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Transuranics and other radionuclides in Bikini Lagoon: Concentration data retrieved from aged coral sections[1]

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Abstract

X-radiography and autoradiography of thin vertical sections were used to estimate the growth rate of a specimen of *Favites virens* from Bikini Lagoon. Discrete bands of radioactivity were identifiable with specific nuclear test series. The coral growth rate of 8.0 mm year^{-1} determined by autoradiography is in good agreement with the rate of 8.1 $\pm$ 2.2 mm year^{-1} derived from the "seasonal" alternating light and dark bands on X-radiographs. With these bands as growth rate indicators, the coral was sectioned into yearly increments and analyzed by low-level, nondestructive gamma spectrometry, radiochemical techniques, and mass spectrometry to reconstruct the variations in the concentration of transuranics and other radionuclides in the marine environment at Bikini since 1954. From the concentration data retained in this indicator species, the exchange rate of radionuclides between the lagoon and the open ocean is computed to be longer than exchange rates based on physical circulation data. There is no constant ratio of plutonium isotopes in the coral growth sections, suggesting that the redistributions of the several plutonium isotopes in the environment may be governed by different biogeochemical processes. Increased levels of ^{210}Po (^{210}Pb) were found in test-year growth sections, contradicting previous arguments that no ^{210}Pb has resulted from weapons testing.

Knutson et al. (1972), Knutson and Buddemeier (1973), and Buddemeier et al. (1974) demonstrated that massive coral colonies from Enewetak Atoll contain radioactive growth inclusions, detectable by autoradiography, that can be correlated with the annual density-banding evident in X-rayed sample sections. Results from these and other samples (Buddemeier et al. 1974) show periodicity in growth, interpreted as density variations in the $CaCO_3$ skeleton, that can be used to estimate growth rates of massive coral colonies. Other recent applications, using natural occurring radionuclides (Dodge and Thomson 1974; Moore and Krishnaswami 1972), have also used radiometric techniques to determine coral growth. Radiometric studies (Dodge and Thomson 1974) have substantiated the relationship between annual growth-band composition and the changing environment in which the coral grows. Previous investigations of the growth rates of fossil and reef coral have been adequately reviewed (Buddemeier et al. 1974; Dodge and Thomson 1974).

Our studies began in an attempt to reconstruct changes in transuranium concentrations in the marine environment of the Pacific test sites through observations of the yearly response of the coral to environmental changes. Although several radiological surveys have been made at the Pacific test sites, no chronological information is available on the behavior of specific radionuclides in these environments. The radiological records retained in the closed aragonite structure of coral could circumvent the need for years of real-time data and provide insight into the behavior of these radioelements in the lagoon. Knutson and Buddemeier (1973) found ^{90}Sr in coral from Enewetak and concluded, on the basis of the observed differential changes in concentration, that the lagoon community acts as a long term source of ^{90}Sr, probably by exchange with and solution of carbonate materials deposited during the period of nuclear testing. It is not clear that a single mechanism can explain all environmental radiological information retained by coral colonies; we need infor-

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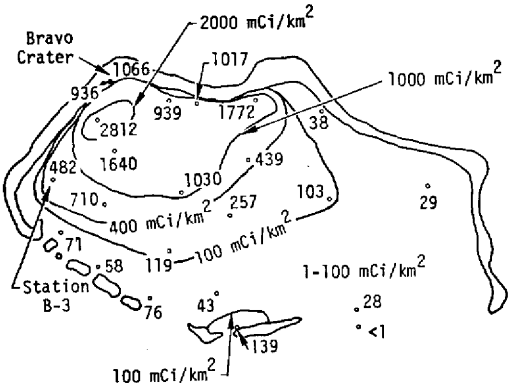

Fig. 1. Bikini Atoll—location of sampling station and Bravo Crater. Isoactivity lines of ^{241}Am (mCi km^{-2}) in the surface (2.5 cm) sediment of Bikini Lagoon.

mation on other possible mechanisms controlling the redistribution of specific radionuclides, principally the transuranium elements.

We prepared autoradiographs and X-radiographs from vertical slices cut from the center of a sample of *Favites virens* collected from Bikini Atoll. Autoradiography revealed three discrete, relatively intense, radioactive bands within the skeletal structure identifiable with the nuclear test series of 1954, 1956, and 1958. Fourteen density bands between the outermost radioactive band (1958) and the coral surface (1972) confirmed the thesis that these are correlated with annual growth. Sections from each annual growth band were analyzed by low-level gamma spectrometry and radiochemical techniques to reconstruct the chronological history of specific radionuclides incorporated in the coral skeleton. Much, if not all, of the observed artificial radioactivity in the coral at Enewetak (Knutson and Buddemeier 1973) was due to ^{90}Sr and its daughter ^{90}Y, but we identified other radionuclides in all growth sections of this Bikini coral (e.g. ^{241}Am, ^{90}Sr, ^{155}Eu, ^{207}Bi, ^{60}Co, ^{210}Po, $^{238,239,240,241}Pu$). We believe that the record recovered in these yearly growth increments is one of the most complete histories of changes in concentration of specific radionuclides in the marine environment of Bikini Atoll. From these data we explain the relative behavior of some specific radionuclides in Bikini Atoll.

This work was part of a cooperative program to investigate the biogeochemical behavior of the transuranium elements at Bikini Atoll. Samples were collected in November 1972 from the RV *Palumbo*, operated by the Puerto Rico Nuclear Center (PRNC). We acknowledge the help provided by the crew of the RV *Palumbo* and specifically thank F. Lowman and W. Schell, cooperative participating investigators.

Sectioning methods and growth results

The coral used in this study was a live specimen of *F. virens* dredged in November 1972 from a depth of 28 m at station B-3, Bikini Atoll (Fig. 1). In the periods of June–July 1946, February–May 1954, May–July 1956, and May–July 1958, Bikini Atoll was the site of a series of nuclear device tests. Many of the events in the last three series were conducted along the inner northern and southern perimeter of the lagoon. The largest test on the reef (1954: 15 megatons) (Telegadas 1961) produced Bravo Crater, also shown in Fig. 1.

Table 1. Concentration of ^{241}Am, ^{155}Eu, ^{60}Co, and ^{207}Bi in the sediment column from location B-3; water depth, 28.3 m; collection date, 8 November 1972; core diameter, 36.3 cm².

Section Thickness (cm)	pCi/g dry ± (error in % of value)*			
	^{241}Am	^{155}Eu	^{60}Co	^{207}Bi
	Coarse fraction > 0.5 mm			
0–5	7.10±3	9.22±2	3.48±4	1.16±10
5–10	5.17±3	6.13±2	4.02±4	2.64±6
10–15	5.93±3	7.01±1	3.89±6	3.67±4
15–20	2.41±5	2.66±4	1.71±6	2.78±4
20–25	0.89±10	1.19±4	0.67±9	0.96±5
25–30	< 0.1	0.15±24	0.29±18	0.39±16
30–35	< 0.1	0.13±30	0.20±25	0.38±16
	Fine fraction < 0.5 mm			
0–5	8.37±2	13.4 ±1	5.06±2	1.38±8
5–10	11.1 ±2	14.8 ±2	6.83±3	2.31±6
10–15	20.3 ±2	22.4 ±1	7.45±3	11.82±2
15–20	15.7 ±2	17.3 ±2	4.78±5	12.41±3
20–25	8.26±3	8.73±2	2.00±6	3.62±5
25–30	1.73±6	2.09±4	0.89±9	1.14±8
30–35	1.58±6	1.89±4	0.60±12	0.68±10

*Based on counting error only.

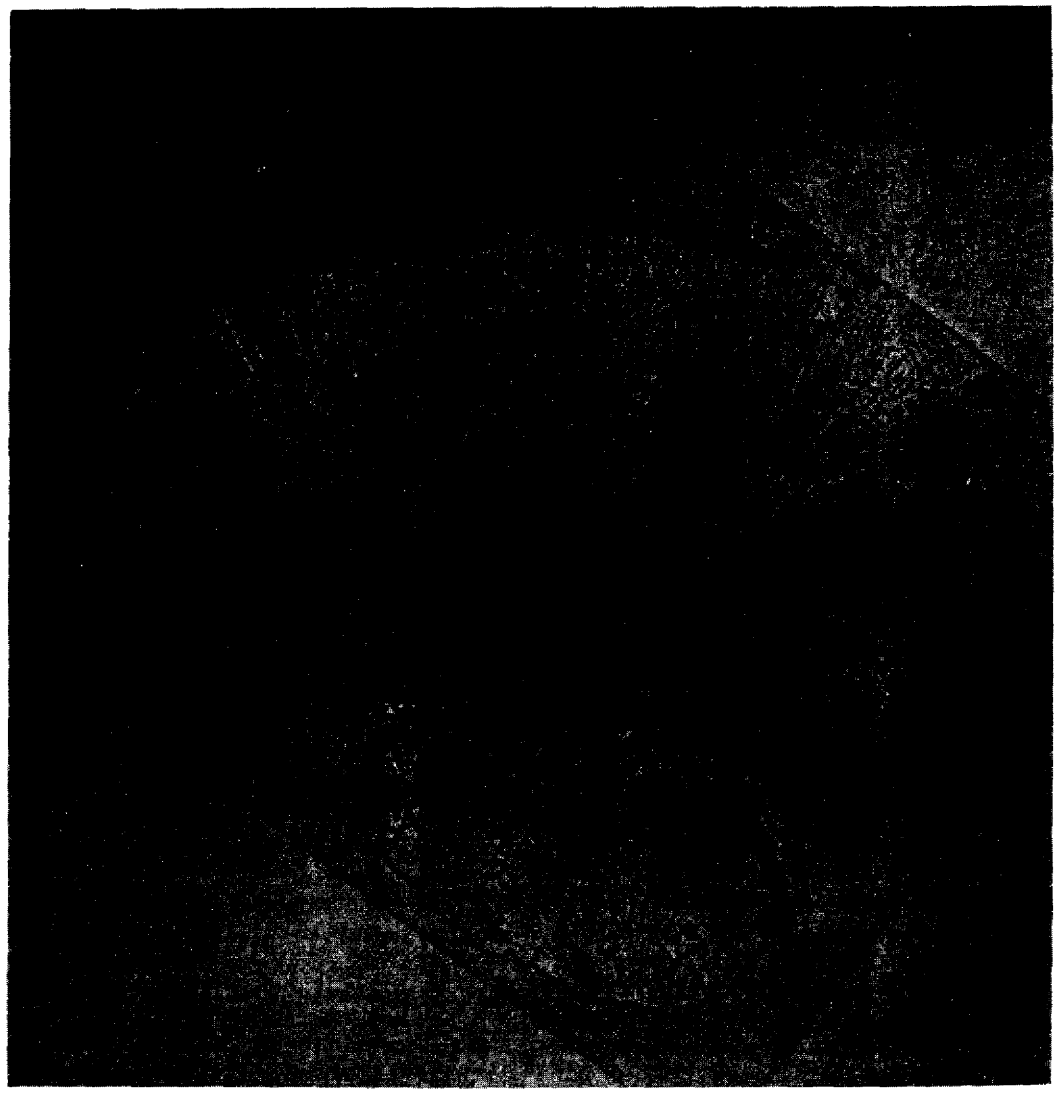

Fig. 2. Autoradiograph of a cut vertical section from the coral sample. Radioactive regions have been retouched for better contrast.

The distributions of specific radionuclides in the lagoon are extremely heterogeneous (Noshkin et al. 1974). For example, the 1972 activity levels and isoactivity lines of ^{241}Am in the surface 2.5 cm of sediment deposit (Fig. 1) show that the region around station B-3 was heavily contaminated with artificial radionuclides from the test series. Table 1 gives the levels of several radionuclides to a depth of 35 cm in a sediment core from station B-3. Levels of radioactivity are relatively high throughout the column. The coral from this location should reflect the changes in activity during its growth cycle in this contaminated region of the lagoon.

The coral surface was washed and the specimen was dried at 110°C for 1 week. The sample was first cut in half vertically from the surface through the base. (We always cut from presumably low-radioactivity sections toward higher level sections to

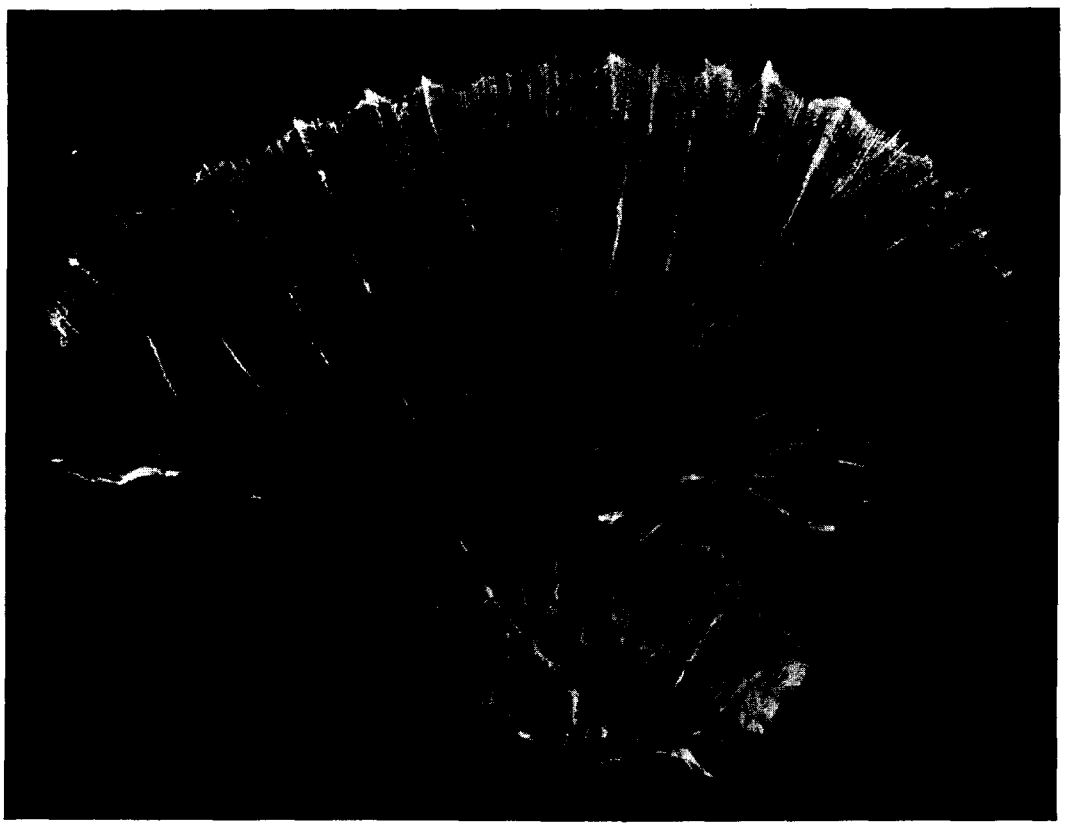

Fig. 3. Demonstration X-radiograph of a coral section. Annual band boundaries (not all necessarily evident on this exposure) are identified. Wedge outlines the section removed for radionuclide analysis.

avoid possible contamination.) Vertical subsections 2 cm thick were cut from the facing coral halves. One slice, chosen for autoradiography, was placed in direct contact with No-screen-X-ray film for 60 days; the adjacent slice was subjected to X-radiography, with several exposures at different energies.

Regions of the developed autoradiography film revealed inclusions of radioactivity (Fig. 2: these bands are darkened for better contrast). Three distinct bands near the base of the coral can be identified with the test series years 1954, 1956, and 1958; they are 15, 14, and 12.5 cm from the coral surface. These measurements gave average growth rates of 0.81, 0.85, and 0.87 cm yr^{-1} between 1954, 1956, 1958, respectively, and 1972. The distance between the 1954 and 1958 bands, however, is only 2.5 cm, yielding an average growth rate during this period of only 0.6 cm yr^{-1}. The annual skeletal layers are not of constant thickness; in assigning the annual boundaries to post-test growth, we relied on the density variations in the exposed X-ray film.

Figure 3 shows one exposed X-ray film of the coral section. Careful examination reveals alternating light and dark bands in the skeletal matrix. Several exposures were examined before a complete selection of annual band "boundaries" could be made.

Buddemeier et al. (1974) concluded that the low-density band (dark X-ray negative) is correlated with a growth period beginning around December or January and ending around July. In our sample, the three bands in the autoradiograph of

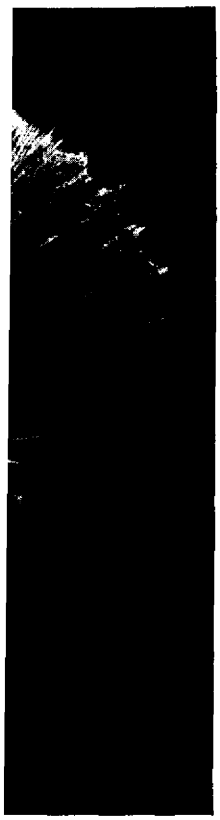

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Table 2. Coral section data.

Growth section from outer edge (mm)	Estimated year of growth	Δmm growth/ year	Δt in years from Nov 72 (collection date)
0-8	1971-1972	4	0.9
8-15	1970	7	2.4
15-21	1969	6	3.4
21-29	1968	8	4.4
29-38	1967	9	5.4
38-46	1966	8	6.4
46-56	1965	10	7.4
56-65	1964	9	8.4
65-75	1963	10	9.4
75-83	1962	8	10.4
83-96	1961	13	11.4
96-108	1960	12	12.4
108-116	1959	8	13.4
116-123	1958	7	14.4
123-129	1957	6	15.4
129-137	1956	8	16.4
137-147	1954-1955	5	17.9
147-156	1953	9	18.4

the Bikini coral are either associated with the low-density bands of the X-ray negative or close to the transition zone between light and dark bands. By correlation of the bands shown by both the autoradiograph and X-ray exposures, the years 1956 through 1958 were identified. The 1954 growth, however, could not be resolved from the 1955 growth on the X-ray negative. From the 1958 band to the coral surface (1972 growth), 14 alternating light and dark areas were identified (Fig. 3). Table 2 gives the thickness of each annual section along with the estimated year of growth.

The varying dimensions of the yearly increments show that growth rates vary from year to year and also that the dimensions follow no predictable trend with time. The mean annual growth from the X-radiograph is 8.1 ± 2.2 mm yr^{-1}, a value in agreement with the autoradiography results. No activity from the 1946 test was detected because it predated the earliest growth of this particular coral.

A wedge, containing the area with the more nearly linear growth record, was removed from the center of the slice (*see wedge outline, Fig. 3*). A bandsaw was used to remove each defined annual growth band; at least 1 mm of material was lost at each cut, so that the coral record between each two sections was lost. Each section was then ground and homogenized in a ball mill and a known weight was transferred to a vial for radionuclide analysis by gamma spectrometry. A low-background, Compton-suppressed, Ge(Li) detector system was used for some of the gamma-emitting radionuclides. The samples were then processed for Pu, ^{90}Sr, and ^{210}Po by chemical separation followed by radioassay with low-background beta detectors and alpha spectrometry. Selected samples were also analyzed by mass spectrometry to determine ^{240}Pu, ^{239}Pu, and ^{241}Pu. The analytical techniques were essentially those described by Wong (1971) and Noshkin and Gatrousis (1974). Stable strontium was determined by atomic absorption. Our discussion here will concentrate primarily on the transuranium elements; however, Tables 3–5 give the results for all the radionuclides analyzed, expressed as pCi g^{-1} (dry wt) and corrected for decay back to the estimated year of growth.

Two sources contribute to the ^{241}Am levels found in the coral. Part is from the radioactive decay of the parent radionuclide ^{241}Pu, while the remainder is unsupported ^{241}Am. Based on the quantities of ^{241}Am and ^{241}Pu, the time between separation, and the age of the coral section, levels of both supported and unsupported ^{241}Am are computed and given in Table 5.

Radionuclide results

General—The coral bands identified with the 1954, 1956, and 1958 test series, as would be expected from the autoradiograph, contain the highest concentrations of radionuclides. A spectrum from 12 g of the 1954 growth section is shown in Fig. 4 with each prominent gamma ray identified. Several gamma-emitting radionuclides with half-lives less than 1–2 years have been identified in earlier surveys (Welander 1969); these were not detected in any coral sections with our spectrometer system. Other radionuclides requiring radiochemi-

Table 3. Transuranium concentrations and plutonium activity ratios.

Estimated year of growth section	pCi/g dry weight - decay corrected to year of growth ± % error				
	$^{239+240}Pu$*	^{238}Pu*	^{241}Am†	$^{240}Pu:^{239}Pu$‡	$^{241}Pu:^{239}Pu$‡
1971-1972	0.13±7	0.005±18	0.09±51	0.799±0.9 (0.216)§	21.0±5.5 (0.0128)§
1970	0.10±5	0.005±11		0.754	28.7
1969	0.10±4	0.004±22	0.09±37	0.740	31.3
1968	0.10±2	0.003±19	0.18±41	0.769	28.6
1967	0.10±5	0.004±11	0.14±39	0.775	32.8
1966	0.09±5	0.006±14	0.14±30	0.770	39.6
1965	0.11±4	0.003±21	0.14±52	0.753±0.9 (0.204)	30.1±6.1 (0.0184)
1964	0.48±4	0.023±6	0.62±13	0.851	36.2
1963	0.14±3	0.007±14	0.18±29	0.779	42.7
1962	0.20±2	0.011±18	0.27±47	0.754±1.3 (0.204)	—
1961	0.60±3	0.022±9	0.50±41	0.821	46.5
1960	1.25±3	0.058±8	0.50±53	0.822	46.9
1959	0.82±3	0.049±5	0.68±33	0.687±0.8 (0.186)	37.5±3.8 (0.0229)
1958	4.50±3	0.266±2	3.29±17	0.879±0.5 (0.238)	36.6±2.2 (0.023)
1957	1.20±3	0.140±3	1.35±28	0.567	35.2
1956	13.19±3	0.900±9	9.00±7	0.838±0.2 (0.227)	36.1±0.8 (0.0222)
1954-1955	38.97±3	2.18 ±2	17.55±13	0.806±0.1 (0.218)	39.1±0.6 (0.0239)

*Determined by alpha spectrometry.
†Determined by gamma spectrometry.
‡Determined by mass spectrometry.
§Atom ration shown within parentheses. $^{240}Pu:^{239}Pu$ activity ratio divided by 3.69; $^{241}Pu:^{239}Pu$ activity ratio divided by 1638.

Table 4. Other gamma-emitting radionuclides detected, decay corrected to year of coral growth.

Estimated year of growth section	pCi/g dry weight ± % error					
	^{155}Eu	^{207}Bi	^{60}Co	^{137}Cs	^{102m}Rh	^{125}Sb
1971-1972	0.11±25	0.06±28	*			
1970						
1969	0.14±28	0.09±14	0.15±24			
1968	0.18±27	0.09±14	0.13±75			
1967	0.18±24	0.10±15	0.11±28			
1966	0.18±26	0.09±15	0.22±14			
1965	0.23±22	0.09±17	0.15±80			
1964	1.22±9	0.23±6	0.37±12			
1963	0.45±80	0.14±37	0.54±43			
1962	0.90±67	0.42±28	0.68±39			
1961	1.80±53	0.50±29	1.08±34			
1960	4.05±41	0.86±29	1.33±22			
1959	5.63±47	0.90±47	2.50±38			
1958	31.5±17	5.13±10	9.90±30	0.40±40		
1957	5.85±60	1.49±33	7.20±32	0.41±49		
1956	117.0±5	13.4±4	30.6±10	1.04±18		
1954-1955	288.0±6	37.6±1	88.2±2	2.16±11	192±20	126±47

*A blank space indicates the data were not collated or the concentration was below detection limits.

th ± % error $^{241}Pu:^{239}Pu$‡
21.0±5.5
(0.0128)§
28.7
31.3
28.6
32.8
39.6
30.1±6.1
(0.0184)
36.2
42.7
—
46.5
46.9
37.5±3.8
(0.0229)
36.6±2.2
(0.023)
35.2
36.1±0.8
(0.0222)
39.1±0.6
(0.0239)

3.69;

r of coral growth.

^{102m}Rh	^{125}Sb

92±20 126±47

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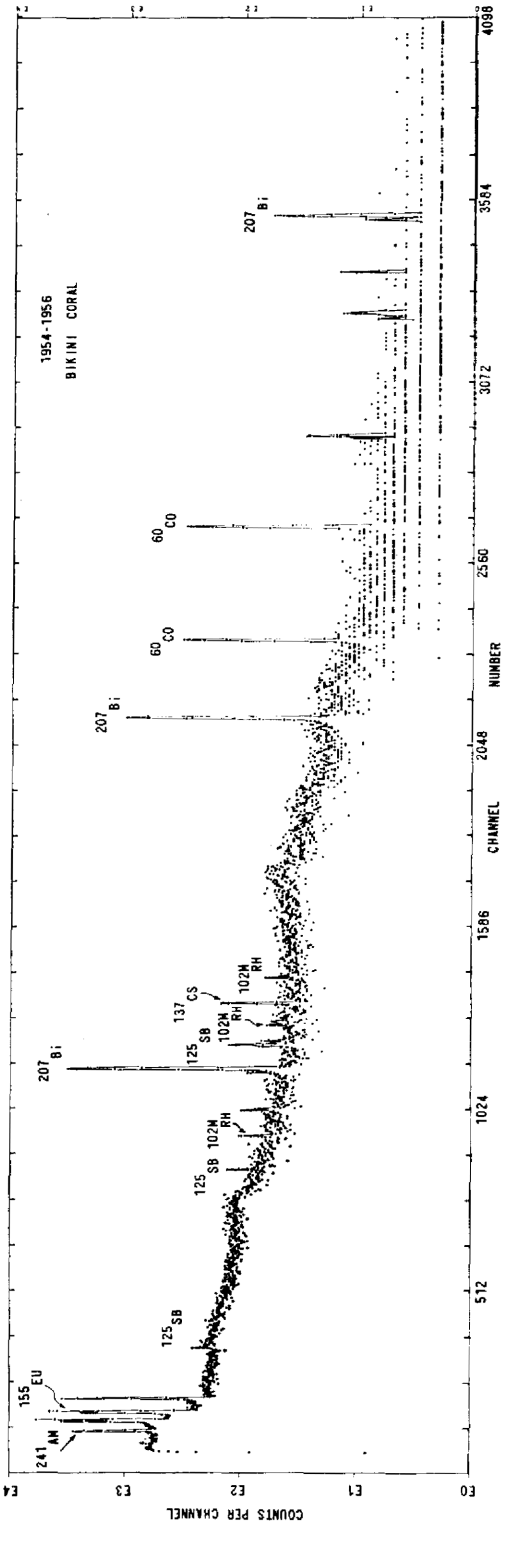

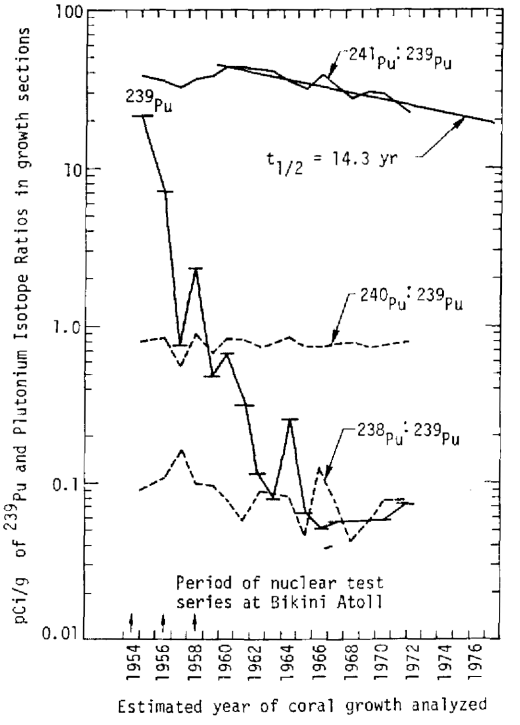

Fig. 5. Concentration of ^{239}Pu and the plutonium isotope activity ratios in each annual section analyzed.

cal separation from the coral (such as ^{55}Fe) and known to be present in atoll samples (Welander 1969) were not determined. The concentration of ^{239}Pu and the plutonium isotope activity ratios in each annual band, decay-corrected to year of coral growth, are shown in Fig. 5.

In surface water samples collected from the north equatorial current or east of Bikini, the $^{239+240}Pu$ concentration averaged 0.4 fCi liter^{-1} (Noshkin et al. 1974); the average in 10 filtered lagoon samples (ranging from 79–4 fCi liter^{-1}) was 40 (Noshkin et al. 1974). The ^{90}Sr concentration in the lagoon averaged 570 fCi liter^{-1} compared to 71 in the surface waters out-

←

Fig. 4. Gamma-ray spectrum of the 1954–1955 growth section. Unidentified photopeaks are from background and naturally occurring radionuclides.

Table 5. ^{90}Sr and ^{210}Po (^{210}Pb) concentrations, decay corrected to year of growth. Unsupported and supported ^{241}Am concentrations.

Estimated year of growth	pCi/g dry weight ± % error			
	^{241}Am*		^{90}Sr†	^{210}Pb‡
	Unsup.	Sup.		
1971-1972	0.09±51	0.00	lost	lost
1970	—	—	0.45±12	0.19±5
1969	0.09±37	0.00	0.78±12	0.15±6
1968	0.17±41	0.00	0.71±4	
1967	0.13±39	0.01	0.69±6	0.19±5
1966	0.12±30	0.02	0.73±6	0.20±5
1965	0.12±52	0.02	0.89±4	0.22±12
1964	0.52±13	0.10	1.15±5	0.19±5
1963	0.07±29	0.11	1.03±5	0.21±9
1962			1.21±4	0.25±25
1961	0.30±41	0.20	1.75±3	0.19±10
1960	0.05±53	0.45	2.38±3	0.24±9
1959	0.40±33	0.28	8.68±3	0.29±22
1958	1.85±17	1.44	16.0±2	0.48±4
1957	0.93±28	0.42	33.3±1	0.25±7
1956	4.4 ±7	4.6	32.2±1	1.04±11
1954-1955	1.4 ±13	16.1	39.3±1	1.35±9

*Unsup: unsupported ^{241}Am. Sup: see ^{241}Am supported by ^{241}Pu decay.
†Determined by beta counting ^{90}Y - daughter of ^{90}Sr on low-level proportional counters.
‡^{210}Pb activity determined by counting ^{210}Po using alpha spectrometry.

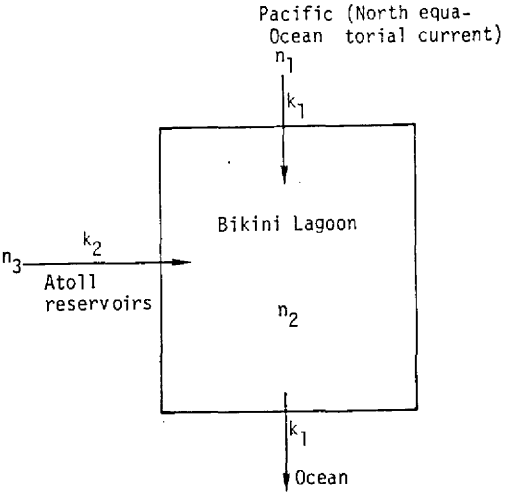

Fig. 6. Box model diagram of Bikini Lagoon describing the general flow and sources of radionuclides into and out of the lagoon.

side and east of the lagoon. Radioelements ^{60}Co, ^{207}Bi, ^{241}Am, and ^{155}Eu were not detected by gamma spectrometry in any water samples collected outside the atoll but were prominent in biota and sediment samples (Fig. 1, Table 1) from the lagoon. We conclude that the atoll is the principal source of radionuclides to the lagoon environment.

Our investigation differs from others attempting to use corals to determine the trace element composition of adjacent waters. We are not comparing concentrations from different species or concentrations in whole specimens from different areas. We expect coral samples from different lagoon locations to have significantly different absolute radionuclide concentrations, and preliminary data have verified this assumption. Having only a single sample from one location, however, we need only assume for each element or radionuclide that each yearly increment of coral growth concentrates the same available fraction from seawater per unit weight of coral; then, the concentration (pCi g^{-1}) in the annual growth band is proportional to the concentration in the adjacent environment. On this basis, we find that all detected radionuclides (except ^{210}Po) have decreased in the lagoon by two to three orders of magnitude since the test years.

The rate at which the radionuclide concentration changes with time is not constant. The rate of change was most rapid after the test series. In some years (e.g. 1964 compared to 1963 and 1965), the concentrations of some radionuclides changed abruptly: for these small but real changes we have no explanation. We tried to correlate the 1964 increase with the peak in fallout deposition in the Northern Hemisphere, but the computed amount deposited in the lagoon in this period was insignificant compared to the observed change. Although these data are derived from only one coral collection, they suggest that unanticipated processes in the atoll may lead to abrupt changes in the concentrations of specific radionuclides in the lagoon environment. However since 1965 the concentrations of many specific radionuclides have decreased only slowly (after correction for decay); this indicates that recycling from sedimentary processes, biological activity,

and surface runoff, or some combination of these or other biogeochemical processes, are responsible for replenishing activity levels of some radionuclides in the lagoon at rates that compensate for the rates of removal.

No model can adequately account for these unanticipated changes in concentration unless the specific mechanisms responsible for the changes are understood. However, even though a degree of uncertainty exists, we can use a simplified model of the lagoon environment based on the diagram in Fig. 6 and the coral data to describe the rate at which the radionuclides are exchanged between the lagoon and open ocean and the rate at which specific radionuclides are recycled from the atoll.

The statement of the mass balance in terms of the change in the amount of a radionuclide, n_2, in the lagoon water with time is

$$\frac{dn_2}{dt} = k_1 n_2 - \lambda n_2 + k_2 n_3 + k_1 n_1, \quad (1)$$

where k_1 is a universal rate constant in yr^{-1} and is independent of the particular radioactive species considered. k_1 is the mean residence time of the lagoon water. λ is the radiological decay constant in yr^{-1} and k_2 is the rate constant in yr^{-1} defining the supply of a particular dissolved species from all diagenetic processes. If n_1, the quantity of species n_2 supplied to the lagoon from the ocean reservoir, is small compared to the amounts contributed by the atoll, as it is for all radionuclides detected in the coral except ^{90}Sr and ^{137}Cs, Eq. 1 reduces to

$$\frac{dn_2}{dt} = -n_2(k_1 + \lambda) + k_2 n_3. \quad (2)$$

The change in n_3, the quantity of species n_2 supplied to the lagoon from diagenetic processes, with time is

$$\frac{dn_3}{dt} = -n_3(\lambda + k_2). \quad (3)$$

Solving Eq. 3 and substituting the solution in 2, the solution for n_2 as a function of time is

Table 6. k_2 and k_0 values computed from Eq. 5 for specific radionuclides.

Radionuclide	k_2	$k_0 = \frac{k_2}{k_1 - k_2} n_{30}$
^{239}Pu	0.07	0.12
^{240}Pu	0.07	0.091
^{241}Pu	0.06	3.90
^{238}Pu	0.13	0.014
^{155}Eu	0.06	1.25
^{207}Bi	0.13	0.39
^{60}Co	0.12	1.52

$$n_2 = n_{20} e^{-\lambda t} e^{-k_1 t} + \frac{k_2}{k_1 - k_2} \times n_{30} e^{-\lambda t}(e^{-k_2 t} - e^{-k_1 t}). \quad (4)$$

We propose that the radionuclide concentrations in each annual growth section are proportional to the amount of species n_2 in the surrounding water environment during the respective year of growth. The last nuclear test series at Bikini was held in 1958. Taking 1958 as t_0, there was an amount n_{30} of species n_3 in the atoll reservoirs. We assume that the rate at which the lagoon is flushed with uncontaminated ocean water is rapid enough so that after 5 years $e^{-k_1 t}$ can be taken to be zero. Equation 4 then reads

$$n_2 \text{ (after 1962–1963)} = k_0 e^{-(k_2+\lambda)t}, \quad (5)$$

where $k_0 = (k_2 n_{30})/(k_1 - k_2)$.

Using the data retained in the coral sections from 1962 to 1972 we can compute a best-fit unique value of k_0 and k_2 from Eq. 5. These values for each radionuclide detected are listed in Table 6. Substituting the values of k_0 and k_2 into Eq. 4, and now using the 1958 and post-test year coral data, we get an average value for k_1 of 1.98 ± 0.14 yr^{-1}.

The lagoon volume along with any dissolved species is exchanged 1.98 times a year with the open ocean. The residence time of the lagoon is 127 to 198 days. From calculated flows into and out of Bikini Lagoon, Von Arx (1954) estimated that during the tradewind season one lagoon vol-

Table 7. Radionuclides in surface sediments at station B-3 compared to levels in most recent coral sections.

	0-5 cm sediment core section at B-3		Recent coral section
	fine	coarse*	
$^{241}Am:^{155}Eu$	0.62±0.03	0.77±0.03	0.83±0.47
$^{60}Co:^{155}Eu$	0.62±0.03	0.38±0.02	4.4 ±1.2
$^{207}Bi:^{155}Eu$	0.10±0.01	0.13±0.01	1.0 ±0.4

*Sedimentary components greater than 0.5 mm.

ume exchanges with the open ocean every 39 days. During the summer the average exchange is about half the winter rate. These rates imply about seven changes per year between the lagoon and open ocean. The coral data show that the rate of natural displacement of any radionuclide from the lagoon water may not be directly assessed from physical circulation estimates alone. The radiological data stored in the yearly growth increments yield longer residence times for this initially contaminated lagoon, or for the region of the lagoon around station B-3, than those predicted by Von Arx (1954). Any chronological assessment of the availability of pollutants to marine organisms, in an aquatic environment where the flow characteristics are similar to those in an atoll, should be treated with these findings in mind.

Table 6 shows that the rate constants for supply of a specific radionuclide from diagenetic processes are smallest for ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{155}Eu. Surprisingly the value of k_2 for ^{238}Pu is about twice that of ^{239}Pu and equivalent to the values for ^{60}Co and ^{207}Bi. ^{238}Pu appears to be released to the lagoon faster than ^{239}Pu and one must conclude that the processes acting on the reservoirs regulating the amount of ^{238}Pu released to the lagoon are different from those regulating ^{239}Pu.

The average concentration of $^{239,240}Pu$ in Bikini Lagoon water during the fall of 1972 was 49 fCi liter^{-1} which converts to a lagoon water inventory of 1.4 Ci. Assuming the $^{240}Pu : ^{239}Pu$ ratio in the water was equivalent to that in the recent coral sections there were 0.8 Ci of ^{239}Pu and 0.6 of ^{240}Pu in the lagoon. Substituting these values into Eq. 5 with $t = 13.5$ years and the respective values of k_1 and k_2 we can compute the initial size of the reservoir supplying ^{239}Pu and ^{240}Pu to the lagoon. The source contributing $^{239,240}Pu$ must have contained, in 1958, at least 97 total Curies of the radionuclides; by 1972, of this amount, 32 Ci have been lost to the lagoon water and advectively transported to the open ocean.

A number of sediment samples from Bikini have been analyzed for ^{241}Am and a few for $^{239,240}Pu$. The total amount of ^{241}Am in the surface 2.5 cm of sediment (*see Fig. 1*) is about 200 Ci and represents on the average only 27% of the total activity in the sediment column (unpublished data). The mean $^{241}Am : ^{239,240}Pu$ ratio in samples of specific sedimentary components from several lagoon locations was 0.74 ± 0.17. Assuming that this ratio is constant over the entire lagoon basin, we estimate there are at least 250 Ci of $^{239,240}Pu$ in the surface 2.5 cm of lagoon sediment. This source alone is more than sufficient to account for the size of the $^{239,240}Pu$ reservoir predicted by the coral data.

Since the estimated size of the reservoir is 97 Ci, or 65 Ci by 1972, substantially less than the amount presently contained in the surface 2.5 cm of surface sediment alone, we can at present only ask whether new or different diagenetic processes will act on this larger reservoir in the future (equivalent to a variable rather than constant k_2) to increase lagoon concentrations, or whether a quantity of $^{239,240}Pu$ in the atoll will forever remain unavailable to the water and the pelagic organisms of the lagoon.

Concentration factors—On the basis of the average activity from the four most recent growth sections and the average water concentrations of $^{239,240}Pu$ and ^{90}Sr given earlier in this report, the concentration factors for these radionuclides in Bikini coral are, respectively, 2.7×10^3 and 1.1×10^3. Stable strontium in the coral sections averaged 8.94 ± 0.35 mg g^{-1}. The specific activity of ^{90}Sr in the coral is 0.072 pCi mg^{-1};

its specific activity in the Bikini Lagoon water, assuming an average of 8 mg liter^{-1} of strontium in seawater (Goldberg et al. 1971), is 0.071 pCi mg^{-1}. The ^{90}Sr therefore is incorporated by the living coral polyps in direct proportion to its concentration in the water and there is no discrimination between ^{90}Sr and stable strontium. The concentration factor for ^{90}Sr is in good agreement with the average specific activity of 37 ± 10 pCi g^{-1} strontium in recent coral surface sections from Enewetak reported by Knutson and Buddemeier (1973). If the stable strontium in coral is 8.9 mg g^{-1}, the ^{90}Sr concentration is 0.33 pCi g^{-1} coral. Based on our average 1972 concentration of ^{90}Sr in water (0.33 pCi liter^{-1}) at Enewetak (Noshkin et al. 1974), the concentration factor in Enewetak coral is 1.0×10^3.

Our computed ^{239,240}Pu concentration factor agrees well with the results of Imai and Sakanoue (1973) who reported a concentration factor of 1 to 2×10^3 for fallout ^{239,240}Pu coral collected from Yoran Island (27°04′N, 128°25′E). The similar values in corals from different environments with different levels of contamination indicate that coral species take up ^{90}Sr and ^{239,240}Pu in proportion to the concentration in the water; therefore they serve as excellent indicators for environmental levels of these radionuclides.

Values of 370 and 9,400 fCi liter^{-1} for ^{90}Sr and ^{137}Cs were reported from a single filtered midlagoon bottom water sample collected at Bikini in August 1964 (Welander et al. 1967). In our November 1972 filtered midlagoon bottom water sample (Noshkin et al. 1974) we detected 315 and 340 fCi liter^{-1} ^{90}Sr and ^{137}Cs. The similarity in the ^{90}Sr values after decay correction shows little change in the lagoon concentration at a specific location over the 8-year period. Our coral record over the same period confirms this observation. Again we must conclude that the mechanisms now releasing ^{90}Sr and, as the coral record indicates, plutonium nuclides and lanthanides as well, are supplying these radionuclides to the lagoon at a rate that will only slowly change the lagoon concentration with time.

Specific radionuclides in the coral section—Precise measurements of ^{239}Pu, ^{240}Pu, and ^{241}Pu were made in nearly all sections by mass spectrometry. Small, but nevertheless significant, changes are noted in the ^{240}Pu : ^{239}Pu activity ratios (Table 3, Fig. 5) in the coral sections. The average ^{240}Pu : ^{239}Pu activity ratio was 0.77 ± 0.07, the range from 0.57 to 0.88. In each post-test series year, 1957 and 1959, the ratio was reduced to an average of 72% of its test year value. In 1960 it increased by 20% over the 1959 ratio and has since changed by no more than ±10% of the 1960 value.

Assuming that the coral does not discriminate between the same chemical forms of ^{241}Pu and ^{239}Pu, we found more variation in the activity ratio, as a function of time, than was anticipated. If both isotopes are released to the environment at the same rates, the ratio change in the coral sections should follow a 14.3-year decay curve (Fig. 5). An inspection of Fig. 5 and Table 3 will show that the ^{241}Pu : ^{239}Pu decay-corrected ratios in the test year growths, 1954, 1956, and 1958, are lower than any extrapolated post-test year ratio would predict. These differences in test and post-test year ratio can only be explained if the plutonium isotopes had different ratios, in unique chemical or physical forms, after production. A smaller amount of soluble ^{241}Pu relative to ^{239}Pu was deposited in the lagoon water during the years of the test series while relatively more ^{241}Pu ended up in the atoll reserviors that now supply both ^{241}Pu and ^{239}Pu to the lagoon.

Even more significant are the variations with time in the ^{238}Pu : ^{239}Pu values. In 17 Bikini water samples collected during 1973 (Noshkin et al. 1974), the average ^{238}Pu : ^{239,240}Pu ratio was 0.018 ± 0.006 (range 0.011–0.026). In the three most recent coral growth sections the average ratio is 0.040 ± 0.005. The plutonium concentration factor based on ^{239}Pu is higher than the computed value using $^{239+240}$Pu concen-

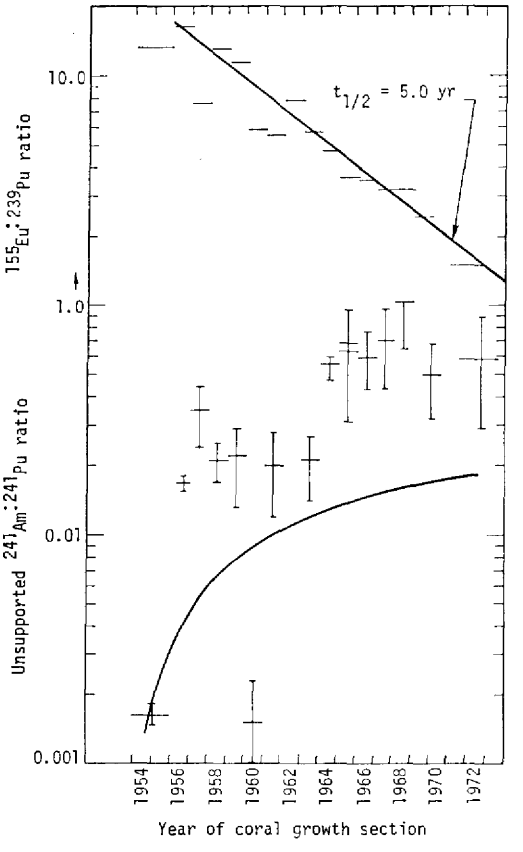

Fig. 7. ^{155}Eu : ^{239}Pu and unsupported ^{241}Am : ^{241}Pu ratios in the yearly coral growth sections.

trations. At this time we can offer no explanation of this difference.

Assessing the possible influence of ^{238}Pu from fallout and from SNAP-9A on lagoon concentrations, we found this input to have little or no effect on the concentration levels during any one year. Consider, for example, that the cumulative deposition of ^{238}Pu up to 1971 has been only 0.009 mCi km^{-2} between 10° and 20° N latitude (Hardy et al. 1973). Comparison of this value with the 1972 average standing level in Bikini Lagoon of 0.033 mCi km^{-2} (mean depth of the lagoon is 46 m) shows that the present lagoon level alone exceeds the maximum possible concentration derived from fallout. The mechanisms regulating the discrimination of these plutonium isotopes are meshed with biogeochemical processes within the lagoon and are as yet not understood. The failure of the data to conform to predictable patterns is a most important feature of this study and complicates the interpretation of the behavior of plutonium radionuclides in this environment. It does not appear possible, with these data, to predict unequivocally the behavior of all plutonium isotopes in the environment from an assessment of a single isotope of the element; a great deal of research is still required to unravel the biogeochemistry of this element.

The ^{239}Pu activity in the coral sections seems to correlate better with ^{155}Eu (Fig. 7) than with ^{238}Pu. The ^{155}Eu : ^{239}Pu ratio in the coral sections decreases in value with a best-fit half-life of 5.1 years which is the radiological decay half-life of ^{155}Eu. Similar geochemical processes appear to govern the fate and behavior of this lanthanide and of ^{239}Pu in the lagoon, and the way in which the change in ratio correlates with time supports the age assessment of each section.

We pointed out earlier that the ^{241}Am in the coral growth sections originates from the decay of ^{241}Pu in the coral and also directly from the environment. If the environmental source of ^{241}Am is from ^{241}Pu decay only, and the coral does not discriminate between these two transuranics, the excess ^{241}Am to ^{241}Pu ratio in the coral should follow a predictable growth curve with time. Plotted in Fig. 7 are the ratios of unsupported ^{241}Am to ^{241}Pu in each coral growth section and a calculated growth curve of ^{241}Am from ^{241}Pu ($t_0 = 1954$). Although the errors are large we find that the ratios in the post-test years, with the exception of the 1960 value, are changing with time in a manner consistent with the predicted curve. However, all the values are displaced above the 1954 curve and would be further removed from a growth curve originating in 1956 or 1958. The ^{241}Am in the 1954–1955 growth section falls on the predicted curve, showing there was essentially no ^{241}Am directly produced during the 1954 test series. If we assume this to be true for the 1956 and 1958 test series, the coral must either have been preferen-

tially enriching ^{241}Am over ^{241}Pu, or the concentration factor of ^{241}Am must be greater than plutonium for this marine organism.

Concentrations of several radionuclides in the sediment from station B-3 are shown in Table 1. The ratios of ^{241}Am, ^{60}Co, and ^{207}Bi to ^{155}Eu in the surface 5 cm of the fine and coarse (>0.5 mm) fractions are compared to the ratios in the most recent coral section in Table 7. Both the ^{241}Am : ^{155}Eu and the ^{60}Co : ^{207}Bi ratios in the coral are similar to those in the sedimentary phases. However, the ^{60}Co : ^{155}Eu and ^{207}Bi : ^{155}Eu values are greater in the recent coral than in the sedimentary environment. If the sediments are a principal source now supplying radionuclides to the lagoon, by dissolution or exchange or other processes, then we find that relative to the sediment the coral does not discriminate between ^{155}Eu and ^{241}Am, but ^{60}Co and ^{207}Bi are greatly enriched relative to ^{155}Eu. This implies that processes governing the fate of ^{155}Eu in the lagoon are similar to those for ^{241}Am. From the above comparisons, and the concentration factor arguments, we may discount the possibility that the source of the radionuclides in this particular coral that we analyzed is in trapped, previously resuspended, sedimentary material. The coral is functioning rather as an indicator of the aquatic environment.

In an attempt to see whether a naturally occurring radionuclide could be used to confirm the age of the coral sections, we separated from the coral and measured the ^{210}Po, the daughter of ^{210}Pb ($t_{1/2} = 22$ yr). Provided that the environmental levels of ^{210}Pb are constant, the amounts taken up by the coral reflect the age of any section relative to the youngest section, because the concentration within the coral changes due to radioactive decay. When the activity levels are corrected to the date of coral growth (as shown in Table 5), the concentration-time relationship should be invariant. The data from 1966 to 1971 fit this model very well. The decay corrected ^{210}Po (^{210}Pb) concentrations during these years averaged 0.20 ± 0.03 pCi g^{-1}. In the sections identified with the test years, however, there are small but definite increases in ^{210}Po (^{210}Pb) concentrations, which correlate in time with the increases noted for the artificial radionuclides. Several investigators (*cited in* Beasley 1969) have discounted the possibility that ^{210}Pb was produced from weapons testing, but the elevated levels recorded in the coral during nuclear test series are at least circumstantial evidence that elevated levels of ^{210}Po (^{210}Pb) were present in the Bikini environment during those periods. It appears, from the later years' growth, that ^{210}Po (^{210}Pb) levels in coral from remote environments may be used as another means to date modern coral growth, confirming the work by Dodge and Thomson (1974) and Moore and Krishnaswami (1972).

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