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The Effect of Carbonate Soil on Transport and Dose Estimates for Long-Lived Radionuclides at a U.S. Pacific Test Site

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Abstract

The United States conducted a series of nuclear tests from 1946 to 1958 at Bikini, a coral atoll, in the Marshall Islands(MI). The aquatic and terrestrial environments of the atoll are still contaminated with several long-lived radionuclides that were generated during testing. The four major radionuclides found in terrestrial plants and soils are Cesium-137 (^{137}Cs), Strontium-90 (^{90}Sr), Plutonium-239+ 240 ($^{239+240}$Pu) and Americium-241 (^{241}Am).

^{137}Cs in the coral soils is more available for uptake by plants than ^{137}Cs associated with continental soils of North America or Europe. Soil-to-plant ^{137}Cs median concentration ratios (CR) (kBq kg^{-1} dry weight plant/kBq kg^{-1} dry weight soil) for tropical fruits and vegetables range between 0.8 and 36, much larger than the range of 0.005 to 0.5 reported for vegetation in temperate zones. Conversely, ^{90}Sr median CRs range from 0.006 to 1.0 at the atoll versus a range from 0.02 to 3.0 for continental silica-based soils. Thus, the relative uptake of ^{137}Cs and ^{90}Sr by plants in carbonate soils is reversed from that observed in silica-based soils. The CRs for $^{239+240}$Pu and ^{241}Am are very similar to those observed in continental soils. Values range from 10^{-6} to 10^{-4} for both $^{239+240}$Pu and ^{241}Am. No significant difference is observed between the two in coral soil.

The uptake of ^{137}Cs by plants is enhanced because of the absence of mineral binding sites and the low concentration of potassium in the coral soil. ^{137}Cs is bound to the organic fraction of the soil, whereas ^{90}Sr, $^{239+240}$Pu and ^{241}Am are primarily bound to soil particles. Assessment of plant

uptake for ^{137}Cs and ^{90}Sr into locally grown food crops was a major contributing factor in (1) reliably predicting the radiological dose for returning residents, and (2) developing a strategy to limit the availability and uptake of ^{137}Cs into locally grown food crops.

Introduction

Bikini Atoll is one of the two sites in the Marshall Islands used by the United States to test nuclear devices from 1946 to 1958. During this period a total of 23 tests, with a total yield of 77 MT, were conducted at the atoll. Bikini Island, the main residence island at the atoll, was contaminated with fallout primarily from the BRAVO test on March 1, 1954.

The Bikini people, since their initial relocation in 1946, have had a continuing desire to return to their homeland. In 1969 a general cleanup of debris and buildings as well as the planting of coconuts, breadfruit, *Pandanus*, papaya and banana trees began at Bikini Island. A few Bikini families moved back to Bikini Island in 1970. In 1978, when the coconuts started producing fruits, whole body counting revealed that ^{137}Cs body burdens were well above the U.S. recommended level. Consequently, in August of 1978 Trust Territory officials arrived at Bikini Island and relocated the people once again.

A research and monitoring program was initiated in 1978 at Bikini Atoll. As part of this continuing program, we determined the contribution of each exposure pathway and radionuclide to the total dose for people resettling the atoll. Also we developed concentration ratios for predictive purposes. More detail on the dose assessment results can be found in Robison et al, 1997[1], but in summary, ^{137}Cs uptake by terrestrial food crops generates about 89% of the estimated total dose to people inhabiting the atoll. Consequently, we focused our effort to find an efficient, cost effective, and environmentally sound method that would reduce the dose from ^{137}Cs via the food chain.[1,2,3]

In this report we discuss the composition of the coral soil at the atolls and it's effect on the relative uptake by plants of ^{137}Cs, ^{90}Sr, $^{239+240}Pu$ and ^{241}Am that ultimately determines the major route of exposure and the radionuclide of most consequence. Concentration ratio data are provided for the major fruit tree crops (coconuts(*Coco nucifera*), Pandanus(*Pandanus spp.*), breadfruit(*Actocarpus altilis*), banana(*Musa spp.*) and papaya(*Carica papaya*)) and for vegetable and grain crops (corn, sorghum, squash, beans, cabbage and okra).

Study Site

Bikini Atoll is located in the northern Marshall Islands (MI), in the equatorial Pacific Ocean (11° 35' N, 165° 25' E). The climate is tropical with the average temperature of about 84° F. There is a dry season between December through March, and a wet season between April through November. The mean rainfall at Bikini Island is approximately 150 cm.

The coral soil at Bikini Atoll, and at other atolls in the Marshall Islands, is made up almost entirely of calcium carbonate ($CaCO_3$), with some $MgCO_3$ and essentially no silicate clay. The soil components includes, by weight, 25% coral, 35% Foraminifera, 15% Halimeda, 15% Lithothamnion, and 10% mollusk shells.[4] The carbonate matrix contains both the calcite and aragonite forms. The soils are low in exchangeable potassium (K) that ranges from 20 to 79 ppm in the top 25 cm. The highest K concentrations are found in the 0-5 cm layer, and originates from sea salt spray and decaying vegetation. The pH of soil-water slurries ranges from 7.7 to 8.8. The organic content of surface soils ranges from 5 to 14%. The organic matter decreases with depth in the soil column and is generally insignificant below 40 to 50 cm depth. The nutrients for plant growth and the radionuclides are contained in this top 50 cm layer of the soil. Chemical and physical properties of two representative soil profiles from Bikini Island are shown in Table 1. The distribution of ^{137}Cs, ^{90}Sr, $^{239+240}Pu$, and ^{241}Am in soil at Bikini Island are shown in Figure 1.

Field Sampling and Analytical Methods

Field trips to collect food crops, vegetation and soils are conducted quarterly or semiannually by the Lawrence Livermore National Laboratory (LLNL). All samples are frozen within 3 to 4 hours after collection and transported to LLNL for processing and radiological analysis.

Coconuts are the most common and abundant food crop on the island. They account for approximately 68% of the total intake for local terrestrial foods. A coconut sample consists of a composite of five to eight "drinking" or copra nuts depending on the maturity of the type of nut present. A drinking coconut is a younger stage in the coconut development cycle. The maturity of the drinking coconut is characterized by the meat's dry to wet weight ratio which ranges from 0.090 to 0.45. The drinking coconut is full of a light, slightly sweet liquid and the "meat" is undeveloped and soft. The copra coconut is older and has developed solid white meat; it is no longer full of fluid and the remaining fluid is unsuitable for drinking. The dry/wet weight ratio for copra nuts ranges from 0.46 to 0.80. The drinking coconut meat data are used for time-dependent, comparative studies. Copra meat and drinking coconut meat and fluid data are all used for dose assessment. The coconuts are husked, the fluid is collected in plastic bottles, and the intact shell containing the coconut meat is bagged and labeled.

Pandanus fruits are another commonly used food in the Marshall Islands. The fruits consists of numerous phalanges, or keys, attached to a central stalk. A bag of keys is collected in the field. The annual food crops of breadfruit, papaya and banana are collected whole. A sample consists of approximately 5 to 10 fruits for a total mass of several kilograms. These food crops are generally collected when ripe.

Soil profiles for tree crops are taken in depth increments of 0-5, 5-10, 10-15, 15-25, 25-40, and 40-60 centimeters. A single trench, or three trenches at 120° intervals, is dug radically from the trees to minimize root damage using a backhoe. A step method is used for sampling which is described in detail in Stuart 1995.[5] Approximately two kilograms of soil are taken for each depth increment. Soil cores associated with experiments using annual food crops are taken to a depth of 20 cm. These plots are usually confined to a relatively small area, and soil-coring tubes are used to minimize soil disturbance.

At LLNL, the samples are dried, homogenized and compressed into containers for analysis by gamma spectroscopy.[5] Each sample is analyzed on a gamma spectrometer coupled to a DEC VAXStation operating under Canberra/Nuclear Data system data acquisition and reduction software.[6] The most prominent gamma-emitting radionuclide detected is ^{137}Cs. After gamma analysis is completed, selected samples are sent for wet chemistry analysis of ^{90}Sr, $^{239+240}Pu$ and ^{241}Am.[7] There is a separate quality control acceptance criteria for each isotope that is evaluated by using blind standards and duplicate samples.[8]

Results and Discussion

^{137}Cs is far more available for plant uptake in coral soils than in continental silicate soils. ^{137}Cs is retained in the clay matrix of silicate soils, especially when the organic content is low, and is relatively unavailable to plants.[9–15] This difference is very evident when comparing the CRs in table 2 and 3 for both coral and continental soils.

The median soil-to-plant CRs for ^{137}Cs on Bikini Island range from 0.8 to 36 for tropical fruits and vegetables (Table 2 and 3). These CRs are significantly higher than those observed in continental silicate soils which range from 0.004 to 0.5 for fruits and vegetables.[16–18] The range of CRs for ^{137}Cs observed in silicate soils does not even overlap the range of CRs observed in coral soil. This difference is attributed to coral soil being composed almost entirely of Calcium carbonate (with some Mg-Sr carbonate), varying but relatively high amounts of organic matter, essentially no clay minerals, and low concentrations of available K. A fraction of the ^{137}Cs in the carbonate-organic complex is mobile and relatively available to plants. Consequently, being chemically similar to potassium (K), ^{137}Cs is transported across the root membrane as an analog for K because of the absence of clay mineral binding sites, and the low concentrations of K in the soil. Thus, the CRs for ^{137}Cs are one to two order of magnitude greater than those observed in continental silica-based soil for similar plant species.

Soil to plant median CRs for ^{90}Sr range from 0.006 to 0.37 at Bikini Island (Table 4). The one exception is *Pandanus*, which has extremely high concentrations of Ca and a CR for ^{90}Sr of about 1. These values are much lower in general than those associated with plants grown in continental soil which range from 0.02 to 3.0.[16] The very large quantities of available Ca in coral soils reduce the uptake of the ^{90}Sr. Moreover, some of the ^{90}Sr can be bound structurally into the $CaCO_3$ matrix. Consequently, the uptake of ^{90}Sr in coconuts in coral soils is a factor of 10 to 100 less than plants grown in silica-based soils. *Pandanus*, breadfruit, and papaya appear to be similar to CRs reported in temperate zones for fruits.

The CRs for $^{239+240}Pu$ and ^{241}Am (Table 5&6) are very similar to those observed in continental(silica-based) soils in spite of the high pH and organic content of the soil. They range from 10^{-6} to 10^{-4} for both $^{239+240}Pu$ and ^{241}Am. The $^{239+240}Pu$ and ^{241}Am are bound to or incorporated in soil particles and are relatively unavailable for plant uptake. Consequently, the concentrations of the transuranium elements is very low in edible foods at the atoll.

A summary of the CRs for Bikini Island is listed in Table 7.

Large quantities of coral surface soil (0-25 cm) have been treated with acetic acid to dissolve the $CaCO_3$ matrix. After total dissolution of the carbonate soil the remaining fraction has a reddish brown color and is comprised of mostly organic material and some calcium phosphate.

The organic remainder contains about 90% of the ^{137}Cs inventory in the soil prior to treatment with acetic acid. Less than 10% of the ^{137}Cs is lost during the acid treatment. Conversely, the ^{90}Sr, $^{239+240}Pu$ and ^{241}Am are not associated with the organic moiety but are found in the leachate. Consequently, the ^{137}Cs is bound in a very different manner than the other three radionuclides. Further experiments are underway to determine the structural component in the organic residue that is controlling the ^{137}Cs availability.

The unique composition of the coral soil provides for a relatively high uptake of ^{137}Cs, and a relatively low uptake of ^{90}Sr, into locally grown food crops which in turn greatly influence the exposure pathway, and radionuclide that contributes most of the dose to people resettling the islands. The following exposure pathways at the atolls have been evaluated in great detail: ingestion of terrestrial and marine foods, external gamma, inhalation of resuspended contaminated soil, and ingestion of catchment water and ground water. The uptake of ^{137}Cs into terrestrial foods, and subsequent consumption of these foods by the people, accounts for about 89% of the total estimated dose.

These results clearly indicated that efforts to reduce the dose to returning populations needed to be directed at reducing ^{137}Cs uptake into locally grown food crops. If this problem could be solved, the dose problem would be solved. Many different approaches were evaluated [2,3] within a framework that included leaching of ^{137}Cs from the soil, immobilization of ^{137}Cs in the soil, removal by continuous cropping, removal of the contaminated soil, and treatment with potassium (K).

The most effective, most easily accomplished, and most environmental friendly method was found to be the application of K (in the form of either KCl or a full fertilizer N,P,K) to the surface soil. Subsequent rainfall solubilizes the K so that it is available for uptake by the roots of plants. The effectiveness of this method lies in the fact that coral soils are very low in available K and ^{137}Cs, being chemically similar to K, is absorbed across the root membrane as an analog for K. Plants exist on the margin of K deficiency in this ecosystem. Consequently, when K is available (K applied at 1000 kg ha^{-1}) the plants absorb large amounts of K, and the ^{137}Cs concentration in edible foods is reduced to about 10% of pretreatment concentrations in one growing season. Second applications of KCl reduce the ^{137}Cs concentration in food crops to about 5% of pretreatment concentrations.

Conclusion

Relative uptake of ^{137}Cs and ^{90}Sr by plants in carbonate soils is reversed from that observed in silica-based soils. Consequently, due to the unique composition and properties of the coral soil, ^{137}Cs uptake in food crops is much higher than ^{90}Sr and thus dominates the estimated dose to people resettling the atolls. This is in stark contrast to the results that would be obtained using CRs observed in continental, silica-based soils, in conjunction with radionuclide concentrations in coral soil.

The CRs for $^{239+240}$Pu and ^{241}Am in coral soil are not statistically different based on our data and moreover are the same as those observed in continental soils.

Because of the unique features of coral soil, treatment of the agricultural area of Bikini island with potassium reduces the uptake of ^{137}Cs (i.e., CR) into food crops, and in turn the dose from ingestion of local food crops, to about 5% of pretreatment levels.

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Table 1. Composition of coral soils from Bikini Island.

Island location and depth (cm)	pH[a]	Total[b] Strontium (%)	Calcium (%)	Magnesium (%)	Phosphorus[c] (%)	Nitrogen (%)	Organic matter[d] (%)	Extractable potassium[e] (mg kg^{-1})	Particles sized <0.5 mm (%)
Bikini No. 1									
0–5	7.7	0.38	30.4	0.95	1.35	0.64	14.4	79	11.5
5–10	7.8	0.39	30.8	0.89	1.28	0.62	13.2	26	11.5
10–15	7.9	0.39	30.9	0.89	1.29	0.63	12.3	20	9.5
15–25	7.9	0.40	31.9	0.86	1.17	0.50	10.6	23	11.7
25–40	8.3	0.39	34.3	1.28	0.67	0.19	4.5	4	6.3
40–60	8.4	0.31	34.5	2.05	0.16	0.11	1.6	3	0.6
Bikini No. 2									
0–5	7.8	0.40	31.0	1.02	0.82	0.49	10.7	50	5.7
5–10	8.0	0.40	32.4	1.09	0.71	0.46	8.5	24	3.71
10–15	7.9	0.38	33.1	1.18	0.56	0.35	7.4	24	3.3
15–40	8.2	0.38	34.7	1.79	0.32	0.11	1.6	6	1.1

[a] pH in water.
[b] Stable cesium was below the detection limit of 1.3 mg kg^{-1}.
[c] High phosphorus values indicate ancient guano deposition.
[d] Organic matter by wet oxidation.
[e] Extractable *N* ammonium acetate.

Table 2. Cesium-137 soil-to-plant concentration ratio (CR[a]) summary for food crops grown in the 0-40 cm root zone in Bikini Island coral soil.

	No. of trees	No. of samples	Median	Arithmetic mean	Minimum	Maximum	Silicate soil median[b]
Drinking Coconut Meat							
CR	106	448	13	16	2.8	64	0.0044
Dry/Wet wt.		448	0.29	0.29	0.090	0.45	
Copra Meat							
CR	55	128	11	14	4.3	57	
Dry/Wet wt.		128	0.58	0.58	0.45	0.80	
Pandanus							
CR	9	39	35	44	11	82	
Dry/Wet wt.		39	0.19	0.19	0.090	0.35	
Breadfruit Meat							
CR	8	35	2.9	3.4	0.44	7.4	
Dry/Wet wt.		35	0.25	0.22	0.11	0.32	
Papaya Meat							
CR	23	49	11	22	2.1	62	
Dry/Wet wt.		49	0.11	0.11	0.070	0.19	
Banana Meat							
CR	5	5	0.78	0.71	0.27	1.2	
Dry/Wet wt.		5	0.28	0.28	0.25	0.31	

[a] CR is calculated from kBq kg^{-1} fruit dry weight versus 0-40 cm kBq kg^{-1} soil dry weight.
[b] Median CRs for fruit(apples) in European Soil from Green 1997.[17]

Table 3. Cesium-137 soil-to-plant concentration ratio (CR[a]) summary for food crops grown in the 0-20 cm root zone in Bikini Island coral soil.

	No. of samples	Median	Arithmetic mean	Minimum	Maximum	Clay median[b]	Sand median[b]	Peat median[b]
Sorghum (Whole)								
CR	55	8.1	10	0.81	24	0.017	0.29	0.30
Dry/Wet wt.	55	0.30	0.30	0.12	0.53			
Sorghum Seed Heads								
CR	15	11	13	0.94	31	0.010	0.026	0.083
Dry/Wet wt.	15	0.74	0.63	0.33	0.84			
Corn Kernels								
CR	41	0.88	1.4	0.20	7.8	0.010	0.026	0.083
Dry/Wet wt.	41	0.31	0.35	0.14	0.58			
Corn Stalks								
CR	27	2.3	2.5	0.49	9.0	0.017	0.29	0.30
Dry/Wet wt.	27	0.18	0.18	0.12	0.31			
Chinese Cabbage								
CR	17	36	41	2.9	144	0.18	0.46	0.26
Dry/Wet wt.	17	0.075	0.077	0.036	0.11			
Squash								
CR	10	2.7	3.6	0.73	9.7	0.18	0.46	0.26
Dry/Wet wt.	10	0.099	0.12	0.064	0.23			
Sweet Potato								
CR	3	3.5	3.0	1.7	3.8	0.070	0.17	0.27
Dry/Wet wt.	3	0.34	0.31	0.25	0.36			
Yard Long Beans								
CR	1	3.9				0.017	0.094	
Dry/Wet wt.	1	0.26						
Okra								
CR	1	1.9				0.18	0.46	0.26
Dry/Wet wt.	1	0.12						

[a] CR is calculated from kBq kg^{-1} fruit dry weight versus 0-20 cm kBq kg^{-1} soil dry weight.
[b] Median CRs in temperate environments from IAEA, 1994.[16]

Table 4. Strontium-90 soil-to-plant concentration ratio (CR[a]) summary for food crops grown in the 0-40 cm root-zone in Bikini Island coral soil.

	No. of trees	No. of samples	Median	Arithmetic mean	Minimum	Maximum	Silicate soil median[b]
Drinking Coconut Meat							
CR	30	48	0.018	0.023	0.0038	0.073	0.20
Dry/Wet wt.		48	0.30	0.30	0.16	0.45	
Copra Meat							
CR	16	28	0.0057	0.0073	0.0012	0.030	
Dry/Wet wt.		28	0.61	0.61	0.52	0.72	
Pandanus							
CR	5	6	1.1	0.95	0.27	1.7	
Dry/Wet wt.		6	0.23	0.21	0.13	0.25	
Breadfruit Meat							
CR	7	11	0.27	0.25	0.036	0.58	
Dry/Wet wt.		11	0.25	0.26	0.21	0.31	
Papaya Meat							
CR	5	5	0.37	0.52	0.23	0.87	
Dry/Wet wt.		5	0.070	0.076	0.050	0.11	

[a] CR is calculated from kBq kg^{-1} fruit dry weight versus 0-40 cm kBq kg^{-1} soil dry weight.

[b] Median CRs for fruit in temperate environments from IAEA, 1994.[16]

Table 5. Plutonium-239+240 soil-to-plant concentration ratio (CR[a]) summary for food crops grown in the 0-40 cm root zone in Bikini Island coral soil.

	No. of trees	Median	Arithmetic mean	Minimum	Maximum	Silicate soil median[b]
Drinking Coconut Meat						
CR	16	5.6×10^{-5}	6.3×10^{-5}	9.7×10^{-6}	1.9×10^{-4}	1.4×10^{-4}
Dry/Wet wt.	16	0.28	0.29	0.26	0.34	
Copra Meat						
CR	13	1.0×10^{-5}	3.2×10^{-5}	4.4×10^{-7}	1.5×10^{-4}	
Dry/Wet wt.	13	0.59	0.59	0.55	0.66	
Pandanus						
CR	2	1.3×10^{-4}	1.3×10^{-4}	1.1×10^{-4}	1.6×10^{-4}	
Dry/Wet wt.	2	0.16	0.16	0.13	0.18	
Breadfruit Meat						
CR	6	6.6×10^{-5}	5.7×10^{-5}	1.0×10^{-5}	1.1×10^{-4}	
Dry/Wet wt.	6	0.25	0.24	0.21	0.25	
Papaya Meat						
CR	3	2.9×10^{-6}	3.3×10^{-5}	1.3×10^{-6}	9.6×10^{-5}	
Dry/Wet wt.	3	0.070	0.077	0.050	0.11	

[a] CR is calculated from kBq kg^{-1} fruit dry weight versus 0-40 cm kBq kg^{-1} soil dry weight.
[b] Median CRs for fruit(apples) in European Soil from Green 1997.[17]

Table 6. Americium-241 soil-to-plant concentration ratio (CR[a]) summary for food crops grown in the 0-40 cm root zone in Bikini Island coral soil.

	No. of trees	Median	Arithmetic mean	Minimum	Maximum	Silicate soil median[b]
Drinking Coconut Meat						
CR	8	5.9×10^{-5}	2.1×10^{-4}	6.9×10^{-6}	1.2×10^{-3}	1.5×10^{-4}
Dry/Wet wt.	8	0.29	0.29	0.26	0.34	
Copra Meat						
CR	11	1.6×10^{-5}	5.3×10^{-5}	3.3×10^{-6}	3.4×10^{-4}	
Dry/Wet wt.	11	0.59	0.59	0.56	0.66	
Breadfruit Meat						
CR	4	6.5×10^{-5}	9.5×10^{-5}	1.6×10^{-5}	2.3×10^{-4}	
Dry/Wet wt.	4	0.24	0.24	0.21	0.25	
Papaya Meat						
CR	2	2.2×10^{-5}	2.2×10^{-5}	1.0×10^{-5}	3.4×10^{-5}	
Dry/Wet wt.	2	0.080	0.080	0.050	0.11	

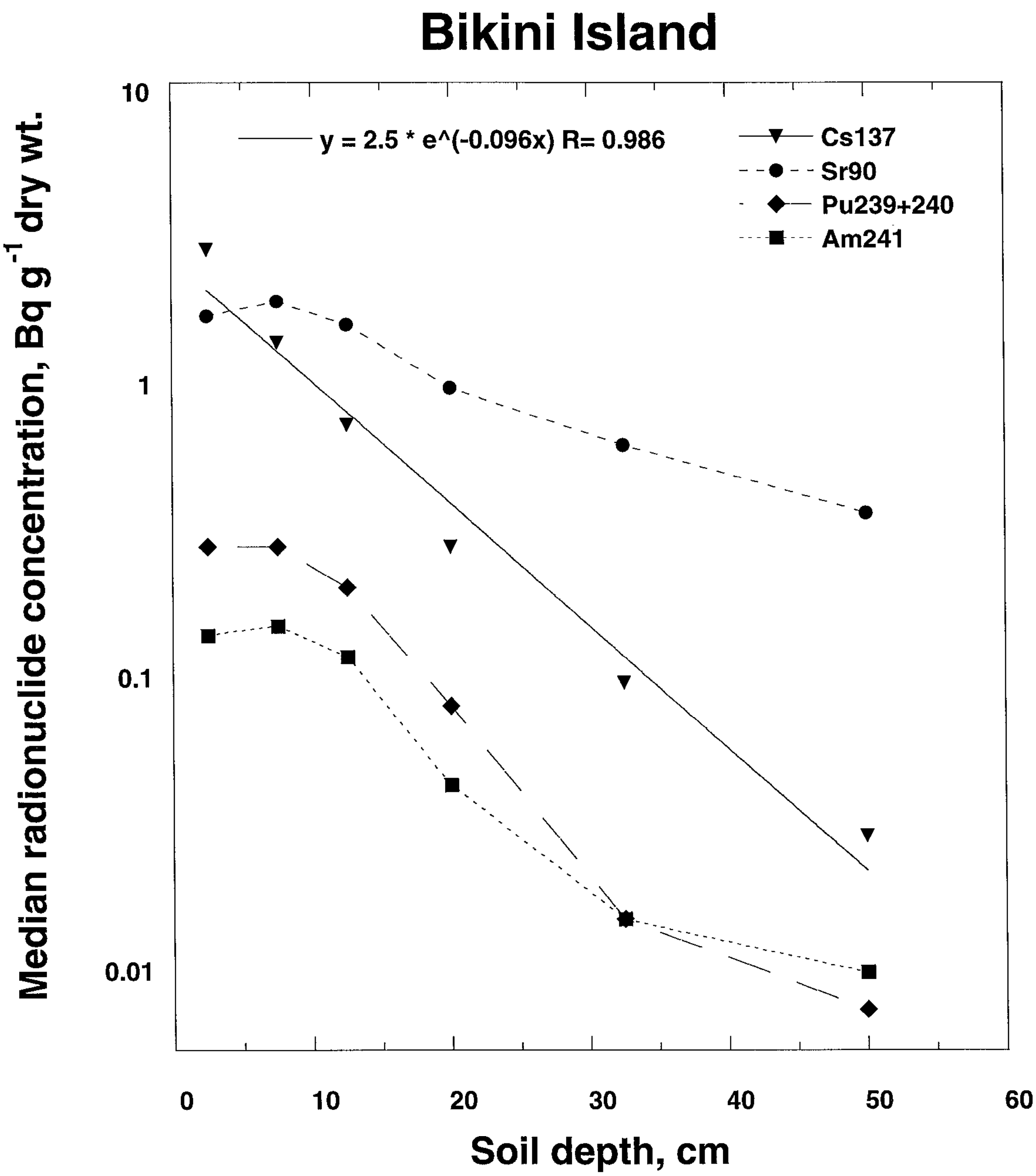
[a] CR is calculated from kBq kg^{-1} fruit dry weight versus 0-40 cm kBq kg^{-1} soil dry weight.

[b] Median CRs for fruit(apples) in European Soil from Green 1997.[17]

Table 7. Soil-to plant concentration ratio summary for food crops grown on Bikini Island.

Food Crops	dry/ wet wt.	^{137}Cs CR median	^{90}Sr CR median	$^{239+240}$Pu CR median	^{241}Am CR median
Drinking Coconut Meat	0.29	13	0.018	5.6×10^{-5}	5.9×10^{-5}
Copra Meat	0.58	11	0.0057	1.0×10^{-5}	1.6×10^{-5}
Pandanus	0.19	35	1.1	1.3×10^{-4}	----
Breadfruit Meat	0.25	2.9	0.27	6.6×10^{-5}	6.5×10^{-5}
Papaya Meat	0.11	11	0.37	2.9×10^{-6}	2.2×10^{-5}
Banana Meat	0.28	0.78	----	----	----
Sorghum (Whole)	0.30	8.1	----	----	----
Sorghum Seed Heads	0.74	11	----	----	----
Corn Kernels	0.31	0.88	----	----	----
Corn Stalks	0.18	2.3	----	----	----
Chinese Cabbage	0.075	36	----	----	----
Squash	0.099	2.7	----	----	----
Sweet Potato	0.34	3.5	----	----	----
Yard Long Beans	0.26	3.9	----	----	----
Okra	0.12	1.9	----	----	----

Figure 1. Bikini Island median radionuclide concentrations in soil.



Bikini Island
y = 2.5 * e^(-0.096x) R= 0.986
Cs137
Sr90
Pu239+240
Am241
10
1
0.1
0.01
Median radionuclide concentration, Bq g-1 dry wt.
0
10
20
30
40
50
60
Soil depth, cm