry, T. E. Ewart, D. E. Hayes, T. S. Heath, R. R. Hessler, C. D. Hollister, K. Kelt, J. M. McGowen, R. W. Rohde, W. P. Schimmel, C. L. Schuster, A. J. Silva, W. H. Smyrl, B. A. Taft, and D. U. Talbert, "Release Pathways for Deep Sea bed Disposal of Radioactive Wastes", Impacts of Nuclear Releases into the Aquatic Environment, IAEA, Vienna, 483 (1975).

4. S. Smith, Hawaii Institute of Marine Biology (Private communication), 1979.

5. W. S. VonArx, "Circulation Systems of Bikini and Rongelap Lagoons", U.S.G.S. Professional Paper No 260-B. Govt. Printing Office, Washington, D.C. (1954)

6. D. M. Nelson and M. B. Lovett, "Oxidation State of Plutonium in the Irish Sea", Nature, 276, 599 (1978).

7. C. P. Hardy, P. W. Krey, and H. L. Volchok, "Global Inventory and Distribution of Fallout Plutonium", Nature, 241, 444 (1973).

TABLE I

Summary of Mean and Range of $^{239+240}Pu$ Concentrations in Seawater From Locations in the North Equatorial Pacific Ocean

Location & Region Sampled	Month/Year Sampled	No. of Locations Sampled	Mean $^{239+240}Pu$ (pCi/m^3)[a] Soluble	Particulate	Total
Enewetak Atoll					
Lagoon surface samples	11/72	29	--	--	29.0(0.4-96)
Lagoon bottom samples	11/72	6	--	--	44.0(10-75)
Lagoon surface samples	8/74	48	23.6(1.4-65)	18.6(0.8-125)	42.2
Lagoon bottom samples	8/74	23	28.5(3-69)	18.9(0.9-36)	47.4
Ocean reef samples	10/75	9	114(3-644)	N.A.[b]	
Lagoon reef samples off N. Yvonne Island	10/75	9	94.0(72-120)	N.A.	
Lagoon surface samples 2 km offshore the inner Atoll perimeter	5/76	19	17.0(2.1-31)	12.6(1.6-67)	29.6
Lagoon bottom samples 2 km offshore the inner Atoll perimeter	5/76	10	14.0(0.7-26)	15.0(2.6-43)	29.0
Ocean reef samples	5/76	6	26.0(10-70)	58.0(6-167)	
Ocean reef samples	10/76	3	55.0(28-94)	220(71-774)	
Lagoon reef samples	11/78	5	17.0(3-46)	N.A.	
Bikini Atoll					
Lagoon surface samples	11/72	10	40.4(3.9-79)	12(0.1-42)	
Lagoon bottom samples	11/72	7	40.0(8.6-64)	71(5-460)	
Lagoon surface samples	2/77	18	52.0(27-84)	N.A.	
Lagoon bottom samples	2/77	8	44.0(13-104)	N.A.	
Lagoon reef samples	11/78	8	29.0(7-60)	N.A.	
Kwajalein Atoll					
Lagoon surface samples	5/75-10/76	10			0.46 ± 0.15
Lagoon bottom samples	5/75-10/76	3			0.59 ± 0.27
Wotho Atoll					
Lagoon reef samples	12/78	3			0.35 ± 0.16
Rongerik Atoll					
Lagoon reef samples	11/78	4			0.32 ± 0.20

TABLE I (Continued)

Location & Region Sampled	Month/Year Sampled	No. of Locations Sampled	Mean $^{239+240}$Pu (pCi/m^3)[a] Soluble	Particulate	Total
Bikar Atoll					
Lagoon reef samples	11/78	2			.32 ± 0.04
Equatorial Pacific					
1-5 miles W & S of Bikini	10/72	4			15 ± 6
26-90 miles W of Bikini	7/78	3			2.0 ± 0.8
1 mile S of wide pass Enewetak	11/72	5			4.8 ± 3.0
1 mile S of wide pass Enewetak	4/76	3			5.3 ± 3.0
2 miles W of Enewetak	10/76	9			1.7 ± 0.4
North equatorial Pacific	10/72-7/78	14			0.38 ± 0.12

a--Values in parenthesis represent the range in concentrations encountered at locations sampled.
b--Not analyzed.

TABLE II

Inventories of $^{239+240}Pu$ (mCi/km^2) in filtrates from water columns at stations in the North Equatorial Pacific.[a]

tation umber	Location	$^{239+240}Pu$ (mCi/km^2) for Various Depths (m)					
		0-1000	0-2000	0-3000	0-4000	0-5000	Bottom
2	270 mi E of Bikini 11° 'N 170°00 'E	1.19 ± 0.07	2.06 ± 0.14	2.38 ± 0.1[illegible]	2.[illegible] ± 0.1[illegible]		0-4300 2.62 ± 0.21
4	300 mi NE of Bikini 13°40 'N 170° 'E	1.20 ± 0.06	1.89 ± 0.11	2.18 ± 0.1[illegible]	2.[illegible] ± 0.23	2.43 ± 0.[illegible]	0-5431 2.48 ± 0.[illegible]
9	95 mi W of Bikini 11°40 'N 163°36 'E	1.23 ± 0.07	2.24 ± 0.[illegible]	2.[illegible] ± 0.20	3.[illegible] ± 0.2[illegible]	3.28 ± 0.[illegible]	0-5204 3.28 ± 0.[illegible]
11	15 mi W of Enewetak 11°37 'N 161°48 'E	1.35 ± 0.09	2.31 ± 0.18	3.[illegible] ± 0.2[illegible]			0-3800 4.44 ± 0.[illegible]
13	212 mi NW of Enewetak 13°30 'N 154°00 'E	1.41 ± 0.1[illegible]	2.[illegible] ± 0.20	2.[illegible] ± 0.1[illegible]	3.[illegible] ± 0.2[illegible]	3.[illegible] ± 0.[illegible]	0-5[illegible] 4.[illegible] ± 0.[illegible]

[a] These inventories are calculated from the activity [illegible] Most [illegible] activity usually found [illegible] inventories.

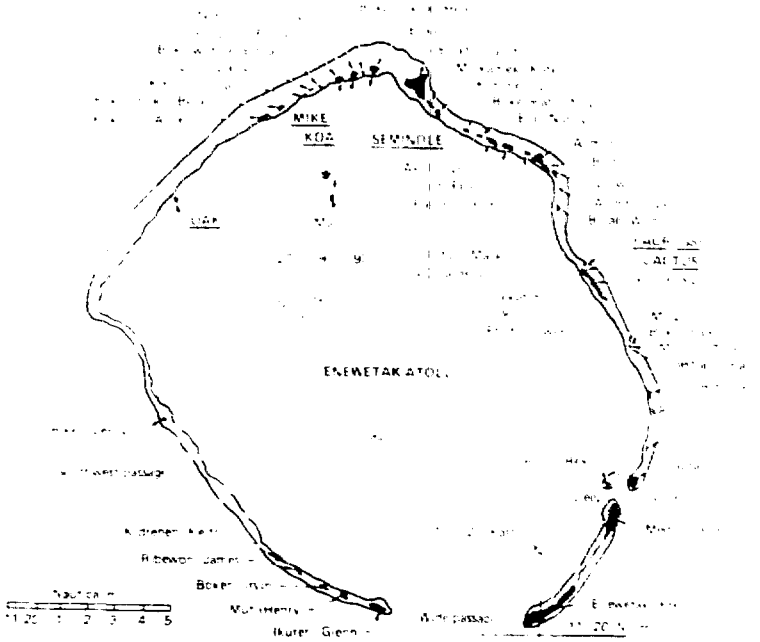

Fig. 1. Enewetak Atoll with names and l[illegible] nuclear craters.