

**TRANSURANICS AT PACIFIC ATOLLS
I. CONCENTRATIONS IN THE WATERS
AT ENEWETAK AND BIKINI**

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

June 26, 1974

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**Division of Biomedical
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TRANSURANICS AT PACIFIC ATOLLS

I. CONCENTRATIONS IN THE WATERS AT ENEWETAK AND BIKINI

Abstract

This report presents the available results on the concentrations and distributions of $^{239,240}\text{Pu}$ and other transuranic radionuclides in the lagoon waters of Enewetak and Bikini Atolls. The data are derived from a series of samples collected during the period October-December 1972. The samples are being radiochemically processed and analyzed for specific radionuclides; the available results for ^{90}Sr , ^{137}Cs , ^{237}Np , ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Pu , and ^{241}Am in specific water samples are presented and discussed.

The two Pacific atolls, sites of nuclear testing series in the 1940s and 1950s, act as sources of radionuclides, contributing quantities greatly exceeding the fallout deposition to the lagoon marine environments. The distributions of plutonium in the two lagoons are very heterogeneous; strong concentration gradients are evident in both lagoons, and the horizontal stratifications define areas of water renewal. The concentrations are affected by wave-driven water transported across

the reefs and by flow through the main channels.

Three to 90% of the $^{239,240}\text{Pu}$ in Bikini lagoon is associated with a particulate phase. $^{239,240}\text{Pu}$ concentrations associated with bottom particulates generally exceed the surface levels, suggesting that resuspension of fine material is an active mechanism for redistributing some plutonium throughout the lagoon. In the soluble phase, however, surface plutonium concentrations generally exceed bottom concentrations, indicating slow vertical mixing in the water column, and suggesting (1) that plutonium is derived from several sources, and (2) that surface and bottom currents independently redistribute different forms of the radionuclide to different areas of the lagoon.

The $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios in the surface waters are not constant, indicating that different sources or mechanisms govern the redistribution of specific radionuclides in the water column of the lagoon.

Introduction

This is one of a series of reports describing the distributions of transuranium elements in the marine environment of Bikini and Enewetak Atolls.

During the fall of 1972, personnel from this Laboratory, the University of

Washington, and the Puerto Rico Nuclear Center jointly initiated a biogeochemical investigation of the transuranics at the Pacific Atolls. Samples were collected at Bikini Atoll during November 1972 aboard the RV Palumbo. LLL investigators,

during this period, also collected samples at Enewetak for this transuranic program and a large-scale radiological survey (Enewetak Marine Program in Vol. 1 of Ref. 1). Each laboratory is to present its own data in a series of in-house reports after which all data on specific subjects will be collated and presented as cooperative or individual publications.

This first report, concerns the data on the transuranics in the lagoon water of Enewetak and Bikini. Future articles from this Laboratory will deal with the transuranics in sediment and biota. The purpose of this report is to make our preliminary data available to the participating groups and other interested agencies. Some of the results presented here for transuranics at Enewetak were first published in Volume 1 of the Enewetak Radiological Survey, the transuranium data for water and sediments are abstracted and will be discussed in this and future

reports. In addition, this Laboratory has continued to collect samples at Enewetak since 1972 and some new data are presented.

Our discussion of Bikini is based only on the data derived from samples processed at this Laboratory. Discussions in reports emanating from the Puerto Rico Nuclear Center and the University of Washington may differ significantly from ours even though they may be based on similar and/or duplicate samples. The reader is advised to carefully assess the reports from the three laboratories.

The analytical techniques are not discussed in these reports; however, the procedures are essentially those reported in several published articles appearing in the bibliography. It is assumed that the reader is familiar with the nuclear test history of the Atolls and the physical geography of the region; those readers requiring background information on these subjects are referred to Refs. 1 and 2.

Transpacific Surface Waters

During the passage of the R. V. Palumbo from San Diego to the Marshall Islands in September-October 1972, periodic samples were taken enroute to provide background information on levels of fallout radionuclides in equatorial Pacific waters (Table 1). Surface seawater samples were collected by bucket within each 10° longitudinal band and transferred to 55-liter black "Deldrum" containers for storage.

In the region bounded by 12° and 14°N latitude and 180°W and 170°E longitude, plutonium concentrations were about 0.4 fCi/kg, and ^{137}Cs averaged 155 fCi/kg. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios were relatively

higher at lower latitudes than at more northern latitudes, reflecting relatively higher depositions of ^{238}Pu from the 1964 SNAP-9A generator burnup. The ratio in the surface water is considerably higher than the cumulative deposition ratio of 0.037 reported³ for fallout at these latitudes. Apparently, the ^{238}Pu deposited more recently has mixed in the surface water with a depleted inventory of $^{239,240}\text{Pu}$.

The $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio has shown no significant hemispheric or latitudinal variations. The average deposition ratio by 1972, on a global basis, is computed from earlier deposition data^{4,5} to be

Table 1. Radionuclide concentrations and concentration ratios in unfiltered Pacific surface waters. Values in parentheses: one standard deviation based on counting statistics.

Sample No.	Sampling location	Date	Radionuclide concentration (fCi/kg)				Concentration ratios	
			$^{239,240}\text{Pu}$	^{238}Pu	^{137}Cs	^{90}Sr	$^{238}\text{Pu}/^{239,240}\text{Pu}$	$^{239,240}\text{Pu}/^{137}\text{Cs}$
RVP-1	32°N 120°W	9/29/72	0.55 (0.25)	n. d. ^a	270 (50)	1.9 ± 0.5		0.002
RVP-2	31°N 130°W	10/1/72	0.90 (0.17)	0.06 (0.02)	430 (25)	1.2 ± 0.3	0.07	0.002
RVP-3	27°N 140°W	10/3/72	0.92 (0.43)	n. d.	220 (50)	1.5 ± 0.7		0.004
RVP-4	24°N 150°W	10/6/72	2.3 (0.5)	n. d.	220 (50)	150 ± 13		0.010
RVP-5	21°N 160°W	10/12/72	1.3 (0.2)	0.46 (0.16)	250 (30)	9.6	0.35	0.005
RVP-6	17°N 173°W	10/15/72	1.5 (0.2)	0.09 (0.08)	130 (30)	71 ± 4	0.06	0.011
RVP-7	14°N 180°W	10/17/72	0.35 (0.05)	0.05 (0.04)	170 (20)	3.7	0.14	0.002
RVP-8	12°N 170°W	10/19/72	0.44 (0.07)	0.07 (0.06)	140 (20)	22.2	0.16	0.003

^an. d., below detection limits.

0.020; the mean ratio for our surface Pacific samples is 0.0044. By 1972 about 75% of the deposited fallout plutonium has separated in the surface waters from ^{137}Cs and must be redistributed by process other than those which are responsible for the movement of the typical "soluble" fallout ^{137}Cs .

These observations are consistent with the view that fallout plutonium is rapidly depleted in surface^{6,7} waters following its

deposition on the sea surface. Its rate of removal must be largely controlled by its ability to associate with sinking particulates.

The average $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio in Pacific surface waters is very similar to values reported in Atlantic surface waters.⁶ The rate of withdrawal of plutonium from the Atlantic water column must be similar to that in the Pacific. It is possible that similar mechanisms control the fate of fallout plutonium in the two oceans.

Enewetak Lagoon

The marine sampling program at Enewetak has been described in detail.¹ Briefly, during October-December 1972, unfiltered, 15-gallon water samples were collected with a pump and hose-line assembly from the locations shown in Fig. 1. Samples were processed and analyzed at Lawrence Livermore Laboratory. The concentrations in water of $^{238,239,240}\text{Pu}$ and ^{137}Cs (reported previously in Ref. 1) and of ^{90}Sr are abstracted in Table 2.

GENERAL RADIONUCLIDE INVENTORY

The average concentrations of ^{238}Pu , $^{239,240}\text{Pu}$, ^{137}Cs , and ^{90}Sr in the lagoon,

weighted by area, are, respectively, 4.7, 39, 317 and 330 fCi/kg. Comparison of the average $^{239,240}\text{Pu}$ concentration to the value of 0.3-0.4 fCi/kg for water east of the lagoon and in the North Equatorial samples shows that the Atoll is the major source for plutonium in the lagoon. The concentrations of ^{137}Cs and ^{90}Sr in the lagoon water are on the average only three times as high as the current levels of fallout in offshore surface waters. If we take the average water concentration and assume the average depth of the lagoon to be 47.4 meters, we obtain the following values :

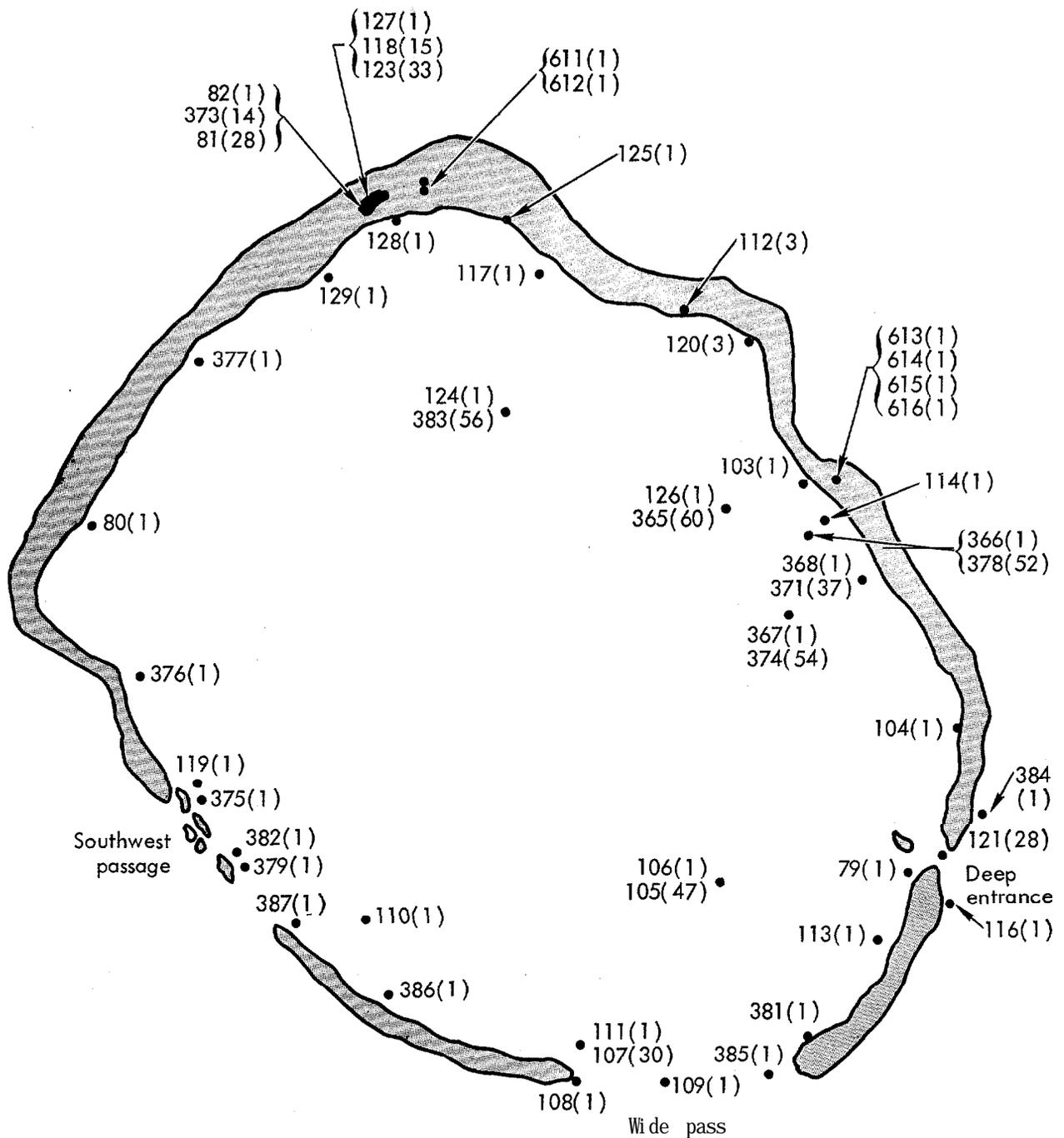


Fig. 1. Enewetak lagoon: water-sampling stations. Values in parentheses are depths sampled in meters.

	$^{239,240}\text{Pu}$	^{90}Sr	^{137}Cs
Water column activity (mCi/km ²)	1.84	15.6	15.0
Lagoon, standing inventory (curies)	1.7	14.6	14.0
Deposition to 10-20°N (mCi/km ²) ⁴	0.24	8.0	12.0

The present standing inventory in the water column alone exceeds the total fallout deposition to these latitudes, indicating again that the Atoll acts as a major contributor of radionuclides to the lagoon water. The total activity in the water column at Enewetak, however, is lower than the total fallout delivered to more northern latitudes (4).

Table 2. Concentrations of $^{239,240}\text{Pu}$, ^{238}Pu , ^{137}Cs , and ^{90}Sr in unfiltered Enewetak lagoon water. Values in parentheses = one standard deviation based on counting statistics.

Station No.	Depth ^a (m)	Collection date	Radionuclide concentration (fCi/kg)			
			$^{239,240}\text{Pu}$	^{238}Pu	^{137}CS	^{90}Sr
79	S	12/9/72	6.0 (1.1)	1.1 (0.3)	296 (19)	166 (14)
80	S	12/12/72	32.5 (3.0)	2.7 (0.5)	471 (22)	420 (34)
103	S	12/2/72	43.6 (1.4)	6.8 (0.3)	486 (17)	468 (42)
104 _b	S	11/29/72	13.1 (0.7)	1.9 (0.2)	241 (18)	184 (17)
105 ^b	47	12/1/72	17.4 (0.7)	2.5 (0.2)	300 (19)	300 (24)
106	S	12/1/72	22.4 (0.7)	2.2 (0.1)	342 (19)	473 (43)
107 ^c	30	10/18/72	9.6 (0.5)	0.9 (0.1)	190 (14)	163 (5)
108	S	10/18/72	10.2 (0.5)	1.1 (0.2)	229 (16)	223 (9)
109	S	10/19/72	9.6 (0.5)	1.0 (0.1)	228 (17)	167 (5)
110	S	10/20/72	28.9 (0.9)	3.8 (0.2)	377 (18)	306 (25)
111	S	10/18/72	11.6 (0.4)	1.4 (0.9)	258 (20)	230 (16)
112	S	12/2/72	15.4 (0.7)	1.9 (0.2)	163 (19)	200 (16)
113	S	11/27/72	4.8 (0.3)	0.6 (0.1)	170 (18)	110 (6)
114 ^d	S	12/2/72	51.9 (1.9)	7.1 (0.4)	462 (17)	505 (36)
116 ^d	S	12/9/72	0.43 (0.25)	0.01 (0.01)	n.d. ^k	96 (8)
117	S	12/5/72	11.8 (0.9)	1.7 (0.2)	107 (30)	248 (20)
119	S	12/10/72	18.0 (0.9)	2.3 (0.2)	290 (17)	315 (13)
120	S	12/2/72	7.4 (0.6)	1.1 (0.1)	228 (14)	170 (5)
121 ^e	28	12/9/72	2.80 (0.7)	0.14 (0.05)	251 (22)	133 (6)
124	S	12/7/72	71.2 (2.3)	10.0 (0.5)	579 (18)	937 (38)
125	S	12/7/72	6.8 (1.0)	1.6 (0.2)	59 (9)	Lost
126	S	12/7/72	30.4 (1.2)	3.9 (0.3)	322 (18)	635 (40)
128	S	12/5/72	33.1 (1.5)	3.0 (0.3)	532 (25)	1 070 (120)
129	S	12/7/72	44.4 (1.7)	4.4 (0.3)	538 (20)	Lost
366	S	12/16/72	77.0 (3.1)	13.3 (0.8)	499 (28)	434 (13)
367	S	12/15/72	66.2 (3.0)	7.9 (0.6)	482 (25)	426 (21)
368 _f	S	12/16/72	96.1 (3.7)	14.9 (0.8)	410 (23)	397 (20)
371 ^f	37	12/16/72	75.2 (3.1)	11.2 (0.7)	305 (20)	410 (37)
374 ^g	54	12/15/72	63.2 (2.8)	9.0 (0.6)	462 (22)	428 (47)
375	S	12/12/72	29.0 (1.7)	3.7 (0.4)	305 (23)	350 (40)
376	S	12/12/72	18.6 (1.2)	2.6 (0.3)	250 (20)	277 (11)
377 ^h	S	12/12/72	62.9 (2.7)	9.7 (0.7)	364 (21)	455 (16)
378 ^h	52	12/16/72	43.1 (1.4)	7.1 (0.3)	497 (25)	455 (17)
379	S	12/12/72	14.5 (0.7)	2.1 (0.2)	246 (19)	184 (11)
381	S	12/9/72	6.8 (0.5)	(0.7) (0.1)	176 (19)	180 (11)
383 ⁱ	56	12/7/72	53.3 (2.0)	4.6 (0.3)	295 (25)	882 (60)
384 ^d	S	12/9/72	0.21 (0.25)	0.0 (0.05)	146 (26)	94 (11)
385	S	12/9/72	1.60 (0.14)	0.5 (0.1)	130 (20)	
386	S	12/10/72	13.9 (0.6)	2.0 (0.2)	291 (30)	324 (10)
387	S	12/10/72	0.38 (0.10)	0.03 (0.01)	109 (32)	116 (6)
612 ^j	S	12/15/72	302 (4)	65 (2)	212 (38)	421 (50)

^aS, surface sample.

^bBottom water, surface station 106.

^cBottom water, surface station 111.

^dEast of atoll; refer to Fig. 1 for location.

^eBottom water, deep pass.

^fBottom water, surface station 368.

^gBottom water, surface station 367.

^hBottom water, surface station 366.

ⁱBottom water, surface station 124.

^jOne-quarter mile north of Seminole Crater, on the ocean side.

^kn.d., below detection limits.

PLUTONIUM, ^{137}Cs , AND ^{90}Sr

Distributions in Surface Waters

The values of the surface concentrations of ^{238}Pu , $^{239,240}\text{Pu}$ and ^{137}Cs are given for each sample location in Figs. 2 through 4, with isolines outlining areas of similar concentration. The distributions of $^{239,240}\text{Pu}$ and ^{137}Cs are very heterogeneous within the lagoon. Plutonium concentrations range from 0.4 to 96 fCi/kg; they are highest in an area extending from Yvonne in the east to the western reef and lowest along the inner perimeter of the southern half and northeast quadrant of the lagoon. Ocean water of low plutonium concentration (0.3 fCi/liter) advects over the northern reef and through the southern channels and mixes with the higher-activity water in the lagoon; the result is concentration gradients increasing in value from both the north and the south toward the center of the lagoon. The ^{238}Pu and $^{239,240}\text{Pu}$ are very similar in areal distribution; ^{137}Cs differs in that concentrations are highest in waters from the northwest quadrant. The ^{137}Cs and plutonium radionuclides must reach the lagoon from different sources. When the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios are mapped and isolines are constructed (Fig. 5), the largest changes in ratio occur at regions on the inner perimeter of the lagoon. The smallest ratios are near channels and passages or at areas of the northern reef, indicating the principal locations of advective lenses of ocean water that exchange with the lagoon. The ratios for samples from the vast central area of the lagoon appear similar in value but not identical, indicating that even the central area of the lagoon is not well mixed with respect to these two

radionuclides. However, although $^{239,240}\text{Pu}$ and ^{137}Cs may have different sources the general wind-driven overturning circulation appears to keep the central water of the lagoon well mixed to a degree, whereas tide-controlled currents greater affect the concentrations levels around the inner perimeter of the lagoon.

The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio in open lagoon water varied only between 0.09 and 0.17; the only discernible gradients in ratio are illustrated in Fig. 6. The mean ratio for 45 samples, excluding crater water values, was 0.12 ± 0.04 , which is five times the mean value for Bikini; this indicates either that ^{238}Pu is more easily leached to the water at Enewetak or that the ratio for total plutonium sources is higher at Enewetak than at Bikini.

To demonstrate the significant differences around the inner perimeter of the lagoon, Fig. 7 gives the ^{137}Cs concentration and the $^{137}\text{Cs}/^{90}\text{Sr}$ ratios and Fig. 8 gives the $^{239,240}\text{Pu}$ concentrations and $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios. Several pairs of samples collected from almost identical locations within a day or two contained considerably different levels of activity. Some mechanism must be operating over a small time scale to produce such rapid changes.

To test the assumption that the concentration levels found around the inner perimeter of the lagoon are affected by tide-controlled currents, the concentrations are plotted in Figs. 9 and 10 over the tidal cycle for the closely spaced stations in three areas: off Yvonne, in the area of the southwest passage, and in the northwest area near Mike and Koa craters. During sampling, the collection time for each sample was recorded and pumping

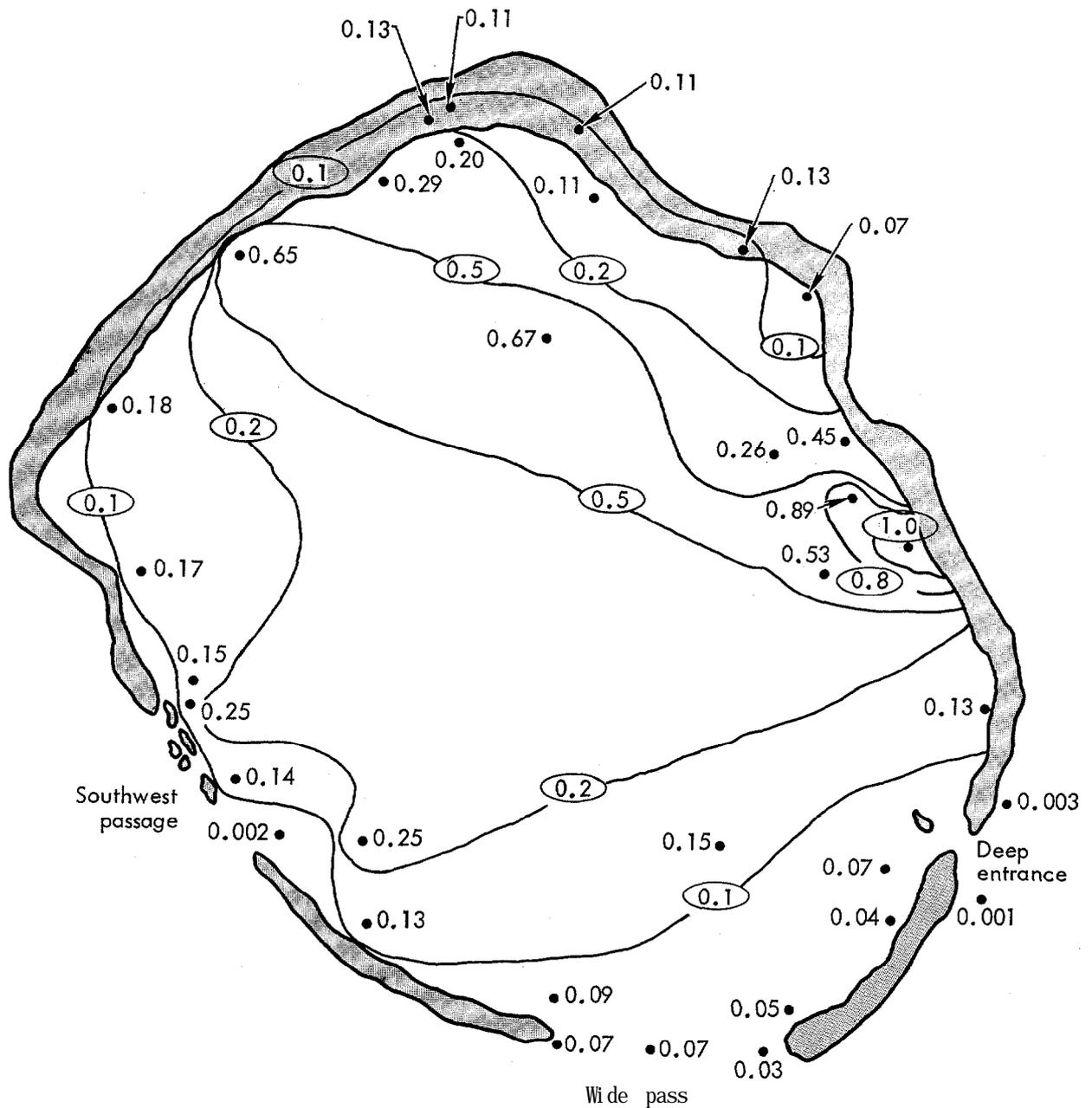


Fig. 2. Enewetak lagoon: surface concentrations and isolines for ^{238}Pu . In this figure only, all values are normalized to highest surface concentration (at Station 368).

time did not exceed 10 minutes; the collection time for each sample was assigned to its specific location in the tidal cycle by reference to a chart of daily high and low tides for Enewetak. In Fig. 9, which plots the levels of $^{239,240}\text{Pu}$ and the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios from stations in water off

Yvonne, flood tide was associated with increases both in the concentration of plutonium and in the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio, and ebb tide was associated with reductions in both concentration and ratio. Wind-driven surface currents over the reef could average as much as 1 km/hr;

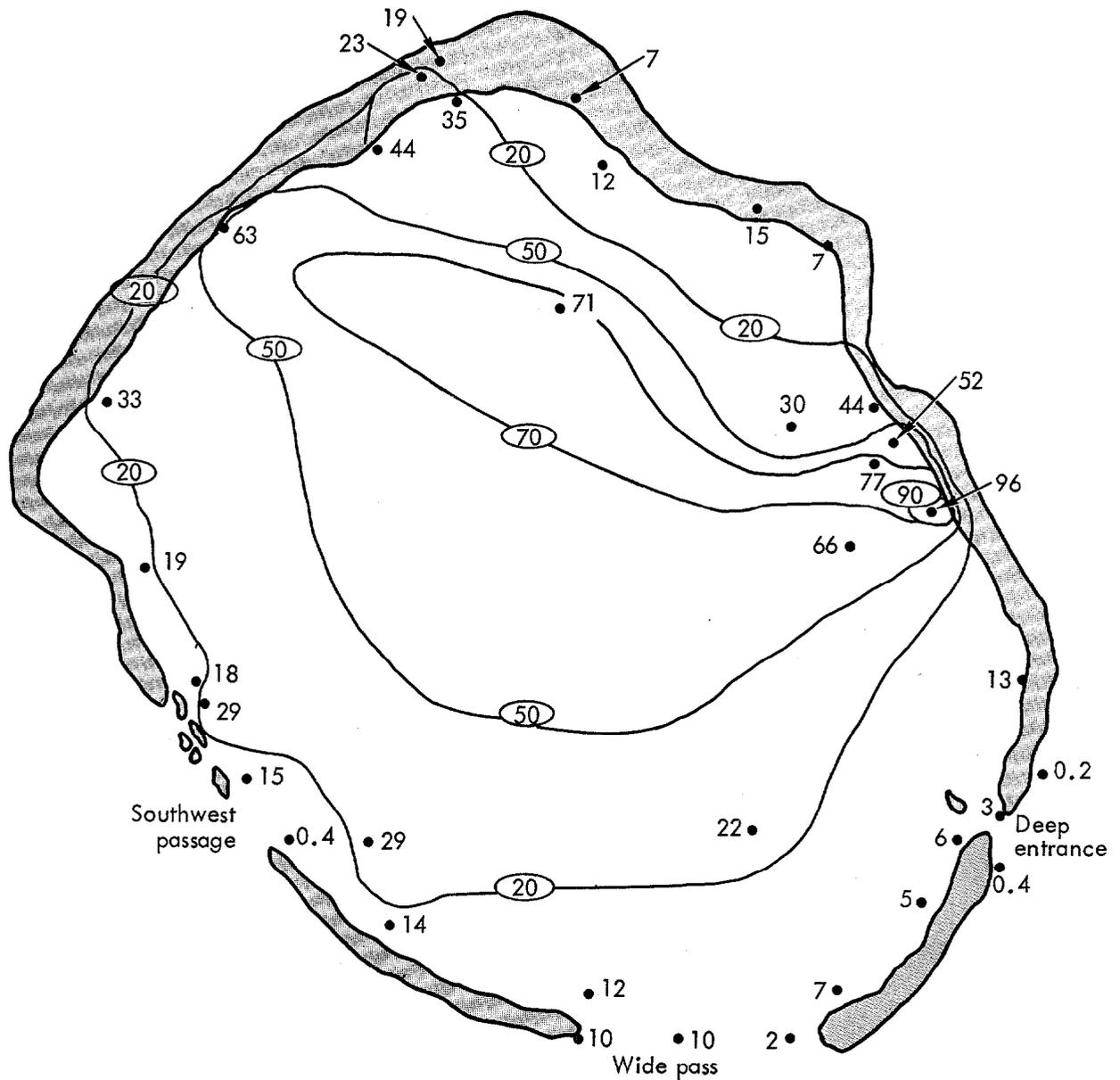


Fig. 3. Enewetak lagoon: surface concentrations and isolines for $^{239,240}\text{Pu}$. All values are in fCi/kg.

hence, concentrations at stations as far as 4 to 5 km away from the reef appear to be influenced by the daily tidal changes. The increased concentration as ocean water passes across the reef and mixes with the surface water of the lagoon, however, indicates that the reef around Yvonne must be acting as a source of plutonium.

Figure 10 presents the contrasting situation at four stations near the southwest passage and at four stations near Mike and Koa. In the surface waters in these areas, both the concentration of $^{239,240}\text{Pu}$ and the $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratio decreased between ebb and flood tide. Tidal action in the area of the southwest passage brings fresh ocean water through the pass

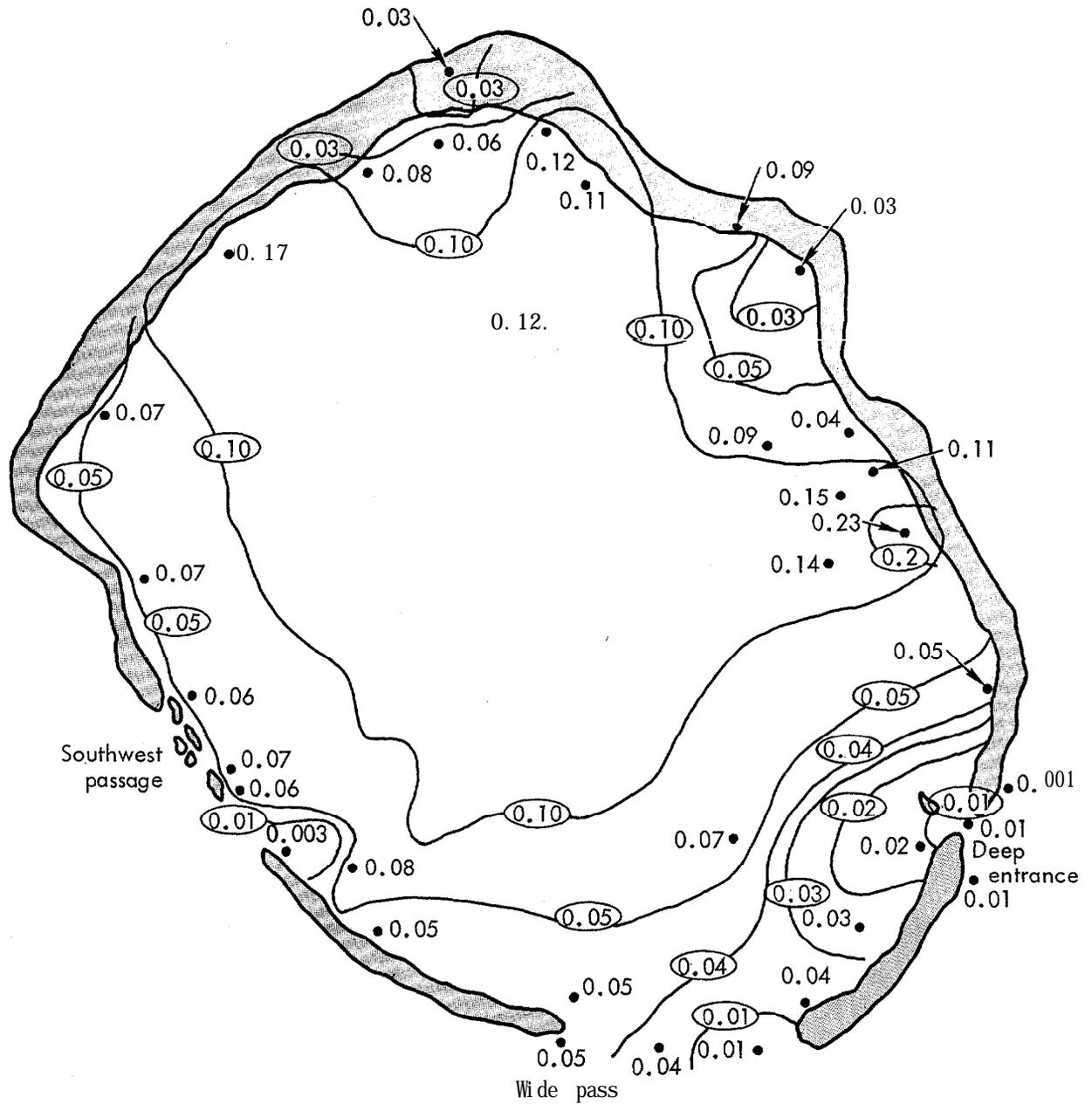


Fig. 5. Enewetak lagoon: $^{239,240}\text{Pu}/^{137}\text{Cs}$ concentration ratios and isolines.

suggest that the concentration of radionuclides in the water around the lagoon perimeter is affected differently at different locations by tide-controlled currents during the winter period. The reef around Yvonne may be a principal contributor of plutonium to the water within the lagoon.

Physical State of Plutonium

The water samples taken from Enewetak lagoon were unfiltered. Data available from Bikini show that at least 3 to 90% of the plutonium in surface water is bound to particulates greater than $1\ \mu\text{m}$ in diameter and that the percentages are even larger

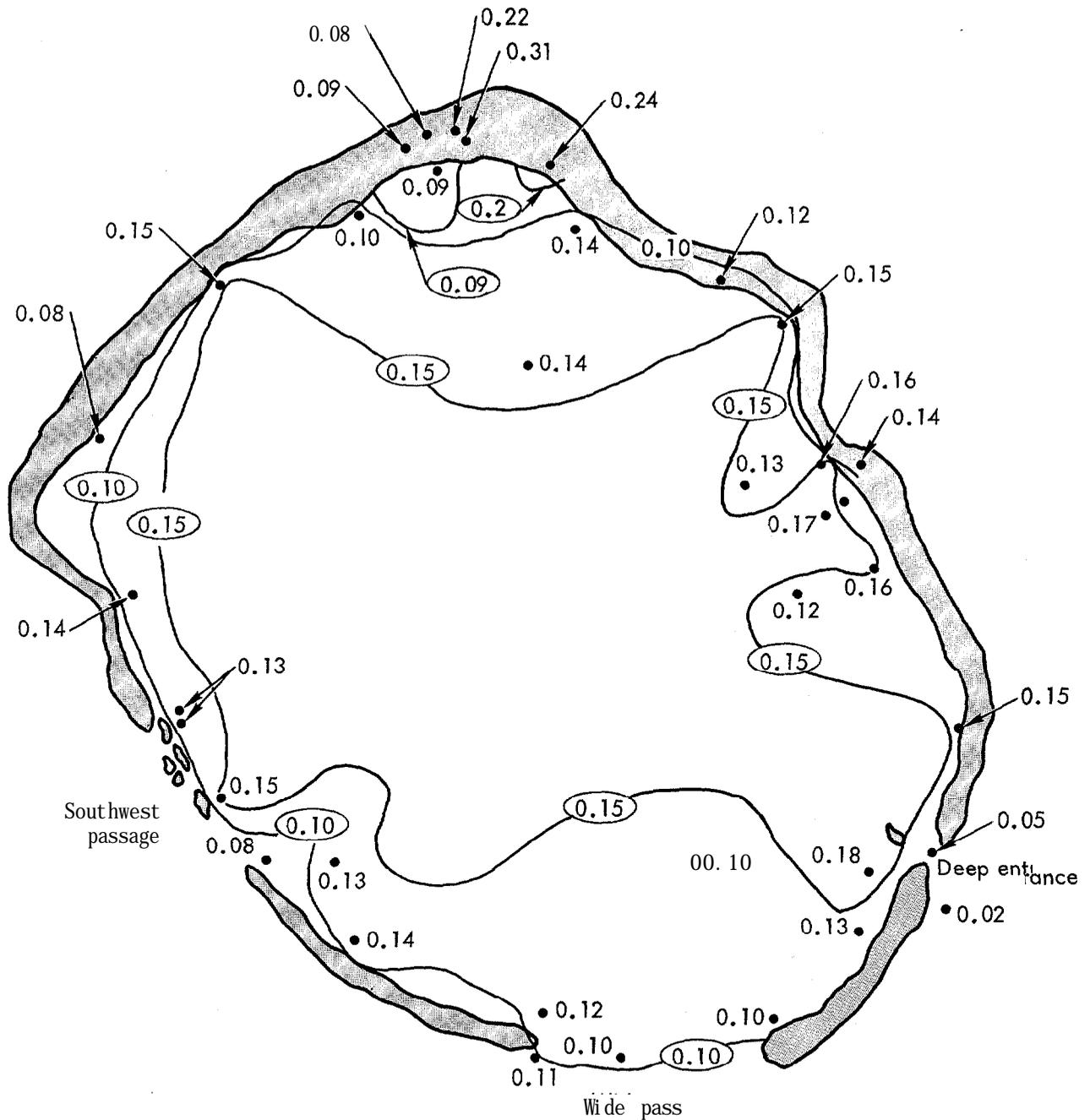


Fig. 6. Enewetak lagoon : $^{238}\text{Pu}/^{239,240}\text{Pu}$ concentration ratios and isolines.

in bottom water. Undoubtedly, plutonium in Enewetak water is distributed between the particulate and soluble fractions and the ratio differs from station to station. The existence of one form of particulate material in the water was established by direct visual observation. A string of underwater buoys spaced 30 ft apart was

placed by R. Nolan of Scripps Institute of Oceanography off the northern tip of Fred. During August 1973, divers from this Laboratory could not see the second buoy from the first, but during February 1974 the third buoy was clearly visible from the first. Thus, at time the lagoon contains appreciable quantities of suspended debris

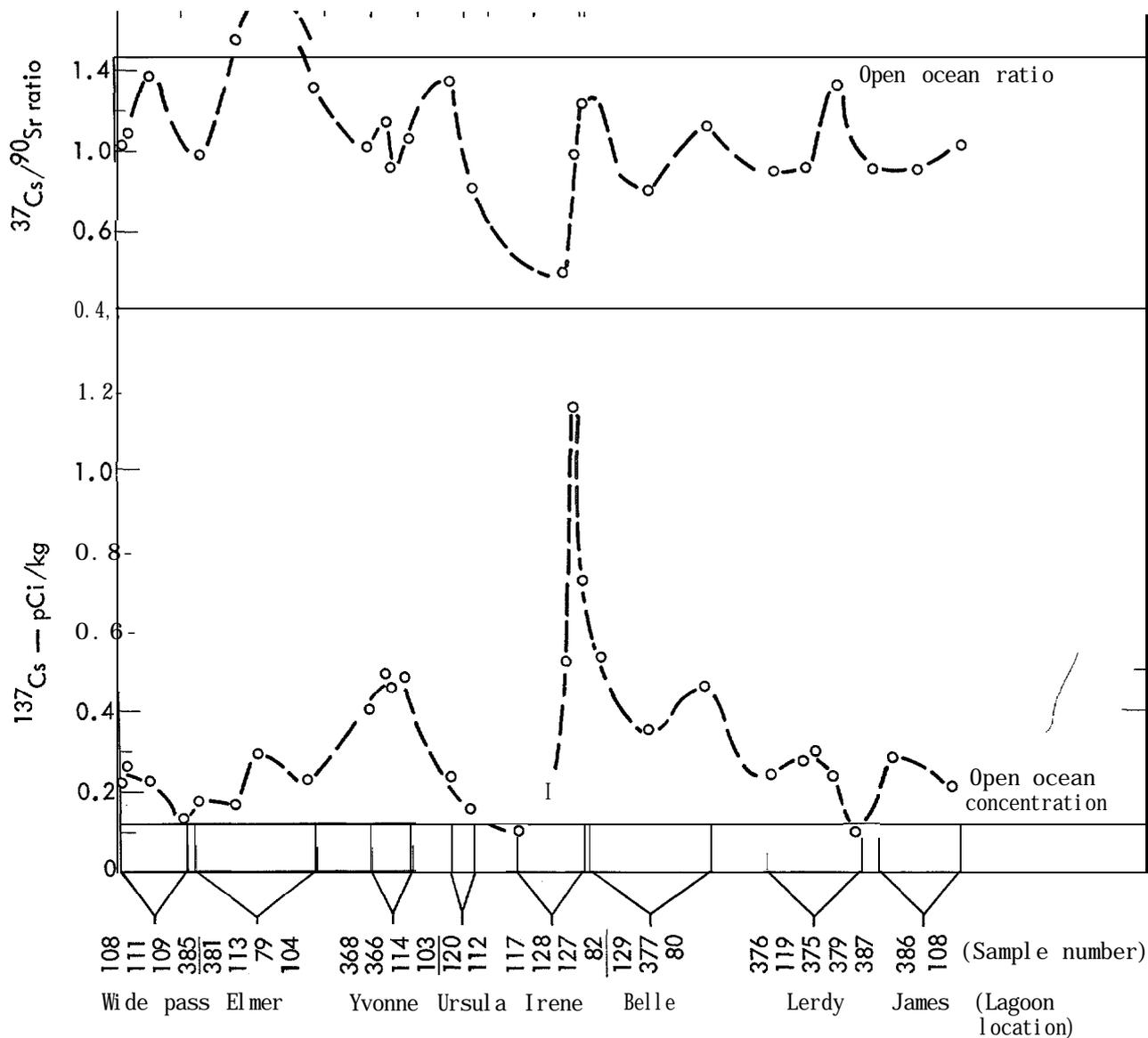


Fig. 7. Enewetak lagoon: ^{137}Cs concentrations (pCi/kg) and $^{137}\text{Cs}/^{90}\text{Sr}$ concentration ratios for the perimeter of the lagoon.

that lowers visibility. Physical observation confirmed that most of the material responsible for the poor visibility during August 1973 was coral floc.

The plankton data will be discussed in greater detail in a future report, but some aspects of our Enewetak results are pertinent to the present discussion. We found that the average concentration factor

for $^{239,240}\text{Pu}$ by lagoon plankton was 10^4 ; this value is approximately 5 times the value derived for planktonic species in the open ocean.⁸ Either the atoll species are uniquely efficient collectors of plutonium or some other particulates collected along with the plankton are contributing to the observed activity. The ^{90}Sr was also measured in several samples; the calculated

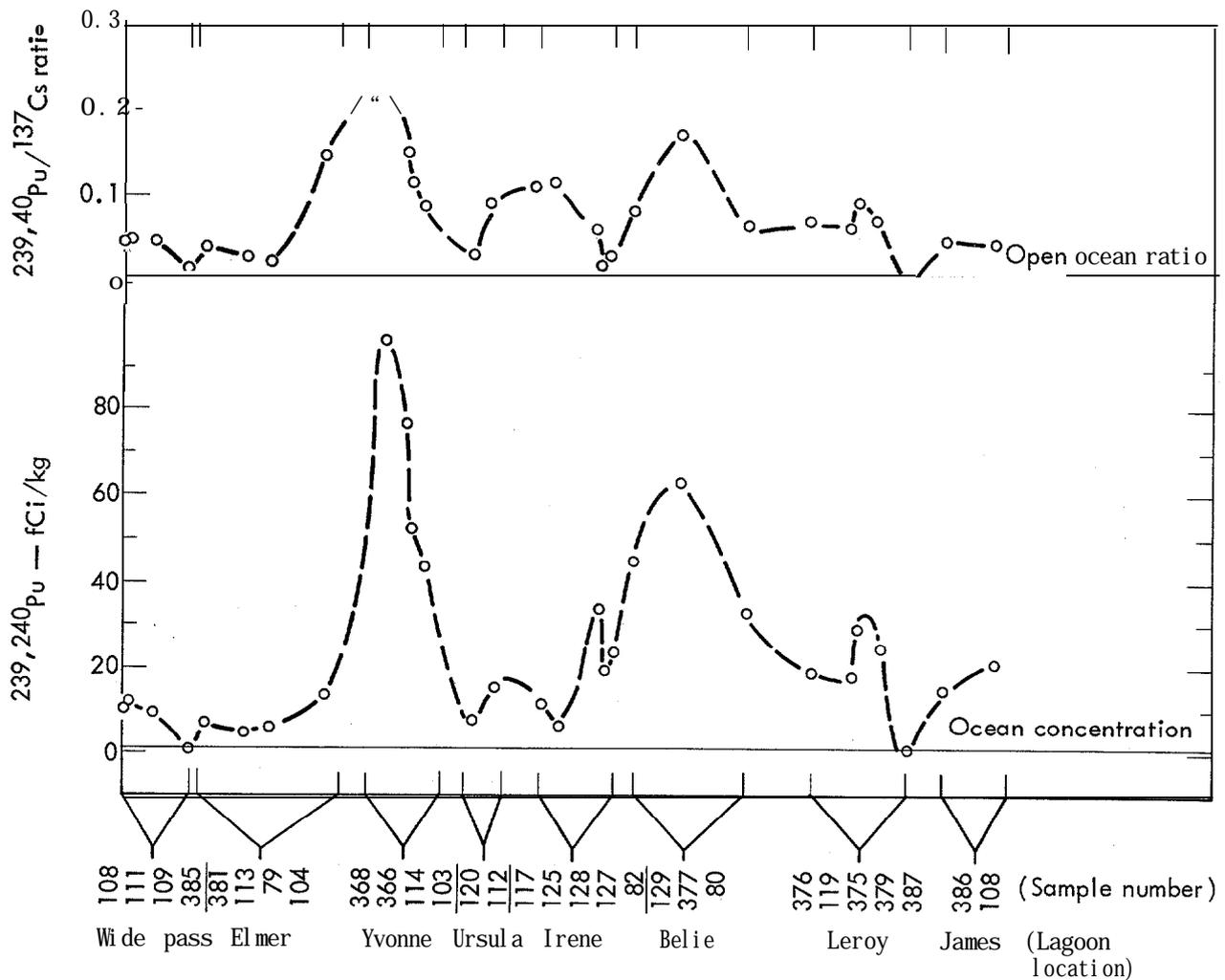


Fig. 8. Enewetak lagoon: $^{239,240}\text{Pu}$ concentrations (fCi/kg) and $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios for the perimeter of the lagoon.

concentration factor was 1.6×10^3 , which is greater than any previously determined value by orders of magnitude. If plankton derive soluble ^{90}Sr from the water, stable strontium must be accumulated in proportion to the ^{90}Sr in the water. The average concentration of stable strontium in sea water is 8 mg/liter; and on the basis of the concentration factor of 1.6×10^3 , the concentrating organisms must contain 1.2% stable strontium. Since the only organism in the lagoon containing approximately this high a concentration of stable strontium is coral,⁹ it can be concluded

that a large fraction of the collected plankton contain quantities of coral fragments. Microscopic examination of several unprocessed samples has since verified this conclusion. The plankton samples from either inside or outside Bikini and Enewetak contain thousands of small coral fragments bound quite often in a matrix of coral floc. Separation of the material from the plankton species is being attempted, but note first, that unprocessed plankton samples will probably yield high concentration factors for many radionuclides, including plutonium, and

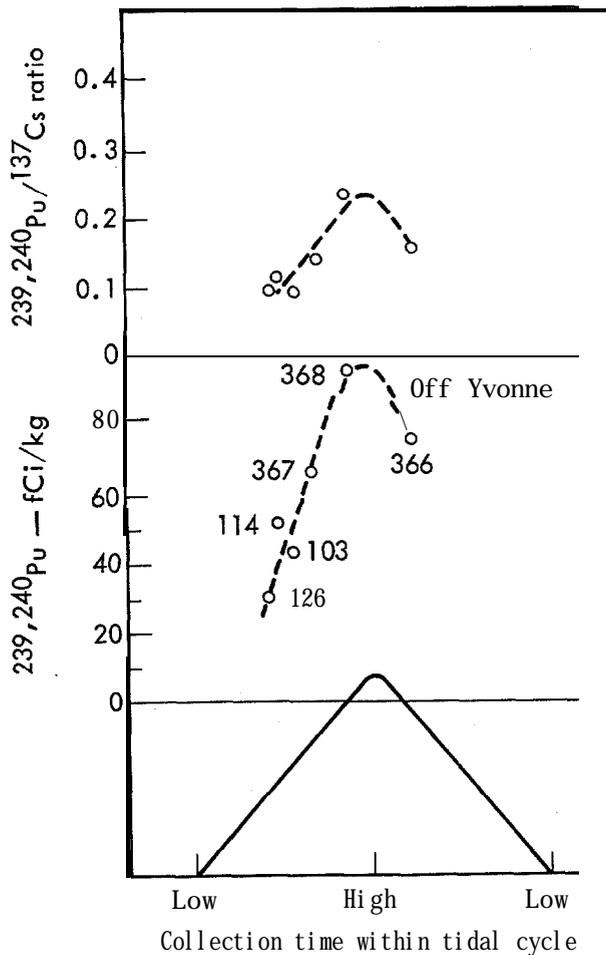


Fig. 9. Changes in $^{239,240}\text{Pu}$ concentrations and $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios during the tidal cycle in water off Yvonne. The station number is given for each data point.

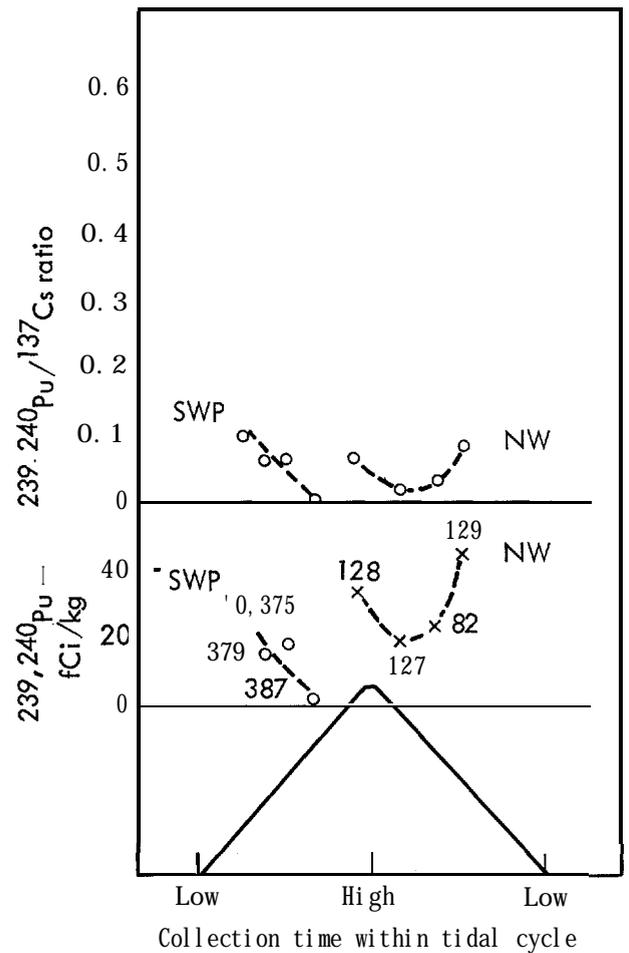


Fig. 10. Changes in $^{239,240}\text{Pu}$ concentrations and $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios during the tidal cycle in water in the area of the southwest passage (SWP) and in the northwest (NW) near Mike and Koa craters. The station number is given for each data point.

second that considerable quantities of radionuclides are bound also in small suspended coral fragments and floc in the water phase. Undoubtedly, many of these fragments are resuspended by upwelling or are formed as a result of wave action on the beaches of the lagoon; however, other processes, could be involved also. Recently, Smith¹⁰ studying organic carbon production and calcification in the Enewetak Reef Flat community concluded that on the

average, the reef calcifies at a rate of 4 kg CaCO_3 per square meter per year. Since there has been no net CaCO_3 accumulation on the reef over the last several thousand years, this rate must be equal also to the rate of decalcification of the reef flat. The reef area of 51.52 km^2 , would yield a total of CaCO_3 to the lagoon of 2.1×10^{11} grams per year.

If the Enewetak lagoon flushes at a rate similar to that at Bikini, its lagoon volume of water is renewed in the tradewind season every 39 days¹¹ yielding a standing inventory of 0.15 mg/liter of either dissolved or particulate CaCO₃ and the associated radioelements derived from the reef breakdown. As the reef builds and breaks down, plutonium and other radionuclides could be eroded in either soluble or particulate forms and could enter the lagoon during flood tides. If this continuous process occurred in an area of the reef containing high quantities of plutonium, it could be one of the principal mechanisms contributing to the plutonium to the lagoon.

Surface-Bottom Relationships

In the open lagoon only six surface-bottom profiles were obtained; four of the stations were in the northeast quadrant of the lagoon and the other two were at surface stations 111 and 106 in the southern half of the lagoon, north and northwest of the Wide Pass. The surface-bottom ratios for ^{239,240}Pu, ¹³⁷Cs and ⁹⁰Sr are given in Table 3.

The circulation of the water in Enewetak lagoon has never been thoroughly investigated, but its main features are probably similar to the general circulation at Bikini and Rongelap.¹¹ A wind-driven surface current of variable thickness moves from east to west. Some sinking of surface water occurs at the leeward reef; this water mixes and returns with the bottom current. Upwelling probably occurs some distance offshore from the eastern reef, and there may be counter-rotating compartments in the secondary bottom flow. During the winter season, these vertical motions should produce

homogeneity in the lagoon water. But the radionuclide data do not confirm this general pattern. First, surface concentrations in the lagoon are extremely variable as indicated earlier. Second, the surface-to-bottom concentration ratios indicate that portions of the lagoon are not well mixed vertically. There is no difference in ⁹⁰Sr between surface-to-bottom samples off Yvonne, but surface concentrations exceed bottom concentrations in the more southern stations. Both ^{239,240}Pu and ¹³⁷Cs are, on the average, 30% higher in all surface-over-bottom samples.

The lower concentrations of ^{239,240}Pu observed in the bottom waters suggest that leaching or resuspension from sediments, although active, is somewhat overshadowed since these two processes would be expected to result in relatively higher concentrations of radionuclides in the bottom than in the surface waters. The higher surface concentrations of ²³⁹Pu support our suggestion that radionuclides are reaching the lagoon water via leaching processes or surface runoff from the exposed reef.

Table 3. Surface/bottom ratios of radionuclide concentrations in Enewetak Lagoon.

Station Nos. (surface /bottom)	Ratio		
	⁹⁰ Sr	^{239, 240} Pu	¹³⁷ Cs
124/383	1.06	1.33	1.96
366/378	0.95	1.78	1.00
367/374	1.00	1.05	1.04
368/371	0.97	1.27	1.34
106/105	1.57	1.29	1.14
111/107	1.41	1.20	1.36

Distributions in Crater Waters

The concentrations of $^{239,240}\text{Pu}$, ^{238}Pu , ^{137}Cs , and ^{90}Sr in Mike, Koa, Seminole, LaCross and Cactus craters are given in Table 4. The crater locations are described in detail in Ref. 6; however, samples can be located by reference to Fig. 1.

Although Cactus and LaCross craters are adjacent, separated by no more than 200 yards at the northern oceanside reef of Yvonne, Cactus contains higher levels of $^{239,240}\text{Pu}$ in its surface waters, on both the leeward and windward sides, than does LaCross. LaCross crater is continuously awash at high and low tides, whereas Cactus is in communication with the open ocean only at high tide. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios for the two craters are similar, and higher than for any other eastern lagoon sample except one value from station 385 in the Wide Pass. Since no high ratios were observed in the lagoon water in the immediate vicinity of the craters, they must directly contribute little plutonium to the lagoon off Yvonne. Most of the soluble and/or suspended

plutonium originating from the craters must mix into the tidal circulation and be swept eastward to sea on receding tides. However, offshore currents could possibly carry diluted plutonium from the crater area southward; mixture in the lagoon and passage through the eastern half of the wide pass might account for the high ratio at station 385.

The plutonium levels in the surface water of Seminole Crater (on Irene) are higher than in any other crater surface water. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio of 0.31 is also higher than for any surface water sampled in the northwest area. However, higher-than-average values are noted at station 125 and station 6 12, 0.25 miles outside Seminole on the seaward side of Helen. At the latter station the plutonium, which appears to have originated from Seminole, is reduced to 302 fCi/kg by mixing and diluting with ocean water; the $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio is also reduced by dilution, to 0.22. The water at station 125 had a relatively high $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio compared to values at nearby stations,

Table 4. Radionuclide concentrations in unfiltered samples from Enewetak craters. Values in parentheses: one standard deviation based on counting statistics.

Crater	Station No.	Depth ^a (m)	Collection date	Radionuclide concentration (fCi/kg)				$^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio
				$^{239,240}\text{Pu}$	^{238}Pu	^{137}Cs	^{90}Sr	
Mike	82	S	12/12/72	23.4 (2.0)	2.0 (0.4)	730 (20)	590 (6)	0.085
	373	14	12/12/72	71.9 (5.8)	7.0 (1.0)	4220 (40)	48 600 (990)	0.097
	81	28	12/12/72	54.6 (3.8)	1.9 (0.4)	3200 (21)	2 090 (21)	0.035
Koa	127	S	12/11/72	19.0 (0.8)	1.7 (0.2)	1170 (19)	1 180 (70)	0.084
	118	15	12/11/72	26.4 (1.4)	3.2 (0.3)	1100 (17)	1 180 (90)	0.12
	123	33	12/11/72	1510 (60)	236 (9)	8900 (40)	11 200 (110)	0.15
Seminole	611	S	12/15/72	1330 (70) 57 (3)	411 (22)	970 (40)	7 480 (38)	0.31
Lacross	613	S	1/8/73	46 (2)	26 (2)	118 (62)	125 (5)	0.46
	615	S	2/8/73		24 (2)	108 (54)	109 (11)	0.52
Cactus	614	S	2/8/73	185 (7)	98 (3)	935 (46)	209 (11)	0.53
	616	S	2/8/73	105 (9)	52 (5)	302 (57)	182 (7)	0.50

^aS, surface sample.

suggesting that small diluted quantities of radionuclides from Seminole may circulate into the lagoon.

Mike and Koa craters are the only two large craters directly in communication with the ocean and lagoon. At both Mike and Koa, the plutonium is higher in concentration in bottom water than in surface water, and at Mike the middepth sample contains more ^{137}Cs and plutonium than the bottom water. The latter observation suggests that resuspended fine particulates may be concentrated in the crater water column at specific depths. In both Mike and Koa the surface concentrations of ^{137}Cs and ^{90}Sr differ, although the samples were collected at locations less than 400 yards apart. These surface samples were collected at different times within the tidal cycle, but it appears that the input rate of these two radionuclides from the local contributing sources at the craters must be high enough to prevail over the horizontal mixing rates.

There is also poor vertical mixing in the craters as well as mixing between the craters. Note for example that the $^{238}\text{Pu}/^{239,240}\text{Pu}$ value for bottom water is 0.035 for Mike and 0.156 for Koa. For Mike, the plutonium ratios are similar for surface

and mid-depth but differ from that for bottom water. These differences both in ratio and concentration indicate that vertical mixing between surface water and bottom crater water is a slow process.

ADDITIONAL TRANSURANICS

Neptunium-237

The ^{237}Np levels were determined in 14 water samples by mass spectrometry (Table 5). Too few data are available to permit any specific conclusions about neptunium. We do find, however, that the ^{237}Np concentrations in the lagoon and crater water are less than 0.2% of the plutonium concentrations. Interestingly, the $^{237}\text{Np}/^{239,240}\text{Pu}$ ratios were higher in the surface water outside the lagoon, probably indicative of world-wide fallout ratios in oceanic surface waters. However, the ^{237}Pu concentrations were significantly higher in Mike and Koa craters than in any lagoon sample thus far analyzed, suggesting that neptunium and plutonium may differ both in source and in mechanisms of redistribution in the lagoon.

All Transuranics

An entire transuranic inventory has been completed for Sample 123. Table 6 gives

Table 5. Concentrations of ^{237}Np in Enewetak water samples. Values in parentheses, one standard deviation from mean.

	No. of samples	Concentration (fCi/kg)	^{237}Np , % of $^{239,240}\text{Pu}$
Av outside atoll	4	0.013 (0.003)	2.2 (0.9)
Av inside atoll	6	0.058 (0.013)	0.20 (0.03)
Craters (Mike, Koa)	4	0.45 (0.22)	0.14 (0.05)
Mean	14	0.16 (0.03)	0.19 (0.14)
Range		0.01 - 1.4	0.09 - 0.49

Table 6. Transuranics in sample 123 bottom of Koa Crater (108-ft depth).

	Concentration (fCi/kg)
^{237}Np	1.42
^{238}Pu	236
^{239}Pu	1 100
^{240}Pu	474
^{241}Pu	1 800
^{241}Am	346

the levels of ^{237}Np , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{238}Pu , and ^{241}Am in this sample, decay-corrected to date of sampling; ^{241}Pu , ^{240}Pu , and ^{239}Pu concentrations were determined by mass spectrometry. In

order of decreasing concentration, the transuranic radioisotopes found **were** ^{241}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , ^{238}Pu , and ^{237}Np .

The activity ratios $^{240}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Pu}/^{239}\text{Pu}$ for sample 123 are, respectively, 0.432 and 1.64; in sample 365 they are 0.289 and 3.30, respectively. These **values** indicate, as did the $^{238}\text{Pu}/^{239,240}\text{Pu}$ values in the lagoon, that plutonium isotopes are nonuniformly mixed within the lagoon, a finding that could be of considerable value in attempts to define specific sources contributing plutonium to the lagoon environment. No significant number of americium values are yet available for the Enewetak water samples; discussion of this transuranic will be deferred to a later report in this series.

Bikini Lagoon and Surrounding Waters

SAMPLING AND INTERLABORATORY COMPARISONS

The sampling program at Bikini during November 1972 is to be described in the report from the Puerto Rico Nuclear Center. Water, sediment and biological samples were collected by each investigator at **some** but not all of the locations shown in Fig. 11.

In the water-sampling, several objectives were emphasized. First, each laboratory collected duplicate samples at several stations for intercomparison. All samples were collected by pumping; each laboratory processed its samples differently. The PRNC personnel concentrated and processed their samples for

plutonium on board immediately after collection. The University of Washington personnel collected particulate plutonium on 0.3 micron filters and soluble plutonium ion-exchange beds, using the large volume water sampler of Battelle Northwest Laboratory. We collected water in 15-gallon "Deldrums" which were returned to LLL for radiochemical processing.

Each of our samples was acidified to pH 1.5 with HCl. We also collected duplicate 1- μm filtered and unfiltered samples from some stations. These samples were used to compare the fraction of plutonium retained on 1- μm filters to that on 0.4- μm filters and to provide some data for evaluation of in-house analytical precision. Our "Micro-Wynd" 0, ACPY

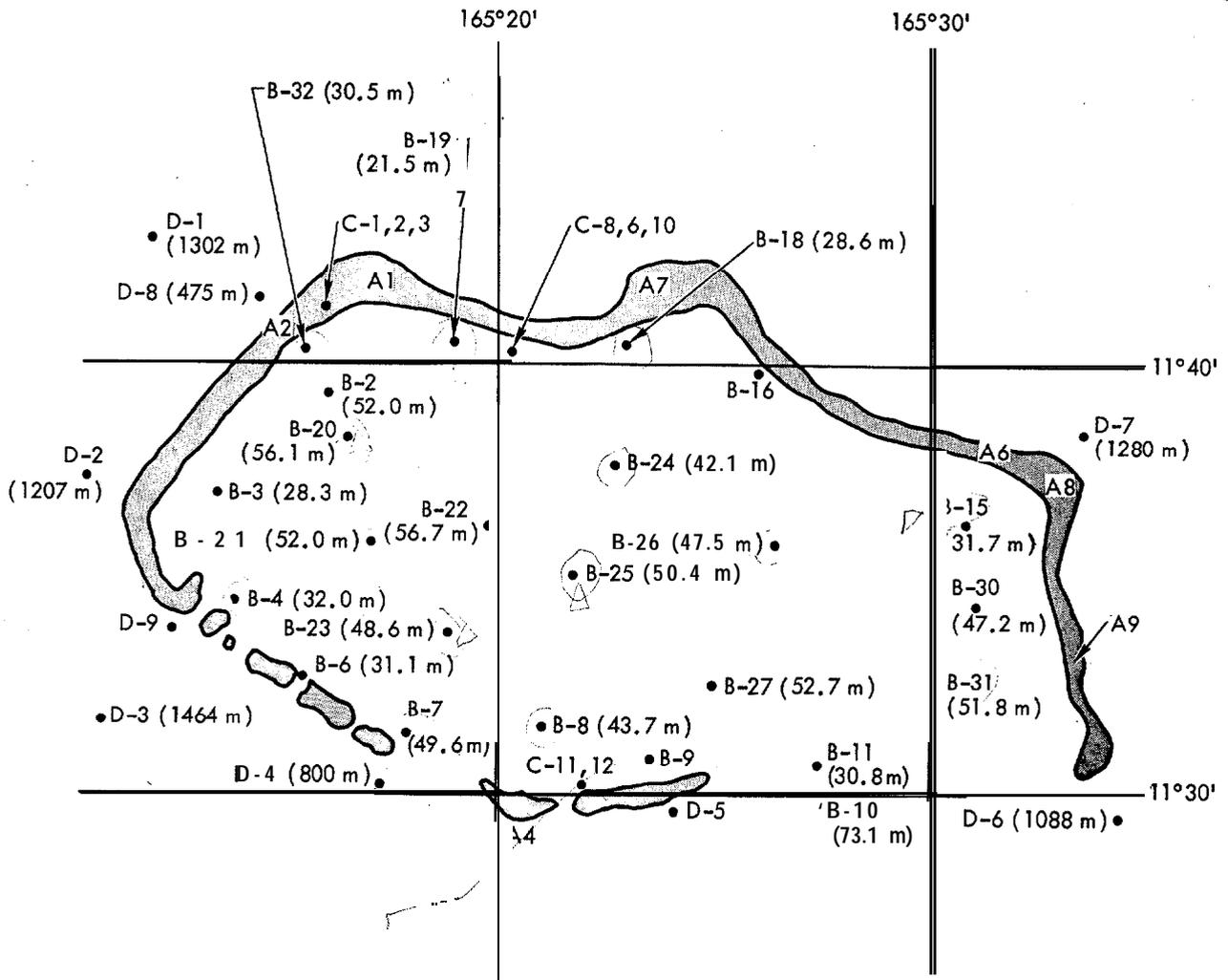


Fig. 11. Bikini lagoon: water-sampling stations. Values in parentheses are bottom depths at station in meters.

filters cartridges were mounted in CFC "Fulflo" containers (Commercial Filter Corporation, Lebanon, Indiana). The usual practice was to collect the filtered 15-gallon sample intermittently while filtering a total volume of water usually exceeding 60 gallons..

A more detailed intercomparison of the analytical results will be made when results from all laboratories are available, but preliminary assessment of the available plutonium data is instructive. Our results for plutonium are shown in Table 7, compared to the available data from

University of Washington (UW) and PRNC. Let us considering first only our results (columns (1), (2), (3), and (4)). First, agreement is good between columns (3) and (4) (duplicate samples); that is, the quantity on the filter plus that in the filtrate is in excellent agreement with the total found in the unfiltered sample. Plutonium bound to particulates in stored water samples is readily exchanged with carriers during processing. The characteristics of the 1- μ m filter are also of interest, since our sampling regime of intermittent collections and the good

Table 7. Laboratory intercomparisons for $^{239,240}\text{Pu}$ analyses. All values are in pCi/m^3 ; values in parentheses are standard deviations.

Station	Depth ^a (m)	LLL				UW			PRNC
		Insoluble ^a (1)	Filtrate from (1) (2)	(1) + (2) (3)	Unfiltered ^b (4)	Insoluble ^c (5)	Filtrate from (5) (6)	(5) + (6) (7)	Unfiltered ^d (8)
B-2	S	28 (2)	79 (3)	107 (4)	93 (6)	30 (2)	40 (4)	69 (4)	98 (7)
	Bottom								
B-15	S	4.7 (0.6)	61 (2)	66 (2)		3.1 (0.2)	30 (5)	33 (5)	49 (4)
	29	5.6 (0.6)	54 (3)	60 (3)	—	6.4 (0.06)	29 (1)	35 (2)	76 (7)
B-19	S		38 (2)			17 (1)	36 (6)	53 (6)	
	19		18 (0.4)			71 (2)	52 (4)	123 (5)	
B-25	S	9.7 (0.9)	69 (3)	79 (3)					67 (4)
	Bottom		29 (2)		64 (3)				127 (9)
B-30	S		49 (2)			2.5 (0.3)	26 (3)	29 (3)	55 (3)
	45		53 (1)			29 (1)	31 (3)	60 (3)	81 (2)
C-3	S	10 (1)	28 (1)	38 (1)	34 (1)	13.6 (0.3)	49 (2)	62 (2)	32 (1)
	22	14.2 (0.6)	17.2 (0.4)	31 (1)	32 (1)				
	44	22 (1)	13 (2)	45 (2)	44 (1)	24 (2)	7 (2)	31 (3)	
C-8	S				47 (4)	14.6 (0.6)	34 (8)	48 (8)	68 (3)
	34				75 (3)	35 (1)	22 (6)	57 (6)	66 (2)
D-1	300				51 (6)				5 (1)
D-7	S				3.5 (0.2)	0.13 (0.06)	3.27 (1.27)	3.4 (1.2)	

^aRetained on 1- μm prefilter corrected for volume filtered.

^bSeparate unfiltered sample taken immediately before or after the filtered sample.

^cRetained on 0.3- μm Millipore filter.

^dCompare with columns (3), (4), and (7).

agreement between filtered and unfiltered samples shows that larger volumes of water than were collected can be effectively filtered for suspended plutonium without breakthrough or obvious loss from the cartridge.

The results from LLL and the University of Washington for particulates (Table 7, columns (1) and (5)) agree very well, indicating that little if any plutonium is associated with particles larger than 0.3 μm but smaller than 1 μm , assuming the 1 μm filter is efficient only in removing particles greater than 1 μm in size. For the lagoon samples, however, there are considerable differences between LLL and the University of Washington in the results for the soluble fractions and between PRNC and the University of Washington in the total values; surprisingly, agreement is much better in both cases for crater water

and for samples collected outside the lagoon. The PRNC total (column 8) and the LLL total (column 3 or 4) appear to agree reasonably well except for the values at stations B-25 bottom and D-1 at 300 m. Some of the disagreement may be attributable to the short sample-collection period for LLL and PRNC samples compared to that required by the University of Washington collection technique. That is, 'some of the discrepancies could be due to concentration changes with time, as at Enewetak where tide-controlled currents greatly influence the concentration levels at any location in the lagoon over a period of time. We are, as well, still investigating other possible sources of error.

General Radionuclide Inventory

Tables 8 and 9 present our available results for $^{239,240}\text{Pu}$, ^{238}Pu , ^{137}Cs , and

Table 8. Radionuclide concentrations in Bikini lagoon and crater waters; filtered through 1- μ m fiber cartridge except where otherwise stated.

Station	Depth ^a (m)	Collection date	Radionuclide concentration (fCi/kg)			
			^{239,240} Pu	²³⁸ Pu	¹³⁷ Cs	⁹⁰ Sr
B-2 ^b	S	11/6/72	93 (6)	1.9 (0.6)	1 040 (40)	
B-2	S	11/6/72	79 (3)	1.0 (0.2)	800 (20)	
B-4 ^b	S	11/6/72	Lost	Lost	660 (30)	
B-4	S	11/6/72	Lost	Lost	580 (30)	
B-7	S	11/6/72	36 (1)	0.76 (0.15)	420 (30)	
B-9	S	11/2/72	6.9 (0.9)	0.1 (0.1)	150 (20)	
B-10	S	11/2/72	3.9 (0.3)	0.04 (0.3)	160 (30)	
B-15	S	10/31/72	61 (2)	0.9 (0.1)	670 (20)	
B-15	15	10/31/72	58 (3)	1.2 (0.3)	630 (30)	
B-19	S	11/8/72	38 (2)	1.0 (0.3)	810 (30)	
B-19	19	11/8/72	18 (0.4)	0.2 (0.05)	470 (20)	
B-20	S	11/8/72	38 (2)	0.8 (0.2)	1 020 (20)	756 (19)
B-20	54	11/8/72	50 (1)	0.85 (0.13)	2 640 (50)	1 730 (35)
B-23	S	11/6/72	20 (1)	0.4 (0.2)	450 (25)	351 (14)
B-23	46	11/6/72	8.6 (0.6)	0.3 (0.1)	240 (60)	
B-25	S	11/5/72	69 (3)	n.d. ^e	760 (30)	630 (19)
B-25	48	11/5/72	29 (2)	0.4 5 (0.23)	340 (30)	315 (13)
B-25 ^b	48	11/5/72	64 (3)	1.1 (0.2)	470 (30)	395 (14)
B-30	S	10/31/72	49 (2)	0.52 (0.11)	690 (30)	581 (20)
B-30	45	10/31/72	53 (1)	1.3 (0.2)	570 (20)	491 (20)
C-3 ^c	S	11/12/72	28 (1)	0.8 (0.1)		
C-3 ^{b,c}	S	11/12/72	34 (1)	1.1 (0.1)	850 (30)	
C-3 ^c	22	11/12/72	17.2 (0.4)	0.69 (0.07)	790 (25)	
C-3 ^{b,c}	22	11/12/72	32 (1)	1.0 (0.1)	810 (20)	
C-3 ^c	44	11/12/72	13 (2)	0.4 (0.4)	990 (30)	
C-3 ^{b,c}	44	11/12/72	40 (1)	1.2 (0.1)	780 (30)	
C-8 ^{b,d}	S	11/12/72	47 (4)	0.8 (0.6)	560 (20)	
C-8 ^{b,d}	17	11/12/72	Lost	Lost	620 (25)	
C-8 ^{b,d}	34	11/12/72	75 (3)	1.0 (0.2)	1 230 (90)	

^aS, surface sample.

^bUnfiltered sample.

^cBravo Crater.

^dTewa Crater

^en. d., below detection limits.

Table 9. $^{239,240}\text{Pu}$ concentrations (fCi/kg) of insoluble fraction collected on 1- μm filters for Bikini lagoon and craters. Values in parentheses: one standard deviation based on counting statistics.

Station	Depth ^a (m)	$^{239,240}\text{Pu}$ (fCi/kg)	% of total ^b
B-2	S	28 (2)	26
B-15	S	4.7 (0.6)	7
	29	5.6 (0.6)	9
B-25	S	9.7 (0.9)	12
	48	7.1 (0.4)	20
c-3	S	10 (1)	26
	22	14.2	45
	44	22 (1)	63
B-7	S	1.3	3
B-10	S	0.13 (0.02)	3
B-19	S	17.3 (0.7)	31
	19	42 (1.4)	70
B-20	S	42 (2)	53
	54	460 (70)	90
B-30	S	2.9 (0.1)	6
	45	13.4 (0.4)	20
B-9	S	1.4 (0.1)	17
B-23	S	8.2 (0.3)	49
	46	8.4 (0.4)	30

^aS, surface sample.

^bPercentage of total activity on particulates.

^{90}Sr at stations in the lagoon and in craters (refer to Fig. 11 for station location). The average concentration of $^{239,240}\text{Pu}$ is 40 ± 25 fCi/kg in 10 filtered lagoon surface-water samples and 35 ± 19 fCi/kg for 6 bottom-water samples. In the surface waters of the lagoon, the average amount of plutonium associated with particulate material is $30 \pm 25\%$ of the total. From these values, an overall lagoon average of approximately 49 fCi/kg can be computed. This is slightly but not significantly higher than the average concentra-

tion of $^{239,240}\text{Pu}$ in Enewetak lagoon. The average concentration of ^{238}Pu (0.7 fCi/kg) is, however, roughly one-seventh the average at Enewetak. This difference is sufficiently great to indicate that Enewetak is a greater source of ^{238}Pu than Bikini but the water columns of the two lagoons contain essentially equal quantities of $^{239,240}\text{Pu}$.

The average concentration of ^{137}Cs at Bikini, including an average 16% on particulates, is 712 fCi/kg, double the Enewetak average. The average $^{137}\text{Cs}/$

^{90}Sr ratio in the water at 8 stations is 1.25 ± 0.14 . Assuming that this ratio is constant over the lagoon, the average ^{90}Sr concentration is 570 fCi/kg. The concentration of ^{90}Sr also is higher at Bikini than at Enewetak. Comparisons of these concentrations with possible values for world wide fallout, discussed in a previous section, indicates that the Atoll contributes radionuclides to the lagoon. The inventories of ^{90}Sr , ^{137}Cs and plutonium radionuclides in Bikini and Enewetak lagoons are summarized in Table 10.

The average fractions in the suspended phase are similar for $^{239,240}\text{Pu}$ (30%) and ^{137}Cs (16%). Because of the different sorption characteristics of these elements, it can be concluded that withdrawal by particulates is not the mechanism whereby these radionuclides become bound to the suspended phase. Rather the particulate phase must be made up of small fragments of coral or other organic materials arising from either beach erosion or upwelling.

PLUTONIUM, ^{137}Cs , AND ^{90}Sr

Distributions in Surface Water

A complete area1 description of the radionuclide concentrations in surface water in Bikini lagoon awaits collation of the data from the three participating laboratories. Our own presently available values for filtered surface water are plotted on lagoon charts (Fig. 12) with isoactivity contours added. The highest plutonium and cesium concentrations are seen in the northwest sector. Strong gradients are evident, consistent with the presence of different water masses. Average wind and wave directions during sampling (Fig. 13) were abstracted from the bridge log of the RV Palumbo. The surface circulation is shown to be strongly influenced by the prevailing wind; the general pattern is circulation east to west with water displaced over the western reef and through the southwest passes. Off the western reef, water movement parallels the reef, Major exchange of ocean water

Table 10. Summary of radionuclide concentrations in Enewetak and Bikini lagoons, Fall of 1972. Values in parentheses are possible fallout levels in the lagoon, determined by "replacing" lagoon water with North equatorial surface water.

	$^{239,240}\text{Pu}$	^{238}Pu	^{137}Cs	^{90}Sr
Av. concentration (fCi/kg)				
Enewetak	39	4.7	317	330
Bikini	49	0.7	712	570
N. equatorial	0.4	0.06	155	71
Water column inventory (mCi/km ²)				
Enewetak (47.4 m depth)	1.84 (0.02)	0.22 (0.003)	15.0 (7.3)	15.6 (3.4)
Bikini (45.6 m depth)	2.23 (0.02)	0.03 (0.003)	32.5 (7.1)	25.9 (3.2)
10°-20°N (total fallout) ^a	0.24	0.009	12.0	8.0
Lagoon inventory (Ci)				
Enewetak (933 km ²)	1.7 (0.02)	0.20	14.0 (6.8)	14.6 (3.1)
Bikini (630 km ²)	1.4 (0.01)	0.02	20.4 (4.5)	16.3 (2.0)

^aDeposition to 10-20° latitude band.⁴

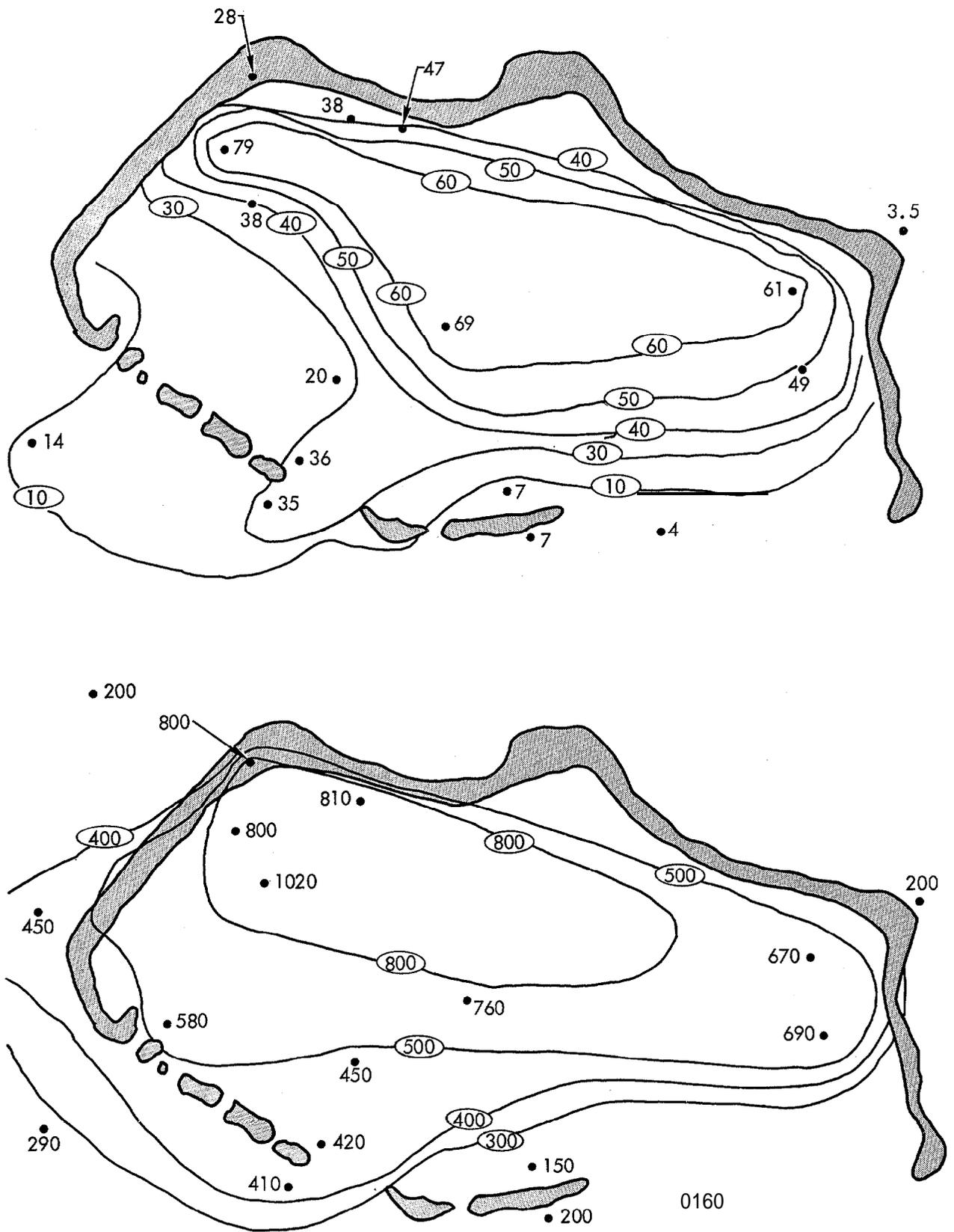


Fig. 12. Bikini lagoon: surface concentrations and isolines of $^{239,240}\text{Pu}$ (upper) and ^{137}Cs (lower). All values are in fCi/kg.

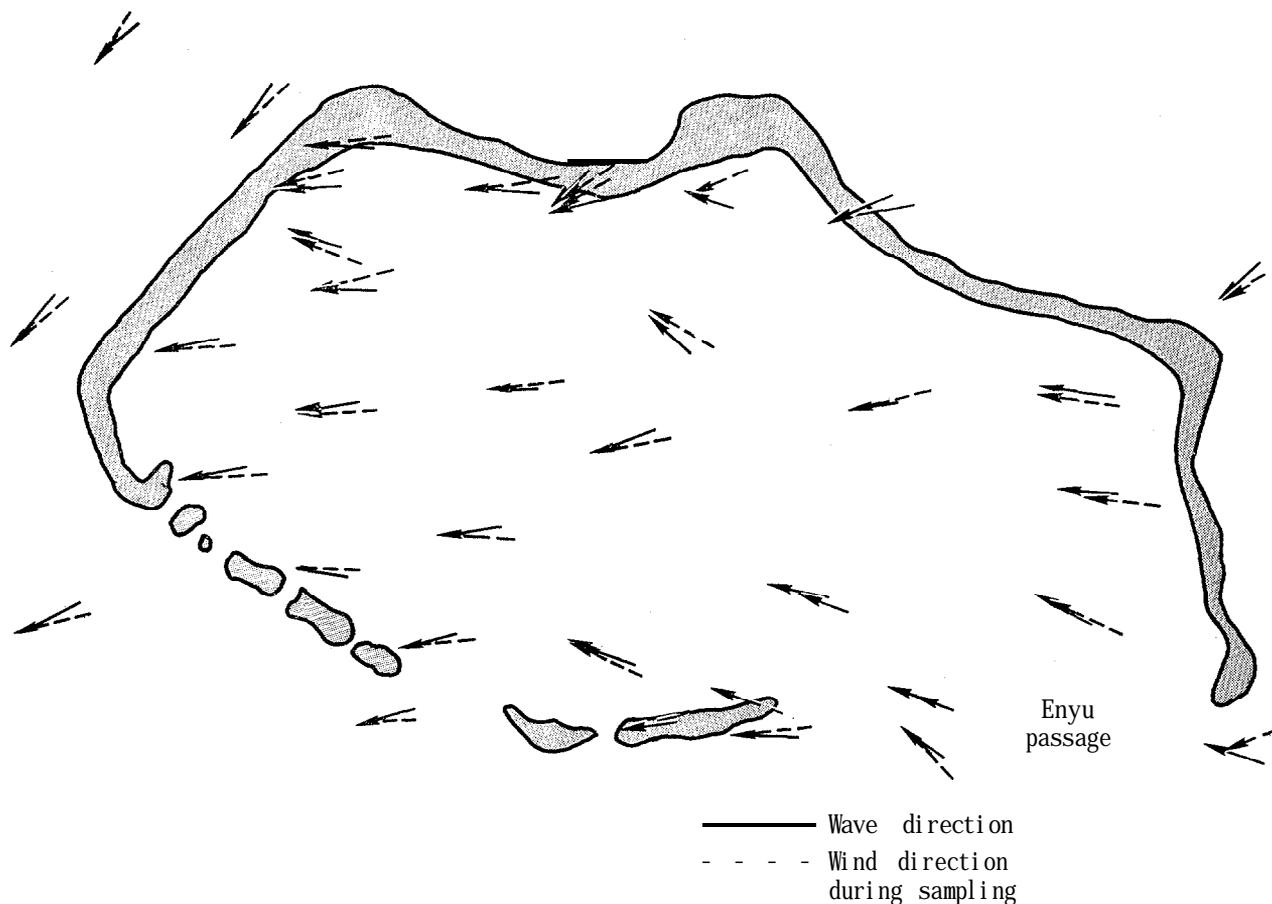


Fig. 13. Bikini lagoon: wind and wave directions measured during sampling program.

must occur by intrusion through the Enyu passage as well as over the eastern and northern reef. This system has the general features of the pattern described by Ford² for the post-Baker days during mid-July 1946. It is indeed surprising that a similar circulation pattern persists in November, when wind and wave conditions are much more active than during the doldrum season. This suggested circulation is supported by our radionuclide data, however. The concentrations in the Enyu pass are similar to those in ocean water east of the Atoll, and the isoactivity lines define the extent of influence of exchanging ocean water in the lagoon. This passage

is of major importance for the exchange of ocean water into lagoon during this period.

Over the surface area of the lagoon, we find changing plutonium/cesium ratios (Fig. 14); they are highest in the central region of the lagoon and range to lower values both to the east and west. The gradients, both for the concentrations (Fig. 12) and for the ratios (Fig. 14) show that the area of most rapid mixing is in the southeast. Both ratios and concentrations decrease relatively little toward the northern and western sectors of the lagoon; the influx of water over the reefs in these regions of the atoll is less significant than

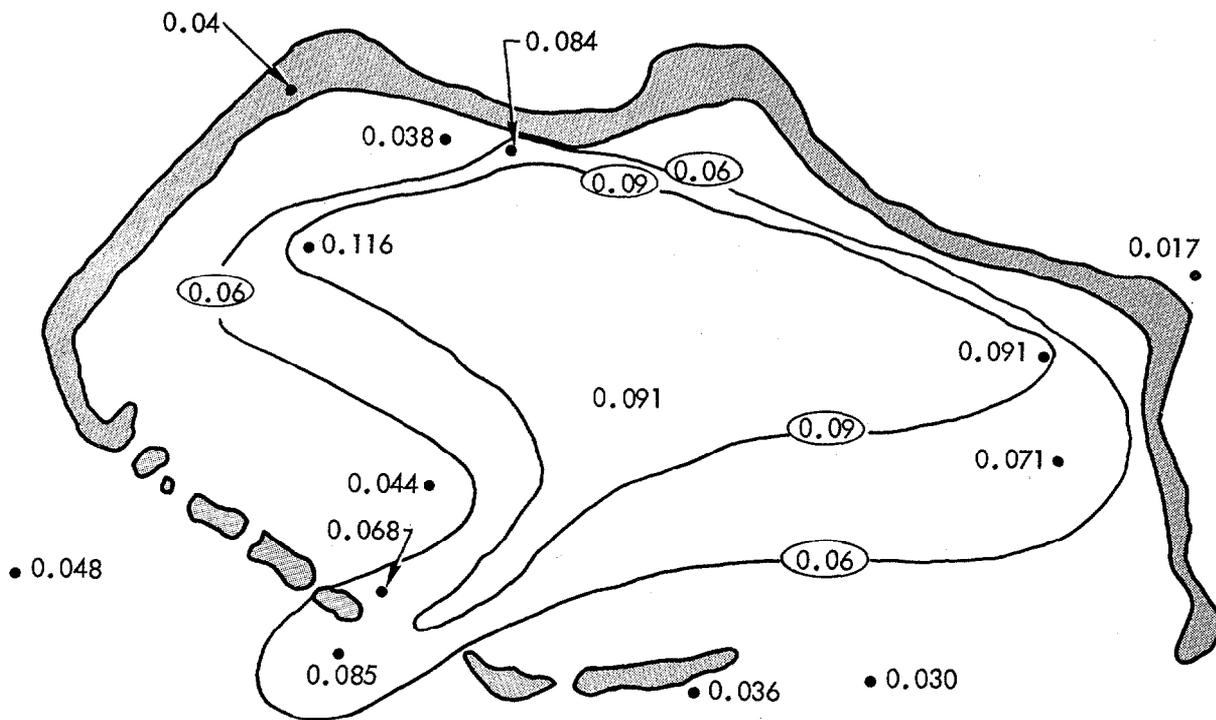


Fig. 14. Bikini lagoon: $^{239,240}\text{Pu}/^{137}\text{Cs}$ concentration ratios and isolines.

the exchange through Enyu channel. In the western lagoon we find high concentrations with low $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios, suggesting that these radionuclides are being contributed to the water column by different sources over the atoll.

The results of the 1964 survey¹² included data reported for ^{90}Sr and ^{137}Cs in one "midlagoon" bottom sample: 420 fCi/kg and 370 fCi/kg for ^{90}Sr and 12 000 fCi/kg and 9 400 fCi/kg for ^{137}Cs , in unfiltered and filtered water, respectively. Compare these values with our "midlagoon" (station B -2 5) bottom values of 395 and 315 fCi/kg for ^{90}Sr and 470 fCi/kg and 340 fCi/kg for ^{137}Cs (unfiltered, filtered); ^{137}Cs levels have decreased significantly in the lagoon since 1964 but ^{90}Sr concentrations are essentially unchanged after correction for decay. The rate of input into the soluble phase must

be equivalent to the rate of loss. This suggests that ^{90}Sr may be continuously recycled in the lagoon by exchange with and solution of carbonate materials, whereas ^{137}Cs , in less critical demand in the carbonate environment, is constantly depleted from the lagoon.

Distributions in Water Outside the Lagoon

The locations of stations sampled outside the outer lagoon perimeter are given in Fig. 11. Our available radionuclide data for these stations are listed in Table 11. Some ^{137}Cs vertical profiles are plotted in Fig. 15.

Recall that only 12 mCi of ^{137}Cs from world-wide fallout was deposited per square kilometer in these latitudes. But only at one station (D-7, NE of Bikini Island) is this quantity found to the depth sampled. At the three stations west of

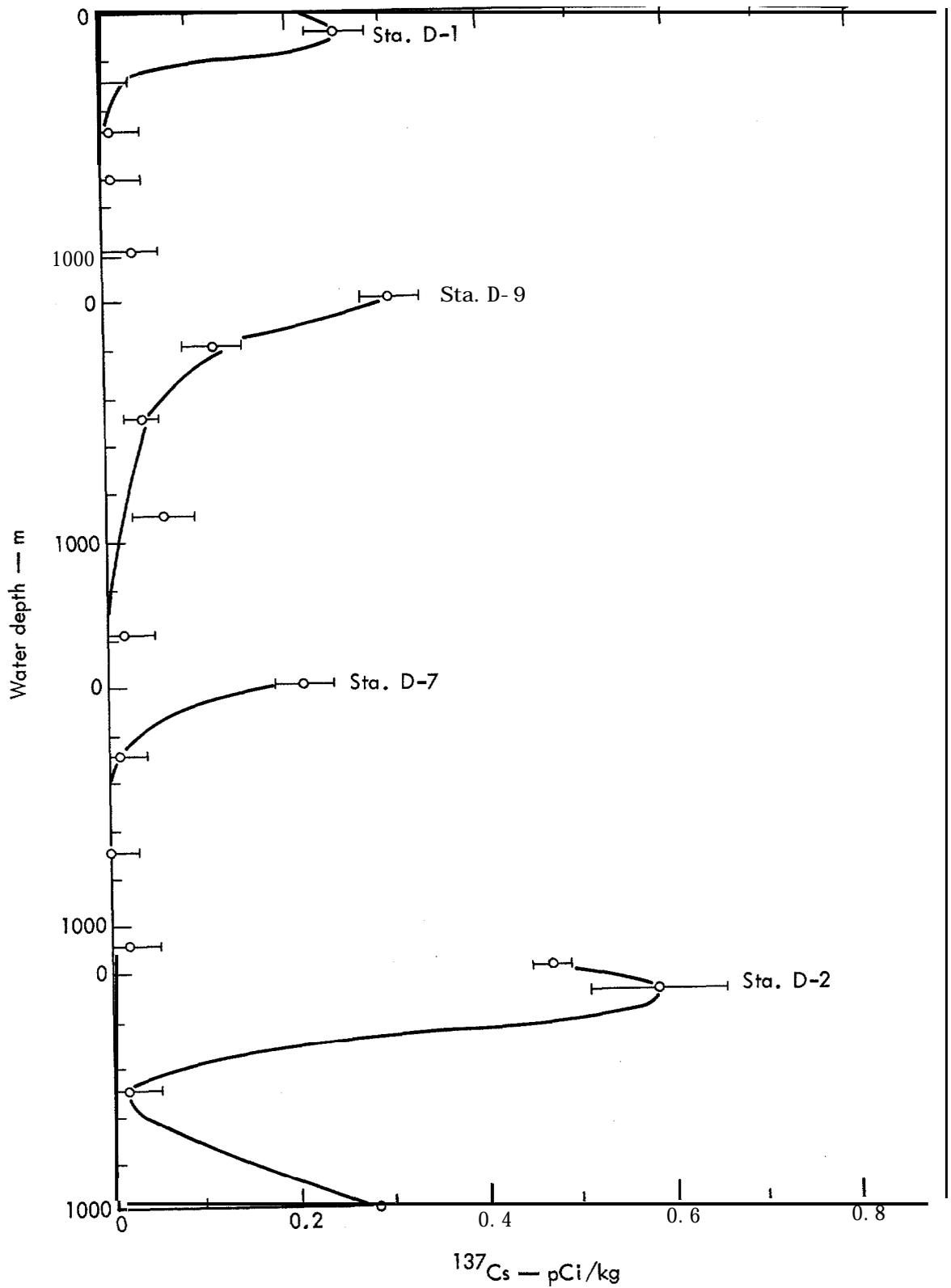


Fig. 15. The ^{137}Cs concentrations (pCi/kg) in the water column at stations outside Bikini lagoon.

Table 11. Total radionuclide concentrations (in fCi/kg, all samples unfiltered) in samples taken outside the outer perimeter of Bikini lagoon.

Station	Depth ^a (m)	Collect ion date	^{239, 240} Pu	²³⁸ Pu	¹³⁷ Cs
D-1		11/12/72			200 (20)
	100	11/12/72			240 (30)
	300	11/12/72	51 (6)		0 (30)
	500	11/12/72	6.7 (1.4)		10 (25)
	700	11/12/72			10 (30)
	1000	11/12/72			30 (30)
D-2		11/12/72			450 (20)
	100	11/12/72			560 (70)
	500	11/12/72			20 (30)
	1000	11/12/72			270 (25)
D-3		11/12/72	14 (4)		290 (30)
	200	11/12/72	1.3 (0.5)		110 (30)
	500	11/12/72			35 (20)
	900	11/12/72			60 (30)
	1400		5.0 (1.1)		20 (30)
D-4		11/11/72	35 (4)	0.7 (0.7)	410 (40)
	350	11/11/72	1.0 (0.3)		0 (30)
	700	11/11/72	1.2 (0.4)		0 (30)
D-5		11/9/72	7.3 (0.3)	0.07 (0.03)	200 (20)
D-7		11/10/72	3.5 (0.2)	0.14 (0.06)	200 (30)
	300	11/10/72	0.2 (0.2)		10 (25)
	700	11/10/72	1.6 (0.8)	-	5 (20)
	1100	11/10/72	39 (1)	0.29 (0.10)	25 (30)

^aS, surface sample.

the Atoll the values are 60 to 220 mCi/km² to the depth sampled; the additional quantities result from continuous advective processes transporting ¹³⁷Cs-burdened water from the Atoll to the open ocean. Assessment of surface concentrations in the outer perimeter samples alone is insufficient to distinguish the areas receiving ¹³⁷Cs from the lagoon. The ¹³⁷Cs concentration

in the surface water at station D-1, (NW of the Atoll) is identical to that at station D-7. The integrated quantity of ¹³⁷Cs at station D-7 is equivalent to fallout levels for these latitudes, whereas both ¹³⁷Cs at station D-1 and ^{239,240}Pu at D-7 exceed them. At D-7, the concentration of ^{239,240}Pu falls from the surface value to near zero at 300 m, then increases

significantly at 700 m. We conclude that tidal processes scouring the eastern reef contribute either soluble or particulate plutonium by mechanisms that are independent of the processes controlling the redistribution of ^{137}Cs .

The total $^{239,240}\text{Pu}$ in the water column to a depth of 700 m at station D-4 is approximately 5.6 mCi/km^2 , assuming a smooth concentration gradient from each sampled depth to the next. This quantity is 23 times as high as the expected fallout level and

2.5 times the water column inventory of the lagoon. At depths in waters from station D-1, D-3 and D-4, $^{239,240}\text{Pu}$ is greatly enriched over ^{137}Cs . These quite unusual profiles show considerable separation of plutonium and cesium in the water masses surrounding the lagoon. We cannot rule out the possibility of subsurface fluxes of plutonium — possibly from sediments expelled during the nuclear test series and now residing on the outer slopes of the Atoll.

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