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VICTOR E. NOSHKIN

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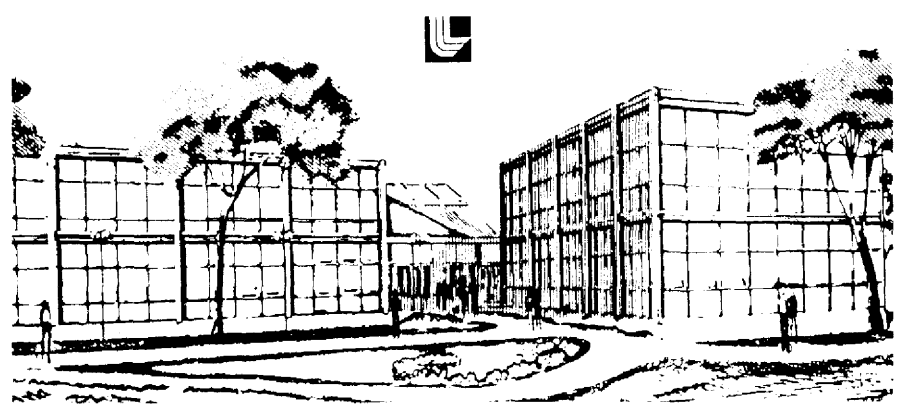

TRANSURANIUM RADIONUCLIDES IN COMPONENTS OF THE BENTHIC ENVIRONMENT OF ENEWETAK ATOLL

V.E. Noshkin

Lawrence Livermore Laboratory

University of California

Environmental Sciences Division

Livermore, California 94550

ABSTRACT

Data on the concentrations and distributions of transuranium radionuclides in the marine environment of Enewetak Atoll are reviewed. The distributions of the transuranics in the lagoon are very heterogeneous. The quantities of transuranics generated during the nuclear test years at the Atoll and now associated with various sediment components are discussed. Whenever possible, concentrations of ^{241}Am and $^{239+240}Pu$ are compared. The lagoon is the largest reservoir of transuranics at the Atoll and radionuclides are remobilized continuously to the hydrosphere from the solid source terms and are cycled with components of the biosphere. Although $^{239+240}Pu$ is associated with filterable material in the water column, the amount that is relocated and redeposited to different areas in the lagoon is small. Barring

catastrophic events, little alteration in the present distribution of transuranics in the sediment is anticipated during the next few decades. The Atoll seems to have reached a chemical steady state in the partitioning of $^{239+240}Pu$ between soluble and insoluble phases of the environment. Using an experimentally determined K_d for $^{239+240}Pu$, the amount of dissolved radionuclides predicted to be in equilibrium with concentrations in the sediment agrees well with recently measured average concentrations in the water at both Enewetak and Bikini Atolls. The remobilized $^{239+240}Pu$ has solute-like characteristics. It passes readily and rapidly through dialysis membranes and can be traced as a solute for considerable distances in the water. We estimate that 50% of the present inventory of $^{239+240}Pu$ in sediment will be remobilized in solution and discharged to the North Equatorial Pacific over the next 250 yr.

INTRODUCTION

Large inventories of several transuranium radionuclides (Ne73) persist in the marine environment of Enewetak Atoll. Forty-three nuclear weapons tests were conducted by the United States at Enewetak between 1948 and 1958. The testing produced close-in fallout debris that was contaminated with transuranics and that entered the aquatic environment of the Atoll. More transuranics were transported westward to Enewetak in airborne debris and water contaminated from nuclear testing at Bikini Atoll. Global fallout deposited a small additional amount of transuranics to the Atoll. Presently the largest inventory of

transuranics introduced from these source terms is associated with components of the benthic environment.

Because of the high level of deposition the Atoll is now its own transuranic source term. Plutonium, for example, is not permanently fixed with the carbonates and other material with which it was originally deposited in the lagoon and reef during nuclear testing. Small amounts of plutonium are now remobilized, resuspended, assimilated, and transferred continuously within the Atoll environment by physical, chemical, and biological processes.

More than half of the United States nuclear tests in the Pacific were conducted at Enewetak Atoll. Surface and tower shots left craters and contaminated scrap on land and generated radioactive debris that was redistributed to the adjacent reef and lagoon. Megaton tests that left underwater craters and barge shots in the lagoon contributed significantly to the present transuranic inventory.

The impact of nuclear testing and the fate of the residual radioactive materials introduced to the aquatic environment at both Enewetak and Bikini Atolls are the subjects of reports too numerous to list herein. However, not until late 1972 when a radiological resurvey of Enewetak Atoll was conducted to gather data for the development of cleanup and rehabilitation procedures for the resettlement of the Enewetak people to their homeland did extensive measurements of transuranics in the Atoll environment begin. The information was published in the survey report (Ne73), which contains data on most long-lived residual radionuclides, including plutonium and americium, in

components of the marine environment. The survey was followed by other more extensive investigations, concentrating on measurement of transuranics to better assess the impact of these radionuclides on the environment and inhabitants of the Atoll and to increase our understanding of the mobilization, reconcentration, and redistribution processes from sources within the environment.

This paper contains a summary of data related to the concentrations of the transuranium elements in components of the benthic and pelagic environment of the Atoll lagoon. Data from the survey report (Ne73), more recent publications, and unpublished results from this laboratory will be discussed. Some published and unpublished data from our studies at Bikini Atoll will be presented when necessary for comparison with Enewetak data and in the absence of Enewetak data, for the clarification of characteristics of transuranic radionuclide concentrations at the Atolls. Whenever possible, the Atoll data will be compared with that from other marine ecosystems.

GEOGRAPHY AND ATOLL TEST HISTORY

Enewetak Atoll, with U.S.-assigned and native names and several landmarks including the locations of craters formed by nuclear tests, is shown in Fig. 1. U.S.-assigned island names will be used throughout this report.

The Atoll consisted of a ring of 42 low islands arranged on a roughly elliptical reef, 40.2 x 32.2 km (Em54), with the elongated axis in the northwesterly direction. Nuclear testing completely destroyed

the islands of Gene and Flora, and only a sand bar now remains to distinguish the island of Helen. Only 39 of the original 42 islands of the Atoll remain and make up a total land area of approximately 6.9 km^2 that is situated on the reef whose area is 84 km^2. The average depth of the lagoon is 47.4 m; the maximum depth is 60 m. The lagoon area is 933 km^2. The sedimentary components in Enewetak Lagoon were studied extensively during the late 1940s (Em54). The main components in the lagoon sediments included foraminifera, coral, Halimeda remains, shells of mollusks, and fine material. Material finer than 0.5 mm in diameter was too fine to identify and was classified as fine debris. Distributions and average abundance of the sedimentary components were described (Em 54). Fine debris made up 57% of the lagoon sediments and was abundant throughout the lagoon to within a few hundred feet from the shore.

A detailed description of the forms, living habits, populations, and specific relationships of the aquatic biological components at the Atoll is beyond the scope of this report. A significant number of articles that were published between 1955 and 1974 and resulted from research conducted at the Enewetak Marine Biological Laboratory were compiled recently in a three-volume report (En76). The individual reports dealing with specific ecological studies at the Atoll, are too numerous to list. The reader is referred to the compilation (En76) for descriptions of the biology and ecology of the Atoll.

The most severe radiological impact on the aquatic environment of Enewetak occurred during the nuclear test years between 1948 and 1958.

The types of nuclear events, shot frequencies, geographical locations, yields, generated particles, conditions after the tests, and other factors determined the resulting distributions of transuranics and influenced the physical and chemical forms of the elements deposited in the benthic environment. A brief historical review of testing at Enewetak, abstracted from several unclassified documents (Ne73, C164, Hi62), explains a few conditions responsible for the transuranic distributions and inventories at the Atoll.

The test series at Enewetak began in 1948 (Operation Sandstone) when 37-, 49-, and 18-kt devices were detonated from 200-ft towers on the islands of Janet, Sally, and Yvonne between 14 April and 14 May. In 1951, testing was resumed (Operation Greenhouse) and four tower shots were conducted during a 47-da interval. The island of Janet was again the location of two ground zeros. In 1952, the first thermonuclear device (Mike) destroyed the island of Flora on the northwest reef. The Mike event was a 10.4-Mt surface detonation occurring on 31 October. Water surging from the point of the explosion sent a wave over adjacent islands including Janet, the site of three previous ground zeros. The original crater where Flora had once been had an irregular outline and was more than 1 mi in diameter, which, before it was partially refilled by the returning rush of coral sediment, was almost 200 ft deep. The crater is presently 90 ft deep. The 1952 series of tests concluded with the King event, a high-yield air drop over Yvonne Island. In 1954 a single device, Nectar, was detonated on a barge located over Mike Crater. Not only did this test greatly disturb the radionuclides

already deposited in the crater sediments, but it also again sent a surge of contaminated water over adjacent islands including Janet. In 1956, the Redwing series began with a tower detonation on Yvonne and included two additional cratering events, LaCross and Seminole. LaCross was a 39.5-kt device detonated on an earth-filled causeway built on the reef off the north end of Yvonne. Seminole, detonated on the island of Irene, was first placed in a 15-ft diameter tank that was itself then placed in a 50-ft-diameter tank filled with water before it was fired. During 1958, the final year of testing at Enewetak, 22 tests of various types were held at different Atoll locations during an 82-da period. The series opened with a 86000-ft ballon shot over the Atoll on 28 April. On 5 May, a 18-kt device produced Cactus Crater on the northwest end of Yvonne and west of LaCross Crater. During May 11 to 12, one of the three tests was the Koa event, a 1.37-Mt nuclear device housed in a tank of water and detonated on the east end of the Gene-Helen island complex. A sizable crater was produced, connected with Mike Crater. On June 8, the Umbrella device was detonated on the floor of the lagoon. Twenty days later, the 8.9-Mt Oak device was fired on a barge 4 mi southwest of Alice off the edge of the reef. The test left a crater that breached to the lagoon. The Quince event on Yvonne Island failed to produce a fission yield so that the plutonium within the device was dispersed by high explosive. Subsequently another nuclear device was successfully detonated over the same area that undoubtedly further dispersed the nonnuclear-generated plutonium. In addition to the nuclear tests, radionuclides were dispersed by plowing on many of the

islands during the test years. Unfortunately, none of the radiological safety reports during these operations provided details to determine the eventual fate of the radioactive debris, e.g., location and quantity of the disposal (Ne73).

From this brief summary, we can assume safely that the transuranic elements were introduced to the aquatic environment not only as complicated carbonate particles fused or condensed with other material from the environment or with devices and associated structures, but also as soluble and particulate species of transuranium oxide.

Despite the complexities in the formation processes, much of the behavior of the transuranics is similar to those determined from investigations of fallout and other aquatic pathways. The results from the Atoll studies therefore have great value in predicting transuranic behavior and fate on a global aquatic scale.

TRANSURANICS IDENTIFIED AT THE PACIFIC TEST SITE ATOLLS SINCE 1972

Neptunium

Concentrations of ^{237}Np in several 1972 samples of unfiltered lagoon and crater water from Enewetak were determined by mass spectrometry (No74). The average concentration in six samples from the lagoon was 0.058 ± 0.013 fCi/l. Water samples from Mike and Koa craters averaged 0.45 ± 0.22 fCi/l. Outside of the lagoon and to the east of the Atoll, concentrations in water samples from the open ocean surface averaged 0.013 ± 0.003 fCi/l. This comparison shows, as do results for all other transuranics, that Atoll sources contribute the major fraction

of the transuranic inventory in the water column of the lagoon. The ^{237}Np concentrations in the lagoon and crater water samples were less than 0.2% of the measured $^{239+240}Pu$ concentrations in those samples.

Plutonium

Many types of samples from the Atoll contain ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu. Most reported values are the sum of ^{239}Pu and ^{240}Pu activities determined by alpha spectrometry. These radionuclides are distributed widely throughout the Atoll and were detected in nearly every type of marine and terrestrial sample analyzed to date. Atoll water samples, sedimentary components (including fine unidentifiable carbonate sands, coral fragments, Halimeda debris, foraminifera, and mollusk shells), living algae, benthic invertebrate tissues, planktonic species, and marine vertebrate tissue all contain $^{239+240}Pu$.

The distribution of ^{238}Pu is as wide among components in the marine environment as is $^{239+240}Pu$, but at lower concentrations. The ^{238}Pu:$^{239+240}Pu$ ratio determined in a variety of aquatic samples from different regions of the lagoon ranges from less than 0.04 to greater than 0.50.

A few activity ratios of ^{240}Pu:^{239}Pu was determined by mass spectrometry. The ratios in two water samples collected from the lagoon during 1972 were 0.432 and 0.289 (No74). Samples of mackeral bone and viscera collected in 1972 near the island of Glenn had ^{240}Pu:^{239}Pu activity ratios of 1.15 $\pm$ 0.25 and 1.27 $\pm$ 0.26, respectively, while goatfish viscera and tridacna tissue from nearby David had ratios of 0.68 + 0.07 and 0.66 $\pm$ 0.19 (Ga 75), respectively. The activity ratios

in 56 soil samples from 7 islands ranged from 0.066 to 1.42 and averaged 0.84 $\pm$ 0.37 (Ga75), while the average ratio in 7 marine water and biota samples was 0.66 $\pm$ 0.40. Neither average value determined in the environmental samples differed greatly from the average of 0.65 $\pm$ 0.05 for global fallout debris (Kr 76). The similar isotopic ratio in mackeral tissue shows no obvious discrimination in uptake of isotopes by tissues of organisms in the Atoll if feeding and living are restricted to specific regions of the Atoll.

The average ^{240}Pu:^{239}Pu ratio in the yearly growth sections of a live sample of Favites virens coral collected from the western basin in Bikini Lagoon was 0.77 $\pm$ 0.07 (No 75). This value is similar to the isotopic ratio in Enewetak samples. However, the mean isotopic concentration ratio in soil and vegetation of Bikini and Eneu Islands is 1.15 (Mo76), somewhat higher than the average in the Bikini coral sample.

Since the ^{240}Pu:^{239}Pu activity ratio in some environmental samples exceeds 1, it seems inappropriate to use the shorthand notation, ^{239}Pu, when referring to the sum of ^{239}Pu and ^{240}Pu activities as has so often been done in the literature. Throughout this report, $^{239+240}Pu$ will refer to the sum of the activities of the two radionuclides, while ^{239}Pu will refer to only that isotope.

In two Enewetak lagoon water samples collected during 1972 (No 74), ^{241}Pu was measured by mass spectrometry. The ^{241}Pu:$^{239+240}Pu$ activity ratios as of December 1972 were 1.14 and 2.56. In the 1972 growth section of the previously mentioned live coral from Bikini, the

$^{241}Pu:^{239+240}Pu$ activity ratio was 11.7 and the $^{241}Pu:^{239}Pu$ ratio was 21.0 ± 1.1 (No 75). Recent values of the $^{241}Pu:^{239}Pu$ ratio in soil samples from Bikini and Eneu Islands and in Bikini island vegetation averaged 22.0 ± 3.3 (Mo 76) as of 1 January 1975. Correcting the ^{241}Pu in the November 1972 coral growth section for decay to 1 January 1975 yields a $^{241}Pu:^{239}Pu$ ratio of 18.9 ± 1.1. Bikini and Eneu Islands and the sedimentary environment from which the coral was obtained were contaminated principally with radioactive debris from the 1954 Bravo event. The good agreement between the ratios determined in the terrestrial and marine samples indicates a lack of discrimination between ^{241}Pu and ^{239}Pu isotopes in processes in these environments. Bikini and Enewetak have very different isotopic ratios and therefore different inventories of plutonium isotopes. The amount of ^{241}Pu in the environment regulates the projected inventory of ^{241}Am through growth and beta decay of the parent radionuclide. The amount of ^{241}Am that will be generated at Bikini from ^{241}Pu decay will exceed the amount produced by this source at Enewetak.

Americium

The distribution of ^{241}Am is also widespread in the aquatic environment of the Atoll. Although the highest concentrations of plutonium and americium are in the same areas of the lagoon at Enewetak, the two transuranics are distributed differently. The $^{241}Am:^{239+240}Pu$ ratio in sediments collected during 1972 from the lagoon ranged from 0.06 to 0.93. Significant errors therefore can be introduced if one transuranic is used to predict the levels of others at any given

location in the lagoon. No other americium isotopes were detected in the aquatic environment of either Enewetak or Bikini.

Curium

No strenuous effort has been made to obtain an inventory of ^{242}Cm or ^{244}Cm in Enewetak by alpha spectrometry. Curium activities were separated and measured in several lagoon water samples; ^{242}Cm activities were less than 0.2 fCi/l. No ^{242}Cm or ^{244}Cm was detected in sediment samples from the Bravo Crater at Bikini Atoll (Be76).

Higher Transuranics

No information is available to this authors knowledge on either Berkelium or Californium in marine samples from Enewetak or Bikini.

TRANSURANICS IN THE BENTHIC ENVIRONMENT OF ENEWETAK ATOLL

Surface Sediment Distributions and Inventories

The distributions of $^{239+240}Pu$ and ^{241}Am activities measured in December 1972 and expressed as mCi/km^2 in the 2.5-cm-thick surface layer of sediment from the lagoon floor are shown in Figs. 2 and 3. Isolines were constructed to distinguish regions of the lagoon having similar concentrations. The mean transuranic inventory in the surface layer and the range of concentrations within the defined areas are shown in the two figures. Figs. 4 and 5 show regions of the surface sediment layer with similar $^{238}Pu:^{239+240}Pu$ and $^{241}Am:^{239+240}Pu$ ratios.

The transuranic concentrations in the surface layer sediments were determined in over 150 ball-milled surface samples of known thickness and in 20 core samples obtained throughout the lagoon. The lagoon was

divided into a grid consisting of a series of 6-km^2 regions; at least one sediment sample was obtained from each region to provide radiological data for areal distributions. All sediments are composed of different quantities of fine and coarse-grained carbonate material, shells, coral fragments, and Halimeda debris. To assess the sediment inventory, no attempt was made to distinguish concentration levels in specific sedimentary components. Figs. 2 through 5 illustrate the main features of the transuranic distributions in the surface layer of the lagoon sediment. Isolated regions of relatively high concentrations of $^{239+240}Pu$ are evident in some lesser contaminated areas of the lagoon; other small regions of high surface radioactivity might have escaped detection. The areal distributions are based on available data from the samples that were collected and analyzed.

The transuranics are distributed nonuniformly over the lagoon floor. Highest surface concentrations are associated with the sediments near, but not necessarily adjacent to, the locations of larger or more numerous nuclear tests. Highest plutonium concentrations are associated with the sediments from the northwest quadrant in a north- and south-oriented elliptical area, roughly 2 to 3 km east of the islands of Alice and Belle and several km southwest of Mike and Koa craters. A second region of relatively high concentration is in sediments off the shore of Yvonne Island. The activity in this region is lower than the concentrations in sediments in the northwest. Most of the transuranic inventory in the surface sediments can be separated roughly from the lesser contaminated deposits by a line extending from the Southwest

Passage to the island of Tom (Munjor), which is south of Yvonne on the eastern reef. The surface $^{239+240}Pu$ concentrations north of this line range between 2 and 170 pCi/g dry weight, while south of this line the surface concentrations were less than 2 pCi/g. All surface sediment samples obtained during and since 1972 contained $^{239+240}Pu$. The inventory in only the top 2.5-cm layer (mCi/km^2) of sediment exceeds the activity deposited to the earth's surface as worldwide fallout in any latitude band in the northern or southern hemisphere (Ha73).

Although the surface distribution of ^{241}Am in the sediments appears similar to that of $^{239+240}Pu$, the ratio of ^{241}Am:$^{239+240}Pu$ activities (Fig. 5) shows that the radionuclides are not well mixed throughout the surface deposits. The ratio in the sediments ranges from 0.06 to 0.93. The mean ratio, however, determined by averaging ^{241}Am:$^{239+240}Pu$ activities from all surface sediment samples, is 0.29 ± 0.17, and the ratio determined from the mean surface concentrations (Table 1) is 0.30 ± 0.06. The average ratio is similar to that found in central Pacific and northeast Atlantic sediments (Li76), which receive only worldwide fallout deposition but have, in contrast, one-half the average concentration ratio of surface sediments at Bikini (Ne75).

The distribution ratio of ^{238}Pu:$^{239+240}Pu$ in the surface sediments, shown in Fig. 4, demonstrates the nonuniformity among plutonium isotopes in components of the sediment in the Atoll environment. There are, however, large geographical regions of the lagoon with similar isotopic ratios in the sediment. On the other hand, small areas of the lagoon, such as a 600-m strip on the lagoon side of Yvonne Island, contain

plutonium with isotopic ratios ranging from 0.05 to 0.38 (Ne73). In Cactus Crater, at the northern end of Yvonne, the isotopic ratio of 0.55 in the sediments is one of the highest at the Atoll. The average concentration ratio in the lagoon sediments determined from the mean surface concentrations is 0.14. The average ratio determined in the lagoon water samples during 1972, 74, and 76 is identical to the sediment ratio. A steady state condition is reached where plutonium isotopes are remobilized to the aqueous phase in proportion to their concentrations in different regions of the sediments and reef environments. One region of the lagoon sediments with lower or higher isotopic ratios, for example, is not the dominate source term supplying plutonium isotopes to the water column.

In 1977 several core samples were obtained from the lagoon basin near stations sampled in 1972. The ^{241}Am concentrations in surface sediment layers sampled in 1972 and 1977 were nearly identical showing that there was little change in the surface concentrations of transuranics at many lagoon locations during those years. Only small quantities of the transuranics were remobilized or reworked to greater depths in the sediment column during these years. Little resuspended material from other areas of the lagoon and with different concentrations of transuranics were transported and deposited to the areas that were resampled.

The largest inventory of transuranics at Enewetak Atoll is associated with the components of the lagoon sediment. The estimated lagoon sediment inventories given in Table 1 were determined from Figs.

2 and 3 by summing the products of the areas in the lagoon by the average inventory of the transuranics present there. There are approximately 250 Ci of $^{239+240}Pu$ and 75 Ci of ^{241}Am unevenly distributed throughout the 2.5-cm-thick surface sediment layer of the lagoon. The total $^{239+240}Pu$ inventory in island soils, sampled to depths of 35 to 150 cm, is estimated from available data (Ne73, No76) at < 25 Ci. Transuranic distributions in surface sediment at Bikini Atoll were constructed and inventories were estimated from published (Ne75, No75) and unpublished data (No78a). Bikini sediment inventories were estimated from substantially less data than were available from Enewetak. Future results from Bikini might change the present estimates of transuranic inventories given in Table 1. Analysis of 25 cores (12 to 21 cm deep) from different locations in Bikini and Enewetak lagoons showed that only 21 $\pm$ 11% of the $^{239+240}Pu$ and 16 $\pm$ 6% of the ^{241}Am (No78a) in the sediment column is associated with components in the top 2.5-cm layer. If the average $^{239+240}Pu$ inventory in the surface sediment is only 21% of the total inventory to a mean depth of 16 cm for the entire lagoon, then the estimated $^{239+240}Pu$ inventory in the sediment column to a 16-cm depth at Enewetak and Bikini are 1.2 and 1.5 kCi, respectively. However, in a few deeper cores that are difficult to obtain from carbonate deposits, $^{239+240}Pu$ and ^{241}Am were detected at depths below 20 cm. The inventories computed to a depth of 16 cm then can be assumed only to represent lower limits. Using the average isotope ratios from samples from the Atoll environment (discussed earlier), an estimate of the concentration for each plutonium isotope

and ^{241}Am in the Atoll sediments is made (see Table 1). Transuranic isotopes deposited from global fallout of weapons debris are estimated from available data (Kr76, Ha73) and are also given in Table 1.

The inventory at the Atolls of transuranics produced by weapons is only a small fraction of the total quantity deposited to the earth surface with global fallout debris. Some specific marine environments were contaminated with substantial quantities of transuranics from other source terms. However, these lagoon sediments are the most contaminated aquatic regions in the world that received transuranic inputs only from nuclear weapons. The estimated ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{241}Am inventories at Bikini exceed the respective isotopic inventories at Enewetak, while ^{238}Pu is higher in Enewetak Lagoon. Inventories of ^{241}Am at Bikini will increase by 25% from ^{241}Pu decay, while only a 10% increase over present ^{241}Am levels is expected at Enewetak from ^{241}Pu decay.

TRANSURANICS ASSOCIATED WITH COMPONENTS IN THE SEDIMENT COLUMN

Using the definition of Emery et al. (Em54) for classifying fines as material less than 0.5 mm in diameter, 23 surface samples and several cores were separated into fine and coarse fractions. The dry weight of the fines ranged from 25 to 80% of the total dry weight of the surface volume (No78a). A similar range of fine material was found in Bikini Atoll sediments. At least 93% of the sediment weight in Mike and Koa crater deposits was fine material. In over 98% of the sediment samples from Enewetak lagoon, the ^{241}Am and $^{239+240}Pu$ concentrations (pCi/g)

associated with the fine sediment components were greater than or equal to the concentrations associated with the coarse fraction. The activity of ^{241}Am and $^{239+240}Pu$ in the fines was 0.6 to more than 10 times that in the coarse fraction. These distributions between size fractions are very unlike those encountered for fallout of $^{239+240}Pu$ in sediments in Buzzards Bay, Mass., where the $^{239+240}Pu$ was not preferentially associated with the fine fractions of sedimentary deposits (Bo76). This difference is perhaps not unexpected, because most of the transuranic inventory deposited to the lagoon environment was probably associated with small particulate carbonates. During the years after nuclear testing, some plutonium has exchanged slowly as a result of chemical reactions with exposed surfaces of the larger sedimentary components.

The transuranic inventory at lagoon locations, however, is dependent on the local abundance of the fine and coarse materials. Table 2 shows the ^{241}Am concentrations in the fine and coarse components of two core samples from mid-lagoon locations at Enewetak. The fraction of the coarse components in the sediment column of core 6 decreases with depth and in core 1 increases with depth. The ^{241}Am concentration associated with the fine fraction in the surface 2-cm section is 2 to 5 times the concentration associated with the coarse fraction; but because the fine material in the surface layer of core 6 accounts for only 25% of the total dry weight of the sediment volume, 58% of the ^{241}Am in the surface 2-cm layer is associated with the coarse fraction. In core 1, on the other hand, 95% of the total ^{241}Am in the surface 2-cm layer is associated with the fine fraction. Although the ^{241}Am concentrations

(pCi/g) associated with the fine material at various depths in the sediment column exceeds the concentrations associated with the coarse components in both cores, the inventory of the radionuclide (pCi/cm^3) within any depth interval associated with the fine and coarse components can be variable throughout the sediment column. Areal transuranic deistributions like those shown in Figs. 2 and 3, but associated with only the fine or only the coarse component of sediment, would differ.

The vertical distributions of the transuranics in the lagoon sediment are very complex. No generalization about the shape of the concentration profile in any region can be made. Table 2, for example, shows a ^{241}Am peak associated with the fine components of core 6 at depths of 25 to 30 cm with little ^{241}Am associated with the coarse components at these depths. In core 1, the highest ^{241}Am concentrations are associated with the fine components between depths of 8 to 10 cm in the sediment column. The ^{241}Am concentrations associated with the coarse component in both cores generally decrease gradually with depth. Transuranic concentrations increase, decrease, or remain constant with depth in sediment cores from other lagoon locations (No78a). The concentrations of $^{239+240}Pu$ and ^{241}Am associated with the carbonate components in four cores taken along a 1.5-km transect across Mike and Koa Craters are shown in Fig. 6. The concentrations in the sediments from the Atolls' largest craters are surprisingly nonhomogenous. Turbulence and large scale mixing of the sediments during and after testing should have produced a much more uniform distribution than that found. The $^{239+240}Pu$ concentration in the sediment column at station

17E is fairly uniform to a depth of 50 cm. At station 16E, the concentration increases with depth to 35 cm. The ^{241}Am concentration in the sediment column at station 16E decreases with depth. No correlation is obvious between the ^{241}Am and $^{239+240}Pu$ concentrations associated with the components of these crater sediments. The craters should act as natural sediment traps but little sedimentation in the Mike and Koa craters has occurred since the bottom depths were redetermined in 1964. In 1964 the maximum bottom depth of Mike Crater was 27.4 m below sea level (Ne73). We have found no measurable change in the depth of the crater bottom during the period 1972-1977. Only small quantities of resuspended or reef-generated particulate material are then transported in the water masses to the western reef. Very little sedimentary material therefore escapes from the lagoon and any resuspended bottom material probably settles out again on the lagoon floor close to its origin. The complex areal and vertical patterns of transuranics detected in this relatively small region of the lagoon where the distributions are expected to be more uniform are but examples of the complex patterns in the lagoon.

Halimeda, shells, coral, and foraminifera fragments were sorted from the coarse fraction of several sediment samples by hand. Table 3 shows the $^{239+240}Pu$ concentrations associated with each component in the surface layer from two locations in Enewetak Lagoon and at various depths in a core from Bikini lagoon. The $^{239+240}Pu$ concentration associated with Halimeda fragments at station 40C only slightly exceeds those in fragments from station 3D. The concentrations associated with

the separated foraminifera and coral fragments from station 40C are, however, at least 2.5 times higher than those associated with their respective components at station 3D. The distribution of $^{239+240}Pu$ is different among components in the sediment from different regions of the lagoon. The fine fraction at these locations contained the highest concentration of $^{239+240}Pu$. To within our analytical precision, the ^{238}Pu:$^{239+240}Pu$ concentration ratio is identical in the components from both stations.

In the sediment column from Bikini station B3, $^{239+240}Pu$ is associated with all components that were separated. At all depths in the sediment column, the highest concentration of $^{239+240}Pu$ in the coarse components was associated with Halimeda fragments. Sedimentation of labelled material to the lagoon occurs at a rate that is too slow to account for the buried activities below a few mm in the sediment column. Although the age of the Halimeda fragments, coral, and other components at depths greater than a few cm must therefore predate the test years, $^{239+240}Pu$ is associated with these components.

The possibility that subsurface remains labelled during testing were buried later in the sediment column by large scale turbulence can be discounted. Coral or Halimeda fragments directly subject to a nuclear explosion probably would not retain their identity. In recent yearly growth increments of a living sample of Favites virens coral from station B3, the $^{239+240}Pu$ concentrations averaged 104 $\pm$ 12 pCi/kg (No75). This value agrees well with the $^{239+240}Pu$ concentrations in dead coral remains in surface layers at station B 3. In no yearly

growth increment from this coral since 1954 was the $^{239+240}Pu$ concentration below 104 $\pm$ 12 pCi/kg. Lower concentrations are associated with coral remains deeper in the sediment column. From the radiological record retained in the skeletal matrix of the Favites virens, coral labelled during 1954 and 1958, for example, should have $^{239+240}Pu$ concentrations of 39×10^3 and 4.5×10^3 pCi/kg, respectively (No75). These concentrations are orders of magnitude larger than those in any subsurface coral remains. This data, therefore, does not support translocation of labelled coral material deeper into the sediment column by physical processes during or after testing. Burrowing organisms could redistribute some fraction of labelled sedimentary components to depths in the sediment column. However, when the $^{239+240}Pu$ activites associated with each component at various depths are compared to the activity in the corresponding component at the surface, the $^{239+240}Pu$ activities differ. For example, between 5 to 10 cm the $^{239+240}Pu$ concentrations associated with the coral, Halimeda, foraminifera, and shells are 1.29, 0.83, 0.43, and 0.34 times the concentrations associated with those components in the surface layer. Burrowing and mixing processes by organisms are not likely to move specific component selectively down through the sediment column.

The data indicate that all plutonium does not remain associated with the sedimentary materail with which it was originally deposited. Small quantities of plutonium are remobilized continuously from the sediments to the lagoon water column by surface exchange mechanisms. Plutonium is also detected in the interstitial water extracted in situ

from sediments (No78b) at higher concentrations than those in the overlying bottom water at Enewetak Atoll. By equilibration, small quantities of $^{239+240}Pu$ from the sediments are exchanged and released. Vertical diffusion moves the radionuclides to the sediment water interface where the plutonium mixes with the lagoon water mass. Remobilized plutonium can then be concentrated by members of the marine food chain. Vertical diffusion can also move the exchanged plutonium in the interstitial fluid deeper into the sediment column. Exchange of plutonium with exposed carbonate surfaces might account for the concentrations associated with material deeper in the sediment column.

TRANSURANICS ASSOCIATED WITH THE CALCAREOUS ALGAE, HALIMEDA

Debris from the calcareous algae, Halimeda, is the second most abundant component of Enewetak Lagoon sediments (Em54) and covers an estimated 26% of the lagoon floor (Em54). Live species were collected by divers and during dredging operations from numerous locations at both Enewetak and Bikini. Because algae were shown previously to concentrate plutonium (No72), the role of this benthic algae is recycling the transuranics at the Atoll should be assessed.

The mean concentration factor for $^{239+240}Pu$ associated with algae species from both Atolls is 6×10^4 and ranges from $1 \times$ to 32×10^4 (No78a). To within the precision of our measurements, the concentration factors for plutonium at the two atolls and of different Halimeda species from both atolls do not differ (No78a). Concentrations of $^{239+240}Pu$ associated with the live algae ranged from 0.4 to 22 pCi/g wet

weight, and the concentrations in the water where the algae were obtained ranged from 10 to 116 fCi/l. Surface sediment concentrations at the stations (No78a, Ne75) were compared to the algae concentrations at these sites. The average ratio of the $^{239+240}Pu$ concentrations associated with the Halimeda species (pCi/g dry weight) to that in the top 2.5-cm sediment layer (pCi/g dry weight) was 0.24 ± 0.13 and the ^{241}Am concentration ratio was 0.32 ± 0.24. Concentrations of $^{239+240}Pu$ and ^{241}Am in the sediment ranged from 9 to 82 pCi/g and from 1.1 to 67 pCi/g, respectively. On an equivalent weight basis the live benthic algae have lower $^{239+240}Pu$ and ^{241}Am levels than sediments in the immediate environment. The average plant to sediment concentration ratio of $^{239+240}Pu$ and ^{241}Am are not statistically different. Thus, there is no discrimination between $^{239+240}Pu$ and ^{241}Am in processes beginning with remobilization of the transuranics from the environment and ending with concentration by the algae.

Table 4 summarizes data on transuranic concentrations in algae, water, and sediment from Cactus Crater at Enewetak. The data show that the ^{238}Pu:$^{239+240}Pu$ ratio in the plants, water, and sediment are identical. In this crater ecosystem marine algae do not discriminate among the plutonium isotopes in the environment. The plant:sediment concentration ratios of $^{239+240}Pu$ and ^{241}Am are nearly identical, which again shows that the processes of environmental release and plant uptake of the two transuranics are similar.

The mean surface sediment inventory of $^{239+240}Pu$ at Enewetak is 249 Ci (Table 1). The lagoon is 933 km^2 in area and the average specific

gravity of the Halimeda and other sediment components is 1.8 g/cm^3 (Em54). Activities of $^{239+240}$Pu associated with the algae are related to the activity in the surface sediment. The mean wet:dry ratio of the Halimeda species is 2.3, and the average wet weight of the plants, without holdfast, is 6.4 ± 3.8 g (No78a). Therefore, the average $^{239+240}$Pu concentration associated with the live Halimeda species at Enewetak is 0.62 pCi/g wet weight. Approximately 4.0 pCi are associated with each plant. If the number of Halimeda plants were known, the mean plutonium inventory associated with the living Halimeda reservoir could be computed. Unfortunately, no estimates of Halimeda biomass at Enewetak are available. During the late 1940s, the mean sedimentation rate of Halimeda at Bikini was estimated at 3.8 mm/yr (Em54). If this sedimentation rate is applicable to Enewetak Atoll, approximatley 1 Ci of $^{239+240}$Pu is deposited annually in the sediments in association with Halimeda detritus. This quantity represents only 0.4% of the surface sediment inventory and yet a smaller fraction of the total inventory in the sediment column. However, if the life span of each plant is 1 yr, for example, a quantity of $^{239+240}$Pu equivalent to half the present sediment inventory, or 125 Ci, could be recycled with the algae in approximately 175 yr. Spies et al. (Sp78) demonstrated that when live Halimeda from Enewetak were cleaned and treated with 1N acetic acid, the acid soluble fraction, or the carbonate material, contained 58% of the total $^{239+240}$Pu, while 42% remained bound to the plant tissues. As the plant decomposes after death, the organic material and associated radioelements are released to the environment leaving the skeletal

carbonate matrix and its associated transuranics in the sedimentary deposits. The transuranics associated with the organic fraction released during decomposition are recycled to the benthic or pelagic environments. Over the long term the algae could play a key role in cycling the transuranics between the sediments and aqueous environment.

PLUTONIUM CONCENTRATIONS IN THE LAGOON SEA WATER

A considerable number of lagoon water samples have been collected for plutonium analysis by this laboratory since 1972 (No73, No74, No76, No78b). Several studies are in progress at the Atoll that require data on concentrations in lagoon water so that the number of samples and the locations sampled are predicated by the current program requirements. Contours of $^{239+240}$Pu concentrations in the water show complex distribution patterns (No74) in various regions of the lagoon. The spatial patterns of surface and bottom $^{239+240}$Pu concentrations in solution and in association with filterable material are very different as are the ^{238}Pu:$^{239+240}$Pu ratios in the water mass. A detailed discussion of the plutonium levels in the pelagic environment of the lagoon is in preparation (No78b). Instead of relating all results from the analysis of lagoon water samples collected since 1972 with hydrological, seasonal, or spatial factors, we will summarize some of the data that is related to remobilization and redistribution of plutonium.

In 1972, 1974, and 1976 a sufficient number of water samples from the lagoon were analyzed for $^{239+240}$Pu to permit an estimate of mean

concentrations in the lagoon. A summary of the mean concentrations is given in Table 5. In 1972 the average $^{239+240}Pu$ concentration in the lagoon was determined for 34 unfiltered surface and bottom samples. A more detailed water sampling program was conducted in 1974. In 1976, a smaller number of water samples were collected around the perimeter of the lagoon 2 km off the shore of the reef. Water samples collected during 1974 and 1976 were filtered through 1-μm filters. In the discussion to follow, the estimated average soluble $^{239+240}Pu$ concentrations shown in Table 5 refers to material passing through a 1-μm filter.

During July 1974, the soluble $^{239+240}Pu$ in the lagoon water ranged in concentration from 2 to 75 fCi/l. The percentage of the total activity associated with the filterable material in the water samples during 1974 and 1976 ranged from 2 to 54% and from 12 to 94%, respectively. The concentrations of plutonium radionuclides in solution above fallout background concentrations in the lagoon water is direct evidence of the remobilization of transuranics from the solid phases of the environment. Dissolved plutonium released from the sediments of Cactus Crater was traced for considerable distances along the reef by a plutonium radionuclide balance, involving the change in the $^{238}Pu:^{239+240}Pu$ ratio in the water, and dyes to trace the crater water (No78c). The dissolved plutonium moves in solution apparently without interacting with the sediments deposits during transport. The dissolved plutonium passes readily through dialysis membranes (No78c). Equilibration between dissolved plutonium in the crater sea water and

low activity sea water contained in dialysis bags is achieved in 3 da (No78c). These characteristics suggest that the plutonium remobilized to the environmental waters has very solute-like characteristics. It is tempting to suggest, considering the environment, that the remobilized chemical species is some form of carbonate complex.

The average concentration of total $^{239+240}Pu$ in the water was essentially the same in 1972 and 1974 but a marked decrease was noted during 1976. In 1976, the average concentration associated with the filterable material in the lagoon doubled over the mean 1974 level and the mean soluble concentration was reduced to half. Forty percent fewer samples were collected in 1976 than in 1974. During the 1974 program, samples were taken at stations throughout the lagoon, whereas the 1976 samplings were restricted to locations only 2 km from the reef. Similar $^{239+240}Pu$ concentrations were found in water samples from the few 1974 locations resampled in 1976, which suggests that any computed mean concentration in the lagoon is contingent on the number and location of samples. The mean $^{238}Pu:^{239+240}Pu$ ratio in the lagoon water samples in 1972, 1974, and 1976 were virtually the same. Differences in sample ratios between soluble and particulate phases were noted sometimes, but the average ratios associated with the two phases from all stations were not significantly different.

During 1974 and 1976, 1.5 and 0.7 Ci of $^{239+240}Pu$, respectively, were found in solution and 0.27 and 0.53 Ci were associated with particulate material. These latter quantities represent less than 0.2% of the plutonium inventory in the surface sediment and less than 0.04%

of the inventory estimated to a 16-cm depth in the sediment column (Table 1). The average quantities of soluble plutonium in the water are also small fractions of the sediment inventory. Therefore, in recent years only small fractions of the Atoll plutonium inventory is either remobilized to the solution phase or resuspended to the water column.

During 1976 zooplankton samples contained less than 1% of the $^{239+240}Pu$ activity in the total material filtered from an equivalent volume of water (No78b). The remaining $^{239+240}Pu$ in the particulate material is therefore associated with other forms of suspended matter. Between 1960 and 1963 Johannes investigated the composition of the suspended particles in the lagoon (Jo67). Progressing from the eastern reef toward the lagoon, suspended benthic algae and sediment particles became less abundant with depth of the water as they settle to the bottom, and suspended macroscopic organic aggregates, consisting largely of mucus released by coral, increased progressively in size and number (Jo67). Often calcareous grains resuspended near the reef, microorganisms, copepod fecal pellets, and other undifferentiated material were incorporated with the aggregates. These materials and other particles produced in the pelagic environment are the most important food components for lagoon zooplankton and certain plankton-feeding fish (Ge74). The small quantities of plutonium ingested with this particulate debris is dispersed over the lagoon by these organisms. Herbivorous fish play a role in the generation of particles in the water column (Sm73). These fish are not efficient assimilators; while satisfying their energetic requirements, they

disturb large quantities of material and release large amounts of unassimilated material containing plutonium in their feces. Moriarty (Mo76) estimates that a 200-g mullet, a species common to Enewetak, which feeds by scooping up bottom material to sift and remove small algae, will pass 50 g of dry sediment through its gut per day.

Bottom particles from the NW quadrant of the lagoon where highest plutonium concentrations in sediment are found usually have high plutonium concentrations, indicating that a fraction of the plutonium in the particulate phase may originate from turbulent resuspension of the sediment components in deep (60 m) water. This resuspended material and associated plutonium is probably not transported for any distance in the lagoon. Previous results indicate that the material is redeposited in the same general area of its origin. Only a few of the variety of active processes capable of generating and moving particulate plutonium in the water mass have been considered. It is remarkable that these and other processes resuspend so little of the plutonium inventory. Barring catastrophic events, the present distribution and inventory of plutonium in the sediments will be only slightly altered during the years by relocation of labelled material from other regions in the lagoon.

Laboratory studies with contaminated sediments and soils from Enewetak show that plutonium is rapidly partitioned between the solid phase and solution, reaching equilibrium after several days with an average distribution coefficient for plutonium of 1.8×10^5. Table 6 shows this and some recent determinations of the distribution coefficient for plutonium in laboratory and field experiments using a

variety of sediments. Considering the difference in the types of environmental samples represented in Table 6, it is striking that the K_d for plutonium differs so little.

Table 1 shows that the mean plutonium inventory associated with the sediment components in the top 2.5 cm at Enewetak is 249 Ci. The lagoon sediment has an average density of 1.8 g/cm^3 (Em54) and occupies an area of 933 km^2. The mean depth of the lagoon is 47.4 m. Using this data and the K_d for plutonium of 1.8×10^5, a simple model can be constructed to predict the average concentration expected in the lagoon water by assuming that the plutonium in solution is in equilibrium with that in the sediments. At any time the amount of plutonium in solution is limited by the saturation of the solution under equilibrium conditions. The rate at which water and the dissolved plutonium is flushed from the lagoon is balenced by input of uncontaminated ocean water, which is rapidly saturated with remobilized plutonium from the Atoll source terms. If plutonium is cycled through an intermediate host such as the Halimeda, the rate at which plutonium is released from decaying plants must be balenced by uptake in the new growth, thereby maintaining a state of equilibrium. Given that equilibrium conditions exist, the mean plutonium inventory in the lagoon water and concentration expected in solution computed from the basic equation relating K_d to water and sediment concentrations are 1.4 Ci and 32 fCi/l, respectively. There is general agreement between the average quantity of $^{239+240}$Pu predicted and measured in solution (see Table 5). In 1976 the computed value differed from the measured mean soluble concentration by a factor of

two. Although this is not a large discrepancy, the average concentration, as was mentioned previously, probably does not represent the real mean for the lagoon at the time sampled. Using the appropriate dimensions for the Bikini Lagoon, sediment data from Table 1, and the K_d for $^{239+240}$Pu, the average inventory in the water column and concentration computed at Bikini are 1.7 Ci and 60 fCi/l, respectively. During December 1972, the mean soluble $^{239+240}$Pu inventory and concentration in the lagoon water were 1.2 Ci and 42 $\pm$ 21 fCi/l, respectively (No74), and in January 1977 the respective values were 1.4 Ci and 49 $\pm$ 21 fCi/l (No78b). These average values also are consistent with the amounts predicted.

For many reasons, it may be argued that some of this agreement is fortuitous. Nevertheless, having found general agreement between computed and twice-measured average concentrations in both lagoons between 1972 and 1977, the general usefulness of this simple equilibrium model in predicting long term average concentrations in lagoon water is demonstrated. From radiological records retained in yearly growth of coral sections (No75, No78a), Bikini and Enewetak Lagoon water along with dissolved species is estimated to be exchanged approximately twice per yr. At this rate of exchange under steady state conditions, slightly more than 250 yr will be required to reduce the plutonium inventory in the sediment by 50%. The rates of the mobilization and migration processes of plutonium away from the Atoll to the equatorial Pacific waters are much faster than the rate of radioactive decay. These figures and results should be considered when the consequences of

disposal methods for transuranic wastes to the oceans are discussed.

Some massive corals collected from the atolls contain well-defined growth bands dating from the collection time to the early 1950s. Each yearly growth concentrates plutonium in proportion to the levels in the environment (No75, No78a). Concentrations of $^{239+240}Pu$ associated with growth increments dated since 1965 in three Enewetak corals from different locations in the lagoon and one Bikini lagoon sample are given in Table 7. The average amount of plutonium concentrated by the coral from 1965 until the year of collection is computed and shown in Table 7.

The average absolute concentrations in the corals are different as expected, and reflect the local environmental concentrations in the region. In only a few growth sections are the $^{239+240}Pu$ concentrations different from the mean by more than a factor of two, and only coral 1 and 2 show this magnitude of variation. Corals 1 and 2 were obtained in the water on the lagoon side of the eastern reef. The patterns of current in this region of the lagoon are variable, and the windward reef community contributes a significant detrital load with associated plutonium to the lagoon. Since growing coral is a point source in the environment, small changes in even the local circulation, to name one of many factors, will greatly alter the plutonium concentration in the vicinity of the coral. It is rather more surprizing that, for the most part, the $^{239+240}Pu$ levels associated with the last 9 yr of growth are nearly constant, which shows that the dissolved $^{239+240}Pu$ levels available to the corals in a specific region have also been similar during the last 9 yr.

These results from coral and other studies demonstrate that Enewetak lagoon has attained a chemical steady state condition with respect to plutonium remobilization from solid components to solution. Not only will the simple equilibrium model explain average concentrations in lagoon water, but it can be used to estimate local concentrations expected in the waters from areas of the Atoll with different levels of contamination. By using appropriate concentration factors for plutonium, the quantities accumulated by marine organisms anywhere in the lagoon can be estimated. The data on biotic concentration can be used to estimate the potential dose to man if part or all of the Atoll were to supply his marine food requirements.

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Table 1. Estimated transuranic sediment inventory: Enewetak and Bikini Atolls, 1 January 1973.

Enewetak Atoll (area, 933 km^2)	$^{239+240}$Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am
Areal activity to 2.5-cm depth (mCi/km^2)	267	38	145	122	493	81
Total radioactivity to 2.5-cm depth (Ci)	249	35	135	114	460	76
Total radioactivity to 16-cm depth (Ci)	1185	167	642	543	2190	475
Bikini Atoll (area, 629 km^2)						
Areal activity to 2.5-cm depth (mCi/km^2)	492	16	229	263	4809	289
Total radioactivity to 2.5-cm depth (Ci)	309	10	144	165	3025	182
Total radioactivity to 16-cm depth (Ci)	1470	76	686	786	14405	1140
Global fallout from weapons testing, Jan. 1971						
Total radioactivity (kCi)	319	22*	192	127	3010	72

*Weapons, 8.6 kCi; fallout debris from SNAP 9A, 13.4 kCi.

Table 2. ^{241}Am associated with components in core samples of sediment.

Core 6	Concentration (pCi/g dry weight)		Inventory (pCi/cm^3)		Radioactivity associated with coarse component	Relative amount of coarse fraction
Depth (cm)	Fine fraction (% σ)*	Coarse fraction (% σ)	Fine fraction	Coarse fraction	(%)	(% dry weight)
0-2	5.97(23)	2.84(28)	0.57	0.78	57.8	74.6
2-4	4.99(7)	1.81(11)	1.54	0.99	39.1	64.0
4-6	6.44(5)	2.08(12)	2.68	0.95	26.1	52.4
6-8	5.51(6)	1.40(11)	2.88	0.63	17.9	46.4
8-10	3.10(6)	0.96(19)	1.74	0.39	18.3	41.6
10-15	0.72(18)	0.20(40)	0.38	0.09	19.1	45.9
15-20	<0.09	<0.09	<0.05	<0.05	-	48.6
20-25	5.83(7)	<0.09	3.75	<0.03	8.0	37.0
25-30	11.1(7)	0.16(42)	6.91	0.06	0.9	39.0
30-35	0.06	<0.08	<0.02	<0.02	-	48.3
35-40	0.06	<0.04	<0.04	<0.03	-	36.6
Core 1						
0-2	42.5(3)	12.9(16)	43.4	2.2	4.8	14.6
2-4	30.4(6)	6.31(5)	24.7	3.7	13.0	41.8
4-6	34.8(5)	4.90(13)	26.1	2.3	8.1	38.0
6-8	41.1(3)	4.91(20)	33.7	2.4	6.6	37.2
8-10	51.0(3)	4.07(14)	37.6	2.4	6.0	44.4
10-15	25.2(3)	0.98(17)	10.5	0.7	6.3	62.8
15-19	lost	0.36(17)		0.2	-	93.0
19-25	3.55(18)	0.23(34)	0.2	0.01	4.7	90.7

*The one sigma counting error expressed as the percent of value listed.

Table 3. Plutonium concentrations associated with sediment components (pCi/g dry weight).

Enewetak Atoll	Sta. 3D			Sta. 40C		
Surface 2.5 cm layer	$^{239+240}Pu$	^{238}Pu: $^{239+240}Pu$		$^{239+240}Pu$	^{238}Pu: $^{239+240}Pu$	
Mollusk shells	0.64±0.06	0.09±0.03		absent in sample		
Dead Halimeda fragments	4.8 ±0.5	0.08±0.01		6.0±0.4	0.08±0.01	
Coral fragments	1.3 ±0.1	0.08±0.03		10±5	0.13±0.07	
Fines (< 0.5mm)	6.8±0.05	0.07±0.01		23.5±0.2	0.10±0.01	
Foraminifera	1.1 ±0.1	0.07±0.03		2.7±0.2	0.10±0.02	

Bikini Core Station B-3

Depth in sediment column (cm)	Halimeda fragments	Foraminifera	Mollusk shells	Coral fragments
0 - 5	1.15±0.06	0.324±.013	0.265±.013	0.101±.004
5 - 10	0.95±0.03	0.154±.008	0.090±.004	0.130±.004
10 - 15	0.62±0.01	0.093±.004	0.058±.003	0.063±.004
15 - 20	0.33±0.01	0.024±.004	0.032±.002	0.073±.004
20 - 25	0.177±.003	0.012±.002	0.018±.002	0.007±.002
25 - 30	0.013±.001	0.002±.001	0.006±.001	0.001±.000
30 - 35	0.009±.001	0.001±.000	0.003±.001	0.008±.003

Table 4. Concentrations of tranuranium radionuclides in Halimeda Algae and Cactus Crater.

Radionuclides	Halimeda monile*	Crater sediment	Crater Water
^{241}Am (pCi/g dry wt)	2.53 ± 0.30	9.2 ± 2.0	
$^{239+240}Pu$ (pCi/g dry wt)	18.68 ± 0.033	82 ± 2	116 ± 62 fCi/l†
^{238}Pu: $^{239+240}Pu$	0.54 ± 0.03	0.54 ± 0.02	0.53 ± 0.02
^{241}Am: $^{239+240}Pu$	0.14 ± 0.02	0.11 ± 0.02	

^{241}Am (Halimeda: sediment)‡ 0.28
$^{239+240}Pu$ (Halimeda: sediment)‡ 0.23
Concentration factor for $^{239+240}Pu$ = 7.5×10^4 (pCi/kg wet wt per pCi/kg H_2O.)

*Wet wt: dry wt = 2.13.

†Average concentration in 8 water samples from the crater bottom collected between 1974 and 1977.

‡pCi/g dry wt Halimeda ÷ pCi/g dry wt sediment.

Table 5. Plutonium concentrations in the water column at Enewetak Atoll.

	December 1972	Sampling time July 1974	April 1976
Mean concentrations in lagoon water (fCi/l)			
Soluble (< 1 um)		35	16
Particulate (> 1 um)		6	12
Total	39	41	28
Mean ^{238}Pu: $^{239+240}Pu$	0.12	0.13	0.13
Water column inventory (mCi/km^2)			
Soluble		1.65	0.76
Particulate		0.29	0.57
Total	1.84	1.94	1.33
Lagoon Inventory (Ci)			
Soluble		1.54	0.71
Particulate		0.27	0.53
Total	1.70	1.81	1.24
Water Inventory - % of Sediment Inventory			
Water Inventory compared to top 2.5 cm of sediment surface (%) inventory			
Soluble		0.62	0.29
Particulate		0.11	0.21
Total	0.68	0.73	0.50
Water Inventory Compared to top 16 cm of sediment (%) inventory			
Soluble		0.13	0.056
Particulate		0.023	0.045
Total	0.14	0.15	0.10

Table 6. Recent determinations of the distribution coefficient for plutonium.

Sediment type	K_d range	K_d average*	Reference
Enewetak, coral soil and sediment (lab desorption study)	$4x10^4$-$3x10^5$	$1.8x10^5$	Unpublished
Enewetak groundwater particulates	$1.4x10^4$-10^6	$2.5x10^5$	No 76
Trombay Harbour, suspended silt	$4.8x10^4$-$1.3x10^5$	$0.9x10^5$	Pi 76
Bikini Tewa Crater sediment (Lab desorption study, oxic-anoxic conditions)	$4x10^4$-$4x10^5$	$2.2x10^5$	Mo76
Windscale area 5%, 50% silt, 45% sand.	$0.6x10^4$-$22x10^4$	$0.5x10^5$	He75
Humboldt Bay, Calif., suspended clay-silt particulates	$4.7x10^4$-$11.8x10^4$	$0.8x10^5$	Unpublished data, this laboratory
Lake Michigan, suspended particulates		$3.0x10^5$	Wa 76
Mediterranean sediment (Lab sorption study)	$1.3x10^4$-$9.4x10^4$	$0.5x10^5$	Du 74
	Mean	$1.4x10^5$ cm^3/g	

*Quantity of $^{239+240}Pu$ bound to the sediment per unit dry weight of sediment divided by the amount of $^{239+240}Pu$ in water per cm^3.

Table 7. Concentrations of $^{239+240}Pu$ in yearly growth sections of Enewetak and Bikini coral (fCi/g dry weight)

Year of growth section	Coral #1 Enewetak	Coral #2 Enewetak	Coral #3 Enewetak	Coral #4 Bikini
1974			7.4(13)	
1973	3.9(12)*	19.6(5)	5.2(14)	
1972	2.3(9)	35.5(4)	6.7(13)	130(7)
1971	0.9(25)	13.5(6)	7.0(10)	130(7)
1970	1.3(14)	5.0(12)	6.3(12)	100(5)
1969	4.9(10)	41.7(4)	5.9(11)	100(4)
1968	7.9(8)	10.4(6)	6.9(10)	100(2)
1967	3.6(6)	12.9(7)	lost	100(5)
1966	5.1(11)	11.2(7)	3.7(13)	90(5)
1965	3.2(9)	10.5(6)	6.0(8)	110(5)
Average concentration (1965 to year of collection)	3.7±2.1	17.8±12.5	6.1±1.1	108±15
Date of collection	Oct. 73	April 74	Aug. 74	Nov. 72
Species	Favia pallida	Conrastrea retiformis	Favia pallida	Favites viren

*One sigma counting error expressed as the percent of the value listed.

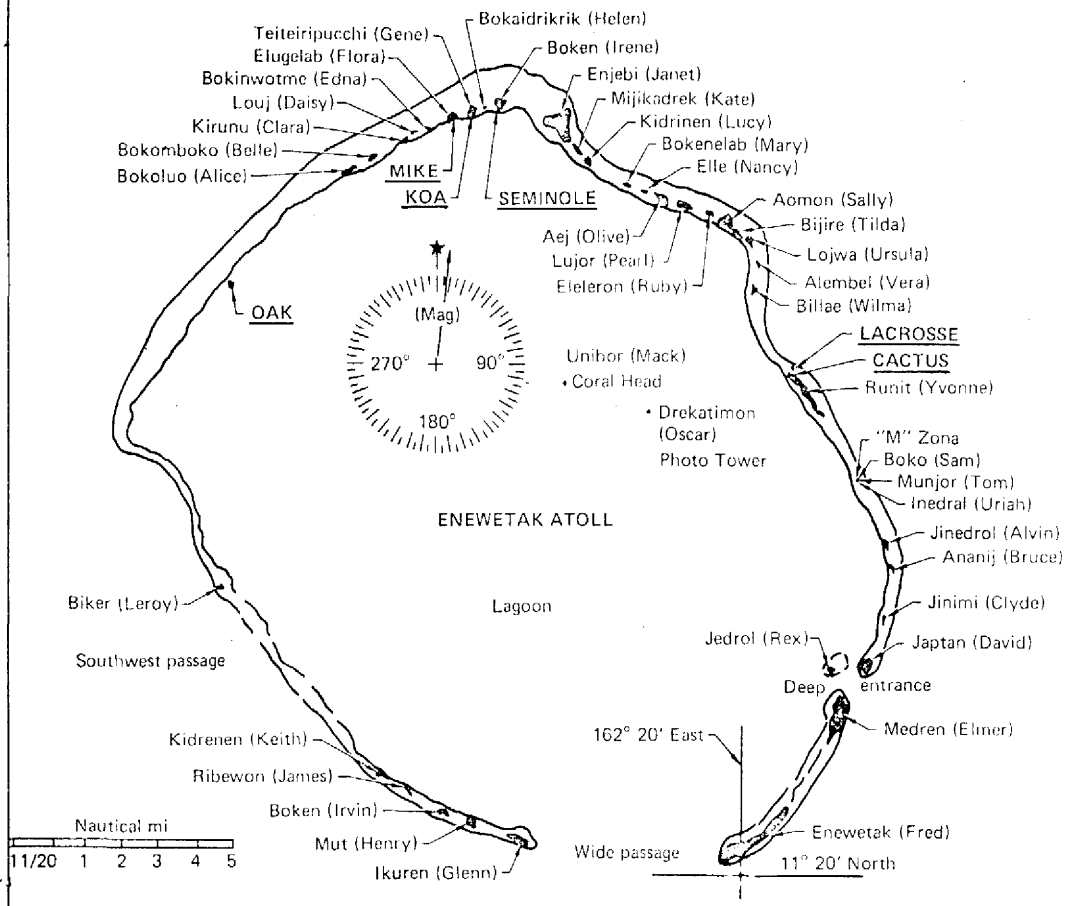

Fig. 1

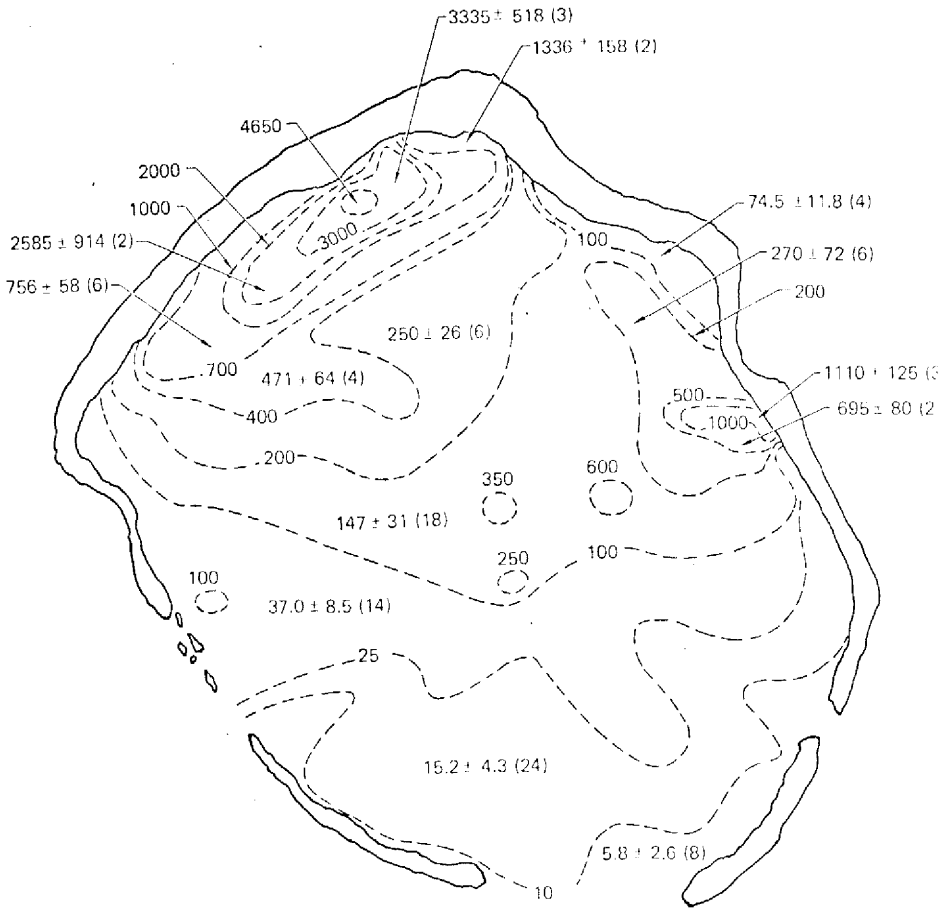

Fig. 2

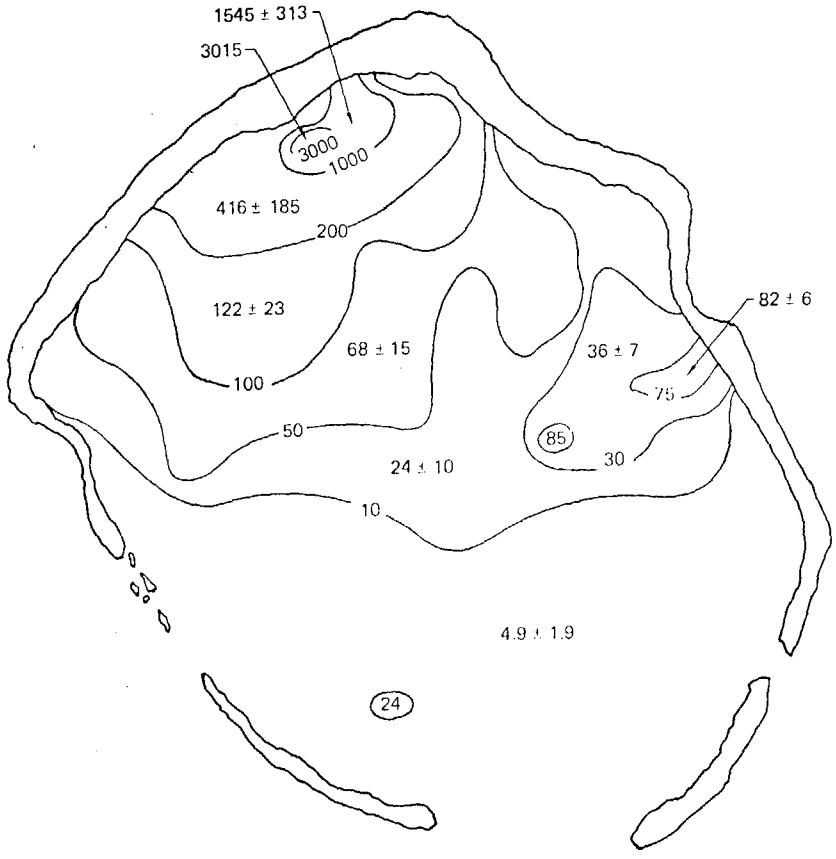

Fig. 3

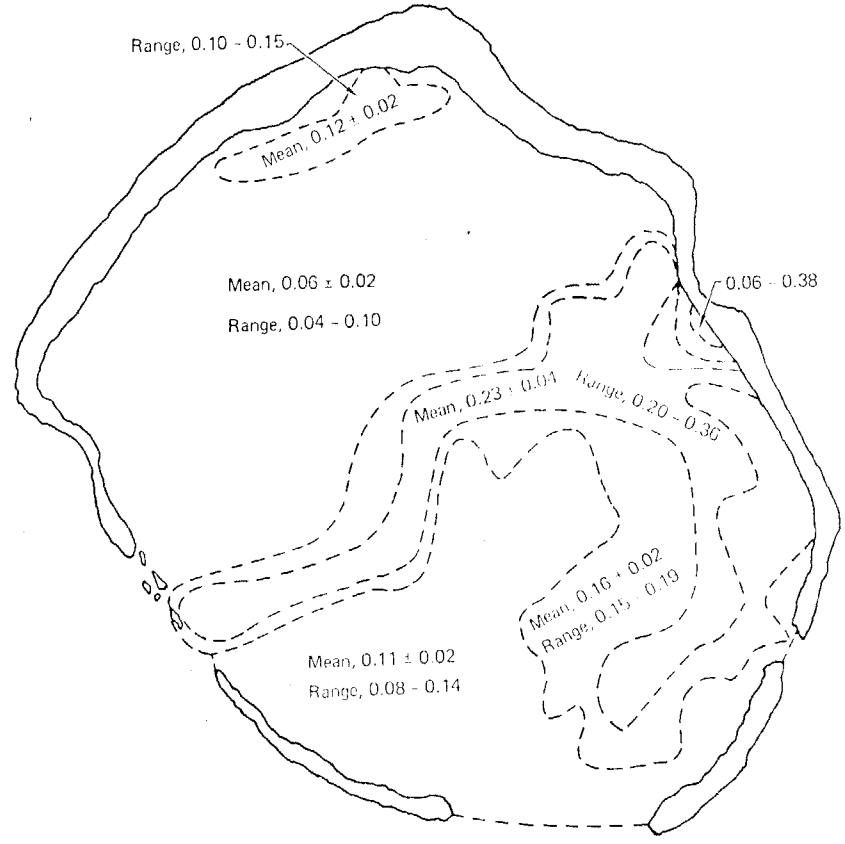

Fig. 4

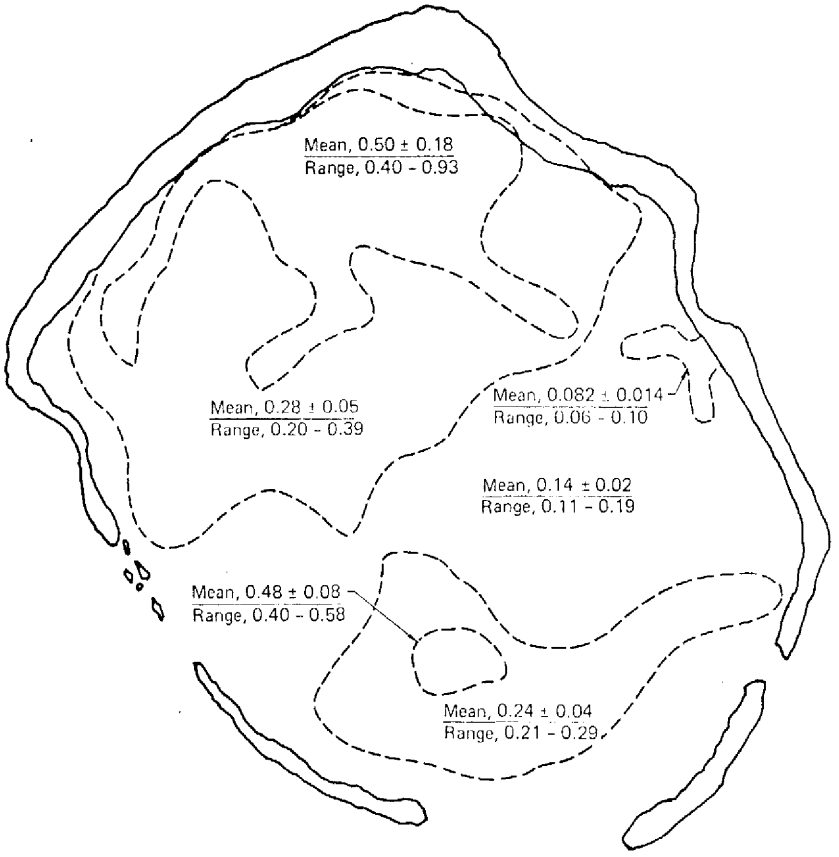

Fig. 5

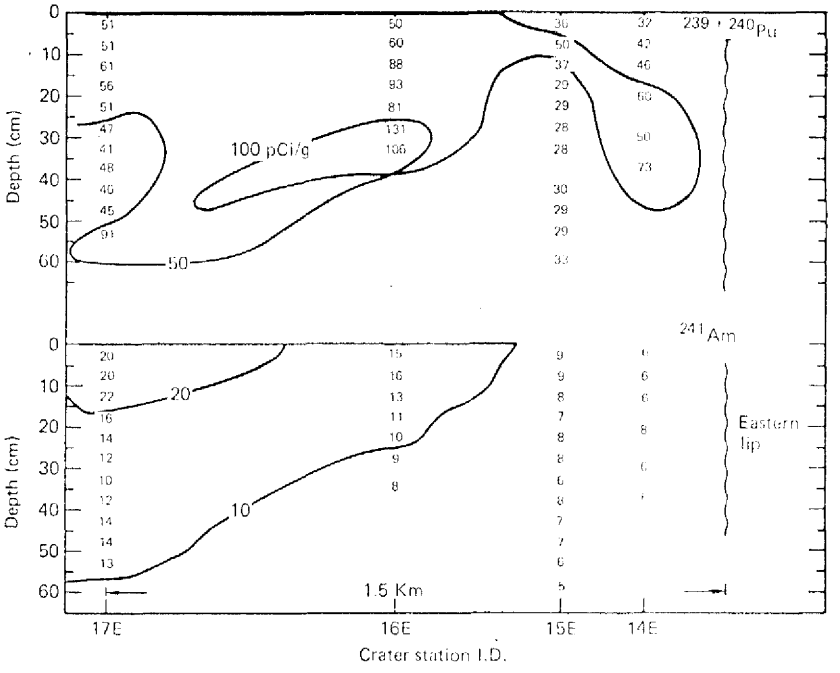

Fig. 6

TABLE CAPTIONS

Table 1. Estimated transuranic sediment inventory: Enewetak and Bikini Atolls, 1 January 1973.

Table 2. ^{241}Am associated with components in core samples of sediment.

Table 3. Plutonium concentrations associated with sediment components (pCi/g dry weight).

Table 4. Concentrations of transuranium radionuclides in Halimeda Algae and Cactus Crater.

Table 5. Plutonium concentrations in the water column at Enewetak Atoll.

Table 6. Recent determinations of the distribution coefficient for plutonium.

Table 7. Concentrations of $^{239+240}Pu$ in yearly growth sections of Enewetak and Bikini coral (fCi/g dry weight).

FIGURE CAPTIONS

Fig. 1 Map of Enewetak Atoll with names and locations of the islands and the 6 nuclear craters.

Fig. 2 Activities of $^{239+240}Pu$ (mCi/km^2) associated with the sediment components in the top 2.5-cm layer of Enewetak Lagoon.

Fig. 3 Activities of ^{241}Am (mCi/km^2) associated with the sediment components in the top 2.5-cm layer of Enewetak Lagoon.

Fig. 4 Activity ratios of ^{238}Pu:$^{239+240}Pu$ in the surface sediments of Enewetak Lagoon.

Fig. 5 Activity ratios of ^{241}Am:$^{239+240}Pu$ in the surface sediments of Enewetak Lagoon.

Fig. 6 Vertical and areal distributions of $^{239+240}Pu$ and ^{241}Am activities in sediments in Mike and Koa Craters (pCi/g).