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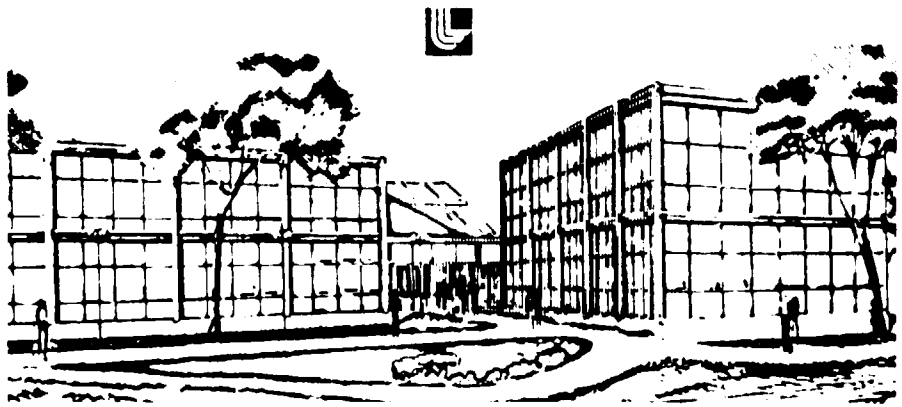

# TRANSURANIUM RADIONUCLIDES IN COMPONENTS OF THE BENTHIC ENVIRONMENT OF ENEWETAK ATOLL

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## 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 $0.058 \pm 0.013$ fCi/l. Water samples from Mike and Koa craters averaged $0.45 \pm 0.22$ fCi/l. Outside of the lagoon and to the east of the Atoll, concentrations in water samples from the open ocean surface averaged $0.013 \pm 0.003$ fCi/l. This comparison shows, as do results for all other transuranics, that Atoll sources contribute the major fraction

REFERENCES

Be76 Beasley T.M., 1976. "Analysis of Windscale and Bikini Atoll Sediments for $^{242}Am$," in Activities of the International Laboratory of Marine Radioactivity 1976 Report, IAEA-187, Vienna.

Bo76 Bowen V.T., Livingston H.D. and Burke J.C., 1976. "Distributions of Transuranium Nuclides in Sediment and Biota of the North Atlantic Ocean," in IAEA Symp. Proc. Transuranium Nuclides in the Environement, IAEA, Vienna, p. 107.

Ci64 Circero L.J. Jr. and Nordyke M.D., 1964. Nuclear Cratering Experience at the Pacific Proving Grounds, Lawrence Livermore Laboratory Rept., UCRL-12172.

Du74 Duursma E.K. and Parsi P., 1974. "Distribution Coefficient of Plutonium between sediment and seawater," in Activities of the International Laboratory of Marine Radioactivity 1974 Report, IAEA-163, Monaco, IAEA, Vienna, p. 94.

Em54 Emery K.O., Tracy J.I., and Ladd H.S., 1954. Geology of Bikini and Nearby Atolls, U.S. Geol. Surv. Prof. Paper 260-A.

En76 Enewetak Marine Biological Laboratory, Mid-Pacific Marine Laboratory, 1976. Contributions 1955-1974, NVO-628-1, Vols. I-III, Mid-Pacific Marine Laboratory, Enewetak, Marshall Islands.



Ga75 Gatrousis C., 1975. Lawrence Livermore Laboratory, private communication.

Ge74 Gerber R.P. and Marshall N., 1974. "Ingestion of Detritus by the Lagoon Pelagic Community at Enewetak Atoll," Limnol. Oceanogr. 19, 815.

Ha73 Hardy E.P., Krey P.W. and Volchok H.L., 1973. "Global Inventory and Distribution of Fallout Plutonium," Nature 241, 444.

He75 Hetherington J.A., Jefferies D.F. and Lovett M.B., 1975. "Some Investigations into the Behaviour of Plutonium in the Marine Environment," in IAEA Symp. Proc., Impacts of Nuclear Releases into the Aquatic Environment, IAEA, Vienna, p. 193.

Hi62 Hines N.O., 1962. Proving Grounds, An Account of the Radiobiological Studies in the Pacific, 1946-1961. Univ. of Washington Press, Seattle, Wash.



Jo67 Johannes R.E., 1967. "Ecology of Organic Aggregates in the Vicinity of a Coral Reef," Limnol. Oceanogr. 12, 189.

Kr76 Krey P.W., Hardy E.P., Pachucki C., Rourke F., Coluzza J. and Benson W.K., 1976. "Mass Isotopic Composition of Global Fallout Plutonium in Soil," in Proc. IAEA Symp. Transuranium Nuclides in the Environment, IAEA, Vienna, p. 671.

Li76 Livingston H.D. and Bowen V.T. 1976. "Americium in the Marine Environment-Relationships to Plutonium," in Environmental Toxicity of Aquatic Radionuclides; Models and Mechanisms (M.W. Miller and J. Newell Stannard, Eds.), Ann

Arbor Science, Publ. Inc., Ann Arbor, Mich. p. 107.

Mo76 Mo T., and Lowman F.G., 1976. "Laboratory Experiments on the Transfer of Plutonium from Marine Sediments to Seawater and to Marine Organisms," in Radioecology and Energy Resources, The Ecological Society of America Special Publ. No. 1 (C.E. Cushing, Jr., Ed.), Dosden, Hutchinson & Ross, Inc., Stroudsburg, Penn., p. 86.

Mor76 Moriarty D.W., 1976. "Quantitative Studies on Bacteria and Algae in the Food of the Mullet, Mugil Cephalus L., and the Prawn, Metapenaeus Bennettae," J. Exp. Mar. Biol. Ecol., 22. 131.

Mou76 Mount M.E., Robison W.L., Thompson S.E., Hamby K.O., Prindle A.L., and Levy H.B., 1976. Analytical Program-1975 Bikini Radiological Survey, Lawrence Livermore Laboratory, Rept. UCRL-51879, part 2.

Ne73 Nevada Operations Office, USAEC, Oct. 1973. Enewetak Radiological Survey, Vols. I-III, NVO-140, Nevada Operations Office, Las Vegas, Nevada.

Ne75 Nevissi A. and Schell W.R., 1975. "Distribution of Plutonium and Americium in Bikini Atoll Lagoon," Health Phys. 28, 539.

No72 Noshkin V.E., 1972. "Ecological Aspects of Plutonium Dissemination in the Aquatic Environment," Health Phys. 22, 537.

No74 Noshkin V.E., Wong K.M., Eagle R.J. and Gatrousis C.,



1974. Transuranics at Pacific Atolls I. Concentrations in the Waters at Enewetak and Bikini, Lawrence Livermore Laboratory, Rept. UCRL-51612.

No75 Noshkin V.E., Wong K.M., Eagle R.J. and Gatrousis C., 1975. "Transuranics and Other Radionuclides in Bikini Lagoon: Concentration Data Retrieved from Aged Coral Sections," Limnol. Oceanogr., 20, 729.

No76 Noshkin V.E., Wong K.M., Marsh K., Eagle R., Holladay G. and Buddemeier R.W., 1976. "Plutonium Radionuclides in the Groundwater at Enewetak Atoll," in IAEA Symp. Proc. Transuranium Nuclides in the Environment, IAEA, Vienna, p. 517.

No78a Noshkin V.E., Wong K.M., Spies R., Eagle R. and Marsh K., 1978. Transuranics at Pacific Atolls-Concentrations in Sediment Components and Benthic Organisms at Enewetak and Bikini, Lawrence Livermore Laboratory, Rept. in preparation.

No78b Noshkin V.E., Wong K.M., Eagle R.J., Jokela T., Brunk J. and Marsh K.V., 1978. Transuranics in Seawater and Pelagic Components at Enewetak and Bikini, Lawrence Livermore Laboratory Rept., in preparation.

No78c Noshkin V.E., Eagle R.J., Wong K.M. and Marsh K.V., 1978. "Remobilization of Plutonium Radionuclides from Cactus Crater Sediment at Enewetak," in preparation.

Pi76 Pillai K.C. and Mathew E., 1976. "Plutonium in the Aquatic Environment, Its Behaviour, Distribution and

Significance," in IAEA Symp. Proc., Transuranium Nuclides in the Environment, IAEA, Vienna, p. 25.

Sm73 Smith S.V., 1973. "Carbon Dioxide Dynamics: A Record of Organic Carbon Production, Respiration and Calcification in the Eniwetok Reef Flat Community," Limnol. Oceanogr. 18, 106.

Sp78 Spies R.B., Marsh K.V. and Colsher J., 1978. "Dynamics of Radionuclide Exchange in the Calcareous Algae, Halimeda," Limnol. Oceanogr., to be submitted.

Wa76 Wahlgren M.A., Alberts J.J., Nelson D.M. and Orlandini K.A., 1976. "Study of the Behaviour of Transuranics and Possible Chemical Homologues in Lake Michigan Water and Biota," in IAEA Symp. Proc., Transuranium Nuclides in the Environment, IAEA, Vienna, p. 9.



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

| | $^{239+240}Pu$ | $^{238}Pu$ | $^{239}Pu$ | $^{240}Pu$ | $^{241}Pu$ | $^{241}Am$ |
|---|---|---|---|---|---|---|
| Enewetak Atoll (area, 933 $km^2$) | | | | | | |
| 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.