

Fig. 88. Alr-sample station location, Yvonne A.

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		Concentration, fCi/m ³ (standard error, %) ^a								
		7 _{Be}	40 _K	⁵⁴ Mn	⁹⁵ Zr	¹⁰⁶ Ru	137Cs	¹⁴⁴ Ce	239, 240 _{Pu}	²³⁸ Pu Other ^b
FRED	UH3	-NDET	-NDET	0.38±10	0.11±20	<0.25 NDET	0.21±19	0.5±17	< 0.003 NDET	-NDET
	UH5	94 ±4	-NDET	0.6±12	0.3±20	1.3 ± 32	0.39 ± 17	1.1±18	0.0067 ± 12	-NDET
	UH6	81±10	4.5±16	0.23 ± 20	0.3 ± 26	1.0±35	0.41±18	1.9±19	0.0086 ± 6	-NDET
	UH7	58±3	-NDET	0.22±14	0.12±18	.12±18 -NDET		0.36 ± 17	-NDET	-NDET
	UH8	40±25	10±32	0.8±20	-NDET	<1.5 NDET <	< 0.17 NDET - NDET <0.036 NDET 0.23±36		0.0032 ± 26	0,0028±22
	UH9	32±10	-NDET	0.14 ± 25	-NDET	< 0.29 NDET			0.0012±13	-NDET
	UH10	95±3	-NDET	-NDET	0.08±14	0.42 ± 20	0.43±5	0.22±11	0.003 ± 21	-NDET
	UH11	110 ± 50	5.4 ± 24	-NDET	-NDET	1.6 ± 32	0.34 ± 30	0.83±26	0.012 ± 20	-NDET
	UH12	6±10	-NDET	-NDET	0.03±22	< 0.2 NDET	0.13±18	0.28±16	<0.03 NDET	-NDET
	VC11	-NDET	-NDET	-NDET	-NDET	-NDET	-NDET	-NDET	-NDET	-NDET -
	VC21	116 ± 50		1.9±42			1.2±41			
	VC12	81±34		4.0±30			-NDET			
	VC22	70±50		1.3±36			2.5±19		¥	
	A11A	52±50		-NDET			-NDET		0.017 ± 22	
	A11B	-NDET	¥						0.005 ± 25	
	A11C		1.6±18						-NDET	
	A11D		-NDET							
	A11E	¥	27 ± 10	V						
	A12A	43±50	-NDET	0.4 ± 28						
	A12B	-NDET	7.7±32	0.4±28						
	A12C		15±14	0.5±23						
	A12D		6.0±40	0.5±25						
	A12E		-NDET	-NDET	¥	¥	¥	¥		¥
DAVID	UH1	38±5	-NDET	0.3±14	-NDET	< 0.29 NDET	0.15±23	-NDET	0.025 ± 6	- NDET
	UH4	-NDET	-NDET	0.4±12	0.2±30	<0.46 NDET	0.17 ± 32	0.4 ± 27	0.024 ± 7	0.008±9

Table 105. Radionuclide concentrations in Enewetak air samples,

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Table 105 (continued).

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		Concentration, fCi/m ³ (standard error, %) ^a									
		⁷ Be	40 _K	⁵⁴ Mn	⁹⁵ Zr	¹⁰⁶ Ru	137 Cs	¹⁴⁴ Ce	239,240 _{Pu}	²³⁸ Pu Other ^b	
JANET	UH21	41±15	-NDET	-NDET	-NDET	<0.45 NDET	2.1±7	-NDET	0.006 ± 16	0.007 ± 11	
	UH22	-NDET	-NDET	-NDET	-NDET	< 1.0 NDET	0.44±33	-NDET	<0.006 NDET	-NDET	
	UH23	22±10	9.2±24	1.3±10	-NDET	< 0.9 NDET	0.71 ± 17	-NDET	<0.008 NDET	-NDET	
SALLY	UH24	53±18	-NDET	-NDET	-NDET	<0.65 NDET	0.66±19	-NDET	0.005±21	-NDET	
	UH25	60±2	-NDET	-NDET	0.2±18	<0.34 NDET	0.34±13	1.5±12	0.0011±19	-NDET	
YVONNE	UH2 6	167±9	-NDET	-NDET	-NDET	<0.86 NDET	0.49±24	2.5±23	1.8±5	0.04 ± 9	
	UH27	193±2	-NDET	-NDET	0.4 ± 6	1.6 ± 22	0.82±5	3.7 ± 7	2.6 ± 13	<0.14 NDET b	
	UH28	143 ± 22	22±25	-NDET	-NDET	< 3.3 NDET	<0.58 NDEF	-NDET	1.1 ± 12	0.13±13 b	
	VC31	-NDET	25±37	1.1 ± 43		-NDET	-NDET	-NDET	0.49±9	- NDET	
	VC41		-NDET	1.5 ± 34					-NDET		
	VC32	*	4.2±23	-NDET					0.033 ± 14		
	VC42	190 ± 50	-NDET	2.1±23	2	★			-NDET		
	A21A	152 ± 50		0.6±34		2.6±66			0.18±25		
	A21B	-NDET	32±11	-NDET		-NDET			-NDET		
•	A21C		16 ± 24								
	A21D		-NDET								
	A21E	V	17 ±1 9	*							
	A22A	41 ± 15	21 ± 12	0.5±30					0.011±22		
	A22B	7.5±60	15±17	0.5 ±35					0.01±18		
	A22C	-NDET	15 ± 25	0.5±43					-NDET		
	A22D		-NDET	-NDET					0.074 ± 9		
	A22E	V	-NDET	-NDET	<u> </u>	<u> </u>	¥		0.022±12		

^aNDET = Not detected (when preceded by a dash, the limit of sensitivity was not established.)

^b241_{Am} (0.30±32); ¹²⁵Sb (0.27±24); ¹⁰³Ru (5.5±17).

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Vuclide	YVONNE	Remainder of Enewetak Atoll	Livermore, Calif., 1972	Balboa, Panama, 9°N 79°W, 1972-1973
Ве	<49-193	< 6-116	90-250	43-143 [°]
4 Mn	< 0.6-2.1	< 0.14-4.0	-	-
⁹⁵ Zr	<0.4-0.4 ^a	0.03-0.3	0.005-0.4	< 0.9-8.5
¹⁰³ Ru	<5.5-5.5 ^a	NDET ^b	0.29-3.4	-
125 _{Sb}	<0.27-0.27 ^a	NDET	0.04-0.23	-
¹⁰⁶ Ru	<0.9-2.6	< 0.2-1.6	0.14-2.9	-
137 _{Cs}	<0.49-0.82	< 0.04-2.5	0.63-3.2	0.09-1.7
¹⁴⁴ Ce	< 2.5-3.7	< 0.22-1.9	0.24-3.1	0.7-11.2
239,240 _{Pu}	< 0.03-2.6	<0.001-0.025	0.01-0.05	<0.001-0.030
²³⁸ Pu	< 0.04-0.13	<0.0028-0.008	0.001-0.005	<0.001-0.003
²⁴¹ Am	<0.3-0.30 ^a	NDET	NDET	NDET

Table 106. Comparison of radionuclides in surface air (fCi/m^d) on Enewetak, Livermore, California, and Balboa, Panama.

^aDetected only one sample.

^bNot detected.

^COct. -Dec. 1972 range.

fCi/1 (see the marine survey data). If we assume the total 40 K (excepting the filter material 40 K) is a normal isotopic constituent of ocean water, then we can calculate an average air mass loading equal to 2 mg/m³. This unusually high mass loading partially clogged the filter media during the sampling. If this total airborne salt is from CACTUS crater, then only 0. 012 fCi/m³ of 239 Pu can be contributed to the 1.1 fCi/m³ found in sample UH28. We must conclude that another surface source exists because the oceanborne contribution cannot be any higher than 0.1% of the total.

Some observations regarding the climatic conditions which existed during the survey may be appropriate at this point. As is shown in Table 104, most of the air samples were taken during the period from November 28 to December 19 (only two samples were taken on FRED and two on DAVID before typhoon OLGA struck). Wind speeds were almost always greater than 10 knots and often greater than 20 knots at all sampling locations. In addition, frequent light rain showers served to keep the ground surface damp. Table 107 presents climatological data which have been published for Enewetak

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'NDET = Not detected (when preceded by a dash, the limit of sensitivity was not established. ^b241Am (0.30±32); ¹²⁵Sb (0.27±24); ¹⁰³Ru (5.5±17). and Kwajalein. It is apparent that December represents a fairly average month so far as total rainfall and rainfall frequency are concerned, while average wind speeds are higher than those observed most of the year.

JANET

Three UHVS 24-hr samples were obtained on JANET from December 4 to 9. Wind speeds ranged from 10 to 20 knots, and rainfall during this sampling period was higher than for most days in the Atoll during the survey. Using information from the aerial survey, a sampling location was chosen in the area of highest activity (Fig. 85). This area contained surface contamination in soil, in pCi/g, as follows: 137 Cs (av 15, range 0.6-180), 239 Pu (av 8.5, range 1-170), and 60 Co (av 2.0, range 0.1-6).

SALLY

Two UHVS 24-hr samples were obtained on SALLY from December 13 to 15. Rainfall was very low in this interval, and winds were stronger than usual. The sampling location was chosen on the basis of the aerial survey (Fig. 86). Subsequent to the sampling, it was learned that the sampling location was an old Radiation Exclusion (Radex) area which contained surface contamination in soil, in pCi/g, as follows: 137 Cs (av 3.7, range 0.4-30), 239 Pu (av 7, range 0.2-130), and 60 Co (av 0.7, range 0.1-69).

YVONNE

Air sampling using UHVS's, VCS's, and ACI's was carried out from December 3 to 19, 1972. The portable UHVS was fielded for three days in the area of highest plutonium surface activity recorded for YVONNE (Figs. 87 and 88). The surface soil has been described previously in connection with the soilsampling program.

Winds were generally high and gusty during the sampling of YVONNE, and light daily rainfall was frequent. Air was sampled downwind from CACTUS crater on December 17-19, using the UHVS.

Results and Conclusions

A number of radionuclides were detected in the surface air of Enewetak Atoll, including 7 Be (53 d), 40 K (1.26 10^9 y), 54 Mn (303 d), 95 Zr (65 d), 103 _{Ru} (39. 6 d), 106 Ru (1 y), 125 Sb (2. 7 y), 137 Cs (30 y), and 239 Pu (2.4 10⁴ y). ²³⁸Pu (86 y), and ²⁴¹Am (458 y). Data for all samples collected are shown in Table 105. ⁷Be and 40 K are naturally occurring activities. ⁵⁴Mn, ⁹⁵Zr, ¹⁰³Ru, 106 Ru, 125 Sb, and 144 Ce are intermediatelived activation and fission products found in current worldwide fallout, but present in Enewetak soils in only very reduced quantities due to radioactive decay over the long period since testing ended. Longer-lived ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu and ²⁴¹Am in air could result from either local resuspension or from worldwide fallout.

The natural ⁷Be provided convenient order-of-magnitude verification of the accuracy of air volume measurement. ⁷Be is formed by cosmic-ray interactions with ¹⁴N in the troposphere^{*} and is found

P. F. Gustafson, M. A. Kerrigan, and S. S. Bar, "Comparison of Be7 and Cs¹³⁷ Radioactivity in Ground Level Air," <u>Nature 191</u>, 454 (July 29, 1961).

			Perc	entage	oi tot	ai tim	ie at e	ach w	ind-sp	eed ir	iterval			
	Jan	Feb	Mar	<u>Apr</u>	<u>May</u>	June	July	Aug	Sept	Oct	Nov	Dec	Av	
Wind speed, knots)													
0-3	1	1	1	0	1	1	6	10	16	9	3	1	4.2	
÷-10	15	12	22	20	2 7	27	49	6 0	59	63	÷2	20	34.7	
::-21	68	80	70	75	69	70	44	29	24	28	53	70	56.7	
22-33	15	7	7	5	3	2	1	1	1	0	2	9	4.4	
<u>~ 33</u>	1	0	0	0	0	0	0	0	0	0	0	0	0	
Prevailing wind														
direction and	NE	NE	NE	NE	NE	NE	E/NE	Е	NE	NE	NE	NE		
frequencyb	86%	87%	81%	77%	6 7 %	64%	36% each	31%	27%	33%	55%	74°°0		
Precipitation ^C										<u> </u>			Yr	Yr. of record
Av. amount, in.	1.02	1.84	1.86	1.28	4.57	3.37	6.45	6.81	6.24	9.09	6.30	2.63	51.46	30
Greatest amount, in.	1.95	10.21	7.33	3.86	8.38	7.03	15.35	14.41	13.17	18.07	17.38	9.18	69.86	13
Least amount, in. Mean number of	0.12	0.40	0.37	0.49	0.37	1.33	1.36	4.22	1.53	2.60	1.94	0.86	24.42	13
days, 0.01 in. or more.	11	10	13	13	16	16	21	21	20	21	21	16	198	10

Table 107. Climatological data for Kwajalein and Enewetak^a.

^aU. S. Hydrographic Office, <u>Sailing Directions for the Pacific Islands</u>, H. O. Pub. No. 82, Vol. 1, Second Edition (1964), updated to Dec. 5, 1970.

^bWind data for Kwaialein.

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^cPrecipitation data for Enewetak.

in surface air in concentrations of approximately 100 fCi/m³.[†] We compared our Enewetak data with Livermore, Calif. data^{**} (typically higher due to slower rainout of the condensation nuclei), and with data taken at Balboa, Panama,

which is at nearly the Enewetak latitude. ^{††} The range of ⁷Be values for Enewetak Atoll, Balboa, Panama, and Livermore, Calif. are in reasonable agreement (Table 106).

[†]One femtocurie (fCi) equals 10⁻¹⁵ curie, or 0.0022 disintegration per minute.

P. H., Gudiksen, C. L. Lindeken, J. W. Meadows, and K. O. Hamby, <u>Environmental Levels of Radioactivity</u> in the Vicinity of the Lawrence Livermore Laboratory, Lawrence Livermore Laboratory, Rept. UCRL-51333 (1973).

^{††}Health and Safety Lab., HASL-276, Appendix, Fallout Program, New York, Oct. 1, 1973.

ENGINEERING SURVEY --RADIOLOGICAL ASPECTS O. D. T. Lynch, Jr. Nevada Operations Office U. S. Atomic Energy Commission Las Vegas, Nevada

Purpose and Scope

As part of the Enewetak precleanup survey, the Defense Nuclear Agency (DNA) contracted with Holmes and Narver, Inc. (Hand N) to conduct an engineering survey of Enewetak Atoll in December 1972. The purpose of this survey was to identify and examine all existing structures now on the Atoll, provide their descriptions, and develop cost estimates for removal of such structures as part of the cleanup effort. An additional purpose was to develop plans for such a cleanup and the necessary support, including the setup of a large camp facility required to implement cleanup activities.

Each island was visited by the engineering team, and each structure was located, examined, categorized, and indicated in the notes and on the drawings. The results of this engineering effort were reported to DNA.^{*}

Radiological support was provided to the engineering survey by a team of AEC and EPA personnel. The purpose of the radiological effort was twofold:

- To provide radiological safety support to the engineering team on those islands which had known or suspected radiological hazards.
- To survey, evaluate, and report the radiological conditions of the structures and scrap on these islands.

Islands Requiring Radiological Support

The islands for which radiological support was required and for which measurements were reported were: ALICE, BELLE, CLARA, DAISY, EDNA IRENE, JANET, PEARL, SALLY, and YVONNE. Of these ten islands, five had surface ground zeros (SGZ) and one, EDNA, was little more than a sandbar. The islands of IRENE, JANET, PEARI. SALLY, and YVONNE had SGZ's and thus had possible radiological hazards. The remaining five islands had received heavy, close-in fallout.

Instrumentation

Since the purpose of the radiological support was to survey structures and scrap, not areas, and to provide radiological safety services, instrumentation specific for that purpose was used. A detailed description of each instrument is provided in the Terrestrial Soil and Radiation Survey chapter of this report and will not be discussed here. However, it is appropriate to identify at this point the instrumentation used and the reason for the selection.

During the engineering survey effort the only alpha survey meter available for field use was the PAC-IS. This instrument was used only on those islands where surface ground zeros were located. Since this survey was performed at the end of the rainy season, alpha emissions were effectively masked by the moisture on structures and scrap surfaces. For this reason, the alpha survey meter was really only useful for personnel monitoring prior to leaving a contaminated island, particularly YVONNE.

Beta-gamma radiation detection was

^{*}Engineering Study for a Cleanup Plan, Enewetak Atoll - Marshall Islands, Holmes and Narver, Inc., Repts. HN-1348.1 and HN-1348.2 (1973).

provided by the E-500B and Ludlum Model 3 survey meter with pancake probe. The E-500B was used for high-range radiation levels (greater than 3mR/hr) and the Ludlum for lower levels of beta and low-energy gamma-radiation emitters on scrap metal and structures.

Low-level gamma exposure rates (less than 3mR/hr) were measured with the Baird-Atomic scintillator.

Execution of the Radiological Support Effort

The engineering survey effort was carried out by Holmes and Narver, Inc., under the direction of Earl Gilmore, Project Manager, H and N, Las Vegas, Nevada. The radiological support effort was directed by O. D. T. Lynch, Jr., NVOO, USAEC who was assisted by William E. Moore, James R. Martin, Rex Price, and Jack Thrall of EPA, Las Vegas.

Radiological survey measurements of structures and scrap metal were recorded directly on as-built drawings provided by H and N. These drawings were also used by the engineering team to locate the structures they were examining.

As a part of the radiological support provided to the engineering survey, single profile soil samples were collected on each of the following islands: IRENE, JANET, PEARL, SALLY, ALICE, BELLE, CLARA, and DAISY. These soil samples were taken to a depth of 40 to 60 cm from contaminated areas noted during the November aerial radiological survey. The results of analyses of these profile samples are included in the Terrestrial Soil and Radiation Survey data. The locations of these special soil sample profiles are indicated on the "f" series of figures in Appendix II.

Radiological Results

As a result of the radiological monitoring and safety support, none of the team members received any significant external exposure to radiation. Subsequent urine samples and whole-body counts from selected members of the monitoring team indicated that no detectable exposure was received due to internal deposition of radionuclides.

Scrap and Structure Survey

Contaminated structures and activated/ contaminated scrap were found on a number of islands. The locations of this scrap and the contact exposure rates measured are indicated on the as-built drawings that follow (Figs. 89 through 113, inclusive). Area exposure rates and approximate isopleths are also shown, so that a simple comparison can be made between scrap radiation levels and the surrounding "background."

In many cases, the contact exposure rate was not significantly different from the surrounding area exposure rate. In this situation, the determination of whether or not the scrap was contaminated was inconclusive. This determination could be made only if the scrap were to be removed from the high background area and resurveyed. Such a procedure was not considered warranted at this time. Rather, it is suggested that the scrap be assumed contaminated if it rests in an area where exposure rates are, say, greater than 100 μ R/hr.

Radioactive scrap conditions are summarized in Table 108, on an island-byisland basis. In general, the scrap found on ALICE, BELLE, CLARA, DAISY, and EDNA is apparently not con-

Island	Relative scrap quantities	Scrap radioactivity	Remarks
ALICE	Significant	Apparently not con- taminated above background.	Background is up to 170 μ R/hr. An M-boat wreck on beach reads 8 mR/hr.
BELLE	Insignificant	Apparently not con- taminated above background.	Background up to 250 $\mu R/hr$.
CLARA	Insignificant	Apparently not con- taminated above background.	Background up to $100 \ \mu R/hr$.
DAISY	Insignificant	Apparently not con- taminated above background.	Background up to 140 $\mu R/hr$.
EDNA	None	Not applicable.	Sandbar
JANET	Large	Up to 8 mR/hr.	Activated scrap metal in all sizes can be found in piles or indi- vidual pieces scattered over the island.
PEARL	Small	Up to 5 mR/hr.	Confined to SGZ area.
SALLY	Large	Scrap metal up to 120 μ R/hr; concrete surfaces, alpha to 10 ³ dpm/50 cm ² .	Most scrap metal is apparently not con- taminated. Several structures contain plutonium-contaminated debris.
YVONNE	Large	Activated/contaminated to 60 mR/hr.	Most scrap metal is activated or contam- inated. Also much plutonium contamina- tion.

Table 108. Radioactive scrap conditions by island.



Fig. 89. Scrap and structure radiation measurements, ALICE, WEST.

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Fig. 90. Scrap and structure radiation measurements, ALICE, EAST.

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Fig. 91. Scrap and structure radiation measurements, BELLE.

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Fig. 92. Scrap and structure radiation measurements, CLARA.

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Fig. 93. Scrap and structure radiation measurements, DAISY.

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Fig. 94. Scrap and structure radiation measurements, HELEN and IRENE.

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Fig. 95. Scrap and structure radiation measurements, HELEN and IRENE.

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Fig. 96. Scrap and structure radiation measurements, HELEN and IRENE.

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Fig. 97. Scrap and structure radiation measurements, JANET.



Fig. 98. Scrap and structure radiation measurements, JANET.

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Fig. 99. Scrap and structure radiation measurements, JANET.

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Fig. 100. Scrap and structure radiation measurements, JANET.

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Fig. 101. Scrap and structure radiation measurements, JANET.

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Fig. 102. Scrap and structure radiation measurements, JANET.

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Fig. 103. Scrap and structure radiation measurements, JANET.

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Fig. 104. Scrap and structure radiation measurements, PEARL.



Fig. 105. Scrap and structure radiation measurements, PEARL.

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Fig. 106. Scrap and structure radiation measurements, RUBY and SALLY.

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Fig. 107. Scrap and structure radiation measurements, RUBY and SALLY.

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Fig. 108. Scrap and structure radiation measurements, RUBY and SALLY.

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Fig. 109. Scrap and structure radiation measurements, YVONNE.

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Fig. 110. Scrap and structure radiation measurements, YVONNE.

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Fig. 111. Scrap and structure radiation measurements, YVONNE.

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Fig. 112. Scrap and structure radiation measurements, YVONNE.

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Fig. 113. Scrap and structure radiation measurements, YVONNE.

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taminated above the area background levels (up to 250 μ R hr), if at all. The only exception appears to be the wrecked M-boat on the east end of the lagoon beach of ALICE. The contact exposure rate measured on this scrap was about § mR/hr.

The island of JANET has large quantities of radioactive scrap metal and debris scattered all over it. Contact exposure rate measurements of up to 8 mR/hr were observed at the old ITEM SGZ on the north end of the island. Near the EASY SGZ, scrap piles and individual pieces of metal read several mR/hr. Structures on the island exhibit some residual surface contamination (below 100μ R/hr) which seems to be on the SGZ-oriented and upper surfaces of the concrete.

PEARL has a small quantity of scrap material, all near the SGZ. The metal is measurably activated, with contact measurements of up to 5 mR/hr. The concrete blocks near the SGZ also exhibit surface contamination of several hundred μ R/hr.

SALLY has large quantities of scrap, most of which is near the shoreline on the northern sides of the islands. Most of this is apparently not contaminated. On the other hand, several small concrete blocks and other structures apparently contain plutonium-contaminated debris. At least six such structures have been identified.

The most contaminated island in the Atoll is YVONNE. Large quantities of scrap metal are found on the beaches, the ocean reef, and in the interior of the island. Nearly all of this scrap is activated and/or contaminated. A very large pile of scrap metal near the ERIE SGZ, just north of the airstrip, exhibited contact readings of 60 mR/hr.

Concrete structures on YVONNE, north of the airstrip, exhibit surface contamination with levels of several hundred $\mu R/hr$. South of the airstrip, scrap metal and structures do not appear to be contaminated.

Limitations on Results

Although the H&N Engineering Survey was thorough, there are several limitations which must be placed on any interpretation of this evaluation of radioactive scrap and structures:

- It must be kept in mind that the survey covered only structures and scrap which were on the surface, visible and accessible. No attempt was made to search for any buried scrap or unknown structures. No known buried contaminated debris was unearthed or surveyed.
- Except for grossly obvious structures, only structures which appeared on the H&N as-built drawings were examined.
- If a precise estimate or evaluation of the amount of radioactive scrap is desired, it must be realized that it was not possible to survey each piece of scrap nor seek out the location of all scrap piles. Therefore, additional radioactive scrap may still be hidden in the dense vegetation.

Conclusion

Scrap-metal debris found on those islands which did not have surface ground zeros is probably not contaminated to any significant degree. The only exception to this would be the wrecked M-boat on ALICE, which presumably drifted there

after a test.

Scrap metal on those islands which had SGZs was found to be radioactive to some degree. Some of this scrap was contaminated or activated to levels as high as 60 mR/hr.

The possibility of having buried scrap is very real on the SGZ islands. It should not be overlooked when developing cleanup estimates for Enewetak Atoll.

ANALYTICAL PROGRAM

R. W. Hoff, J. W. Meadows, H. D. Wilson, A. L. Prindle, R. Gunnink, and K. O. Hamby Lawrence Livermore Laboratory Livermore, California

Introduction

More than 5000 samples were collected during field operations of this survey, and approximately 4500 samples were selected for analysis. This chapter describes the analytical program required to provide measurements of significant nuclides in these samples. A breakdown of the samples submitted for analysis is given in Table 109a.

Sample treatment can be described in a general way as consisting of three phases: initial processing, gamma counting, and so-called "wet-chemistry" analyses. The latter phase involved dissolution of a sample, followed by chemical isolation of specific elements and radiation counting of elemental samples.

For most samples, initial processing consisted of selecting appropriate parts of a field sample, drying, homogenizing, and packaging. The selection process was provided by dissection of fish, rats, birds, etc., whereas it was unnecessary for soil samples. Drying was accomplished by heating in ordinary ovens for soils, vegetation, and fish specimens, or by the freeze-dry process for rat, bird, and crab samples. During initial processing, samples were converted to forms appropriate for gamma counting.

All of the samples were counted on Ge(Li) detector systems to determine their gamma-emitting constituents.

Most of the samples were put through a wet chemical analysis, either by destructive analysis of a sample which had been gamma-counted first (as was the case for most of the fish, vegetation, animal and air-filter samples) or by submission for wet chemistry of a separate aliquot of sample (as was the case for most of the soil samples). The latter approach required reasonably homogeneous samples at the end of initial preparation. Minor exceptions to this general scheme of treatment are the seawater samples. where extensive chemical separation procedures were required during initial preparation before any gamma counting could be performed. Wet chemical analyses were needed to measure concentrations of certain nuclides that cannot be detected with acceptable sensitivity by gamma counting; examples of such nuclides are ²³⁹Pu and ²⁴⁰Pu, predominantly alpha emitters (with almost identical energies), and ${}^{90}\text{Sr}{}^{-90}\text{Y}$, both beta emitters with no accompanying gamma radiation.

Because complete analysis of these 4500 samples was a very large undertaking, scientists from a number of organizations participated in the analytical program. A listing of these organizations and some of the scientists who were

after a test.

Scrap metal on those islands which had SGZs was found to be radioactive to some degree. Some of this scrap was contaminated or activated to levels as high as 60 mR/hr.

The possibility of having buried scrap is very real on the SGZ islands. It should not be overlooked when developing cleanup estimates for Enewetak Atoll.

ANALYTICAL PROGRAM

R. W. Hoff, J. W. Meadows, H. D. Wilson, A. L. Prindle, R. Gunnink, and K. O. Hamby Lawrence Livermore Laboratory Livermore, California

Introduction

More than 5000 samples were collected during field operations of this survey, and approximately 4500 samples were selected for analysis. This chapter describes the analytical program required to provide measurements of significant nuclides in these samples. A breakdown of the samples submitted for analysis is given in Table 109a.

Sample treatment can be described in a general way as consisting of three phases: initial processing, gamma counting, and so-called "wet-chemistry" analyses. The latter phase involved dissolution of a sample, followed by chemical isolation of specific elements and radiation counting of elemental samples.

For most samples, initial processing consisted of selecting appropriate parts of a field sample, drying, homogenizing, and packaging. The selection process was provided by dissection of fish, rats, birds, etc., whereas it was unnecessary for soil samples. Drying was accomplished by heating in ordinary ovens for soils, vegetation, and fish specimens, or by the freeze-dry process for rat, bird, and crab samples. During initial processing, samples were converted to forms appropriate for gamma counting.

All of the samples were counted on Ge(Li) detector systems to determine their gamma-emitting constituents.

Most of the samples were put through a wet chemical analysis, either by destructive analysis of a sample which had been gamma-counted first (as was the case for most of the fish, vegetation. animal and air-filter samples) or by submission for wet chemistry of a separate aliquot of sample (as was the case for most of the soil samples). The latter approach required reasonably homogeneous samples at the end of initial preparation. Minor exceptions to this general scheme of treatment are the seawater samples, where extensive chemical separation procedures were required during initial preparation before any gamma counting could be performed. Wet chemical analyses were needed to measure concentrations of certain nuclides that cannot be detected with acceptable sensitivity by gamma counting; examples of such nuclides are ²³⁹ Pu and ²⁴⁰ Pu. predominantly alpha emitters (with almost identical energies), and 90 Sr- 90 Y, both beta emitters with no accompanying gamma radiation.

Because complete analysis of these 4500 samples was a very large undertaking, scientists from a number of organizations participated in the analytical program. A listing of these organizations and some of the scientists who were w.

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responsible for this analytical program are as follows: Messrs, E. L. Geiger and E. A. Sanchez, Eberline Instrument Corporation (EIC), Santa Fe, New Mexico: Messrs, W. J. Major, R. A. Wessman. and L. Leventhal, Laboratory for Electronics, Environmental Analysis Laboratories Division (LFE), Richmond, California; Lawrence Livermore Laboratory (LLL), Livermore, California; Drs. V. A. Nelson, W. R. Schell, and A. H. Seymour, Laboratory of Radiation Ecology (LRE), University of Washington, Seattle, Washington; and Colonel R. C. McBryde, Major W. A. Myers, Major W. A. Rush, Captain J. R. Baca, and Captain H. T. Hawkins, McClellan Central Laboratory (MCL), Sacramento, California (a U. S. Air Force organization). A listing of laboratory participation in each phase of the analytical program is given in Table 109a. Initial processing of samples was performed at LLL and at LRE. All gamma counting was done at LLL. The wetchemistry effort was split between MCL.

LFE, EIC, and LRE, with some special analyses performed at LLL.

Samples were initially numbered at Enewetak at the time of collection. In addition, personnel working at a samplereceiving station on Enewetak assigned each sample a survey ID number according to the scheme listed in Table 109b. The first two digits of this number indicate type of sample, the next four digits are sequential and identify a sample uniquely, and the last two digits identify the island (or area nearby) from which a sample was collected. Thus, each sample usually had two numbers associated with it. the survey ID number and a field-collection number. The latter category is comprised of numbers chosen by different fieldcollection teams and with varying formats. Considerable care was exercised in identifying samples; instances of confusion were relatively rare. The analytical data for each sample are identified by the appropriate survey ID number. Data for each sample are listed in Appendix II on microfiche.

Sample type	Numb er of samples analyzed	Initial processing	Gamma counting	Wet chemistry
Soils, total		LLL	All samples	MCL, LFE, EIC
Surface, 0-15 cm (and 0-5 cm)	870		were gamma counted at LLL.	-
Profile samples	2135			
TLD samples	14			
Biota group samples	20			
Standards, backgrou samples	nd 18			
Sediments	} 345	LLL		MCL, LFE, EI(
Cores	} 345	LLL		MCL, LFE, EI(
Marine samples	410	LRE		MCL, LFE, LR
Invertebrates and vertebrates				MCL, LPL, LK
Algae	3	LRE		LFE
Plankton	16	LLL		MCL
Vegetation (terrestrial)	216	LLL		MCL, LFE
Animals (terrestrial)	274	LLL		MCL, LFE
Rats, crabs, birds, eggs, etc.				
Seawater	54	LLL		LLL, LFE
Hydroxide fraction, Cs fraction— gamma counting				
Pu, Sr fractions	-			
Freshwater	4	LLL		MCL, LFE
Water samples, distillation, plant residue				
Air s a mples	67	LLL		MCL
High volume (20), low volume (23), Anderson cascade impactor (24)	e			
Seawater fil t ers, UW	28	LRE		None required
Total samples analyzed	4,474			

Table 109a. Enewetak sample analysis — sample listing and laboratories.

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Table 109b. Survey ID number scheme.

General f	orm:	AB-1	XXX	Χ-	CD
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where AB are two digits which indicate sample type.

- XXXX are four digits which were assigned sequentially and which identify the sample uniquely, and
- CD are two digits which indicate location of sample collection.

Specific example: 09-0577-20

- where 09 indicates a marine sample, vertebrate (in fact, the sample is bone from a snapper fish),
 - 0577 identifies this sample, and
 - 20 indicates collection in proximity of TILDA.

Sample-type identifier, first two digits:

01 General soil classification, used where depth information is not known or for biota soil samples.

Soil samples, classified according to depth:

29	0-10 cm	44	75-85 cm	74	40-50 cm
30	0-2 cm	45	85-95 cm	75	50-60 cm
31	0-5 cm	46	95-105 cm	76	60-70 cm
32	0-15 cm	47	105-115 cm	77	70-80 cm
33	2-5 cm	48	115-125 cm	78	80-90 cm
34	5-10 cm	49	125-135 cm	79	90-100 cm
35	1 0- 15 cm	50	135-145 c m	80	100-110 cm
36	15-20 cm	51	145-155 cm	81	110-120 cm
37	15~25 cm	52	155-165 cm	82	120-130 cm
38	20-25 cm	53	165-175 cm	83	130-140 cm
39	25-35 cm	54	175-185 cm	84	1 40-150 cm
40	35-45 cm	70	0-10 cm	85	150-160 cm
41	45-55 cm	71	10-20 cm	86	160-170 cm
42	55-65 cm	72	20-30 cm	87	170-180 cm
43	65-75 cm	73	30-40 cm	88	180-190 cm

Table 109b (continued)

- 02 Algae
- 03 (designation not in use)
- 04 Plankton
- 05 Samples from lagoon floor, sediments, cores, dredge samples, etc.
- 06 Seawater
- 07 Coral (pieces broken from living coral heads)
- 08 Marine, invertebrate
- 09 Marine, vertebrate
- 10 Vegetation
- 11 Animal, terrestrial
- 12 Potable water
- 13 Air, high-volume sampler
- 14 Air, low-volume sampler
- 15 Air, Anderson cascade impactor

Location identifier, last two digits:

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01	ALICE	19	SALLY	37	FRED
02	BELLE	20	TILDA	38	GLENN
03	CLARA	21	URSULA	39	HENRY
04	DAISY	22	VERA	40	IRWIN
05	EDNA	23	WILMA	41	JAMES
06	FLORA	24	YVONNE	42	KEITH
07	(Mike crater) GENE	2 5	(not in use)	43	LEROY
08	HENRY	26	SAM	44	МАСК
09	IRENE	27	ТОМ	45	OSC AR
10	JANET	28	URIAH	46	LLL Whaler
11	KATE	29	VAN	47	LCU, Navy vessel
12	LUCY	30	ALVIN	48-51	(not in use)
13	PERCY	31	BRUCE	52	Palumbo, AEC research vessel
14	MARY	32	CLYDE	53	Wide passage
15	NANCY	33	DAVID	54	Deep passage
16	OLIVE	34	REX	60	Kwajalein
17	PEARL	35	ELMER	61	Meck-Kwajalein
18	RUBY	36	WALT	62	Enewetak-Kwajalein
				70	Midway island
				77	Ujilang atoll
 				78	Sacramento, California

Initial Processing of Field Samples

Soil and Sediment Samples

This class of sample, by far the largest category, was treated at LLL in a part of Building 412 devoted entirely to this task. The treatment consisted of drying, pulverizing, blending, packaging, and doing a preliminary gamma count. With sufficient sample, three packages were produced, an aluminum "tuna can" containing 300-350 g and two vials containing 50 g each.

The facility was set up and equipped in the following manner. Since the area used for this work is adjacent to a hotcell facility, and although this area had been used very little in the past two years. the laboratory space was carefully surveyed for possible radioactive contamination. Swipe samples were taken from the floors, and particulate matter in the air was collected on small filters. These samples were checked for 60 Co, 137 Cs, and $^{239+240}$ Pu content: there was no detectable contamination. The area was considered suitable for initial processing of soils. This monitoring program was continued throughout operation of the facility; activity above background levels was detected in only a few instances.

Drying ovens were designed and built to permit initial drying of samples at ~70°C. Two ovens were constructed of asbestos board with steel shelves inside; two 300-W air heaters were used to blow warm air into each unit, along with a fan in the vent pipe. Final drying was accomplished in a large commercial drying oven at 150°C.

Grinding of samples was accomplished by placing a sample in a 1-gal paint can along with a number of 1-in. steel balls. The cover of each can was taped securely; then the entire can was covered with a galvanized-steel jacket which was held in place by two large rubber "O" rings. This arrangement eliminated problems encountered early in the operation when can lids fell off during ball milling. Machines were built to permit rolling of 48 samples at a time and were usually operated overnight to provide 15-24 hr of grinding.

Packaging, weighing, and labeling of samples were performed by hand. Within the laboratory space there were three hoods which provided a flow of air into and up the hood. All work with finely divided soil was performed in these hoods. Before each sample was packaged, clean paper was laid out on the hood bench. Care was taken to prevent crosscontamination of samples. A series of low-level coral soils was treated at various times during operation of the facility; results of these background samples are presented in the quality control section of this chapter.

The following is a detailed description of the operations in the Building 412 facility:

• The samples were first unpackaged from the shipping container and logged. Notes were taken on the appearance of each sample (e.g., amount of organic matter, color, presence of large chunks, etc.). If samples contained appreciable water (e.g., certain sediments were quite wet), the solid material was allowed to settle, and the water was carefully decanted without loss of fine particles.

- The samples were then transferred to a disposable aluminum cake pan and covered with aluminum foil. Holes were punched in the top of the foil to permit evaporation.
- The samples were then transferred to preliminary drying ovens that were designed to handle about 200 samples. These ovens were set at a temperature of ~70°C and run continuously. The average residence time per sample was 48 hr.
- To assure that the samples reached complete dryness, a second oven was used. This oven was set at ~150°C, and the sample residence time averaged ~ 3 hr.
- The samples were then transferred to a 1-gal paint can and a dry weight established. The weights of the samples varied from 100 g to 2 kg.
- The samples were then ball-milled using eight 1-in, steel grinding balls. The average sample residence time in the ball mill was $\sim 15-24$ hr.
- The finely ground soil^{*} was then prepared for gamma spectrometry and wet-chemistry analysis using two different containers. The gammaspectrometry samples consisted of tightly sealed tuna cans made of 0.25-mm thick aluminum. The large can was 3.9 cm high, 8.3 cm in diameter, with a cross-sectional area of 53.8 cm² and a volume of 210 cm³. The small can was 3.3 cm high, 6.0 cm in diameter,

with a cross-sectional area of 28.5 and a volume of 95 cc. Soil-sample weights in these cans ranged from 100 to 375 g.

The wet chemical samples consisted of two vials, each containing soil weighing ~50 g. One of the vials was shipped ou for chemical analysis, and one held as a backup sample.

The gamma-spectroscopy "tuna cans" were counted for gross gammas with a 3 × 3-in. NaI detector; a 512-channel NaI gamma spectrum was measured for those samples which exceeded 100 counts/min. These preliminary NaI data served as a guide in scheduling more precise measurements with Ge(Li) detectors.

Approximately 3400 samples were processed in the soil-preparation facility between November 15, 1972 and June 1, 1973 by an average working force of 4-1/2 people. We wish to acknowledge the dedicated effort of Messrs. Bern J. Qualheim and James S. Schweiger in supervising operation of the Building 412 facility. In addition to LLL personnel, two experienced technicians were supplied by Reynolds Electrical and Engineering Company (NTS) for this work.

Other Samples and Specimens

Initial processing of other samples. marine specimens, algae, plankton. vegetation, terrestrial animals, and air filters has been described in chapters which also describe collection of these samples in the field. For each type of sample, the product of this processing was a package suitable for gamma counting — either an aluminum "tuna can"

^{*}Finely ground soil is a goal which was not always attained. In a few cases, the presence of chunk of coral over 1 cm in diameter was reported by the participating analytical facilities.

where sufficient sample was available or, in the case of small samples, a plastic package which was a right circular cylinder. Because of the versatility of our gamma-spectroscopy data processing code, it was unnecessary to require that all of the small plastic packages be of standardized dimensions and they could be varied according to sample size.

Gamma Spectrometry

Gamma-spectrometric measurements were made on all samples at LLL. The work was accomplished by personnel in the Radiochemistry and Biomedical Divisions. In the Radiochemistry Division effort, 4100 samples (90%) were counted with eight Ge(Li) detector systems, three of which included an automatic samplechange feature. In the Biomedical Division effort, 400 samples (10%) were counted with four Ge(Li) detector systems. The latter systems were devoted to counting marine, vegetation, and animal samples, all of which required long counts (1/2 - 1 day each). Most of the largest sample category, soils, were counted with the Radiochemistry systems which had automatic sample changers; counting times were a minimum of 133 minutes each for the soils. All data were taken with single detectors; no anti-coincidence shielded detector systems were used to count samples in this survey.

Description of Equipment

The gamma counting in Radiochemistry Division was accomplished with a variety of Ge(Li)-diode detector systems which are listed in Table 110a. The diodes varied in volume from 19 to 50 cc. Three of the counting systems were automated. The automated systems, interfaced to a PDP-8 computer, were capable of handling 16 samples per system, thus allowing 24-hr/day counter use. The remainder of the systems could analyze one sample at a time, and the data were removed either by a manual dump onto a PDP-8 computer or by paper tape output. All data were transferred to magnetic tape and analyzed on a CDC-7600 computer as described later in this section under Identification of Nuclides.

The Biomedical Division Ge(Li) detector systems are listed in Table 110b. Data taken with these systems were transferred from memory storage in a pulse-height analyzer to magnetic tape. Analysis of the data was performed on a CDC-7600 computer with a separate code (ANALYSE 5) whose operation has been described by Phelps and Hamby^{*}.

Calibration of Detectors

It was necessary to calibrate each of the detector systems used on an absolute basis. During the course of the Enewetak survey, more than 20 different geometries were encountered. Several of these containers were checked for calibration on an individual basis, while others were submitted to the GAMANAL code (see paragraph below on Identification of Nuclides) as right circular cylinders. GAMANAL is capable of making the proper corrections on cylindrical geometries.

The majority of the samples were packaged in aluminum cans with nominal volumes of 95 and 210 cc. To check the

^{*}P. L. Phelps and K. O. Hamby, "Experience in the Use of an Anticoincidence Shielded Ge(Li) Gamma-Ray Spectrometer for Low Level Environmental Radionuclide Analysis", <u>IEEE</u> <u>Transactions on Nuclear Science</u>, <u>NS-19</u> (1), 155, (1972).

by Radi		
Detector and system identification	Description	
C2	Canberra 45-cc "down-looker," automatic counting chamber No. 2, PDP-8 control and dump.	-10 mbe
N1	Nuclear Diodes 40-cc "down-looker," automatic counting chamber No, 1, PDP-8 control and dump,	10
Τ3	Princeton Gamma Tech 50-cc "down-looker," automatic counting chamber No. 3, PDP-8 control and dump.	• 0 •••••
54	Nuclear Diodes 48-cc "up-looker, " manual change, PDP-8 control and dump.	N al
M5	LLL 19-cc "down-looker, " manual change, paper tape output.	
U8	Nuclear Diodes 25-cc "up-looker, " manual change, paper tape output.	carb mat€
W7	Nuclear Diodes 48-cc "side-looker, " manual change, paper tape output.	A sp good of ea
V7	Nuclear Diodes 48-cc "side-looker, " manual change, paper tape output,	a col with the b
	All systems except U use 4096-channel analyzers. System U uses a 2048- channel analyzer.	were count with
		ca lib

Table 110a. Summary of Ge(Li) detectors and systems used for gamma-counting Enewetak samples

aluminum can geometry, a solution containing accurately known amounts of ⁶⁰Co, $106_{\rm Ru}$, $137_{\rm Cs}$, $152_{\rm Eu}$, $155_{\rm Eu}$, and $241_{\rm Am}$ was prepared. Aliquots of this solution were dried and mixed with powdered coral from Midway Atoll. The powdered coral was packaged in the aluminum cans and used as a calibration standard. All of the standard solutions used for calibration were cross-checked with standards from the International Atomic Energy Authority

and Laboratoire de Métrologie des Rayonnements Ionisants. Some standard samples for the dried marine materials were supplied by the University of Washington.

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Since large, fairly dense samples were being counted, it was necessary to derive self-absorption parameters. Selfabsorption is a function of mass and atomic composition. The Enewetak survey samples were primarily calcium
etector and system Jentification	Description
M7	18-cc detector operated at two different gains; 1 keV/channel, 2048 channels, and 0.25 keV/channel, 1024 channels.
W 1	16.5-cc detector, gain 0.5 keV/channel, 4096 channels.
20	17-cc detector, gain 0.5 keV/channel, 4096 channels.
28	19-cc detector, gain 1 keV/channel, 2048 channels.
	All of the above detectors were housed in 4-in. lead shields.

Table 110b. Summary of Ge(Li) detectors and systems used for gamma-counting Enewetak samples by Biomedical Division, LLL.

carbonate and the composition was approximated as a mixture of calcium and water. A special counter was built to obtain a good approximation of the calcium content of each sample. The counter consisted of a collimated 60-keV gamma-ray beam with a thin NaI scintillation detector in the beam path. Count-rate measurements were made on the unattenuated beam. A count-rate measurement was then made with a sample in the beam path. With calibration samples of known composition and with a sample of known density, the count rates with and without the sample in place could be used to calculate the percentage of calcium in the sample.

All of the background peaks in each detector system were loaded into a special subroutine in GAMANAL crossreferenced by detector. The computer then subtracted backgrounds at these energies before proceeding with the regular analysis described below under Identification of Nuclides. During and at

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the end of the Enewetak program, detailed backgrounds were again measured to verify that counters had not been contaminated during the program.

Sensitivity of Counters

The sensitivity of a counter for a given nuclide depends on the sample size, counter efficiency, the branching intensity of the gamma rays, the length of the count, and the counter background. On a typical Enewetak coral sample (100-375 g) we found that a count time of 133 min on the larger diodes was sufficient to establish a limit of less than 1 dpm/g for most gamma-emitting nuclides. In the case of marine, vegetation, animal, and airfilter samples, the sensitivity was limited by sample size. For these samples the minimum count time was 666 min and ranged up to several thousand minutes.

Identification of Nuclides - Interference Between Various Nuclides

A general-purpose computer program

called GAMANAL was used for the data reduction and interpretation of all Radiochemistry-generated spectra. It examines the pulse-height data for "background" and "peak" regions, fits these peaks with the proper shape functions, and corrects for the effects of geometry, attenuation, and detector efficiency in evaluating the photon emission rate. The program then proceeds to search a "library" of decayscheme information in order to make tentative assignments for each of the observed peaks. A matrix of equations is formed so that the intensity of each peak is described as a linear addition of the identified nuclides present. The quantitative value, as well as the degree of interference, is the result of a leastsquares solution of this set of equations. Unlikely components are also weeded out in this process. A more complete description is given in UCRL-51061. Volume 1.*

For the Enewetak program, a special library of nuclides was loaded into GAMANAL. Table 111 lists the halflives, energies, and branching intensities for these nuclides. These nuclides were chosen because they are longlived products of nuclear explosions or are naturally occurring radionuclides.

Uncertainties

There are many sources of error in the measurement of gamma spectra; generally only a few dominate and determine the accuracy of the reported values. Sources of error in the interpretation of gamma-ray spectra include the intensity of the observed peaks, the level of the surrounding background, interference of neighboring peaks, natural background activities in the counting chambers. attenuation of gamma rays in the sample matrix or container, the calibration of detectors, the effects of sample geometry and positioning, and decay-scheme information. For low-activity-level samples, the dominant factor contributing to the error is the low net count in the observed peaks. In assessing the error on the net counts of the observed peaks. GAMANAL takes into account the background level, interference problems. and attenuation of the radiations by the sample matrix and container. No additional error is added due to incorrect calibrations or to the effects of geometry and positioning. Since the samples were counted in very "close-in" geometry. the last-mentioned sources of error can be appreciable. All errors which could be determined were added in quadrature. Again, a more complete description is presented in UCRL-51061, Vol. 1.

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To establish the relationship between uncertainties in the input parameters for the GAMANAL code and the final answers as output from the computer, a series of tests was made. Each of the input values (density, atomic composition, geometry, and weight) was purposely changed by ±5%. In no case did this alter the final answer by more than $\pm 10\%$.

Method for Setting Upper Limits on Detection of a Given Nuclide

A request was made to calculate an upper-limit amount for certain nuclides,

^{*}R. Gunnink and J. B. Niday, Computerized Quantitative Analysis by Gamma-Ray Spectrometry, Vol. 1. Description of the Gamanal Program, Lawrence Livermore Laboratory, Rept. UCRL-51061, Vol. 1 (1972).





based on those spectra regions where signals would be seen if the species were present in detectable quantities.

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In order to describe the process by which GAMANAL calculates an upper limit for the presence of a given photopeak, we must discuss the method used in detecting photopeaks. The method of detecting peaks cannot be described explicitly because it cannot be described by a simple algorithm. However, the process can be described qualitatively with the aid of Fig. 114. GAMANAL first finds a "background" continuum line. It then proceeds to locate each peak grouping by searching for a minimum of two successive data points which are greater than 1.8 standard deviations above this background. This value was obtained as a result of experience and insures that most of the reported peaks are real. Figure 114 illustrates a case where a peak was detected by GAMANAL and reported as 51 counts with an assigned error of 63%. This peak is just above the threshold of detection.

If the peak in Fig. 114 had not been detected and an upper limit analysis was requested, the calculation would be done as follows: First, the peak region would be located and a number of data points proportional to the expected peak width (seven in this case) would have been integrated. Then the upper-limit area would have been calculated to be twice the square root of this count. In the illustration, this upper-limit value would have been 36 counts. In practice, spectral regions of two gamma rays per nuclide were investigated (if two were available), and limits were calculated for each. Only the lower of the values was retained.

The method used by ANALYSE 5 (Biomedical Division) to calculate peak areas is to specify channel intervals that define both the peak and the baseline on either side of the peak. The latter is used to make a baseline subtraction from the gross peak area. If the net peak area is negative or zero or if one standard deviation is greater than 50%, ANALYSE 5 calculates an upper limit equal to twice the square root of the gross peak area.

Comments on Identification and Measurement of Each Nuclide in the Complete Sample Set

The GAMANAL code searched each spectrum for photopeaks from all of the nuclides listed in Table 111 and reported all positive signals. In addition, in the case of nonobservation of certain nuclides, upper limits were calculated according to the procedure given in the previous section of this chapter. The nuclides for which upper limits were calculated routinely are: ⁶⁰Co, ^{102m}Rh, ¹⁰⁶Ru, ¹²⁵Sb, ¹³³Ba, ¹³⁷Cs, ¹⁵²Eu, ¹⁵⁵Eu, 207_{Bi} , 235_{U} , and 241_{Am} .

In looking over the nuclides listed in Table 111, one finds entries with relatively short half-lives, some even shorter

than 1 yr. Remembering that Table 111 lists a library of possible nuclides, the following comments summarize the question of observation of gamma emitters listed in the library.

- ⁷Be $t_{\frac{1}{2}} = 53.5$ da Observation: Measured in 32 vegetation samples and in 30 air samples. Comments: Commonly occurs in air due to cosmic ray interactions. ²²Na $t_{\frac{1}{2}}$ = 942 da = 2.58 yr Observation: No ²²Na identified in any sample. 40 K t₁ = 1.26 × 10⁹ yr in a large fraction of the samples. Naturally occurring radio-
 - **Öbservation:** Measured and reported activity; origin not related to weapons

testing.

- 54 Mn t_{$\frac{1}{2}$} = 312 da
 - Observation: Identified in a Tridacna kidney sample (08-0556-11) taken from KATE at $0.61 \pm 0.23 \text{ pCi/g}$ (collected December 8, 1972) and in a Guettarda sample (10-2250-23) taken on WILMA at $0.05 \pm 0.02 \text{ pCi/g}$ (collected January 1973). Also observed in 29 air samples. Comments: Existence in air
 - samples and short half-life suggests the origin of this nuclide is worldwide fallout.
- 60 Co $t_{\frac{1}{2}}$ = 1,920 da = 5.26 yr Observation: Positive signals in a large fraction of the samples; upper limits set for all remaining samples.

⁶⁵ Zn	t1	=	245	da
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- Observation: Identified in a few samples, one soil, 16 marine samples at 0.4-5 pCi/g (collected November-December 1972) with uncertainties 6-44%, and in five animal samples at 0.22-0.55 pCi/g (collected January 1973) with uncertainties 23-42%.
- ${}^{95}Zr$ $t_{\frac{1}{2}} = 65$ da Observation: Identified only in highvolume air samples (9). Comment: Origin is recent

weapons-test debris which is transported as worldwide fallout.

- 103 Ru t₁ = 40 da
 - Observation: Identified in only one sample, a high-volume air filter (13-1189-24).
 - Comment: Origin is presumably worldwide fallout.
- 106 Ru t_{1/2} = 369 da = 1.01 yr
 - Observation: Identified only in six air samples; five are from high-volume samplers. Comment: Origin is presumably
 - worldwide fallout.
- ¹⁰¹Rh $t_{\frac{1}{2}}$ = 1100 da = 3.01 yr
 - Observation: Identified in 24 soil samples scattered over the northern half of the Atoll, in 58 sediment samples, and in two marine samples, <u>Tridacna</u> viscera and kidney (08-0504-02) at $0.12 \pm 0.03 \text{ pCi/g}$ (collected December 11 1972) and Tridacna viscera (08-0536-02) at 0.18 \pm 0.03 pCi/g (collected November 29, 1972), with both samples taken near BELLE.

- $102m_{\rm Rh}$ t₁ = 1060 da = 2.90 yr
 - Observation: Identified in 218 soil samples scattered over the northern half of the Atoll, in 12 samples from LEROY, in 162 sediment samples, and in the following seven marine samples:
- 08-0476-01, Tridacna kidney, 0.8 ± 0.2 pCi/g (collected December 11, 1972), ALICE.
- 08-0504-02, <u>Tridacna</u> viscera and kidney, 0.51±0.14 pCi/g (collected December 11, 1972), BELLE.
- 08-0535-02, Tridacna kidney, 1.0 ± 0.3 pCi/g (collected November 29, 1972), BELLE.
- 08-0536-02, Tridacna viscera, 0.54 ± 0.06 pCi/g (collected November 29, 1972), BELLE.
- 08-0789-10, Tridacna viscera, 0.14 ± 0.05 pCi/g (collected December 4, 1972), JANET.
- 08-0676-10, Tridacna kidney, 3.0 ± 0.9 pCi/g (collected December 5, 1972), JANET.
- 09-8048-24, Goatfish viscera, 0.11 \pm 0.02 pCi/g (collected December 6, 1972), YVONNE.
- $108 \text{ m}_{\text{Ag}} \text{ t}_{\frac{1}{2}} = 127 \text{ yr}$ Observation: Identified in the following three marine samples:
- 08-0348-38, Tridacna, muscle and mantle, $0.05 \pm 0.01 \text{ pCi/g}$, GLENN.
- 09-0466-37, Sea turtle, liver, 0.56 ± 0.09 pCi/g, FRED.
- 09-0264-53, Bonito, liver, 0.28 ± 0.04 pCi/g, wide passage.
- ^{110m}Ag $t_{\frac{1}{2}}$ = 253 da Observation: Not identified in any sample.
- ¹²⁵Sb $t_{\frac{1}{2}}$ = 1010 da = 2.77 yr Observation: Identified in a large fraction of soil samples, predominantly from northern

half of the Atoll. Also identified in 130 sediment samples and in the following eight marine, one vegetation, and one airfilter samples:

- 08-0359-38, Sea cucumber, viscera and gut content, 1.55 ± 0.15 pCi/g (collected October 18, 1972), GLENN.
- 09-8018-24, Parrotfish, viscera, 0.35 \pm 0.11 pCi/g (collected December 4, 1972), YVONNE.
- 09-0376-37, Goatfish, eviscerated whole, 1.58 \pm 0.10 pCi/g (collected December 1, 1972), FRED.
- 09-0466-37, Sea turtle, liver, 1.85 ± 0.31 pCi/g (collected December 9, 1972), FRED.
- 09-0467-37, Turtle, 1.24 ± 0.20 pCi/g (collected December 9, 1972), FRED.
- 09-0344-43, Mullet, muscle, 1.72 ± 0.18 pCi/g (collected October 20, 1972), LEROY.
- 09-0346-43, Mullet, viscera, 1.83 ± 0.25 pCi/g (collected October 20, 1972), LEROY.
- 09-0591-61, Yellowfin tuna, muscle, 1.11 \pm 0.15 pCi/g (collected December 9, 1972), Kwajalein.
- 10-0085-38, Scaevola leaf, 0.12 ± 0.05 pCi/g (collected January 1973), GLENN.
- 13-1189-24, High-volume air filter, 0.27 ± 0.06fCi/m³ (collected December 1972), YVONNE.
- ¹³³Ba t₁ = 2630 da = 7.21 yr
- Observation: Identified in 34 soil samples from JANET (7), PEARL (6), SALLY (13), and YVONNE (8).

 134 Cs $t_{\frac{1}{2}}$ = 745 da = 2.04 yr

Observation: Identified in seven soil samples (BELLE, IRENE, LUCY, GLENN, and HENRY), one sediment sample, and the following marine and animal samples:

- 09-8041-24, Convict surgeon, viscera, 0.53 \pm 0.07 pCi/g (collected December 6, 1972), YVONNE
- 09-0466-37, Sea turtle, liver, 0.44 ± 0.12 pCi/g (collected December 9, 1972), FRED
- 11-9118-24, Roof rat, viscera, 1.3 ± 0.3 pCi/g (collected January 15, 1973), YVONNE.
- 11-9135-24, Roof rat, lung, 1.0 ± 0.3 pCi/g (collected February 3, 1973), YVONNE.
- 11-9167-24, Roof rat, bone, 0.8 ± 0.2 pCi/g (collected January 15, 1973), YVONNE.
- 11-9168-24, Roof rat, bone, 1.0 ± 0.2 pCi/g (collected February 3, 1973), YVONNE.
- 11-9269-33, Sooty tern, bone, 0.29 ± 0.06 pCi/g (collected January 15, 1973), DAVID.
- 137 Cs $t_{\frac{1}{2}}$ = 30.0 yr
 - Observation: Positive signals in a large fraction of the total samples; upper limits set for all remaining samples.
- ^{144}C $t_{\frac{1}{2}} = 285 \text{ da}$
 - Observation: Identified in soil (7), sediment (26), marine (11), vegetation (10), and air-filter (12) samples. With the exception of the air filters (all from high-volume samplers), all observations are considered questionable since they are based upon the observation of a single gamma ray at 133 keV. Confirmation of these data would require chemical separation of cerium and further counting. The airfilter data are considered authentic.

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al	half of soil samples from northern half of the Atoll and in about a third of the sedi-
ra,	ments. Also identi-
ected NNE	fied in the following
MIL.	samples:
cted	08-0556-11, <u>Tridacna</u> , kidney,
ED.	0.52 ± 0.11 pCi/g, KATE.
- •	$_{09}$ -0494-02, Mullet, viscera, 0.33 ± 0.10 pCi/g, BELLE.
ed	
NNE.	$_{09}$ -0496-02, Mullet, viscera, 0.26 \pm 0.06 pCi/g, BELLE.
	0.20 1 0.00 per/g, billin.
ed	
NNE.	154 Eu t ₁ = 2860 da = 7.83 yr
•	Observation: Identified in 101 soil
ed NNE.	samples from
THE.	ALIČE (7), BEILE (14), CLARA (4), DAISY (4),
ed	IRENE (6), JANET (3),
NNE.	PEARL (28), SALLY (12),
	and YVONNE (23),
cted	and in 36 sediment samples. There were
TD.	no other authenticated
	observations.
	155
	155 Eu t ₁ = 1850 da = 5.08 yr
in a	Observation: Identified in a large
the	fraction of the soil samples and in most
per re-	of the sediment
•	samples. Also identi-
	fied in 7 (of 16) plank-
	ton samples, in 10 (of 54) seawater
(7),	samples, in 68 (of
lar- lon	410) marine samples,
er	in 3 (of 216) vegetation
ʻi th	samples, and 1 (of 274) animal samples.
he	
om :plers),	$\frac{10-2265-02}{0.06 \pm 0.02 \text{ pCi/g}}$ (collected
are	January 1973), BELLE.
ion-	10-1892-11, Messerschmidia,
re	0.05 ± 0.02 pCi/g (collected
oser-	January 1973), KATE.
3 keV.	10-3275-24, <u>Scaevola</u> , 0.05 ± 0.02 pCi/g
hese	(collected January 1973),
re ion	YVONNE.
cther	11-9082-17, Rice rat, hide,
r-	0.22 ± 0.07 pCi/g (collected January 1973), PEARL.
on-	
2.	207 Bi $t_{\frac{1}{2}} = 32.0 \text{ yr}$
	Observation: Identified in soil
ghly	Observation: Identified in soli
0 -	

samples in localized regions, especially on DAISY, EDNA, IRENE, JANET, IRWIN, JAMES, and KEITH. Also identified in most of the sediment samples, in 14 (of 16) plankton samples, in 22 (of 54) seawater samples, in approximately half of the marine samples, in the following five vegetation samples and four animal samples:

- 10-2455-04, Coconuts, 0.06 ± 0.03 pCi/g, DAISY.
- 10-3700-10, <u>Pandanus</u>, 0.11 ± 0.03 pCi/g, JANET.
- 10-0199-33, <u>Messerschmidia</u>, 0.05 ± 0.02 pCi/g, DAVID.
- 10-0081-38, <u>Pisonia</u> leaf, 0.06 ± 0.03 pCi/g, GLENN.
- 10-2430-42, <u>Messerschmidia</u>, 0.07 ± 0.03 pCi/g, KEITH.
- 10-2434-42, <u>Pandanus</u>, 0.04 ± 0.01 pCi/g, KEITH.
- 11-9133-21, Rice rat, liver, 0.38 ± 0.11 pCi/g, URSULA.
- 11-9150-21, Rice rat, lung, 0.90 ± 0.32 pCi/g, URSULA.
- 11-9087-24, Roof rat, hide, 0.21 ± 0.08 pCi/g, YVONNE.
- 11-9026-38, Hermit crab, pancreas and gonad, 0.15 ± 0.06 pCi/g, GLENN.
- 226 Ra $t_{\frac{1}{2}} = 1620$ yr Observation: Identified in 102 soil samples spread throughout the entire Atoll and 130 sediment samples.
- ²²⁸Th $t_{\frac{1}{2}}$ = 698 da = 1.91 yr Observation: Identified locally in YVONNE soil samples (32). No other positive identification.

²³⁵U $t_{\frac{1}{2}} = 7.13 \times 10^8$ yr Observation: Identified fairly infre-

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quently (about 6.5%) among soil samples. Measured concentrations in the samples averaged in the range 0.014-0.026 ppm²³⁵U. If one assumed the 235 U abundance to be that of natural uranium (not necessarily a valid assumption at Enewetak), these averages correspond to a natural uranium content of 1.9-3.6 ppm. Also identified in 96 sediment samples. There was no positive identification in any other type of sample.

²³⁹Pu $t_{\frac{1}{2}}$ = 24,360 yr

Observation: Identified by gamma detection in just two samples (sensitivity for detection is greatly increased in wetchemistry analysis). Comparison of gamma counting and wetchemistry results is given below:

	²³⁹ Pu.	239+240 _{Pu} .
Sample	dpm/g (gamma)	
73-5235-24	469±145	1020 ± 50
05-1096-24	290±160	714 ± 28

Note that gamma counting measures only 239 Pu whereas alpha counting measures the sum of 239 Pu and 240 Pu activity. One can deduce 240/239 atom ratios

from the above data which have values of 0.32 \pm 0.19 and 0.40 \pm 0.38, respectively. The precision of these values is poor; mass-spectrometric analyses of 240/239 ratio yield more precise data.

²⁴¹Pu $t_{\frac{1}{2}}$ = 5110 da = 14.0 yr Observation: No ²⁴¹Pu detected in any sample by gamma counting.

 241 Am t $_{\frac{1}{2}}$ = 433 yr

Observation: Identified in many of the soil samples from most of the islands. although in greatest abundance and frequency in the northern half of the Atoll. Also prevalent in sediments. Identified elsewhere in 5 (of 16) plankton samples, 7 (of 54) seawater samples, in 38 (of 410) marine samples, in 2 (of 216) vegetation samples, and in 1 (of 67) air filters. There was no positive identification in animal samples.

Comments: Comparison of ²⁴¹ Am determination by gamma counting with that by wet-chemistry analysis is given in the last section of this chapter.

	Nuclide	Half-life days,	Energy, keV	Branching intensity
1.	4 BE 7	5.350E+01	477.400	1.03OE-01
2.	11 NA 22	9.417E+02	511.000 1274.550	1.800E+00 1.000E+00
3.	19 K 40	4.610E+11	1460.760	1.083E-01
4.	25 MN 54	3.123E+02	834.823	$1.000 \pm +00$
5.	27 CO 60	1.922E+03	1173.230 1332.510	1.000E+00 1.000E+00
6.	30 ZN 65	2.450E+02	1115.520	4.900E-01
7.	40 ZR 95	6.500E+01	724.200 756.720	4.300E+01 5.460E-01
8.	41 NB 95	3.510E+01	765,800	9.900E-01
9.	44 RU 103	2.960E+01	$497.090 \\ 610.310$	9.000E-01 5.600E-02
10.	45 RH 101	100E+03	127,200 197,900 325,100	8.400E-01 9.000E-01 1.800E-01
11.	45 RH 106	3.514E-04	511.800 616.300 622.100 873.800 1050.700	2.050E-01 8.100E-03 9.800E-02 4.400E-03 1.400E-02
12.	45 RH 102M	1.059E+03	$\begin{array}{r} 418.800\\ 475.100\\ 628.200\\ 631.400\\ 697.600\\ 767.000\\ 1046.800\\ 1103.300\\ 1112.900 \end{array}$	1.120E-01 9.300E-01 7.000E-02 5.200E-01 4.320E-01 3.300E-01 3.100E-01 4.500E-02 1.800E-01
13.	47 AG 108M	4.635E+ 0 4	$\begin{array}{c} 79.120 \\ 433.610 \\ 614.040 \\ 632.740 \\ 722.730 \end{array}$	5.200E-02 9.200E-01 9.200E-01 1.140E-03 9.200E-01
14.	47 AG 110M	2.530E+02	$\begin{array}{c} 446.200\\ 620.100\\ 657.600\\ 677.500\\ 686.800\\ 706.600\\ 744.200\\ 763.800\\ 817.900\\ 884.500\\ 937.300 \end{array}$	3.500E-02 2.500E-02 9.300E-01 1.220E-01 7.500E-02 1.600E-01 4.330E-02 2.200E-01 6.950E-02 7.100E-01 3.360E-01

Table 111. Half-lives, energies, and branching intensities for nuclides loaded into GAMANAL.

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Table 111 (continued)

	Nuclide	Half-life days,	Energy, keV	Branching intensity
14.	47 AG 110M	2.530E+02	1384.300 1475.900 1505.200 1562.500	2.400E-01 3.700E-02 1.260E-01 1.100E-02
15.	51 SB 125	1.012E+03	176.430380.440427.880463.380600.600606.700635.920671.410	7.200E-02 1.520E-02 3.040E-01 1.070E-01 1.810E-01 5.150E-02 1.150E-01 1.820E-02
16.	55 CS 134	7.450E+02	$\begin{array}{r} 475.340\\ 563.220\\ 569.330\\ 604.700\\ 795.790\\ 801.870\\ 1038.610\\ 1167.910\\ 1365.130\end{array}$	1.540E-02 8.820E-02 1.580E-01 9.800E-01 8.900E-01 9.500E-02 1.060E-02 1.850E-02 3.000E-02
17.	55 CS 137	1.096E+04	661.646	8.500E-01
18.	56 BA 133	2.630E+03	$53.170 \\79.590 \\81.010 \\160.620 \\276.290 \\302.710 \\355.860 \\383.700$	1.950E-02 3.040E-02 3.600E-01 7.600E-03 7.500E-02 1.960E-01 6.700E-01 9.400E-02
19.	58 CE 144	2.846E+02	80.100 133.500 696.500	1.480E-02 1.100E-01 1.330E-02
20.	63 EU 152	5.117E+03	121.780 244.700 295.970 329.300 344.270 367.760 411.100 416.000 443.940 488.700 503.450 586.200 656.400 674.350 678.600 688.800 712.900	3.010E-01 7.740E-02 4.700E-03 1.490E-03 2.740E-01 9.000E-03 2.270E-02 1.140E-03 3.200E-02 3.900E-03 1.500E-03 4.400E-03 1.660E-03 4.400E-03 8.500E-03 1.000E-03

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Table 111 (continued)

	Nuclide	Half-life days,	Energy, keV	Branching intensity
20.	63 EU 152	5.117E+03	$\begin{array}{c} 764.900\\ 778.850\\ 810.240\\ 841.400\\ 867.300\\ 919.100\\ 964.000\\ 1005.100\\ 1085.700\\ 1085.700\\ 1089.500\\ 1111.900\\ 1212.800\\ 1249.700\\ 1292.600\\ 1298.970\\ 1407.920\\ 1457.600\\ 1528.200 \end{array}$	$\begin{array}{c} 1.400 \pm -03\\ 1.300 \pm -01\\ 3.150 \pm -03\\ 2.100 \pm -03\\ 4.260 \pm -02\\ 4.000 \pm -03\\ 1.480 \pm -01\\ 6.500 \pm -03\\ 1.480 \pm -01\\ 6.500 \pm -03\\ 1.025 \pm -01\\ 1.750 \pm -02\\ 1.400 \pm -01\\ 1.400 \pm -02\\ 2.030 \pm -03\\ 1.140 \pm -03\\ 1.640 \pm -02\\ 2.150 \pm -01\\ 5.000 \pm -03\\ 2.830 \pm -03\end{array}$
21.	63 EU 154	2.863E+03	123.140 248.040 591.740 692.410 723.300 756.870 873.190 996.320 1004.760 1274.390 1596.480	4.050E-01 6.590E-02 4.840E-02 1.696E-02 1.970E-01 4.340E-02 1.150E-01 1.030E-01 1.730E-01 3.350E-01 1.670E-02
22.	63 EU 155	1.855E+03	60.010 86.550 105.320	1.320E-02 3.220E-01 2.280E-01
23.	83 BI 207	1.169E+04	569.620 1063.650 1770.180	9.800E-01 7.700E-01 7.150E-02
24.	88 RA 226	5.917E+05	$186.140 \\ 241.960 \\ 295.200 \\ 351.920 \\ 609.270 \\ 665.400 \\ 742.480 \\ 768.350 \\ 785.800 \\ 806.160 \\ 934.060 \\ 1120.280 \\ 1155.170 \\ 1238.130 \\ 1280.980 \\ \end{array}$	4.000E-02 7.900E-02 2.020E-01 4.010E-01 4.840E-01 1.650E-02 PAIR PEA 5.320E-02 1.210E-02 1.310E-02 3.340E-02 1.600E-01 1.820E-02 6.200E-02 1.560E-02

	Nuclide	Half-life days,	Energy, keV	Branching intensity
24.	88 RA 226	5.917E+05	1401.440 1407.980 1509.220 1661.240 1729.550 1764.490 1838.330 1847.440 2118.520 2204.140 2447.630	1.440E-02 2.600E-02 2.300E-02 1.210E-02 3.070E-02 1.660E-01 4.100E-03 2.200E-02 1.230E-02 5.300E-02 1.650E-02
25.	90 TH 228	6.976E+02	$\begin{array}{c} 74.817\\ 74.970\\ 77.108\\ 84.380\\ 84.450\\ 86.830\\ 87.350\\ 238.626\\ 240.982\\ 277.340\\ 300.110\\ 510.720\\ 583.139\\ 727.270\\ 785.460\\ 860.490\\ 1592.690\\ 1620.620\\ 2614.710\\ \end{array}$	9.530E-02 1.303E-01 1.620E-01 1.330E-02 1.580E-03 1.930E-02 3.600E-02 4.480E-01 4.140E-02 2.300E-02 3.420E-02 3.420E-02 3.990E-01 6.650E-02 1.100E-02 4.530E-02 PAIR PEAH 1.510E-02 3.596E-01
26.	92 U 235	2.604E+11	$143.770 \\ 163.370 \\ 185.720 \\ 202.100 \\ 205.330 \\ \end{array}$	1.070E-01 4.850E-02 5.610E-01 1.070E-02 4.870E-02
27.	94 PU 239	8.908E+06	94.665 98.439 129.280 203.520 375.020 413.690	9.830E-05 1.900E-04 6.420E-05 5.630E-06 1.585E-05 1.506E-05
28.	94 PU 241	5.110E+03	148.600 164.590 207.970	1.900E-06 4.500E-07 5.120E-06
29.	95 AM 241	1.582E+05	59.536 99.000 103.000	3.590E-01 2.100E-04 2.020E-04

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Table 111 (continued)

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Wet-Chemistry Analyses

Nuclides Measured; Laboratories Making Measurements

Nuclides that could not be detected by gamma spectroscopy and that were judged to be of potential significance to this survey were analyzed for by dissolution of a sample, chemical separation of a desired element, and quantification by an appropriate radiation-counting technique. An integral part of this technique is the addition of a known amount of elemental carrier or tracer at the beginning of the procedure to permit determination of chemical yield in the final sample. These nuclides, their half-lives, principal radiation, and technique for counting are listed in Table 112. In this list, the nuclides analyzed for most generally were ⁹⁰Sr. ²³⁸Pu. ^{239, 240}Pu. and ⁵⁵Fe. Wetchemistry analysis for ²⁴¹ Am was performed on a small fraction of the samples; gamma counting was the major method used to determine this nuclide. The purposes of wet-chemistry analyses for ²⁴¹ Am were either to check results obtained by gamma counting or, in the case of some marine, vegetation, and animal samples, to extend the sensitivity for 241 Am detection to lower levels. The remaining nuclides in Table 112 were measured in relatively small numbers of samples to provide an approximate indication of levels existing in various biological samples. In addition to the radioactive species, analyses for stable iron, calcium, and iodine were required on certain samples. The kinds and numbers of analyses performed at each laboratory are listed in Table 113.

Separation Schemes

For application to coralline soils and sediments, chemical dissolution, separation, and purification schemes for ⁹⁰Sr and Pu determination as performed at MCL, LFE, and EIC are shown in Tables 114, 115, and 116. These procedures are given in basic outline form; no details on manipulation, quantities of reagents, or fine points of analytical technique are included. Each laboratory received 50-g samples of finely divided coral soil. At MCL, quantities of 10-20 g were dissolved, while at LFE and EIC, entire 50-g samples were put in solution.

Some variation in dissolving technique is seen among the laboratories; each reported coralline soil to be readily soluble in appropriate mineral acids. Both MCL and LFE procedures feature a sequential separation of Sr-Y and Pu from a single aliquot, while EIC chose to isolate these elements from separate aliquots.

Determination of ⁹⁰Sr and Pu in other types of samples required some modifications of procedures given in Tables 114 and 115. For MCL, the required variations are summarized in Table 117. Corresponding procedures in use at LFE are summarized in Table 118.

The isolation of 55 Fe was based upon the extraction of iron carrier into diethyl ether from 6 <u>M</u> HCl solution at all four laboratories. Following further purification, samples were electrodeposited in preparation for gamma counting. An aliquot of each sample to which no carrier had been added was reserved for determination of stable iron via atomic absorption spectrometry. This information

able 112.	Nuclides measur	ed in wet-chemistry analy	ses.
Nuclide	T _{1/2}	Principal radiation	Type of detection
Fe	2.7 y	5.95 keV x ray	Gamma counting: NaI(Tℓ), Ge(Li) detectors.
)Sr	28.5 y	³ particle of ⁹⁰ Y daughter (E _{max} = 2.27 MeV)	Beta counting: gas-filled proportional counter.
³⁹ ,240 _{Pu}	24,400 y (239)	5.16 MeV α	Alpha pulse-height analysis,
	6,540 y (240)		(solid state, Frisch-grid chamber), mass spectrome- try.
³⁸ Pu	8 7. 8 y	5.50 MeV α	Alpha pulse height analysis.
¹¹ Am	433 y	5.49 MeV α	Alpha pulse height analysis.
H	12.35 y	β particle (E _{max} = 18.5 keV)	Gas-filled proportional counter.
¹ C	5,730 y	β particle (E _{max} = 156 keV)	Liquid scintillation counter.
³ Ni	9 2 y	β particle (E _{max} = 65.9 keV)	Liquid scintillation counter.
⁵ Zn	245 d	Gamma ray (1.116 MeV)	Gamma spectrometry of separated samples.
^{13m} Cd	14 y	β particle (E _{max} = 580 keV)	Beta counting: gas-filled proportional counter.
²⁹ I	1.57 × 10 ⁷ y	β particle (E _{max} = 150 keV) Xenon K x rays (29.7, 33.7 keV)	Liquid scintillation counter, x-ray detection (Si diode).
¹⁴ Ce	285 d	β particle of ¹⁴⁴ Pr daughter (E _{max} = 2.99 MeV)	Beta counting: gas-filled proportional counter.
⁴⁷ Pm	2.62 y	β particle (E _{max} = 225 keV)	Beta counting: gas-filled proportional counter.
⁵¹ Sm	87 y	β particle (E _{max} = 76 keV)	Liquid scintillation counter.
^r e, Ca, I,	Sr stable	None	(Atomic absorption)

Analyses performed

Summary of wet-chemistry analyses.

Table 113.

 $65_{Z_{\rm ID}}$ (4) $113m_{\rm Cd}$ (9) $144_{\rm Ce}$ (4) 11(2), $241_{\rm Am}(24)$

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<u>Maior nuclides</u> (⁵⁵Fe, ⁹⁰Sr, ²³⁸, ²³⁹, ²⁴⁰Pu)

> Laboratory, number of samples

> > Type of samples

able 112. Nuclides measured in wet-chemistry analyses.

			Analys	ses performed
Type of samples	Labor number o	ratory, f samples	<u>Major nuclides</u> (⁵⁵ Fe, ⁹⁰ Sr, ^{238, 239, 240} Pu)	$\frac{\text{Minor nuclides}}{({}^{3}\text{H},{}^{14}\text{C},{}^{63}\text{Ni},{}^{65}\text{Zn},{}^{113}\text{m}\text{Cd},{}^{144}\text{Ce},{}^{147}\text{Pm},{}^{151}\text{Sin},{}^{241}\text{Am})}$
Soil, sediment, core	MCL	1923	$^{90}{ m Sr}$, $^{238}{ m Pu}$, $^{239, 240}{ m Pu}$ (all), $^{55}{ m Fe}$ (2)	65 Zn (4), 113m Cd (9), 144 Ce (4), U (2), 241 Am(24)
	LFE	1007	⁹⁰ Sr, ^{239,240} Pu (all), ²³⁸ Pu (29)	
	EIC .	486	⁹⁰ Sr, ^{239, 240} Pu (all)	
Marine	MCL	121	55 Fe, 90 Sr, 238 Pu, 239,240 Pu (all)	63 Ni (9), 65 Zn (2), 113m Cd(14), 147 Pm(9),
				151Sm (9), 241 Am(9)
	LFE	196	55 Fe, 90 Sr, 239,240 Pu (all)	63 Ni(4), 113m Cd(3), 147 Pm(4), 151 Sm(4), 241 Am(8)
	LRE	114	55 Fe, 90 Sr, 239,240 Pu (all), 238 Pu (all)	·
	LLL	10		3 H(10), 14 C(10)
Plankton	MCL	16	90 Sr, 238 Pu, 239,240 Pu (all)	
Algae	LFE	3	55 Fe(1), 90 Sr(1), 239,240 Pu(2)	
Vegetation	MCL	130	55 Fe(28), 90 Sr, 238 Pu, 239,240 Pu (all)	63 Ni(8), 113m Cd(8), 147 Pm(8), 151 Sm(7), 241 Am(8)
	LFE	51	⁵⁵ Fe(11), ⁹⁰ Sr, ^{239,240} Pu (all)	
	LLL	11		3 H(10), 14 C(11)
Animal	MCL	53	55 Fe(53), 90 Sr, 238 Pu, 239,240 Pu (all)	113mCd(4)
	LFE	163	⁵⁵ Fe(116), ⁹⁰ Sr, ^{239,240} Pu (all)	63 Ni(3), 113 mCd(2), 147 Pm(5), 151 Sm(5), 241 Am(6)
	LLL	15		³ II(15)
Seawater	LLL	47	²³⁸ Pu, ^{239,240} Pu (47)	
	LFE	62	⁹⁰ Sr(62)	
Air filter	MCL	58	238 _{Pu} , 239,240 _{Pu} (all)	

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Table 113. Summary of wet-chemistry analyses.

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Dissolution	Fire coral at 950°C for 8 hr.
	Dissolve 12 <u>M</u> HCl + 5.5 <u>M</u> HI; dilute with H_2O . ^a
	Working solution for aliquots, combined Sr-Pu.
Separation	To aliquot, add Y carrier, 236 Pu or 242 Pu tracer. ^b
	Ppt Y(OH) ₃ by adding NH_4OH . (Note Sr-Y separation time).
	Wash ppt H_2O ; dissolve $16 M$ HNO ₃ , dilute with H_2O .
	Ppt Y(OH) ₃ by adding NH ₄ OH.
	Wash ppt H_2O ; dissolve satd HCl + few drops HNO ₃ .
	Load on Dowex 1×8 column (Pu-Y separation).
	Wash column 12 \underline{M} HCl. (Load and wash to Y purification).
	Elute Pu with 12 \underline{M} HCl + satd NH ₄ I. (To Pu purification).
Y purification	Evaporate column load and wash fractions to dryness.
	Dissolve in 0.1 \underline{M} HCl.
	Extract twice with 10% HDEHP (toluene).
	Back-extract 3 M HCl.
	Ppt $Y(OH)_3$ by adding NH_4OH .
	Wash H_2O ; dissolve 12 <u>M</u> HCl + H_2O , filter.
	Ppt Y oxalate by adding satd oxalic acid, digestion.
	Filter ppt, dry, fire to Y2O3 at 900°C, 1 hr.
	Weigh, beta count ⁹⁰ Y.
Pu purification	To column eluant, add 5 <u>M</u> NH ₂ OH HCl, LaCl ₃ carrier, satd NH ₄ I, ZrO(NO ₃) ₂ carrier.
	Boil to reduce volume.
	Ppt LaF ₃ by adding HF.
	Dissolve $HNO_3 + H_3BO_3$.
	Ppt La(OH) ₃ by adding NH_4OH .
	Dissolve 16 \underline{M} HNO ₃ , boil.
	Ppt $La(OH)_3$ by adding NH_4OH .
	Wash H ₂ O; dissolve 12 <u>M</u> HCl + few drops HNO ₃ .

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Determine Pu by either α pulse-height analysis or mass- spectrometric analysis.
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^aThe addition of HI is necessary to insure equilibration of plutonium tracer with the plutonium in the aliquot of the working solution.
^b236_{Pu} was used as an alpha-PHA tracer; ²⁴²_{Pu} could be used either as an alpha-PHA tracer or as a mass tracer. Note that ²³⁸_{Pu} could be determined only on those samples which were assayed via alpha-PHA.

Table 115.	Laboratory for Electronics - Environmental Analysis Laboratories.
	Chemistry scheme for determination of ⁹⁰ Sr and Pu in coralline soils
	and sediments.

Dissolution	Fire coral at 900°C for 12 hr.	
	Add Y carrier, ²³⁶ Pu tracer.	
	Dissolve 6 <u>N</u> HNO ₃ , adjust solution to 0.05 <u>M</u> HF.	Sr-Y:
Separation	Evaporate near dryness, add H ₃ BO ₃ .	
	Evaporate near dryness, add 6 <u>N</u> HNO ₃ + 30% H_2O_2 .	
	Boil down, add more 6 \underline{N} HNO ₃ , cool, add 5% NaNO ₂ .	
	Load on Dowex 1 × 4 column. (Pu, Sr-Y separation).	
	Wash column 6 <u>N</u> HNO ₃ . (Load and wash to Sr-Y purification).	
	Elute Pu with $4 \text{ N} \text{ HNO}_3 - 0.1 \text{ N} \text{ HF}$. (To Pu purification).	
Sr-Y purification	Evaporate column load and wash near dryness.	
	Add H ₂ O; adjust to pH 1.	
	Extract with 20% HDEHP (toluene). (Note Sr-Y separation time).	
	Wash three times with 0.5 N HCl.	
	Back-extract three times with 12 M HCl.	
	Evaporate to dryness (adding fuming HNO ₃).	
	Dissolve 12 <u>M</u> HCl + H_2O .	
	Ppt YF ₃ by adding HF.	
	Dissolve $HNO_3 + H_3BO_3$.	Du oli
	Ppt Y(OH) ₃ by adding NH_4OH .	<u>Pu als</u>
	Dissolve 6 <u>N</u> HCl + H ₂ O.	
	Ppt Y(OH) ₃ by adding NH ₄ OH.	
	Wash twice with H_2O ; dissolve min. 6 <u>N</u> HCl.	
	Ppt Y oxalate by adding satd oxalic acid + $H_{2}O$, digestion.	
	Filter, fire to Y_2O_3 .	
	Weigh, beta count 90 Y.	
Pu purification	Evaporate eluant to low volume, adding H ₃ BO ₃ + Fe carrier.	
	Ppt $Fe(OH)_3$ by adding NH_4OH .	
	Wash dilute NH_4OH .	
	Dissolve 6 \underline{N} HNO ₃ ; cool, add 5% Na ₂ NO ₂ .	
	Load on Dowex 1×4 column.	
	Wash 6 <u>N</u> HNO ₃ , 12 <u>M</u> HCl.	
	Elute Pu with 12 M HCl-HI.	
	Evaporate solution.	
	Electrodeposit on stainless steel disk for alpha pulse-height analysis.	
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Dissolution	Fire coral at 500°C for 12 hr.
DISSOIUTION	Dissolve 8 \underline{N} HNO ₃ , filter.
	Working solution for aliquots, separate Sr and Pu.
Sr-Y aliquot	Add ⁸⁵ Sr tracer, evaporate dry.
	Dissolve 0.08 \underline{N} HCl.
	Extract twice with 20% HDEHP (toluene). (Note Sr-Y separation time)
	Discard organic each time.
	Add Y carrier to aqueous.
	Count sample for 85 Sr with gamma spectrometer.
	Store sample 2 wk (90 Y growth period).
	Extract 5% HDEHP (toluene).
	Wash 0.08 \underline{N} HCl.
	Back-extract 3 \underline{N} HNO ₃ .
	Ppt Y(OH) ₃ by adding NH_4 OH.
	Dissolve 1 \underline{N} HCl.
	Ppt Y oxalate by adding H_2O , NH_4 oxalate, digestion.
	Filter; wash ppt with H ₂ O, alcohol.
	Dry, cool, weigh.
	Beta count ⁹⁰ Y.
Pu aliquot	Add 236 Pu tracer, few drops 25% Na $_2$ NO $_2$.
	Extract with Aliquat 336 (quaternary amine).
	Wash twice with 8 <u>N</u> HNO ₃ , four times with 10 <u>M</u> HCl-H ₂ O ₂ .
	Back-extract Pu twice with HClO ₄ -oxalic acid solution.
	Add NaHSO ₄ , evaporate dry.
	Add 12 \underline{M} HCl, evaporate dry.
	Electroplate from $HC\ell$ -NH ₄ oxalate solution.
	Wash H ₂ O, dry.
	Determine Pu by alpha pulse-height analysis.

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Table 117. McClellan Central Laboratory: Supplemental chemistry schemes required to process biological samples.

1. F	ish, bird, rat samples - bones present:
	After ashing at 900°C and dissolution in 12 <u>M</u> HCl - 5.5 <u>M</u> HI, the amounts of phosphate present prevented effective carrying on a hydroxide precipitate. The following procedure was followed: Evaporate HCl-HI dry.
	Add 12 M HCl, evaporate dry.
	Dissolve 12 M HCl, centrifuge insolubles.
	Load on Dowex 1×8 column in 12 <u>M</u> HCl + few drops HNO ₃ .
	Wash column with 12 M HCl.
	Combine load, wash, insolubles; evaporate dry, proceed with Y purifica- tion shown in Table 114.
	Elute Pu from column with 12 <u>M</u> HCl + satd NH_4I . Proceed with Pu purification shown in Table 114.
2.	Fish, bird, rat samples - muscles, kidneys, liver, viscera (no bones present). Ash at 900°C, dissolve in 12 <u>M</u> HCl - 5.5 <u>M</u> HI. Proceed with Y(OH) ₃ pptn as shown in Table 114.
3.	Bird eggs: Ash, process as with coralline soils.
4.	Plankton: Ash, process as with coralline soils.
5.	Vegetation samples, coconut meat only: Ash at 600°C. Dissolve 12 <u>M</u> HCl + 5.5 <u>M</u> HI. Treat insolubles with HClO ₄ -HF. Proceed as with soil procedure in Table 114.
6.	Vegetation samples, all others: Dissolve in HNO ₃ and HClO ₄ . Bake to dryness. Dilute with 6 <u>M</u> HCl plus 2 ml HI. Proceed as with soil procedure in Table 114.
7.	Polystyrene air filters: Distill styrene off at 450°C. Dissolve residue in 12 <u>M</u> HCl + 5.5 <u>M</u> HI. Proceed as with soil procedure in Table 114.

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Table 118.	Laboratory for Electronics - Environmental Analysis Laboratories:
	Supplemental chemistry schemes required to process biological samples.

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	1. Fish, crab :	samples.
	Dissolution	Ash at 600°C for 12 hr.
		Dissolve ash in 12 \underline{M} HCl, filter.
		Ash filter plus solids at 600°C.
		Dissolve in 12 \underline{M} HCl; combine with filtrate.
		Add 236 Pu and appropriate carriers (Y, Fe, others).
		Add few drops HF; evaporate to near dryness.
		Add HNO_3 , H_3BO_3 ; evaporate to near dryness.
		Add HNO_3 , H_2O_2 ; evaporate to near dryness.
		Dissolve in 12 \underline{M} HCl.
	Separation	Load on Dowex 1×4 column.
l		Column load to Sr-Y purification.
		Elute Fe with 6 <u>N</u> HNO ₃ . (Fe separation - to Fe purification).
		Elute Pu with 4 \underline{N} HNO ₃ -0.1 \underline{N} HF. (To Pu purification).
	2. Vegetation,	bird, and egg samples.
	Dissolution	Ash at 600°C for 12 hr.
		Dissolve ash in 12 M HCl, filter.
		Ash filter plus solids at 600°C.
		Dissolve in 12 M HCl; combine with filtrate.
		Add ²³⁶ Pu and appropriate carriers.
		Add few drops HF; evaporate to near dryness.
		Add HNO ₃ , H ₃ BO ₃ ; evaporate to near dryness.
		Add HNO_3 , H_2O_2 ; evaporate to near dryness.
		Dissolve in 6 <u>N</u> HNO ₃ .
	Separation	Evaporate to near dryness.
		Add 6 <u>N</u> HNO ₃ and NH_4NO_3 until saturated.
		Extract Pu with hexone. (Pu separation).
		Back-extract with 0.1 \underline{N} HNO ₃ .
		Evaporate to dryness; dissolve in 6 <u>N</u> HNO ₃ .
		Load on Dowex 1×4 column.
		Proceed with Pu purification at appropriate step in procedure given in Table 115.
		Adjust aqueous phase to pH 1 with NH ₄ OH.
		Extract Y with 20% HDEHP (toluene). (Sr-Y separation).
		Proceed with Y purification at appropriate step in procedure given in Table 115.

permitted correction to the recovered amount of iron carrier for any iron originally present in the sample.

Chemically bound tritium measurements were made on 35 selected marine, vegetation, and animal samples. A 10-g sample of dried material was taken for each determination. The chemical procedure for this determination is as follows: The sample is ignited in the presence of 300-psi O_2 in a Parr bomb. Water formed by this oxidation is distilled in vacuum into a methanol-dry ice trap. After the sample is warmed, NaOH is added to neutralize the solution. The sample is redistilled. The water is then reduced with magnesium at 600°C to produce hydrogen gas which is put through a molecular sieve trap and then collected on a charcoal trap at liquid nitrogen temperature. The sample is ready for counting.

Carbon-14 measurements were made on 21 selected marine and vegetation samples. The desired sensitivity was obtainable with a sample size of only 300 mg of dried material. The chemical procedure for this determination is as follows: The sample is placed in a 2-liter, heavy-walled flask which has been flushed with oxygen. Following ignition of the sample and complete oxidation, the flask is cooled to freeze out water. A trapping solution (15 ml) of phenylethylamine, toluene, and methanol is introduced into the flask to quantitatively absorb any CO_2 present. Aliquots of this solution are taken for counting.

Various chemical procedures were devised at MCL and LFE to isolate and purify fractions containing ⁶³Ni, ^{113m}Cd,

¹⁴⁷Pm, ¹⁵¹Sm, and ²⁴¹Am. Chemical yields were determined by the addition of known amounts of either carrier solutions (Ni, Cd, Sm) or tracers $({}^{143,144,146}P_{m})$ ²⁴³Am). Several precipitations of nickel dimethylglyoxime were the key purification steps for a nickel fraction. Cadmium was isolated by precipitation of CdS and purified by absorption on (in 2 M HCl) and elution from (in 1.5 $\underline{M} H_2 SO_4$) a Dowex 1×8 column. The two rare earths and americium were carried through common chemistry; elemental separation was achieved by use of a Dowex 50 column eluted with α -hydroxyisobutyric acid. a standard technique for intra-group separation of lanthanides and actinides.

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Counting Techniques

Techniques for measuring nuclides in samples which were purified by wet chemistry are summarized in Table 112.

Although all of the nuclides of interest (Table 112) have half-lives long enough that decay of a counting sample cannot be used conveniently as a means of identification, determination of 90 Sr by chemically "milking" the 64-hr 90 Y daughter does permit one to follow decay of the energetic Y beta particles. Interference from other radioactivities can be readily identified and correction made. It is a highly specific technique for 90 Sr.⁹⁰Y. All laboratories used this method to determine 90 Sr.

Plutonium-238, plutonium-(239, 240), and americium-241 were determined by the addition of an appropriate tracer, ²³⁶Pu, ²⁴²Pu, or ²⁴³Am, at the beginning of analysis, and by measurement of an isotope ratio in a purified sample. Isotope ratios were usually determined _{ey a co}mparison of areas under alpha groups of characteristic energies from . _{pulse}-height analysis data, although some of the plutonium samples were spiked with 242 Pu for measurement by mass spectrometry at MCL. These methods, observation of a characteristic alpha energy or a characteristic massto-charge ratio for an ion, are highly specific for the nuclides in question. Most of the ²³⁸Pu data were measured at MCL where it was determined routinely. Some additional ²³⁸Pu measurements were made at LFE and LRE. Measurements of 238 Pu were made on about 60% of the samples.

Determination of ⁵⁵Fe was based upon detection of a 6-keV Mn K x ray which arises from the electron capture of ⁵⁵Fe. Most often, samples were counted with thin NaI(T1) detectors, although some were measured with planar Ge(Li) diode detectors. Pulse-height analysis was used to provide energy discrimination. Good chemical purity of an iron fraction is required to eliminate interference from other nuclides. The quantitative determination of the 6-keV manganese K x ray also required a correction to account for self-absorption in the iron carrier. This correction varies from sample to sample and may lead to difficulties in comparing data from different laboratories.

Tritium was counted at LLL by introducing the purified sample, as hydrogen gas, into an evacuated proportional counter (2.6-liter volume), adding 380-mm CH₄ and raising the absolute pressure to 1500 mm with tritium-free hydrogen. Acceptable pulses are determined by discrimination in rise time and anticoincidence with a guard counter arranged coaxially around the tritium sample counter.

Counting of ¹⁴C samples was performed at LLL in the following manner: Two equal aliquots of purified CO₂ from a sample, absorbed in a mixture of phenylethylamine, toluene, and methanol, were mixed with liquid scintillator solution (dimethyl POPOP and PPO in toluene). Samples were counted at an optimum ¹⁴C channel on an LS spectrometer for 100 min each. Energy discrimination was used to screen out low-energy betas. e.g., those from tritium decay. When a sample count rate did not exceed background within statistical limits, an upper limit was set at twice the value of the standard deviation.

Another group of nuclides, ⁶³Ni, 113 mCd. 147 Pm, and 151 Sm, all longlived beta emitters, were determined by measuring beta activity in purified samples. Those nuclides with less energetic betas, ⁶³Ni and ¹⁵¹Sm. were measured by liquid scintillation counting. Aliquots of the purified sample were added to a scintillation mixture (dimethyl POPOP and PPO in toluene); each sample was counted with a Tracerlab scintillation spectrometer. The ^{113m}Cd and ¹⁴⁷Pm samples were measured with gas-flow (pure methane) proportional counters. The ¹⁴⁷Pm samples required gamma counting [Ge(Li) detector] as well to provide chemical yield data from the 143, 144, 146 Pm tracer. Because this group of nuclides has long half-lives, so that samples cannot be conveniently followed for decay, preparation of the samples was done with considerable care to insure good chemical purity.

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Soil Samples

An initial set of soil samples was distributed to the laboratories for calibration purposes. These samples were produced by blending varying proportions of two coral batches with different specific activities. Analytical results for 239,240 Pu and 90 Sr are listed in Table 119. The plutonium content of these samples ranged from 0.41 to 17.8 dpm/g; the 90 Sr content was uniform in all samples, with a mean value of 3.96 ± 0.10 dpm/g. Plutonium analyses for sample 4 showed a large spread and replicates did not agree; presumably, this sample was nonhomogeneous due to inadequate blending. This finding is consistent with the 90Sr results because 90Sr was present only in one of the two ingredients, that which comprised the major portion of each sample. (Included in the table are entries for MCL with the date, 5/73. These three samples were blind standards submitted for analysis in the middle of the analytical program.)

The plutonium results for sample 1 measured in January 1973 are quite con-

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Table 119.	Interlaboratory calibration,	coral soil samples,	239, 240 Pu and ⁹⁰ Sr.
	Co	incentration dom/a	

	Concentration, dpm/g				
	<u>No. 0001</u>	No. 0002	No. 0003	No. 0004	<u>No. 0005</u>
239, 240 _{Pu}					
LLL 1/73	18.0±1.7	0.51±0.04	0.45±0.02	2.52±0.91	0.41±0.02
MCL 1/73	17.6±1.0	0.46±0.02	0.47±0.05	1.90 ± 0.11	0.41±0.02
MCL 5/73	14.6 ± 0.8	0.48±0.02		1.54±0.08	~~~~
LFE 1/73	18.7±0.8			3.01±0.12	0.44 ± 0.03
EIC 1/73	1 6 .9±1.1	0.60±0.15	0.54 ± 0.14	1.48±0.16	0.53±0.14
⁹⁰ Sr					
LLL 1/73					3.40±0.17
MCL 1/73	4.13±0.12	4.14±0.58	4.15±0.23	3.90±0.12	4.00±0.12
MCL 5/73	4.04±0.40	3.64 ± 0.17		4.26±0.21	
LFE 1/73	3.43 ± 0.21	3.49±0.21	3.52±0.18	3.68±0.18	3.58±0.21
EIC 1/73	4.83±0.39	3.90 ±0 .33	4.9 2±0. 29	4.29±0.33	3.92±0.29
	⁹⁰ Sr				
		results) = 3.	96±0.43		
	LLL (av				86 (1 sample)
	MCL (ar			L/mean = 1.	
	LFE (av		.54±0.10; LF	,	
	EIC (av)) = 4.	.37±0.49; EIC	/mean = 1.	10

sistent and give a mean value, 17.8 ± 0.4 dpm/g, with 10% spread. Laboratory comparisons for samples 2, 3, and 5 showed a greater spread than for sample 1, with EIC reporting results systematically higher than the mean of LLL, MCL, and LFE by 17-26%, although the relatively large error limits set by EIC encompass the mean of the other laboratories. The ⁹⁰Sr data from all laboratories showed acceptable agreement.

Another plutonium interlaboratory calibration was performed by distributing a standard solution to each laboratory. The results are listed in Table 120. The best value for the solution concentration is probably given by the mean derived from equally weighted values, 1278 ± 14 dpm/ml, because the massspectrometric data from LLL and MCL

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Table 120.	Interlaboratory calibration,	²³⁵ Pu standard	solution (No.	1100).

	Concentration of		
<u>Date</u>	239,240 Pu, dpm/ml	Technique	Reference tracer
LLL			
6/4/73	1303±28	Direct assay, counter efficiency = 49.6%	Counting standard: H.E. ²⁴¹ Am II
6/4/73	1320±20	Pulse-height analysis	²⁴² Pu: Environmental
6/4/73	1265±5	Mass spectrometry	standard ²⁴² Pu: Mass spectro- metry standard
MCL			
10/9/73	1255±15	Pulse-height analysis	²³⁶ Pu
5/9/73	1272 ± 6	Mass spectrometry	²⁴² Pu
LFE			
4/25/73	1330 ± 27	Pulse-height analysis	²³⁶ Pu
LRE			
6/29/73	1273±64	Pulse-height analysis	²³⁶ Pu (HASL calibration)
EIC			
3/6/73	1207±54	Pulse-height analysis	²³⁶ Pu (LLL calibration)
	Mean (equiv wt)	= 1278 ± 14 σ (single det σ (mean)	n) = ± 39 = ± 14
	Mean (weighted)	= 1270±4 σ (single det σ (mean)	n) = ± 12 = ± 4

may have unrealistically low errors quoted. These errors are derived only from counting statistics and do not include any estimates of error from calibration of ²⁴²Pu tracers. Differences between pulse-height analysis of plutonium alpha activity and mass-spectrometric analyses with ²⁴²Pu tracer would include any error in the half-life of ²⁴²Pu. The total spread of the determinations was 9.7%.

Pairs of soil aliquots from common field samples were distributed to MCL, LFE, and EIC over the course of the analytical program. Thus, performance of a given laboratory relative to the others was monitored. In most cases, LFE and EIC results were compared with MCL results. It should be noted that in this comparison nonhomogeneity of any given sample could cause an observed difference between laboratories. However, we have some confidence in this question of homogeneity based upon results obtained by gamma-counting 137 Cs. Analysis of the data for 24 pairs of soil aliquots shows an estimated difference of 6% between duplicate soil aliquots which can be ascribed to lack of homogeneity.

Data for laboratory comparisons of ^{239,240}Pu and ⁹⁰Sr results are given in Tables 121 and 122. Entries in these tables include measured concentrations and errors from each laboratory and a ratio of concentrations with the error on the ratio derived by propagating the measurement errors. Entries in Tables 121 and 122 in parentheses are results which have been discarded before calculating laboratory ratios. The ratios are listed again in Tables 123-126 according to laboratory and nuclide, along with a statistical analysis of each set. Logarithms of the ratios were averaged to produce a mean value for a given set of data. The significance of a mean value differing from unity (i.e., indication of possible bias), was tested by calculating the standard deviation, $s_{\bar{\mu}}$, of the mean, $\bar{\mu}$ (logarithmic mean), multiplying $s_{\bar{\mu}}$ by a factor t which is based upon the 95% confidence level and is obtained from standard tables, and comparing the value of t $\cdot s_{\bar{\mu}}$ with $\bar{\mu}$. If the logarithmic means exceeds t $\cdot s_{\bar{\mu}}$, the observed bias is said to be significant with a 95% level of confidence.

In the LFE/MCL comparisons, the plutonium results (Table 123) show that if one includes all samples (29), a mean laboratory ratio of 1.06 \pm 0.04 is calculated with no evidence for significant bias. If we exclude the two most deviant members of the set, a mean laboratory ratio of 1.02 ± 0.03 is calculated; again, there is no evidence for significant bias. The ⁹⁰Sr comparison (Table 124) shows a mean laboratory ratio (LFE/MCL) of 0.943 ± 0.033 if one includes all samples: a statistical test indicates the bias is not significant in this set of data (the value of t.s_{μ} exceeds that of $\bar{\mu}$). If we exclude the two most deviant members of the set, a pair of ratios which are nearly twice the mean value, a mean laboratory ratio of 0.901 \pm 0.012 is calculated. This reduced set of ratios exhibits much less variation; statistically, the observed bias is significant at the 95% confidence level.

In the EIC/MCL comparisons, the plutonium data (Table 125) have had only one very low ratio excluded; the remainder produces a mean laboratory ratio of 0.85 ± 0.02 , with statistically significant bias indicated. The range of values and

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(Concentration of	239, 240 _{Pu, dpm/g}		Concentration o	d^{90} Sr, dpm/g	
Sample No.	LFE data	MCL data	LEE/MCL	LFE data	MCL data	LFE/MCL
BELLE 100 p	rofile					
30-0834-02	524±21	480 ± 10	1.09 ± 0.05	1940 ± 19	$2140\pm\!39$	0.91±0.02
33-0835-02	132 ± 3	$123\pm\!12$	7 ,1.06±0.03	$868\pm\!\!87$	$959\pm\!\!19$	0.93±0.09
	(196±8)	124 ± 2^{a}			900 ± 16^{3}	
		126 ± 5^{a}			928 ± 15^{41}	
		Av 124±2			$Av/929\pm$	17
34-0836-02	20.3±0.2	19.7±2.2	1.03±0.11	5 1 5±5	586±8	0.8810.01
35-0837-02	5.77 ± 0.12	7.85±0.34	0.69±0.06	222+2	234 ± 1	0.9210.02
	(15.4±0.5)	9.12 ± 0.35^{a}			243 ± 2^{41}	
		8.24 ± 0.31^{a}			24212^{a}	
		Av 8.40±0	.65		Av 2401	5
36-0838-02	1.87 ± 0.06	2.15 ± 0.11	0.85±0.04	13813	75,111.3	1.7710.10
	(1.58±0.03)	2.27 ± 0.15			80.8E1.0	
		Av 2.21±0	.09		Av 78.0	14.0
38-0839-02	1.01 ± 0.04	1.02 ± 0.03^{a}	0.9710.05	31,540,3	33,6±0,4	0.9110.01
	(1.49 ± 0.03)	1.07 ± 0.04^{a}			35.0±0.4 ^a	
		(0.75±0.03)			$34.840.4^{a}$	-
		Av 1.04±0.	.03		Av 34.5	10.3
39-0840-02	0.68±0.01	0.57 ± 0.02^{a}	1.26 ± 0.07	13.1±0.1	13.0±0.1	0.9810.01
		0.52 ± 0.02^{a}			13.6 ± 0.2^{a}	
		(0.4110.03)			13.3 ± 0.2^{4}	
		Av 0.54±0.	. 03		Av 13.3	£0.2
40-0841-02	0.28±0.09	0.145 ± 0.013	1,93±0,63	4,75±0,09	2.73±0.10	1.67±0.10
		(0.064 ± 0.015)			2.96 ± 0.21	
					Av 2,85:	10.16
41-0842-02	0.077 ± 0.003	0.046±0.005	1,4810,23	1.44 ± 0.72	1.42:0.71	1,0110.07
		0.057 ± 0.008				
		Av 0.052±0	0.008			

Table 121. Interlaboratory comparison, 239,240 Pu and 90 Sr data for soil samples, LFE vs MCL.

^aMCL replicates on 0835, 0837, 0839, and 0840 were determined from a new soil sample.

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Table	121 (continued).	
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	Concentration of	239, 240 _{Pu, dpm/g}		Concentration of	of ⁹⁰ Sr, dpm/g	
Sample No.	LFE data	MCL data	LFE/MCL	LFE data	MCL, data	LFE/MCL
CLARA 100 pi	rofile		·			
30-0843-03	191±6	164 ± 18	1.16±0.13	305±3	355±14	0.86±0.04
33-0844-03	163±15	143±12	1.14±0.14	247 ± 5	255 ± 6	0.97±0.03
34-0845-03	62.8±2.5	58.2±5.6	1.08 ± 0.11	220 ± 9	242±4	0.91±0.04
35-0846-03	15,8±0,5	14.2±1.0	1.11±0.09	125±6	138 ± 1	0,9140.09
					138±1	
					Av 138:	± 1
37-0847-03	3.69±0.15	3,57±0.19	1,03±0.09	150 ± 5	166±1	0,90±0.0:
39-0848-03	1.10±0.09	0,84±0,05	1.21±0.18	50.1±1.5	54,4±0,6	0,9410,04
		0,99±0.08			51.9 ± 0.7	
		Av 0.91±0), 11		Av 53.1	2±1.8
DAISY 100 pro	ofile					
30-0852-04	380 ± 11	413±8	0,92±0,03	1400±30	1700±30	0,82:0.0;
33-0853-04	91.5 ± 3.7	86.6±5.8	1.06 ± 0.08	431±9	509±8	0.8510.03
34-0854-04	44.6 ± 1.8	46.8±6.1	1.05 ± 0.15	221 ± 4	239 ± 6	0,92±0,0
		38.5±1.7				
		Av 42.6±5	5.9			
35-0855-04	23, 3±0, 7	25,111.6	0,9340,07		1911-19	
36-0856-04	6,21±0,19	7,6910,43	0,81±0,05	103 ± 1	12015	0,8610,0
38-0857-04	2,97±0,06	2,5240.19	1,18±0.09	71,743.6	73,812,5	0,9710,00
39-0858-04	0.80±0.02	0.86±0.05	0,93±0.06	45.010.9	47.340.6	0,9540,03
0-0859-04	0.33±0.01	0.33±0.03	1,00±0,10	37.2±0.4	41,9±0,4	0.89±0.0
		(0.57±0.16)				

Table 121 (continued).			
<u>Concentration of 239, 240 Pu, dpm/g</u> Sample No. LFE data <u>MCL data</u>	LFE/MCL	Concentration of ⁹⁰ Sr, dpm/g LFE data MCL data	LFE/MCL_

<u>_C</u>	oncentration of	239, 240 _{Pu, dpm/g}		Concentration	of 90 Sr, dpm/g	
Sample No.	LFE data	MCL data	LFE/MCL	LFE data	MCL data	LFE/MCL
JANET surfac	e samples					
32-3793-10	37.7±1.5	36.0±0.4	1.05 ± 0.04	115 ± 2	153±3	0.7510.02
JAN 051-000	-015					
31-3850-10	79,9±3.2	71.1±0.7	1.12±0.05	302 ± 6	35819	0.84±0.00
JAN 066-000	-005					
32-3856-10	45.3±2.3	39.3±0.4	1.15±0.06	196 ± 2	211 ± 7	0,93±0,01
JAN 069-000	-015					
32-4514-10	39.5 ± 2.0	38,2+0.7	1,03±0,05	136 ± 4	150 £5	0,9140,04
JAN 084-000	-015					
32-3922-10	49.0±1.0	54.0±0.5	0,91±0,02	271 ± 5	$336\pm\!9$	0.8110.03
JAN 101-000	-015					
32-3926-10	49.0±1.5	48.0±2.0	1.02±0.05	215 ± 11	232±7	0.9340.06
JAN 103-000	-015					

Table 122,		of 239, 240 _{Pu, dpm/g}		Concententian	of 90 Sr, dpm/g	
					$\frac{\text{or Sr, dpm/g}}{\text{MCL data}}$	EIC/MCL
Sample No.	EIC data	MCL data	EIC/MCL	EIC data	MCL data	
ALICE 100 pro	ofile					
33-0826-01	58.0±2.7	58.7±2.9	0.99 ± 0.07	290 ± 14	336 ± 3	0.88±0.0
				299 ± 6		
				Av 29	5±11	
34-0827-01	2.32±0.14	2.46 ± 0.18	0.91±0.08	96,9±7,2	101 ± 1	0.90±0.0
		2.64±0.23		90,7±3.6		
		Av 2.55±0.	. 16	84.6±3.2		
				Av 90	.7±3.6	
35-0828-01	0.65±0.06	0.66 ± 0.03	1.00±0.11	83,0±5,7	89.210.6	0.86±0.0
		0.65±0.05		72,2±3,1		
		Av 0.65±0.	. 04	75.1±2.8		
				Av 76	.8±3.2	
37-0829-01	0.36±0.05	0.30 ± 0.03	1,20±0,21	64.0±4.8	74.810.4	0.86 ±0.0
				61.5 ± 2.6		
				66.9±3.0		
				Av 64	.1±3.6	
39-0830-01	0.12±0.02	0.073±0.015	1.64 ± 0.34	46.9±2.7	50.0±0.3	0.94±0.0
				48,8±1.9		
				Av 47	.9±2.3	
40-0831-01	0.09±0.03	0.092 ± 0.018	0,98±0,38	30,2±2,2	37.0:0.3	0.82±0.0
IRENE 050 pr	ofile					
30-4720-09	 23.3±1.0	20.3±1.8	1.15±0.11	93,1±1.0	105 ± 4	0,89±0,0
33-4721-09	24.1±1.5	17.2±1.2	1.40±0.13	82,5±0,9	87.413.3	0.94±0.0
34-4722-09	14.2±0.6	16.2±1.2	0.8810.08	76.4 ± 0.8	90.2 ± 2.5	0.85+0.0
35-4723-09	16,5±0,7	21.4±0.3	0.77±0.04	95,4±1,0	114±5	0,84±0.0
37-4724-09	15.2±0.7	19.8±0.3	0.77±0.04	85.0±0.9	114 ± 2	0.7510.0
39-4725-09	24.1±1.0	26.3±0.3	0.92 ± 0.04	104 ± 1	107±6	0,97±0.0
40-4726-09	16.8±0.9	19,3±0.3	0.87±0.05	90,3±1.0	91.3±7.9	0,99±0,0
41-4727-09	7.32±0.52	7.73±0.71	0.95 ± 0.11	53.4 ± 0.6	60.0±2.5	0.77±0.0
42-4728-09	4,09±0,29	6,03±0,60	0,68±0,08	57.2±0.6	54.1±2.5	1.06±0.05

Table 122. Interlaboratory comparison, 239,240 Pu and 90 Sr data for soil samples, EIC vs MCL.

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Table 122 (continued).

Concentration of 239, 240 Pu. dpm/g

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Table 122	(continued).
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	Concentration of	239, 240 Pu, dpm/g		Concentration	of ⁹⁰ Sr, dpm/1	4
Sample No.	EIC data	MCL data	EIC/MCL	EIC data	MCL data	EIC/MCL
PEARL 101 p	rofile					
33-0876-17	269±5	350±33	0.72±0.17	116 ± 1	149 ± 7	0,85±0,1
	206 ± 5	356 ± 9		115 ± 1	122±21	
	289 ± 7					
	Av 255	±43 Av 353±9		Av 11	6±1 A.	v 136±19
34-0877-17	11.0±0.5	11.4±0. 9	1.04 ± 0.13	47.2±0. 5	51.6 ± 2.1	0,91±0,04
	12.6 ± 0.5					
	Av 11.	8±1.1				
35-0878-17	9.82±0.47	12.8±1.2	0.80±0.06	21.6 ± 0.4	68.9±2.7	0.37 ±0.00
	(4.49±0.30)	11.8±0.8		25.9±0.3	60.1±1.0	
	(5.91±0.36)	Av 12.3±0). 7	Av 23	A.8±3.0	v 64.5±6.2
IRENE 100 pr	ofile					
33-0861-09	9.05±0.27	231±17	0.038 ± 0.003	206 ± 2	315±8	0,65±0,0
	8,79±0,46	219±11			31549	
	7.96±0.50	Av 225±9				< 31516
		0±0.57				
34-0862-09	26719	375129	0.74±0.04	672+7	509412	1,2040,1/
	26917	348 ± 14		570±11	522118	
	Av 268	±6 Av 362±19)	Av 62	1172 Av	× 516±9
35-0863-09	$1486\pm\!18$	2100±120	0.74 ± 0.04	3980±80	2960±50	1.3410.04
		1920±90			2950 ±220	
		Av 2010±1	30		Ax	2960±50
37-0864-09	$1367\pm\!\!18$	19 00± 110	0.75 ± 0.04	2260 ± 23	977+32	2,0510.39
		1760±35		1967 ± 20	1420±90	
		Av 1830±9	0	Av 21	15±210 Av	7 1030±140
39-0865-09	535±16	719±40	0.76±0.04	1135 ± 11	731 ± 12	1,55±0,08
		691±32		1075 ± 21	697 ± 20	
		Av 705±25		Av 11	05140 Av	7 7 14 ±24
10-0866-09	305±9	385±25	0,80±0,04	1785±18		1,4110,22
		379±17		1809 ± 36	1140±18	
		Av 382±14		Av 18		1280±200

Table 122 (continued).

	Concentration of	^{239, 240} Pu, dpm/g		Concentration	of ⁹⁰ Sr, dpm/g	
Sample No.	EIC data	MCL data	EIC/MCL	EIC data	MCL data	EIC/MCL
JANET 100 p	rofile					
30-0978-10	128±4	147 ± 5	0.87 ± 0.04	1027±8	1220±20	0,85±0,0
	129±2			1043±8	(332±4)	
	Av 12	8±2		Av 10	035±8	
33-0979-10	44.2±1.5	47.0±1.7	0.92±0.03	498±5	471 ± 9	1.03±0.03
	42,5±0,8			475±5		
	Av 43	.3±0.9		Av 48	6±12	
34-0980-10	5.43±0.37	5.45±0.25	0.94 ± 0.07	17 0±3	168 ± 1	1.01+0.02
	4.76±0.18					
	Av 5.	10±0.33				
PEARL 100 p	rofile					
30-0867-17	18.1±0.8	22.8±1.1	0.81±0.05	25.2±1.2	23.3±2.1	1,06±0.10
	18.9±0.8			24.4 ± 1.0	(7.92 ± 1.74)	
	Av 18	.5±0.6		Av 24	.8±0.6	
33-0868-17	9.03±0.43	8.65±0.31	1.14±0.10	25.5±1.1	18.0±0.8	1.15±0.29
	10.69±0.58			18,2±0.8	19,8±1,5	
	Av 9.	36±0.83		Av 21	.8±5.2 Av 18.9:	±1.3
34-0869-17	8,45±0.48	8.91±0.41	1.14 ± 0.20	31.7±1.3	33.5±2.8	0.95±0.09
	11.89±0.84					
	Av 10.	17±1.72				
SALLY 200 pr	ofile					
30-0884-19	9.09±0.48	11.1±0.4	0.82±0.04	71.5±2.3	71.2±0.9	1.00±0.03
	9.09±0.27					,
	Av 9.0	9 ±0.27				
30-0 885-1 9	12.75±0.73	12,9±0.6	0,99±0.05	104 ±2	81±5	1.29±0.08
	12.75±0.37				(143±5)	
	Av 12.	75±0.37				

Table 122	(continued).					
	Concentration o	f ^{239,240} Pu, dpm/g		Concentration	of ⁹⁰ Sr, dpm/g	
Sample No.	EIC data	MCL data	EIC/MCL	EIC data	MCL data	EIC/MCI

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	Concentration o	f ^{239,240} Pu, dpm/g		Concentration	of 90 Sr, dpm/g	
Sample No.	EIC da ta	MCL data	EIC/MCL	EIC data	MCL data	EIC/MCL
ALICE 024 pr	ofile					
30-2044-01	148±5	208±11	0.71±0.04	1230 ± 12	1360 (41	0,9010,0
33-2045-01	377 ± 12	6 37 ±7 0	0.59 ± 0.07	2210±22	2500 ± 98	0.8810.04
34-2046-01	109±4	139±7	0,78±0,05	751 ± 8	7 97±25	0,9410.03
IRENE 047 pr	ofile					
30-4693-09	50.6 ± 1.0	65,2±3,2	0,78±0,04	23612	20517	1,0140,20
				179 ± 2		
				Av 20	18±40	
33-4694-09	55.0 ± 1.0	82.6±3.9	0.67±0.03	18942	21547	0.8310.07
				169±2		
				Av 17	9±14	
34-4695-09	50.0 ± 0.9	77.4±7.1	0 ,65±0,06	159 ± 2	82,3±5,6	2,02±0,18
				173 ± 2		
				Av 16	6±10	
35-4696-09	57.6±1.2	88.7±5.0	0.65±0.04	265 ± 3	95.0±5.1	2,6310,25
				236 ± 2		
				Av 25	0.120	
34-4697-09	89.4±2.1	141±9	0.63±0.04	300±3	122+7	2, 5440, 19
				320±3		
				Av 31	0 ± 14	
39-4698-09	294 ±6	379±33	0.78±0.07	576 ± 6	59 2 ±27	0.91±0.11
				4.96 ± 5		
				Av 53	6±57	
40-4699-09	197 ± 4	280±1 6	0.70±0.04	402±4	$433{\pm}21$	0.84±0.14
				322±3		
				Av 36	2±57	
41-4700-09	139 ± 3	21 9±10	0,63±0,03	27 0±3	370±13	0.67±0.08
				226 ± 2		
				Av 24	8±30	
12-4701-09	175±4	272±14	0.67±0.04	283 ± 3	312±11	0.81±0.14
	189±5			222±2		
	Av 182	2±5		Av 25	2 ±4 3	

Table 122 (continued).

F	latios of me <mark>asure</mark> d	2 ^{39,240} Pu concentra	ations, LFE/MCL
BELLE 100 profile	1.09±0.05	JANET surface	1.05±0.04
	1.06±0.03		1.12±0.05
	1.03±0.11		1.15±0.06
	0.69 ± 0.06		1.03±0.05
	0.85±0.04		0.91±0.02
	0.97 ± 0.05		1.02 ± 0.05
	1.26 ± 0.07		
	(1.93 ± 0.63)	DAISY 100 profile	0.92±0.03
	(1.48 ± 0.23)		1.06±0.08
			1.05 ± 0.15
CLARA 100 profile	1.16 ± 0.13		0.93±0.07
	1.14 ± 0.14		0.81±0.05
	1.08 ± 0.11		1.18±0.09
	1.11±0.09		0.93±0.06
	1.03±0.09		1.00 ± 0.10
	1.21±0.18		

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1 able 123.	Interlaboratory	comparison,	Enewetak so	11 samples.

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No.	=	29	27
$ar{\mu}$	=	0.05731	0.02268
s ₁	=	0.1846	0.1315
s_1^2	=	0.03408	0.01729
s_ µ	=	0.03428	0.02530
t•s	÷	0.07010	0.05192
$\mathrm{e}^{ar{\mu}}$	=	1.06 ± 0.04	1.02 ± 0.03
Range		0.69-1.93	0.69-1.26
Significant bias	?	No	No

	asured ^{SU} Sr concentration	5, <u>ET E MCE</u>
ELLE 100 profile 0.91±0.02	JANET surface	0.75±0.02
0.93±0.09		0.84±0.03
0,88±0.01		0.93±0.03
0.92±0.02	!	0.91 ± 0.04
(1.77 ± 0.10))	0.81±0.03
0.91±0.01		0.93±0.06
0.98±0.01		
(1.67 ± 0.10)) DAISY 100 profile	0.82±0.02
1.01 ± 0.07		0.85±0.02
		0.02±0.03
LARA 100 profile 0.86±0.04		0.86±0.04
0.97±0.03	;	0.07±0.06
0.91 ± 0.04		0.95±0.02
0.91±0.05	i i i i i i i i i i i i i i i i i i i	0.89±0.01
0.90±0.03		
0.94 ± 0.04		
No. = 28	26	
$\bar{\mu}$ = - 0.05863	-0.1048	
$s_1 = 0.1811$	0.06548	
$s_1^2 = 0.03280$	0.004287	
	0.01284	
$s_{\bar{\mu}} = 0.03423$ t·s_{\bar{\mu}} = 0.07010	0.02640	
	0.901 ± 0.012	
Range = 0.75-1.77 gnificant bias? No	0.75-1.01 Yes	

Table 124. Interlaboratory comparison, Enewetak soil samples.

Ratio	os of measured	239,240 Pu concentrati	ons, EIC/MCL
ALICE 100 profile	0.99±0.07	JANET 100 profile	0.87±0.04
	0.91±0.08		0.92±0.03
	1.00±0.11		0.94 ± 0.07
	1.20 ± 0.21		
	1.64 ± 0.34	SALLY 200 profile	0.82 ± 0.04
	0,98±0.38		0.99±0.05
ALICE 024 profile	0.71±0.04	PEARL 100 profile	0.81±0.05
	0.59±0.07		1.14±0.10
	0.78±0.05		1.14±0.20
PEARL 101 profile	0.72±0.17	IRENE 050 profile	1.15±0.11
	1.04±0.13		1.40±0.13
	0.80±0.06		0.88±0.08
			0.77 ± 0.04
IRENE 047 profile	0.78±0.04		0.77 ± 0.04
	0.67 ± 0.03		0.92 ± 0.04
	0.65±0.06		0.87 ± 0.05
	0.65±0.04		0.95 ± 0.11
	0.63 ± 0.04		0.68±0.08
	0.78 ± 0.07		
	0.70 ± 0.04	IRENE 100 profile	(0.038 ± 0.003)
	0.63±0.03		0.74 ± 0.04
	0.67 ± 0.04		0.74 ± 0.04
			0.75 ± 0.04
			0.76 ± 0.04
			0.80±0.04
No. =	43		
<i>μ</i> =	0.1670		
s = _s 2 =	0.2223		
	0.0491		
S _µ =	0.03390		
t•s =	0.06846		

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Table 125. Interlaboratory calibration, Enewetak soil samples.

eμ

Range

Significant bias?

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0.85±0.02

0.59-1.64

Yes
R	atios of measur	ed ⁹⁰ Sr concentrations,	EIC/MCL
ALICE 100 profile	0.88±0.03	IRENE 100 profile	0.65±0.02
	0.90±0.04		1.20±0.14
	0.86±0.04		1.34 ± 0.04
	0.86±0.05		(2.05 ± 0.35)
	0.94 ± 0.05		1.55±0.08
	0.82±0.05		1.41±0.22
ALICE 024 profile	0.90±0.03	IRENE 047 profile	1.01±0.20
	0.88±0.04		0.83±0.07
	0.94 ± 0.03		(2.02±0.18)
			(2.63±0.25)
PEARL 101 profile	0.85±0.12		(2.54±0.19)
	0.91±0.04		0.91±0.11
	(0.37±0.06)		0.84±0.14
			0.67±0.08
JANET 100 profile	0.85±0.02		0.81±0.14
	1.03±0.03		
	1.01±0.02	IRENE 050 profile	0.89±0.04
			0.94 ± 0.04
PEARL 100 profile	1.06 ± 0.10		0.85±0.03
	1.15±0.29		0.84±0.04
	0.95±0.09		0.75±0.02
			0.97±0.05
SALLY 200 profile	1.00 ± 0.03		0.99±0.09
	1.29 ± 0.08		0.77±0.03
			1.06±0.05
No. = 44	1	39	
μ = - (0.0004	-0.0601	
s ₁ = (0.3486	0.1837	
s ₁ ² = (0.1215	0.03375	
	0.05256	0.02942	
	0.1061	0.05942	
$\mu = 1$	00±0.05	0.94 ± 0.03	
	. 37-2.63	0.65-1.55	
Significant bias?	No	Yes	

Table 126. Interlaboratory calibration, Enewetak soil samples.

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samples

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the standard deviation of a single member of the set are both comparable with values derived for the LFE/MCL plutonium comparisons; however, this EIC/MCL set . exhibits significant bias. The EIC/MCL ⁹⁰Sr comparison (Table 126) is characterized by somewhat greater variation in values than observed in the preceding tables. If we include all values listed in Table 126, the mean laboratory ratio is 1.00 ± 0.05 , with no indication of significant bias. If we exclude the five most deviant ratios, the range of values and the standard deviation of a single member of the set are reduced to values comparable to those encountered in the other comparisons. The mean laboratory ratio for this reduced set is 0.94 ± 0.03 , with statistically significant bias indicated.

Of the four comparisons just discussed, only the LFE/MCL plutonium data showed both satisfactory mean values and no evidence for bias. If one wants to derive a most consistent set of data. the derived values for interlaboratory bias can be used to adjust all of the plutonium and strontium soil data to a common calibration. However, considering the urgency for completion of the survey report and the general nature of the schedule of the analytical program, we have not made any arbitrary adjustment of data. If one considers ≤10% bias as acceptable for the purposes of this survey, only the EIC/MCL plutonium comparison remains a problem. Although the data presented in this report are as reported by each laboratory, we have identified the source of wet-chemistry analyses for each sample in the general data bank given in Appendix II, if there is need for data adjustment.

Marine Samples

Among the marine samples, interlaboratory comparison was made by distributing aliquots of dried and homogenized material to more than one laboratory. These comparisons were made to check laboratory results for bias, recognizing that some samples may present difficulties due to incomplete homogenization, e.g., eviscerated whole fish which show pieces of bone, etc. Comparisons between LRE and MCL for ^{239,240}Pu, ⁹⁰Sr, and ⁵⁵Fe are listed in Table 127. In many cases a comparison is labeled either consistent or inconsistent. Consistent comparisons usually involve an upper limit set by one laboratory and an actual measurement by the other laboratory, where the measured concentration is lower than the upper limit. Those comparisons where a measured concentration exceeded an upper limit are labeled inconsistent. The results are summarized in Table 128.

The effectiveness of this comparison is reduced because ^{239,240}Pu and ⁹⁰Sr results from MCL for many samples are upper-limit values. Positive signals from both laboratories were obtained for only a few samples. One factor which led to this situation was generally low concentrations of plutonium and strontium in marine samples; given additional time and effort, additional measurements for interlaboratory calibration would be appropriate.

For ^{239,240}Pu, five valid comparisons gave a mean laboratory ratio, LRE/MCL^{-1} 1.01 $\frac{+0.06}{-0.05}$, with no evidence of significant bias. There were four comparisons where the results were inconsistent; the worst of these cases was for 09-0381-37

Sample		Concentration of 239, 240 Pu, dpm	Concentration of ⁹⁰ Sr, dpm/f	Concentration of ⁹⁰ Sr, dpm/g dry		
08-0787-10 Tridacna,	LRE	0.129±0.007	LRE	0.16±0.04	LRE	4.49±0.06 4.65±0.10 4.57±0.11 av
mantle	MCL	0.122±0.017 0.130±0.007 0.126±0.007 av	MCL	< 0.37	MCL	4.24+0.93 4.78±0.32 4.51±0.73 av
		LRE/MCL = 1.02±0.07		Consistent		LRE/MCL = 1.01±0.16
09-0427-20	LRE	0.07 ± 0.007	LRE	0.123±0.012	LRE	21.7±0.2
Goatfish,	MCL	< 0.036	MCL	< 0.55	MCL	24.9±2.5
eviscerated whole		Inconsistent		Consistent		LRE/MCL = 0.8740.09
09-0374-37	LRE	0.013 ± 0.003	LRE	0,029±0,008	LRE	13.0±0.1
Goatfish,				0.020±0.018		13.9±2.1 13.0±0.1 av
eviscerated whole	MCL	< 0.019	MCL	< 0.66	MCI.	(54.943.7) 14.441.6 12.841.4 13.641.1 av
		Consistent	Consistent		$LRE/MCL = 0.96\pm0.08$	
09-0376-37	LRE	0.026±0.013	LRE	0.090±0.011	LRE	5.21 ± 0.06
Goatfish, eviscerated whole	MCL	< 0.016	MCL	0.58±0.08	MCI.	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
		Consistent		LRE/MCL = 0.16±0.03		LRE/MCL = 0.4310.02
09-0381-37 Convict surgeon,	LRE	0.406±0.090 0.311±0.079 0.358±0.084 av	LRE	< 0.07	LRE	1.4910.10
eviscerated whole	MCL	< 0.013 < 0.005	MCL	< 0. 27	MCL.	< 4.3
		Inconsistent		Consistent		Consistent
09-0383-37	LRE	0.088±0.008	LRE	< 0.02	LRE	0.68±0.03
Convict surgeon,	MCL	< 0.014	MCL	< 0.71	MCL.	15.4
eviscerated whole		Inconsistent		Consistent		Consistent
09-0385-37	LRE	0.040±0.004	LRE	0.03±0.01	LRE	0.60±0.08
Convict surgeon,	MCL	< 0.016	MCL	< 0,68	MCL	< 5.6
eviscerated whole		Inconsistent		Consistent		Consistent

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Table 127 (continued).

Sample		Concentration of ^{239,240} Pu, dpm/g dry		Concentration of 90 Sr, dpm/g	g dry	Concentration of 55 Fe, dpm/g dr	
09-0493-02	LRE	0.141±0.024	LRE	0.300±0.020	LRE	18.9±0.3	
Mullet,						16.8 ± 0.2 17.9±1.5 av	
eviscerated whole	MCL	0.031±0.003	MCL	0.394 ± 0.034	MCL	17.911.0	
		LRE/MCL = 4.5±0.9		LRE/MCL = 0.7610.08		LRE/MCL = 1.0040.10	
09-0497-02	LRE		LRE	0.443±0.013	LRE	21.8±0.20	
Mullet,	MCL	0.042±0.008	MCL	0.654 ± 0.044	MCL.	21.7±0.54	
eviscerated whole				$LRE/MCL = 0.68\pm0.05$		$LRE/MCL = 1.00\pm0.03$	
09-0498-02	LRE	19.5±0.6	LRE	17.9±0.2	LRE	184±1	
Mullet, viscera	MCL	18.6±0.7	MCL	< 2.6	MCL	40 2 ±6	
		$LRE/MCL = 1.05\pm0.05$		Inconsistent		$LRE/MCL = 0.46\pm0.01$	
09-0499-02	LRE	0.013±0.003	LRE	0.049 ± 0.015	LRE	4.61±0.04	
Mullet, muscle	MCL	0.012±0.002	MCL	< 0.22	MCL	18.7±0.5	
		$LRE/MCL = 1.12\pm0.31$		Consistent		$LRE/MCL \approx 0.25\pm0.01$	
09-0448-09	LRE		LRE		LRE	104±1	
Snapper	MCL	0.146 ± 0.012	MCL	1.84±0.08	MCL	170 ± 4	
						$LRE/MCL = 0.61\pm0.02$	
09-0451-09	LRE	0.045±0.007	LRE	0,097±0,016	LRE	33.510.4	
Mullet, muscle	MCL	0.54 ± 0.08	MCL	< 0.33	MCL.	39.2±1.6	
		$LRE/MCL = 0.083\pm0.018$		Consistent		$LRE/MCL = 0.85\pm0.04$	
09-0453-09	LRE	0.415±0.031	LRE	2.05±0.03	LRE	26.9±0.2	
Mullet	MCL	0.388±0.028	MCL	1,96±0.10	MCL	37.4±1.4	
		$LRE/MCL = 1.07\pm0.11$		$LRE/MCL = 1.05\pm0.06$		LRE/MCL = 0.72±0.03	
09-0511-10	LRE		LRE	0.022±0.004	LRE	3.08±0.03	
Mullet,	MCL	< 0,064	MCL	< 0.68	MCL	< 4.0	
eviscerated whole				Consistent		Consistent	
09-0575-20	LRE		LRE		LRE	10.4 ± 0.1	
Snapper, muscle	MCL	< 0.004	MCL	< 0.24	MCL	13.3±0.9	
						$LRE/MCL = 0.78\pm0.05$	

Table 127 (continu

Sample		Concentration of 239,240 Pu, d	om/g dry	Concentration of ⁹⁰ Sr,	, dpm/g dry	Concentration of ⁵⁵ Fe, dpm/g dry
09-0516-21	LRE	< 0.13	LRE	0,224±0,021	LRE	5.39±0.04
Convict surgeon,	MCL	0.037 ± 0.010	MCL	0.28	NIC L	4.63±0.42
eviscerated whole		Consistent		Consistent		$LRE/MCL = 1.16\pm0.10$
09-0481-36	LRE		LRE		LRE	19142
Skipjack, light	MCL	< 0.009	MCL	~ 0.22	MCL	203±3
muscle						$LRE/MCL = 0.94\pm0.02$
09-0685-37	LRE	< 0.043	LRE	0.05±0.02	LRE	4.17±0.05
Parrotfish,	MCL	< 0.038	MCL	< 0.54	MCL	7.45:1.79
muscle		Consistent		Consistent		$LRE/MCL \geq 0.56\pm 0.13$
09-0687-37	LRE	0.40±0.05	LRE	0.16±0.04	LRE	289(3
Parrotfish,				0.0910.02		
viscera	MCL	0.49±0.09	MCL	0.99	MCL.	473 ± 32
		$LRE/MCL = 0.82\pm0.19$		Consistent		LR4E/MC12 0,6110,04
09-0531-47	LRE	0.016 ± 0.003	LRE	× 0.10	LRE	116±2
Skipjack,						131 ± 2 124±11 av
light muscle	MCL	< 0.014	MCL	·· 0, 17	MCT.	142±1
-		< 0.011				137±3 142±5 av
						14714
		Consistent		Consistent		$LRE/MCL = 0.87\pm0.10$
09-0693-54	LRE	< 0.0025	LRE	0.14	LRE	6,8810,10
Yellowfin tuna,	MCL	< 0.0048	MCL	0.75	MCL	8.84±0.13
light muscle		Consistent		Consistent		LRE/MCL = 0.7840.02

LIVE MCL.			-	
239,240 _{Pu}	90 _{Sr}	⁵⁵ Fe	⁵⁵ Fe	
1.02±0.07	(0.16 ± 0.03)	1.01±0.16	0.85±0.04	-
(4.5±0.9)	0.76±0.08	0.87±0.09	0.72±0.03	
1.05±0.05	0.68±0.05	0.96±0.08	0.78±0.05	,
1.12±0.31	1.05 ± 0.06	0.43±0.02	1.16 ± 0.10	
(0.083±0.018)		1.00±0.10	0.94 ± 0.02	
1.07±0.11		1.00±0.03	0.56 ± 0.13	
0.82±0.19		0.46 ± 0.01	0.61 ± 0.04	
		0.25±0.01	0.87 ± 0.10	
		0.61±0.02	0.78±0.02	
No. = 5	3	18		
$\bar{\mu}$ = 0.01023	-0.2038	-0.3214		
s = 0.1215	0.2257	0.3855		
$s^2 = 0.01476$	0.05093	0.1486		
$s_{\bar{\mu}} = 0.05434$	0.1303	0.09086		
$t \cdot s_{\bar{\mu}} = 0.1397$	0.560	0.1909		
$e^{\vec{\mu}} = 1.01^{+0.06}_{-0.05}$	$0.82^{+0.11}_{-0.10}$	0.72±0.07		
No significa	ant No significant	Significant bias		
bias	bias			

Table 128. Interlaboratory calibration, marine samples, ratios of concentrations, LRE/MCL.

(convict surgeon, eviscerated whole) where the observed plutonium concentration at LRE was 0.36 ± 0.08 dpm/g and upper limits set at MCL were < 0.013 and < 0.005 dpm/g. Duplicate samples were run at both laboratories. There is no obvious explanation for this discrepancy. Various possibilities, such as a nonhomogeneous sample, mixup in sample designation, plutonium contamination in the laboratory, or loss of plutonium in chemistry may be invoked as explanations. In the other three instances, measured plutonium concentration values from LRE exceeded upper limits set at MCL by factors of 2, 2.5, and 6.

For 90 Sr, much of the data were consistent but very often LRE reported a low-level measurement while MCL tended to report an upper limit at some higher concentration. Four valid comparisons were obtained, three of which gave moderate agreement between laboratories. The gen laboratory ratio, LRE/MCL, derived from these three samples was 0.82 ± 0.11 ; the evidence is insufficient for determining significant bias. For the fourth sample, a laboratory ratio of 0.16 ± 0.03 was obtained. Also, one sample (09-0498-02, mullet viscera) produced :01 inconsistent results: LRE reported ิษมไ 17.9 \pm 0.2 dpm/g ⁹⁰Sr and MCL set a 1 50

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limit of > 2.6 dpm/g. Comments regarding possible explanations for inconsistent results as stated in the previous paragraph apply here as well.

For ⁵⁵Fe, the largest set of interlaboratory comparison data for marine samples was obtained. From 18 samples, a mean laboratory ratio of LRE/MCL = $0.72 \stackrel{+}{=} \stackrel{0}{_{-}} \stackrel{1}{_{-}} \stackrel{0}{_{-}} \stackrel{1}{_{-}} \stackrel{0}{_{-}} \stackrel{1}{_{-}} \stackrel{0}{_{-}} \stackrel{1}{_{-}} \stackrel{1}{_{$ of significant bias. No inconsistent results were observed. Additional effort on calibration of detectors at LRE and MCL for counting 6-keV x rays from ⁵⁵Fe decay is going on at this time. Relative to calibration data included with a standard ⁵⁵Fe solution obtained from Amersham, the MCL detector is in agreement, while the LRE detector calibration is apparently low. Although this investigation of interlaboratory bias is not finished, the data could be made more self-consistent (and, apparently, more accurate) by adjusting the LRE ⁵⁵Fe results upward by 1/0.72 (or 1.39). Incidentally, interlaboratory calibrations between LFE and MCL carried out in 1969 and 1971 for other programs have shown good agreement for ⁵⁵Fe results (LFE/MCL = 0.968). However, data for ⁵⁵Fe in marine samples from LRE listed in the data bank (Appendix II) have not been given any arbitrary adjustment because: (1) The problem has not been entirely resolved as to which measurements are most accurate. and (2) 55 Fe plays an insignificant role in dose estimates for the marine food chain; the major sources of biological dose in this pathway are 137 Cs, 60 Co, and 90 Sr. We concluded that adjustment of the LRE ⁵⁵Fe data would cause no detectable

change in dose estimates from the marine pathway.

Comparison was made between LFE and MCL on four marine samples; the results are listed in Table 129. The first sample in the table (08-0782-20, viscera and gut contents of sea cucumber) was judged to be quite nonhomogeneous. based upon the 90Sr data and the amount of coral found as gut content. The second sample (09-0393-33, convict surgeon, eviscerated whole) did not produce agreement between laboratories; concentrations of 239,240 Pu and 90 Sr measured at LFE are factors of 5-7 times higher than those measured at MCL. Consistent results were observed for the last two entries for 239,240 Pu and 90 Sr and moderately good agreement was obtained between laboratories for ⁵⁵Fe.

Replicate Analyses

A question of importance to the analytical program was whether soil samples, having been dried, ground, and blended, were homogeneous enough to allow meaningful comparison of analyses from separate aliquots of finely divided soil. In order to investigate this question, about 100 pairs of replicate soil samples were prepared and gamma-counted. Measured concentrations of 60 Co and 137 Cs in each pair were compared. From this set of data, a group of 24 pairs was selected where both samples of a given pair were counted with the same Ge(Li) detector system. Estimates of the variance for this set of 24 replicate analyses were calculated in two different ways. The first, s_1^2 , is derived from the spread of replicate measurements and is calculated as indicated in Table 130.

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Sample		Concentration of ^{239,240} Pu, dpr	n/g dry	Concentration of 90 Sr, dpm/g	dry	Concentration of ⁵⁵ Fe, dpm/g dry
08-0782-20	LFE	1.18±0.01	\mathbf{LFE}	4.15±0.17	LFE	1,77±0,30
Sea cucumber, viscera and gut contents	MCL	0.93±0.06 1.42±0.05 LFE/MCL = 1.00±0.30	MCL	1.85±0.26; 0.12±0.03 LFE/MCL = 2.240.3; 35	MCL	<19.2 LFE/MCL: Consistent
09-0393-33	LFE	0.096 ± 0.003	LFE	0.5240.02	$\mathbf{L}\mathbf{F}\mathbf{E}$	4.0240.28
Convict surgeon,		0.013±0.002	MCL	$< 0.77; 0.11 \pm 0.02$	MCL	< 8.5
eviscerated whole		$LFE/MCL = 7.2 \pm 1.1$		$LFE/MCL = 4.7\pm0.9$		LFE/MCL: Consistent
09-0300-43	LFE	0.0046 ± 0.0006	LFE	0.20±0.01	LFE	49.2±0.5
Goatfish,	-	< 0.032	MCL	< 0.85	MCI.	53.3±4.0
eviscerated whole	men	LFE/MCL: Consistent		LFE/MCL: Consistent		$LFE/MCL = 0.92\pm0.07$
09-0345-43	LFE	0.011 ± 0.001	LFE	0.15±0.01	LFE	9.3±0.6
-		< 0.024	MCL	< 0.65	MCL	13.7 ± 2.7
Mullet, eviscerated whole		LFE/MCL: Consistent		LFE/MCL: Consistent		LFE/MCL = 0.68±0.14

Table 129. Interlaboratory comparison, marine samples - LFE and MCL.

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		⁶⁰ C	0	¹³⁷ C	s
		Uncorrected		Uncorrected	
$s_1^2 = \frac{\sum \left[\frac{2(x_i - y_i)}{(x_i + y_i)}\right]^2}{2n}$	-	0.00408	0.00464	0.00475	0.00468
s ₁	=	0.0639	0.0681	0.0689	0.0684
$s_2^2 = \frac{\sum_{i=1}^{n} (\sigma_{x_i}^2 + \sigma_{y_i}^2)}{2n}$	=	0.00321		0.00119	
s ₂	Ξ	0.0567		0.0345	
n	=	24		24	
s_1^2/s_2^2	=	1.27	1.44	4.0	3.9
F	=	2.0		2.0	
$\begin{bmatrix} s_1^2 & - & s_2^2 \end{bmatrix} \frac{1/2}{}$	=	0.030		0.060	0.059

Table 130. Gamma-spectroscopy replication, A vs D samples. Two soil aliquots counted with same detector system.

The second, s_2^{2} , is derived from experimental uncertainties quoted for each measurement (σ_{x_i} and σ_{y_i} are expressed as percentage uncertainty) and is calculated as indicated in Table 130.

The two estimates of variance are compared to see if there is a statistically significant difference between them. Should this be the case — and assuming that s_1^2 will be greater than s_2^2 — the source of the difference can be ascribed to nonhomogeneity of samples. If the samples are homogeneous, s_1^2 and s_2^2 should be the same statistically. In order to test this question, we perform an F-test on the ratio, $F = s_1^2/s_2^2$, where s_1^2 has n_1 degrees of freedom and s_2^2 has n_2 degrees of freedom. Standard tables of values for F as a function of n_1 , n_2 , and confidence level are available. Should the ratio, s_1^2/s_2^2 , exceed a value of F extracted from the table, the estimates of variance differ in a statistically significant way.

Tests were made with our data at a 95% confidence level. Results are shown in Table 130. For 60 Co, with data in the first column marked "Uncorrected," we find that s_1^2 values do not differ significantly.

A refinement to the data was added because some of the replicate pairs were packaged in different-sized cans due to lack of sample. The counting results for these pairs were corrected for any difference in counting efficiency between can sizes observed when a series of standard soil samples was counted at the beginning of the program. The data, with these corrections, are listed in columns labeled "Corrected." For ⁶⁰Co, there was little change in the result; in fact, corrected results showed slightly greater imprecision.

The ¹³⁷Cs data, which exhibit greater precision in experimental uncertainties due to higher concentrations than the ⁶⁰Co data, indicate a statistically significant difference in estimates of variance. Standard deviation for a single pair in the set is 6.9%, based upon the spread of duplicates; standard deviation based upon quoted experimental uncertainties is 3.5%. An estimate of imprecision due to nonhomogeneity is derived by taking the square root of $(s_1^2 - s_2^2)$. We obtain an average contribution of 6% imprecision in duplicates due to sample inhomogeneity. There is no difference between results for corrected and uncorrected data. Thus, we have an estimate of effectiveness in the sample homogenization procedure.

Measurement of ²⁴¹Am Concentration – Alpha Detection vs Gamma Detection

In addition to measurement of ²⁴¹ Am content in every sample by gamma assay, wet-chemistry analyses were performed on a selected number of samples, and ²⁴¹ Am concentrations were determined by alpha counting. In practice, ²⁴³ Am tracer is added to permit measurement of chemical yield, and a 241/243 activity ratio is measured by pulse-height analysis techniques. These measurements served two purposes: (1) To permit comparison of two different methods for measuring ²⁴¹Am, and, for other samples, (2) to provide greater sensitivity for detecting ²⁴¹Am than available from routine gamma counting.

Comparison data for 24 soil samples are shown in Table 131. A mean value for the ratio between wet-chemistry determination and gamma counting, MCL/LIL is 1.20 ± 0.05 , with evidence for significant bias. Since relatively large errors. 25-35%, were quoted for about half of these samples, we have calculated a value for the MCL/LLL ratio, 1.21 ± 0.04 based upon samples with more precise 241 Am data (first 11 entries in table). As with the entire set, there is evidence for significant bias. Comparison of a variance for this set of 11 calculated from variation of samples from the mean (s_1^2) with a variance calculated from experimental uncertainties quoted for individual measurements shows no significant difference between them (based upon an F test at a 95% confidence level). Thus, the variation of values in this set of 11 results can be accounted for by experimental uncertainties.

Accurate determination of ²⁴¹ Am in soil samples of nominal 300-g mass and at counting geometries (flush against the detector housing face) typical of this program's gamma spectrometry is judged to be difficult. Questions of selfabsorption, changes in geometry due to settling of soil in the can, incomplete grinding of certain samples, lack of homogeneity, and other problems can be raised in discussing ultimate accuracy of the method. Chemical isolation of americium and assay via alpha pulse-height analysis is expected to be more accurate for this kind of sample. Thus, it appears that

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	Alpha count	Gamma spectrometry	
Sample No.	(MCL), dpm g	(LLL), dpm g	MCL_LLL
33-2039-01	4.13±0.38	4.32±0.20	0.96 ± 0.11
34-2040-01	5.55 \pm 0.45	4.28±0.37	1.30±0.15
37-2042-01	7.06 ± 0.48	5.42 ± 0.35	1.30 ± 0.12
32 - 2141 - 01	4.56 ± 0.33	4.18±0.32	1.09 ± 0.11
35-2017-02	51.2±4.3	39.6±3.7	1.29 ± 0.16
32-2113-02	49.7±3.2	43.7±3. 3	1.14±0.11
32-2115-02	$64.4{\pm}4.6$	55.5±3.6	1.16 ± 0.08
32-2850-03	27.3±3.0	19.3±1.4	1.41 ± 0.13
35 - 2716 - 05	22.3±2.7	17.2±1.÷	1.30±0.19
37-2717-05	25.6±3.3	19.0±1.3	1.35 ± 0.20
3 2-2 856-05	14.1±1.0	12.7±0.0	1.11 ± 0.11
32-2860-05	30.4 ± 6.1	17.9±1.3	1.70±0.36
32-2189-01	66.0±13.2	43.7±2.7	1.51±0.32
30-4720-0 9	7.68 ± 0.41	7.12±2.58	1.08 ± 0.40
33-4721- 09	5.57 \pm 0.22	3,95±1,26	1.41 ± 0.45
34-4722-09	5.17 \pm 0.23	3.88±1.04	1.33±0.36
35-4723-09	7.18 ± 0.48	6.70 ± 1.72	1.07 ± 0.23
37-4724-09	7.22±0.46	5.17 ± 1.41	1.40±0.39
39-4725-09	10.2 ± 0.7	10.3±3.1	0.99±0.31
40-4726-09	8.96±0.66	10.0±2.9	0.90±0.27
41-4727-09	3.61 ± 0.27	3.31±0.08	1.09 ± 0.33
42-4728-09	2.73±0.20	3.48±1.20	0.78±0.28
31-3720-10	40.6 ± 2.6	27.7±5.0	1.47 ± 0.33
31-3333-14	82.7±3.9	70.3±14.4	1.18±0.25
 μ =	0.1496	$t \cdot s_{\bar{u}} = 0.1315$	
s ₁ =	0.2090	$t \cdot s_{\bar{\mu}} = 0.1315$ $e^{\bar{\mu}} = 1.16 \pm 0.07$	
1	0.04369	$s_2^2 = 0.08393$	
-		4	
^s _µ =	0.06034	$s_1^2/s_2^2 = 1.57$	
		$\mathbf{F} = 2.7$	

Table 131. Comparison of alpha count and gamma-spectrometric analyses for 241 Am in separate batches of soil (MCL vs LLL).

Sample No.		Alpha count, dpm/g	Gamma spectrometry (LLL), dpm/g	MCL/LLL or LFE/LLI
08-0504-02	MCL	1.53±0.09	0.73 ± 0.42	2.0±1.1
09-0449-09	MCL	7.8±0.4	8.3±1.0	0.94 ± 0.13
09-0473-02	MCL	3.3±0.2	3.2±1.6	1.0±0.5
09-0506-02	MCL	1.86±0.09	1.62±0.51	1.15±0.36
09-0494-02	LFE	6.0±0.1	10.0±1.0	0.60±0.06
				Factor of improvement
				in sensitivity or value
				of limit
08-0476-01	MCL	0.50 ± 0.04	< 1.5	3.0
08-0535-02	MCL	0.92±0.05	< 1.2	1.3
09-0597-09	MCL	0.52±0.03	< 0.63	1.2
09-0326-33	MCL	<0.019	< 0.61	32
09-0483-36	MCL	< 0.031	1.85±0.20	Inconsistent
08-0358-38	LFE	< 2.9	< 0,063	No improvement
08-0304-39	LFE	< 0.48	< 1.9	4.0
08-0353-39	LFE	0.19±0.04	< 2.5	13
09-0271-39	LFE	0.092±0.023	< 0.35	3.8
09-0312-43	LFE	0.32 ± 0.04	< 2.4	7.5
09-0338-43	LFE	7.0±3.7	< 0.88	Inconsistent
	(23	^{9,240} Pu = 0.47	dpm/g)	
09-0462-60	LFE	0.16 ± 0.02	< 1.0	6.2

Table 132. Comparison of alpha count and gamma-spectrometric analyses for 241 Am in marine samples (MCL vs LLL and LFE vs LLL).

the body of 241 Am data for soil samples reported in this survey may be systematically low by about 20%. However, since this check for bias involved a relatively small number of samples and the magnitude of the bias is somewhat uncertain, the body of 241 Am data for soils is just as reported, based upon gamma spectrometry. A possible bias of 20% in the 241 Am data for soils has negligible effect on estimated external dose due to gamma emitters in soil since 241 Am contributed a very small fraction of the total dose.

Another comparison of $^{241}\mathrm{Am}$ deter-

minations by alpha counting and gamma counting was made on a group of 17 marine samples. The data are listed in Table 132. Ratios which compare methods were obtained for the first five entries in the table. The ratio for the most precisely measured sample, 09-0494-02, is 0.60 ± 0.06 . The other four show satisfactory agreement between methods; the ratios have large enough uncertainties that none varies significantly from unity. Data for the remaining samples in Table 132 demonstrate improvement in sensitivity with factors in the range, 1. 2-32. 11

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Sample No.	Alpha count (MCL), dpm/g	Gamma spectrometry (LLL), dpm_g	Factor of improvement in sensitivity	
Vegetation				
samples				
10-2256-03	0,10	< 0.26	2.6	
10-3293-09	0.010=0.002	0.16	16	
10-3297-09	0.020±0.003	0.32 ± 0.14	MCL/LLL = 0.062±0.02	
10-2438-15	0.012±0.002	< 0.074	6.2	
10-2447-16	0.025±0.003	~ 0.19	7.6	
10-3275-24	0.325±0.016	0.25 ± 0.11	MCL/LLL = 1.33±0.58	
10-0205-33	< 0.006	< 0.19	32	
10-0081-33	0.006	0.17	28	
Sample No.	Alpha count (LFE), dpm/g	Gamma spectrometry (LLL), dpm/g	Factor of improvement in sensitivity	
Animal samples				
11-9094-10	0.009 ± 0.006	< 0.97	110	
11-9130-10	< 0.025	<4.3	170	
11-9099-24	< 0.017	< 1.3	80	
11-9136-24	< 0.016	< 0.21	13	
			0.1	
11-9127-10	0.11±0.09	< 1.0	9.1	

Table 133. Comparison of alpha count and gamma-spectrometric analyses for ²⁴¹Am vegetation and animal samples (MCL vs LLL and LFE vs LLL).

Other comparable ²⁴¹Am data for vegetation and animal samples are given in Table 133. The data emphasize improvements in sensitivity available from wet-chemical analysis and alpha counting. For vegetation samples, increases in sensitivity were factors in the range, 2.6-32, while for animal samples, increases were factors in the range, 9-580.

Background Samples

A series of coral soil samples for background determination were put

through the entire system, from initial processing to wet-chemistry analysis, at varying intervals during the processing period. These samples were given identical treatment to neighboring samples as they were prepared and analyzed. Results from this series, given in Table 134, were expected to bear on questions of crosscontamination in the analytical sequence. Since the main sample load was processed in approximate sequence from low-level contamination to higher levels, crosscontamination if detectable at all, would

				Concentration, pCi/g					
Sample No.	Processed with samples from island	Wet chemistry performed by	⁶⁰ Co	90 _{Sr}	¹³⁷ Cs	239, 240 _{Pu}			
01-1305+70	LUCY	LFE	< 0.014		< 0.010	0.046 ± 0.002			
01-1306-70	JANET	MCL	< 0.015	< 0.32	< 0.010	0,010±0.002			
01-1307-70	FRED	MCL	< 0.034	< 0, 34	< 0.016	0.013±0.004			
01-1308-70		LFE		0.018±0.002		0.012±0.001			
01-1309-70		LFE		0.016±0.006		0.013±0.001			
01-1310-70	IRENE	LFE	< 0.018	0.042±0.009	< 0.017	0.012±0.002			
01-1314-70	YVONNE	LFE	< 0.018	0.024 ± 0.006	< 0.014	0.015 ± 0.001			
01-1315-70	YVONNE	LFE	< 0.008	0.023 ± 0.003	< 0.007	0.118+0.005			
	(YVONNE 122 pro	file)							
01-1316-70	YVONNE	MCL	< 0.003	< 0.43	0.012±0.003	0.013 ± 0.004			
01-1317-70	YVONNE	MCL	< 0.010	< 0.27	< 0.007	0.011 ± 0.002			
	(YVONNE 145 pro	file)							
01-1318-70		LFE							
	Mean value		< 0.015	0.025	< 0.012	0.012 (exclude 1305,1315			
	Range		<(0.003-0.034)	0.016-0.042	<(0.007-0.017)	0.010-0.015			

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become more obvious in later samples of the series. The material used for background measurements came from a single batch of coral sand taken from Midway Island and was supplied to us by Major W. A. Myers of MCL. It was known not to be significantly contaminated.

The data in Table 134 show very little evidence for any cross-contamination with batches of highly contaminated Enewetak coral. The gamma emitters give no evidence of contamination. No 60 Co was detected in any of the samples; an average upper limit value was <0.015 pCi/g. For 137 Cs. one sample vielded detectable cesium at 0.012 \pm 0.003 pCi/g, while the remainder gave upper limits averaging < 0.012 pCi/g. For ⁹⁰Sr. MCL reported upper limits ranging from < 0.27 to < 0.43 pCi/g. while LFE reported five measurements with a mean value of 0.025 pCi/g and a range of 0.016-0.042 pCi/g. For ^{239,240}Pu, two of the results from LFE showed very slightly elevated levels; the values were 0.046 \pm 0.002 and $0.118 \pm 0.005 \text{ pCi/g}$. The remaining analyses, from both LFE and MCL, produce a tightly clustered set with a mean of 0.012 pCi/g and a range of 0.010-0.015 pCi/g. Sample 1315 was processed with samples from YVONNE which contained high levels of plutonium, ranging up to 500 pCi/g. These plutonium results for 1305 and 1315 are the only indication of cross-contamination given by the data for background samples. The constant levels of ⁹⁰Sr and ^{239,240}Pu measured for most of the samples can be ascribed to fallout on Midway Island during the years since atmospheric testing began. Since this batch of soil was collected on a beach and has been subjected to the leaching action of seawater, one should not try to read any significance into the absolute amounts of strontium and plutonium observed.

We are grateful for guidance from Dr. H. B. Levy in the statistical treatment of data.

ENEWETAK SURVEY RADIOLOGICAL CONTROLS

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Introduction

In the planning of the precleanup survey effort, it was recognized that radiation fields and radioactive contamination existed on various islands in the Atoll: however, at the time the survey began, the radiological conditions of all of the islands had not been evaluated. From previous surveys, it was known that the island of YVONNE had significant plutonium contamination problems, while SALLY, IRENE, and JANET were known to have activated/contaminated scrap metal or buried plutonium-contaminated debris. The radiological conditions on KATE, LUCY, MARY, NANCY, OLIVE, PEARL, VERA, and WILMA were unknown but could be inferred. PEARL had a surface ground zero and the other islands were near contaminated locations.

General Radiological Controls

For the safety of the survey personnel, general radiological safety requirements were formally established by Roger Ray, Survey Manager. These requirements provided for personnel dosimetry and radiological support for all surveyrelated personnel.

All survey personnel leaving FRED (Enewetak Island) for other areas within Enewetak Atoll and personnel utilizing radioactive materials or handling potentially contaminated soil samples on FRED were required to wear a personnel dosimeter (TLD). These dosimeters were issued upon arrival at the Atoll and were returned to the LLL Hazards Control representative upon departure from the Atoll. The personnel dosimeter utilized by the survey was the LLL TLD unit, which consisted of three TLD chips [Harshaw TLD 100 (LiF), TLD 200 (CaF₂), and TLD 700 (LiF, depleted in Li,] mounted separately in a numbered plastic disk. An aluminum beta-shield was provided with an open window over the TLD 100 chip. Control TLDs were carried to the Atoll and returned, with the personnel dosimeters, to LLL for reading and interpretation.

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Because of the possibility that contaminated soil samples might be brought back to FRED, all sample processing, counting, and storage areas were monitored with portable survey instruments for beta-gamma and alpha emitters. Swipe tests were also made; the swipes were counted on portable swipe counters fielded for that purpose.

A monitor accompanied all survey parties landing on the islands designated by the Survey Manager as requiring radiological support. Portable survey instruments specifically designed to detect alpha, beta-gamma, and gamma-only emissions were used. The high humidity at Enewetak Atoll makes alpha detection a problem because condensation not only masks alpha emitters in soil, but also causes electrical problems with all portable survey instruments.

• PAC-1S. Early in the survey, and during previous surveys, the principal alpha-radiation detection instrument was the PAC-1S. Aircraft restrictions prohibited carrying of gas for the PAC-4G, a more sensitive and desirabk instrument, leaving the PAC-1S as the

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only alternative. This instrument uses an alpha spintillation detector with an active area of 59 cm² and an aluminized Mylar window with a thickness of 1.5 mg/cm^2 . The spintillation crystal is silver-activated zinc sulfide.

- LLL "Blue Alpha Meter." As the field effort progressed, an LLL modification of a well-known, air-chamber-type alpha survey instrument became available to the Enewetak survey teams. Although not ruggedized like the PAC-1S, the instrument was much more sensitive; it had an air chamber with an effective area of 100 cm^2 and an aluminized (on both sides) Mylar window with an effective thickness of 0.85 mg/cm^2 . Also, the probe guard was half as thick as the PAC-1S probe enabling near-contact measurements of surfaces to be made. The air chamber was subject to damage by sharp objects, more so than the PAC-1S; however, unless a large hole had been torn in the Mylar, it was still serviceable.
- Both the PAC-1S and the LLL "Blue Alpha Meter" were calibrated on ²³⁹Pu. Alpha sources attached to the survey instruments were used for field checking.

The following beta-gamma detectors Are used to obtain contact readings on maminated activated radioactive scrap: •E-400B. This instrument is a portable Geiger counter used for conducting beta-gamma radiation surveys. A tube sensitive to gamma and beta radiation is located in the external probe. Discrimination between the two types of radiation is made by means of a rotary shield on the probe. The probe has an energy cutoff at approximately 0.31 MeV and was calibrated with the 662-keV radiation from 137 Cs.

• Ludlum Model 3, with Model 44-9 "pancake" probe. This "thin-window" detector was used for low-energy gamma- and beta-radiation detection on scrap. The survey meter itself was used only as a relative indicator of contamination levels. The Model 44-9 "pancake" probe uses an effective window thickness of 1.5 to 2 mg/cm^2 (mica) and diameter of 1.75 in. The window thickness was increased to 7 mg/cm^2 by applying plastic tape to the probe face. This instrument was calibrated for gamma radiation using 60 Co and for beta emissions using 90 Sr, but it was intended to be for low-energy beta-gamma.

The following gamma detectors were used:

• NE-148 Scintillation Monitor 904-148. This is the Baird-Atomic portable survey instrument which was used in the soil survey and terrestrial radiation measurement program. The Model 904-148 scintillation monitor is a highly sensitive instrument capable of measuring extremely fine gradations of gamma-radiation levels in three ranges, up to 3 mR/hr. The detecting element is a smaller version $(1 \times 1 \text{ in.})$ of the NaI(T1) crystal scintillator used in the aerial radiation survey. The instrument was calibrated using ¹³⁷Cs for gamma fields. It proved to be a reliable device which was very rugged and well suited for use in the field.

• Fidler probe. The Fidler, a low-

energy x-ray detector, consists of a 5-in. diam \times 1 16 in. -thick NaI(T1) crystal optically coupled through a quartz light pipe to a selected RCA 8055 photomultiplier tube. The detector entrance window is 10-mil beryllium, and the entire assembly is encased in a 5/32-in. stainless steel can. A portable regulated high-voltage power supply and single-channel analyzer with integral count rate meter are used for the detection of plutonium x rays and the americium gamma ray. Field calibration was accomplished with 241 Am.

Results of General Radiological Controls

A total of 125 TLD personnel dosimeters were issued to 84 people between October 13, 1972 and February 17, 1973. Only one dosimeter indicated any positive exposure above the minimum detectable dose (10 mrem). This dosimeter was not turned in when the individual to whom it was assigned left the Atoll. When recovered by mail in the continental United States, it indicated a total exposure of 18 mrem (8 mrem above the minimum detectable dose). The 8-mrem difference is comparable to the exposure one would receive during a high-altitude passenger flight in a commercial aircraft flying from Enewetak to the continental United States and was probably accumulated during this individual's flight and subsequent mailing of the dosimeter.

The monitoring program conducted on FRED to prevent and control crosscontamination of soil samples was apparently successful. All swipes were negative, and no significant contamination was detected in the sample areas by portable survey instruments.

Special Radiological Controls

Special controls were established for the soil-collection and terrestrialradiation survey efforts on the northern half of YVONNE, where pieces of plutonium were known to be randomly distributed on or near the surface north of the Tower Bunker (HARDTACK Station 1310). In addition, the area immediately south of the CACTUS crater was known to have



Fig. 115. Locations of the soil-sampling RADEX area and radiological control operation, YVONNE (Runit).

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The L monitor 1 exposure dividuals YVONNE _{jumma} levels of approximately 1 mR hr, the highest in the Atoll.

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For the purpose of this survey, a radiation exclusion (RADEX) area was established from the Tower Bunker north to the CACTUS crater (Fig. 115). Complete radiation safety controls were in effect within this RADEX area. A decontamination pad for equipment was set up on the ocean side of this RADEX area immediately adjacent to the hot line. A personnel decontamination facility and hot line were established on the east side of the tower; they controlled the sole access route for personnel going into the survey area.

Air Monitoring

Air samples were taken continously in the area immediately downwind from the soil-disturbing activities in order to detect any resuspension of radioactivity. The airborne contaminants were collected on filters which were changed at the end of each work day. Additionally, two lapeltype, low-volume air samplers were used. One was placed on the backhoe operator, and the other on a profile monitor for more realistic evaluation of breathing zone concentrations. Samples were counted at Enewetak at the end of the day for gross activity and forwarded to LLL for further analysis. Daily gross results were available to the sampling team leaders.

Personnel Monitoring

The LLL TLD system was used to monitor personnel on YVONNE for external ^{exp}osure to beta-gamma radiation. All individuals engaged in survey activites on YVONNE were issued the TLD packet which was worn on the upper body in the same manner as a film badge.

Swipe tests using disks of filter paper were used to evaluate and detect removable surface contamination on equipment and area surfaces.

Contamination Controls

Every effort was made to prevent radioactive contamination of personnel. All personnel entering the YVONNE RADEN area or working at the hot line were suited out in full anticontamination (anti-C) clothing, consisting of one pair of coveralls, totes, cotton gloves, and cloth hood. All seams were taped. Those personnel collecting soil samples, displacing soil, or downwind from soil displacing activities, wore an Acme full-face mask, or equivalent, equipped with an Acme OAPR 282 high-efficiency canister.

All personnel left the area through the hot-line station and were monitored for alpha and beta-gamma contamination before, during, and after removing anti-C gear. Nose swipes were collected from selected individuals after all anti-C gear was removed. Decontamination capability was available at the hot line. Smoking and eating were not permitted in the RADEX area.

All contaminated anti-C gear was removed and suitably packaged at the hot line. Contaminated waste produced by the survey effort was collected, bagged, and buried on YVONNE in a marked area just east of the Tower Bunker.

All equipment used in the RADEX area was monitored with portable survey instruments and swipe tested. It was decontaminated when necessary.

Soil samples taken from the northern

part of YVONNE could have contained considerable amounts of ²³⁹Pu, therefore care was taken to prevent not only crosscontamination between individual samples but also contamination of personnel, equipment, and storage areas used in the recovery and processing of these samples.

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Profile samples, collected from pits dug by backhoe (4- to 6-ft-deep profiles), were carefully removed from the pits with special side-wall sampling tools in approximately 10-cm-depth increments. Each individual sample was bagged and numbered at the time of collection. The sample was then bagged once more in a heavy plastic bag to guard against breakage of bags and leakage of material. All samples from a single profile location were again bagged in a single large plastic bag to keep all contents together for transfer to Enewetak.

To reduce airborne contamination during this windy and relatively dry season, all soil collection areas were wet down with salt water prior to any soildisturbing activities.

All samples passing over the hot line on YVONNE were monitored for external alpha contamination. Samples found contaminated were bagged again and marked as having surface contamination on the inner bags. Soil samples from the northern portion of YVONNE were segregated from samples taken from other locations and kept in a special storage area where the floors were covered with a plastic sheet to provide for easy removal of contamination in case of leakage.

Decontamination

A personnel decontamination facility, which included a freshwater shower and

washstand, was provided at the hot line. Personnel were monitored crossing the hot line and were decontaminated as necessary.

A final equipment decontamination pad was set up on the concrete landing ramp. Saltwater washdown capability was provided. All digging equipment used in the RADEX area was decontaminated at the landing ramp. Effluent flowed back into the ground and into the lagoon.

Although every effort was made to prevent personnel contamination, a comprehensive bioassay program was followed to ve ascertain any internal contamination, to document either its absence or presence for the record, and to evaluate the effectiveness of control measures. Samples were taken in the following ways:

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- Nose swipes were taken from selected personnel working in the area of airborne contamination immediately after the end of the work period when they removed their anti-C apparel.
- A 24-hr collection sample of urine was submitted by each YVONNE survey participant at the completion of the survey effort. These samples were forwarded to the United States for analysis.
- Selected individuals known to be involved in the YVONNE sampling effort had base-line whole-body (lung) counts prior to their arrival on Enewetak. These individuals were whole-body counted again upon the completion of the survey to evaluate any internal deposition acquired during the survey effort.

Results

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Only one individual required personal decontamination during the YVONNE soilsampling effort. In this case, simple washing with soap and water removed the alpha contamination (less than 100 dpm). Anticontamination clothing did become contaminated with low-level alpha emitters (a few hundred dpm).

Equipment used to dig the sampling trenches also became contaminated. The low-level alpha contamination was removed by high-pressure saltwater spray before the equipment left the island. Vehicles used for transportation within the RADEX area did not need decontamination.

Air samplers, running continuously during the sampling operation showed positive indications of plutonium activity. The maximum observed values were 1.6 pCi/m^3 on one lapel monitor and $9.5 \times 10^{-2} \text{ pCi/m}^3$ on the downwind lowvolume air sampler, approximately 10 ft from the sampling trench. The background high-volume air sampler, operated behind the hot line, indicated a maximum of $7.4 \times 10^{-3} \text{ pCi/m}^3$.

TLD personnel dosimeters did not indicate any significant beta-gamma exposures due to this final soil-collection effort. The results of analysis of urine samples and whole-body counts, taken after the field effort, were negative.

It is concluded that the radiological safety controls used for the Enewetak precleanup survey field effort were adequate and effective.

Introduction

The potential dosages to the returning population on Enewetak Atoll are developed and discussed in this chapter. The data base for the analysis is derived from all the information collected during the survey portion of the program. Four major pathways are considered: (1) external gamma exposure, (2) inhalation, (3) marine food chain, and (4) terrestrial food chain. Models used for assessment of each pathway are described in the section of the chapter dealing with that specific pathway. Living patterns, i.e., dietary habits, location of villages, and daily habits which influence time distributions at various geographical locations in the Atoll, are of primary importance in determining the relative significance of each exposure pathway to the total dose. Six living patterns have been constructed and evaluated to determine the sensitivity of these factors and the possible range of dosages. Each section describes the relative impact of the complete living patterns, and specific components within living patterns, upon the dose contribution via the four pathways. The chapter is organized as follows:

- (1) Dietary and living patterns
- (2) External dose pathway
- (3) Inhalation pathway
- (4) Marine food chain
- (5) Terrestrial food chain
- (6) Summary of dose assessment and evaluation



Fig. 116. Islands (those circled) requested as village locations by the Enewetak people.

Dietary and Living Patterns

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Living Patterns

The Enewetak people have expressed a desire to make Parry-Enewetak and Engebi the residence islands for the two Enewetakese socio-political groups (see chapter on Enewetak). Figure 116 shows the Atoll and the islands chosen for village locations. The separation of the two socio-political groups was the life-style prior to evacuation, with the Engebi people and their chief headquartered on Engebi and the Enewetak people and their chief headquartered in the southern part of the Atoll. Our dose estimates are therefore in ne:

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Table 135. Living patterns describing the geographical locations for activities involved in daily living.

Pattern IPattern IIResidenceEnewetak, ParryEnewetak, ParryAgricultureALVIN through KEITHKATE through WILMA + LEROYFishingEntire AtollEntire Atoll

Pattern III

Residence Agriculture Fishing

JANET JANET Entire Atoll

Pattern IV

BELLE BELLE Entire Atoll

Pattern V

ResidenceJANETAgricultureKATE through WILMA + LEROYFishingEntire Atoll

Pattern VI

JANET ALICE through IRENE Entire Atoll

based upon these islands as the village areas, with visits to other islands for planting and collection of food. Generally, people living on Engebi own land on the neighboring islands, i.e., in the northern half of the Atoll, while those living on Parry and Enewetak own land on the islands in the southern half of the Atoll. These nearby islands would be used for additional agriculture and food collection by the two respective groups.

As a result of the above-mentioned de-

sires of the Enewetak people, the six different living patterns shown in Table 135 have been synthesized for estimating the potential dose to the returning population. For estimating the dose via the terrestrial food chain, islands are grouped according to a common range of external exposures and radionuclide concentrations in the soil; these groups are shown in Fig. 117. JANET and YVONNE are listed individually (Groups II and IV) and LEROY is included in the Group III islands, KATE through WILMA.





The living patterns are designed to include the most probable circumstances which will occur when the inhabitants return (patterns I, II, III, V, and VI), as well as a more extreme exposure situation which could occur (pattern IV). The effects on total dose of these various living patterns will be discussed in the sections dealing with the external exposure, the exposure via food chains and in the section describing the total doses via all pathways.

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The distribution of time between village, interior, beach, lagoon, and other islands is shown in Tables 136 and 137. The breakdown is based upon reports by Jack Tobin from his years of experience in the Marshall Islands and from Ken

	Village area	Beaches	Interior	Lagoon	Other islands
len	50	5	15	10	20
_{Vomen}	60	10	10	0	20
hildren	55	10	15	5	15
lfants	85	5	0	0	10

Table 136. Estimated time distribution (in percent) for men, women, children, and infants, with emphasis on residence island. Pattern A.

Table 137.Estimated time distribution (in percent) for men, women, children,
and infants with emphasis on additional time spent on nonresidence
islands. Pattern B.

	Village area	Beaches	Interior	Lagoon	Other islands
Men	40	5	20	10	25
Women	50	5	15	5	25
Children	50	5	15	10	20
Infants	70	5	5	0	20

Table 138. Population distribution of Enewetak.

Age groups	Per	centage of total population
Infants (0-5 yr)	Male	12
	Female	10
Children (6-18 yr)	Male	21
	Female	21
Adults (19-50 yr)	Male	18
	Female	14
Adults (over 50)	Male	2
	Female	2

	Diet,	g/day
Food item	At time of return	10 yr after return
Fish	600	600
Domestic meat	60	100
Pandanus fruit	0	200
Breadfruit	0	150
Wild birds	100	20
Bird eggs	20	10
Arrowroot	0	40
Coconut	100	100
Coconut milk	100	300
Coconut crabs	25	25
Clams	25	25
Garden vegetables	0	0
Imports	200-1000	200-1000
	1030 plus impor	rts 1570 plus imports

Table 139. Postulated diet for the returning adult Enewetak population for time of return and for 10 yr after initial return.

Marsh who observed the Enewetak people on Ujilang and interviewed them as to their probable habits upon return to Enewetak Atoll.

Table 137 differs from Table 136 in that it increases the time spent in locations other than the residence island; this may be the situation during the first years of return while the inhabitants are cultivating and reestablishing the agricultural islands.

The population distribution of the Enewetak people, as determined by a census conducted by Dr. Jack Tobin in the fall of 1973, is shown in Table 138.

Diet

The composition of the diet shown in Table 139, both at the time of initial reinhabitation of the Atoll and 10 yr after return, was compiled from reports and interviews of Jack Tobin of the Trust Territories, Ken Marsh of LLL, Vic Nelson of the University of Washington, and Dr. Mary Murai, a nutritionist at the University of California, Berkeley, who spent a number of years living on Majuro and visited several of the atolls in the Marshall Islands. The reports by Tobin, Marsh, and Nelson are included in this report. Dr. Murai contributed through private discussion and through her publication "Nutrition Study in Micronesia," Atoll Research Bulletin No. 27 (1954), issued by the Pacific Science Board National Academy of Sciences — National Research Council.

The diets listed are intended to represent the average diet if the Atoll resources and Atoll agriculture are pursued in a manner similar to that prior to removal of the people from the Atoll for the testing program. The diet from 0 to 10 yr reflects the current lack of significant

quant COCOL plante 7- to contr TŁ listed flour, salme proba of the accus to est most would tions showr total ' a bri€ listed

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<u>Dc</u> likely than 1 suffic livest it wil chick

Pa than at pro starte quantities of pandanus, breadfruit and coconuts, all of which will have to be planted upon return and all of which have 7- to 10-yr development periods before contributing edible fruit to the diet.

The general opinion of all the sources listed above is that imports of rice, flour, tea, canned fish (tuna, mackerel, salmon, etc.) and canned meats will probably be major components in the diet of the Enewetakese because they are now accustomed to such living. They prefer to establish a cash economy and purchase most of their needed food items. It would therefore appear that dose calculations based upon the dietary intake per se shown in Table 139 may overestimate the total dose via the food chains; however, a brief comment on each of the items listed for both diets is in order.

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<u>Fish</u> — The 600-g/day average intake, both initially and 10 yr after return, is probably a high estimate in view of the dietary and living habits of the people today. Bones and viscera of fish, dark muscle of tuna, and invertebrates such as sea cucumbers are not eaten by the natives.

<u>Domestic Meat</u> — This commodity is likely to be in shorter supply upon return than later because the people do not have sufficient room to raise many head of livestock where they presently live and it will take time to increase the pig and chicken population.

<u>Pandanus Fruit</u> — There are fewer than 10 pandanus plants on the entire Atoll at present. New plants will have to be started and will bear fruit about 8-10 yr after planting.

<u>Breadiruit</u> — No breadfruit trees were encountered in the survey. Again a program of replanting will be necessary, with a subsequent period of approximately 8-10 yr before edible fruit is available.

<u>Wild Birds and Bird Eggs</u> — Wild birds are plentiful now, but the population will probably be depleted when the 400 people return.

<u>Arrowroot</u> — Very small patches of arrowroot were observed on the southern islands, with one larger patch observed on DAVID. Arrowroot will therefore have to be planted. However, the people have indicated that they much prefer imported white wheat flour and would only use arrowroot as a second choice.

<u>Coconut and Coconut Milk</u> — Islands in the southern half of the Atoll, especially Parry, FRED, and GLENN, have a large enough coconut crop to supply the people who first return. More coconut groves will be established on the other islands; however it takes approximately 8-10 yr for coconut trees to become productive and useful.

<u>Coconut Crabs</u> — The crab population now parallels that of the coconut trees in distribution. As more trees are planted, the coconut crab can be reintroduced to the other islands. The only question may be how rapidly the returning people harvest the available crabs and how well they practice conservation of this species. The people consider the crab a delicacy and could easily decimate the population. <u>Clams</u> — Giant clams (<u>Tridacna</u>) are eaten raw and almost exclusively by the men during fishing trips. If the clams are not harvested under controlled conditions, they could be depleted to a stage where there would be few available in 10 yr.

Garden Vegetables - Very few garden vegetables will probably be available if current agricultural practices are continued. Terrestrial plants and garden crops, if planted, are allowed to grow in natural surroundings with very little organized gardening, i.e., no fencing or protected areas. The people prefer not to have high-maintenance agricultural situations. At the same time, their livestock, mostly pigs and chickens, are allowed complete freedom to roam as they please. The combination of low-intensity open agriculture and the uncontrolled meandering of the livestock tends to make fresh garden vegetables an unlikely component of the diet.

<u>Imports</u> — A large part of the diet is expected to consist of imported products. It is possible that imports will supply nearly the whole diet, with local marine and terrestrial products serving only in a limited way and on special festive occasions. If this should be the case, then the doses incurred by the returning population via the marine and terrestrial food chains will be far below those listed in this report.

External Dose Determination

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As described in the earlier sections on the EG&G aerial survey and photography, the terrestrial soil and radiation survey, and external dose estimates, gamma-ray exposure rates have been measured in this program by aerial survey, hand-held instruments, and thermoluminescent dosimeters. The three techniques yield the same results within \pm 10%; in the section on external dose estimates it was stated that the aerial-survey data would be used for those estimates. Average dose rates as obtained from the aerial survey are summarized in Table 140.

Even though wide variations in gammaray exposure rates were measured throughout the northern islands, it was necessary, for the purpose of the dose calculation, to derive the most reasonable values of the current mean exposure rates for each specific geographical area under consideration. These values are shown in Table 141 for the living patterns of Table 135. The mean exposure rates for specific areas of JANET were obtained by examination of the 137 Cs and 60 Co isoexposure-rate contour maps provided by the aerial survey. The village area was assumed to lie along the lagoon side of the island. The mean values given for the northern islands were obtained by weighting the mean exposure rates for each individual island with the area of each island. Since the minor contamination of the southern islands is relatively uniform, the mean $^{137}\mathrm{Cs}$ and $^{60}\mathrm{Co}\,\mathrm{ex}^-$

	Averag	e dose rate, $\mu R/hr$	at 1 m ^a	
Island	¹³⁷ Cs	⁶⁰ Co	Total γ (0-3 MeV)	Range ^b
ALICE	42	36	81	4-170
BELLE	61	50	115	5-200
CLARA	20	19	42	5-100
DAISY	6.8	14.4	21.3	5-140
EDNA	2.8	2.4	6	5-8
IRENE	14	63	80	3-560
JANET	25	13	40	2-150
KATE	11	7	1.9	3-22
LUCY	6	7	14	1-20
PERCY	2	2	5	2-11
MARY	5.5	4	10	2-12
NANCY	6	5	12	1 - 50
OLIVE	6.5	4.5	11	1 - 1 5
PEARL	12	45	70	1-400
RUBY	2	12	14	1-42
SALLY	3.5	3	7	3-110
TILDA	4	2	6	2-11
URSULA	3	1.8	5	1-7
VERA	2.8	2	5	1-6
WILMA	1	1	2	1 - 3
YVONNE	5.6	22.4	33	1-750
SAM	<0.3 (0.20)	<0.6 (0.11)	<0.9	0 - 1
ТОМ	<0.3 (0.18)	<0.6 (0.13)	<0.9	0 - 1
URIAH	<0.3 (0.06)	<0.6 (0.43)	<0.9	0 - 1
VAN	<0.3 (0.08)	<0.6 (0.25)	<0.9	0-1
ALVIN	N.D.(0.06)	<0.6 (0.25)	<0.9	0-1
BRUCE	0.4 (0.22)	0.8 (0.34)	1.2	0-1
CLYDE	<0.3 (0.04)	<0.6 (0.11)	<0.9	0-1
DAVID	N.D.(0.21)	N.D.(0.10)	<0.9	0-5
REX	<0.3 (0.28)	<0.6 (0.25)	<0.9	0-1
ELMER	N.D.(0.19)	N.D.(0.12)	<0.9	0-2
WALT	<0.3 (0.08)	<0.6 (0.10)	<0.9	0-1
FRED	N.D.(0.14)	N.D.(0.12)	<0.9	0-1
GLENN	0.4 (0.33)	<0.6 (0.20)	<0.9	0-1
HENRY	<0.3 (0.14)	<0.6 (0.20)	<0.9	0-1

Table 140. Summary of average dose rates for islands in Enewetak Atoll.

Table 140 (continued).

	Avera	age dose rate, $\mu R/F$	ir at 1 m ^a		Major g
Island	¹³⁷ Cs	⁶⁰ Co	Total γ (0-3 MeV)	Range ^b	JANET
IRWIN	<0.3 (0.08)	<0.6 (0.46)	<0.9	0-1	
JAMES	<0.3 (0.05)	2.8	3.0	0-5	
KEITH	<0.3 (0.15)	<0.6 (0.49)	<0.9	0-2	RELLE
LEROY	2.8	4.8	7.6	3-8	2 2

^aAverage dose rates given are derived from aerial-survey data. On islands where activity levels are at the lower limit of sensitivity of the aerial-survey equipment, dose rates derived from the soil sample data are given in parentheses.

^bAs measured with the Baird-Atomic instrument.

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		Exposure rate, $\mu R/hr$		
Major geographical area	Source	Village	Interior	Beach
ANET	¹³⁷ Cs	9.0	33	1.0
HUD -	⁶⁰ Co	5.0	14	0.5
	Cosmic and natural	3.5	3.5	3.5
3ELLE	¹³⁷ Cs	61	61	1.0
30000	⁶⁰ Co	50	50	0.5
	Cosmic and natural	3.5	3.5	3.5
RED, ELMER, or DAVID	¹³⁷ Cs	0.2	0.2	0.2
	⁶⁰ Со	0.1	0.1	0.1
	Cosmic and natural	3.5	3.5	3.5
Lagoon	Cosmic and natural	3.5	3.5	3.5
		Area-we	eighted mean rates, µR/l	
Northern islands	¹³⁷ Cs		14	
ALICE-WILMA, but	⁶⁰ Co		21	
excluding JANET)	Cosmic and natural	3.5		
Northern islands	¹³⁷ Cs		15	
	⁶⁰ Co			
ALICE-WILMA, but	⁶⁰ Co Cosmic and natural		16 3.5	
ALICE-WILMA, but excluding BELLE)	⁶⁰ Co Cosmic and natural		16 3.5	
ALICE-WILMA, but excluding BELLE) Northern islands	${}^{60}Co$ Cosmic and natural ${}^{137}Cs$		16 3.5 34	
ALICE-WILMA, but excluding BELLE) Northern islands	${}^{60}Co}_{Co}$ Cosmic and natural ${}^{137}Cs}_{Co}$		16 3.5 34 47	
ALICE-WILMA, but excluding BELLE) Northern islands ALICE-IRENE)	${}^{60}Co$ Cosmic and natural ${}^{137}Cs$ ${}^{60}Co$ Cosmic and natural		16 3.5 34	
ALICE-WILMA, but excluding BELLE) Northern islands ALICE-IRENE)	${}^{60}Co}$ Cosmic and natural ${}^{137}Cs}_{Co}$ Cosmic and natural ${}^{137}Cs}$		16 3.5 34 47	
ALICE-WILMA, but excluding BELLE) Northern islands ALICE-IRENE) Northern islands	${}^{60}Co$ Cosmic and natural ${}^{137}Cs$ ${}^{60}Co$ Cosmic and natural		16 3.5 34 47 3.5	
ALICE-WILMA, but excluding BELLE) Northern islands ALICE-IRENE) Northern islands	${}^{60}Co}$ Cosmic and natural ${}^{137}Cs}_{Co}$ Cosmic and natural ${}^{137}Cs}$		16 3.5 34 47 3.5 5.9	
ALICE-WILMA, but excluding BELLE) Northern islands ALICE-IRENE) Northern islands KATE-WILMA)	${}^{60}Co}$ Cosmic and natural ${}^{137}Cs}_{Co}$ Cosmic and natural ${}^{137}Cs}_{60}Co}$ Cosmic and natural		16 3.5 34 47 3.5 5.9 11 3.5	
ALICE-WILMA, but excluding BELLE) Northern islands	${}^{60}Co}$ Cosmic and natural ${}^{137}Cs}_{Co}$ Cosmic and natural ${}^{137}Cs}_{60}Co}$		16 3.5 34 47 3.5 5.9 11	

Table 141. Estimated mean exposure rates ($\mu R/hr$) used for dose calculations.

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posure rates were chosen by inspection of the individual aerial-survey contour maps and of the soil data. The cosmicray contribution was estimated to be $3.3 \ \mu$ R/hr at this latitude, and the naturally occurring radionuclides in the soil and sea water were expected to contribute an additional $0.2 \ \mu$ R/hr.

The relative gamma-ray exposure rate contributions from 60 Co and 137 Cs obtained from the aerial survey agrees well with values independently inferred from the soil activity-depth profile measurements. Although the soil measurements indicate trace amounts of other gamma emitters, such as 125 Sb, 155 Eu, and $^{241}\mathrm{Am}$, calculations of exposure rates based upon the observed soil activities indicate that these radionuclides contribute at most an additional 3 to 5% of the total exposure rate. The contribution due to these radionuclides was therefore neglected. Thus, the mean exposure rates shown in Table 141 are felt to be the most reasonable values available for computing integrated dose values. In fact, these mean values may be somewhat conservative, even though the aerialsurvey data agree well with the TLD data, because the latter may have slightly overestimated the exposure rates due to the minimal beta-ray shielding afforded by the TLD badges.

Integral 5-, 10-, 30-, and 70-yr gamma-ray doses for each age group were calculated for each case or living pattern described in Table 135. The results were then combined by "folding" in the present population distribution shown in Table 138. Corrections were made for radioactive decay but not for possible weathering and subsequent deeper penetration of the radionuclides in the soil. The results of these calculations are given in Table 142 and are labeled "unmodified." Additional calculations were made to ascertain the effect of reasonable attempts to reduce the exposure rates on the Atoll.

The first modification, labeled "village graveled" in Table 142, reflects the effect of covering the village areas with about 2 in. of coral gravel - a common practice throughout Micronesia.[®] This action can be expected to reduce the gamma exposure rates in the village area by approximately a factor of two. The second and third modifications are based upon the assumption that clearing the islands for agricultural use and housing will result in some mixing of the topsoil. It appears that it would be practical during this period to also plow many of the more contaminated islands to a depth of 1 ft. Assuming that plowing results in mixing rather than burying the topsoil, an average reduction in exposure rates of about a factor of three may be obtained. This reduction factor is based upon the present 3- to 5-cm relaxation lengths (the depth at which the activity is e^{-1} . or 37% of the surface activity) for activity depth distribution in the uppermost soil layers of the more contaminated areas. This value, however, is highly variable from site to site. In Table 142 modification (2) indicates the effect of plowing only JANET or BELLE, while modification (3) reflects the additional effect of plowing all the northern islands. Deeper plowing or turning over the soil rather than mixing

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^{*}J. A. Tobin, private communication, 1973.

Table 112. Estimated integral external free all gumm	G	amma (lose, ra	d
	r	ſime in	terval,	yr
Case Living pattern	5	10	30	70
I Village: Enewetak/Parry				
Visits to ALVIN-KEITH				
Time distribution: Table 137				
Unmodified	0.14	0.28	0.83	1.92
II Village: Enewetak/Parry				
Visits to ALICE-WILMA				
Time distribution: Table 137				
Unmodified	0.38	0.68	1.59	2.97
3. Northern islands plowed	(0.22)	(0.41)	(1.08)	(2.26)
III Village: JANET				
No visits to other islands				
Time distribution: Table 137 with "other				
islands" time spent in interior of JANET				
Unmodified	0.94	1.71	3.95	6.66
1. Village graveled	(0.82)	(1.49)	(3.48)	(5.96)
2. JANET plowed	(0.36)	(0.68)	(1.70)	(3.24)
IV Village: BELLE				
Visits to ALICE-WILMA				
Time distribution: Table 137				
Unmodified	2.72	4.78	10.06	15.50
1. Village graveled	(1.78)	(3.14)	(6.69)	(10.53)
2. Plus BELLE plowed	(0.83)	(1.47)	(3.26)	(5.47)
3. Plus northern islands plowed	(0.68)	(1.23)	(2.77)	(4.76)
V Village: JANET				
Visits to KATE-WILMA				
Time distribution: Table 137				
Unmodified	0.71	1.28	2.94	5.06
1. Village graveled	(0.59)	(1.07)	(2.48)	(4.36)
2. Plus JANET plowed	(0.36)	(0.66)	(1.59)	(3.02)
3. Plus KATE-WILMA plowed	(0.29)	(0.54)	(1.36)	(2,71)

Table 142. Estimated integral external free-air gamma doses.

Table 142 (continued).

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		lamma Time in	dose, r:	ad
Case Living pattern	5	10	<u>30</u>	
VI Village: JANET				70
Visits to ALICE-IRENE				
Time distribution: Table 137				
Unmodified	1.15	2.03	4.39	7, 13
1. Village graveled	(1.02)	(1.81)		(6.47)
2. Plus JANET plowed	(0.80)	(1.41)	(3.05)	(5.00)
3. Plus ALICE-IRENE plowed	(0.43)	(0.78)	(1.85)	(3, 39)
VIa Village: JANET				
Visits to ALICE-WILMA	•			
Time distribution: Table 136				
Unmodified	0.76	1.37	3.12	5,33
1. Village graveled	(0.62)	(1.12)		
2. Plus JANET plowed		(0.75)		
3. Plus northern islands plowed		(0.56)		
VIb Village: JANET				
Visits to ALVIN-KEITH				
Time distribution: Table 136				
Unmodified	0.60	1.10	2.60	4.60
1. Village graveled	(0.48)	(0.88)	(2.14)	(3.90)
2. Plus JANET plowed	(0.25)	(0.48)	(1.26)	(2,56)
Mean population dose				
(Average of Cases I, II, III, V, and VI)				
Unmodified	0.66	1.20	2.74	4.75
1. Village graveled	(0.59)	(1.07)	(2.46)	(4.33)
2. Plus JANET plowed	(0.41)	(0.74)	(1.75)	(3.25)
3. Plus all northern islands plowed	(0.29)	(0.54)	(1.36)	(2.70)
Sea level, U.S.A.				
(80 mrad/yr) Typical	0.40	0.80	2.40	5.60

 $_{\rm it\ would}$, of course, result in even greater exposure-rate reductions. For example, mixing to a depth of 2 ft would reduce the exposure rates by an additional factor of $_{\mathrm{two,}}$ while covering the sources with approximately 1 ft of uncontaminated soil would essentially reduce the exposure rates to negligible values similar to those observed on the southern islands. Removing the top 6 in. of soil, which often contains about two-thirds of the activity, would result in a threefold reduction in the exposure rates. The advantages of plowing or removing the topsoil should, however, be considered on a case-bycase basis because of the highly variable distributions of activity with depth. In fact. plowing IRENE could possibly increase the exposure rates in specific areas due to the elevated activity levels beneath the surface.

A review of Table 142 reveals that extensive modifications may not be required in order to reduce the dose levels to values comparable to typical U.S. values. Keeping in mind that Cases I, II, III, V, and VI represent approximations to the most likely living patterns, one observes that even for Cases V and VI, the unmodified 70-yr integral doses are comparable to U.S. values*, while Cases I and II lead to considerably lower doses. The mean integrated doses to the entire population, shown in Table 142, were derived by averaging those for Cases I, II, III, V, and VI. This implies that half of the returning population live on JANET, and the other half live on FRED, ELMER, or

DAVID, and that trips to the northern or southern islands are equally likely for both groups. The unmodified mean population doses are all quite comparable to U.S. values. At most, implementation of modifications 1 and 2 should be sufficient to assure mean population exposures well below the U.S. levels. Case IV represents a "worst credible" type of living pattern which, of course, leads to appreciably higher doses. However, even in this situation, the modifications can bring the levels down to the range of U.S. values.

Because of the low amount of natural radioactivity normally present in the coral atolls, the external dose levels calculated for Cases III, V, and VI are still appreciably higher than corresponding levels found elsewhere in the Marshall Islands (essentially Case I). The results for Cases I and II indicate that restricting the permanent villages to "clean" southern islands at least temporarily would result in lower exposures. Note that for Case IIb almost as much exposure is accumulated in the first 10 years as in the succeeding 20 years.

As illustrated in Table 143 for Case VIa, the differences in radiation exposure of the various population groups are minor, particularly for the longer time periods. Similar results were obtained for the other living patterns, indicating that the exact breakdown among age groups is not highly important. Table 144 illustrates the distribution of dose with respect to geographical area for Cases I, II, V, VIa, and VIb. The large fraction received while working in the interior or on other islands reflects, of course, the higher exposure rates present in these areas.

^{*}H. L. Beck, W. J. Lowder, B. G. Bennett, and W. J. Condon, <u>Further</u> <u>Studies of External Environmental</u> <u>Radiation</u>, USAEC, Rept. HASL-170 (1966).

		Total i	ntegrated dose	, rad
Group	5 yr	10 yr	30 yr	70 yr
nfants	0.64	1.15	2.66	4.63
nildren	0.79	1.43	3.24	5.52
en	0.82	1.47	3.32	5.61
omen	0.79	1.42	3.20	5.42

Table 143. Illustration of dose breakdown among population groups (Case VIa - unmodified).

Table 144. Percentage of unmodified exposure received from various locales.^a

Village	Beach	Interior	Lagoon	Other islands
47	2	27	1	23
36	1	33	2	28
22	2	8	4	64
58	2	33	1	5
50	5	17	8	20
	47 36 22 58	47 2 36 1 22 2 58 2	47 2 27 36 1 33 22 2 8 58 2 33	$\begin{array}{cccccccccccccccccccccccccccccccccccc$

^aFor 30-yr intervals averaged over population distribution. Percentages for other time periods are similar.

All of the doses discussed so far are due to free-air gamma plus cosmic-ray exposures. The effect of shielding by structures or the body itself on gonadal or bone doses has been ignored. To convert from free-air dose (rads) to gonadal dose (rem), a body-shielding factor of 0.8 may be used^{*}.

The free-air dose will be additionally enhanced by the presence of beta rays, originating primarily from 90 Sr and 90 Y in the soil. In radiation fields produced by global fallout, where the 90 Sr/ 137 Cs activity ratio in the soil is normally about 0.67, the free-air beta dose at 1 m above the ground is expected to be about four times that due to the 137 Cs gamma rays. At Enewetak, however, the 90 Sr/ 137 Cs activity ratios in the soil samples showed a wide range of values, with an average ratio of about three. Thus, the free-air beta dose rates may average about 600 μ rad/hr in the interior of JANET and about 200 μ rad/hr in the village area. The resulting beta-ray doses to the skin, eye lenses, and gonads will be about 50, 25, and 1%, respectively, of the free-air values[†]. Thus, appreciable ti T

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^{*}Report of the United Nations Scientific Committee on <u>The Effects of Atomic</u> <u>Radiation</u>, 27th Session, Vol. 1, Suppl. No. 25 (1973).

K. O' Brian, Health and Safety Laboratory, USAEC, New York, private communication (1973).
increases in skin and eye-lens doses due to the beta contribution could be expected. The gonadal dose, on the other hand, would be insignificant.

Very little information is available to verify these calculated beta-ray air doses, but indications are that they may be unrealistically high. This is based upon data obtained from two LiF TLD badges that were equipped with aluminum shields, one of which was situated within the interior of JANET. These shielded badges only showed an approximate 10% reduction in exposure rates from those measured by the unshielded badges at the same location, thus leading one to suspect that the beta air doses are considerably less than the calculated values.

Evaluation of the Inhalation Pathway

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The purpose of this analysis is to determine the significance of radioactivity in soils of the Atoll for their potential to inhalation exposures and to provide data necessary for making decisions regarding cleanup and future habitation. It has been well documented that radioactivity in soils can resuspend in the atmosphere and be available for inhalation. For most radionuclides this pathway contributes an insignificant dosage compared with dosages derived from other pathways. For example, analysis of dosages from ¹³⁷Cs in the U.S. environment has shown that, as an upper limit, resuspension is no more than 0.003% of the infinite-time dosage from all pathways to an individual^{*}. The results of

such an analysis would be similar for ⁶⁰Co and ⁹⁰Sr, but not for plutonium and other actinide elements. These latter radionuclides contribute negligibly to external exposures to the whole body and move inefficiently through food chains, resulting in an increased relative importance of the inhalation pathway.

Comparison of the maximum permissible concentrations in air $(MPC_a's)$, shows that ${}^{60}Co$, ${}^{90}Sr$, and ${}^{137}Cs$ contribute little to the dosages derived from inhalation as compared with plutonium (see Table 145). The MPC for 90 Sr in the soluble form comes closest to being significant. The ratio of the MPC_a for soluble 90 Sr to insoluble 239 Pu is 40, for example. These radionuclides would be of equal significance if ⁹⁰Sr levels in soil are 40 times those of 239 Pu. 90 Sr levels in the Atoll are generally only 2-3 times those of ²³⁹Pu. The ratio will decrease with time due to more rapid radioactive decay of ⁹⁰Sr and its preferential runoff to the aquatic environment. ⁹⁰Sr, ¹³⁷Cs, and ⁶⁰Co dosages via inhalation are expected to contribute less than 5% to the total inhalation exposure.

Average values for 241 Am on the islands ranged from 0.07 to 8.2 pCi/g in the top 15 cm of soil (see chapter on terrestrial soil and radiation survey). The 241 Am to $^{239, 240}$ Pu ratio, island by island, ranged from approximately 0.25 to 3.5. Highest 241 Am values were associated with the lowest $^{239, 240}$ Pu

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^{*}Y. Ng, W. Robison, and D. Wilson, "Modeling Radiation Exposures to Populations from Radioactivity Released to the Environment," in <u>IAEA/NEA/WHO</u> <u>Symposium on Environmental Behavior</u> of Radionuclides Released in the Nuclear <u>Industry, Aix-en-Provence, France,</u> May 14-18, 1973.

	N	MPC _a , μCi/cm ³		
Radionuclide	Soluble form	Insoluble form	Reference	·····
⁶⁰ Co	1×10^{-7} (total body)	$3 imes 10^{-9}$ (lung)	2	in the second se
⁹⁰ Sr	$4 imes 10^{-10}$ (bone)	$2 imes 10^{-9}$ (lung)	3	t (a)
¹³⁷ Cs	2×10^{-8} (total body)	5×10^{-9} (lung)	2	
238 _{Pu}	$7 imes 10^{-13}$ (bone)	1×10^{-11} (lung)	2	
239 _{Pu}	6×10^{-13} (bone)	1×10^{-11} (lung)	2	
²⁴¹ Am	2×10^{-12} (bone, kidney)	4×10^{-11} (lung)	2	

Table 145. MPC_a for radionuclides found in Enewetak soils.^a

^aThe concentration in air computed as leading to the maximum allowable dose rate in the organ of reference, noted in the table, when an individual is continuously exposed to the contaminent in air.

values on an island-by-island basis. On the basis of the MPC_a values and these soil data, it is concluded that ²⁴¹Am presently in soil would be a small contributor to the inhalation dosages. An additional amount of ²⁴¹Am activity will grow in from further decay of ²⁴¹Pu. Since the testing period, the ²⁴¹Pu has gone through almost two half-lives. Therefore, ²⁴¹Am values measured now are almost as high as will be obtained by further decay of the remaining ²⁴¹Pu.

Three approaches may be used to evaluate the inhalation pathway:

• Consider only the results of the air sampling during the survey as the basis for the evaluation.

Air sampling of the Atoll during the survey showed resuspension levels to be so low as to be masked in the "background" of atmospheric radioactivity present from fallout of stratospheric origin (see chapter on air-sampling program). The only exceptions were noted on northern YVONNE. These findings are encouraging since they show that the atmosphere of the Atoll, in general, is not influenced by the burden of radioactivity present in the soils. dei tri shi by uni

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Air-sampling data taken during the survey may be unrepresentative of air levels under actual living conditions since they were obtained on uninhabited islands. A high level of human activity can be expected to alter the levels of resuspended activity, particularly near the individuals who create the disturbances of the soil surface. Such circumstances could not easily be simulated during the survey.

• <u>Use measured soil concentrations and</u> published resuspension factors.

A considerable amount of information has been reported on resuspension factors, e.g., the ratio of air concentration expressed as activity per unit volume, to soil concentration expressed as activity per unit area (see Appendix A of this Chapter). Resuspension factors, so calculated, show a wide range of variation, temonstrating that soil-atmosphere disuributions depend upon complex relationships not accounted for very accurately by a bulk measurement of activity per unit area in soil.

• Use measured soil concentrations and estimates of maximum dust loading in the air.

The problems of using resuspension factors derived from data on radioactivity are circumvented by an approach which uses measurements of the amounts of dust in the nonurban atmosphere. The origin of the dust loading of the nonurban atmosphere is assumed to be from soil and to consist of particles in the respirable range of particle size (for our calculations we assume a mean particle of 0.4 µm aerodynamic diameter). An additional assumption is required, namely that radioactivity in the soil will behave similarly to the resuspendable soil surface. This set of assumptions allows one to predict the ambient levels of radioactivity in air, knowing the concentration of radioactivity in the soil and the amount of soil in air:

$$C_{2} = C_{2} \cdot L_{2}. \tag{1}$$

In Eq. (1), the air concentration (C_a) in pCi/m³, equals the concentration in soil, (C_s) in pCi/g, times the concentration of dust in the atmosphere (L_a) in g/m³.

Estimation of dosage, D_t , as a function of soil levels can be made by combining dose conversion factors, R_t , with Eq. (1), yielding:

$$D_{t} = C_{s} \cdot L_{a} \cdot R_{t}.$$
 (2)

Using Eq. (2) to predict dosages requires:

- Knowledge of the activity in soil (C_g) in pCi/g.
- Knowledge of the dust loading in the atmosphere (L_a) in g/m^3 .
- Dose conversion factors (R_t) in rems per pCi/m³, for cumulative dose to organs of the body through the inhalation pathway for t = 5, 10, 30, 50, and 70 yr of continuous exposure.

Plutonium in Enewetak Soils

The soil-sampling program provides information on the plutonium activity in the top 15 cm of soil on each island and. to a somewhat lesser extent, measurements of the vertical distribution of activity. Soil-profile data indicate that soil concentrations decrease with depth, although there are exceptions to this generalization. Two sets of soil-concentration data will be used in the inhalation-pathway evaluation (see Table 146). The activity in the top 2 cm of soil will be used to calculate dosages for conditions of the "unmodified" environment. It is expected that this environment will not exist to a large extent in the rehabilitated Atoll, but will exist in the transition period between now and the time when construction and agricultural rehabilitation is completed. At that time, much of the land will have been turned over by plowing, and large areas will be covered with buildings, coral, and cultivated vegetation. Under these conditions, it is expected that the soil surface radioactivity will be no greater than the average found now in the top 15 cm of soil. The median values for 239 Pu in soil are considered to be representative of this latter case. This evaluation is based on the survey data for 239 Pu

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	In top 15 cm		<u> </u>	In top	2 cm	• in t
Island	Median	Range	Median	Range	No. of samples	•
ALICE	12	3.9-68	56	3.9-105	5	- 10
BELLE			96	12-230	5	
Dense	26	7.2-130				
Sparse	11	5.8-26				
CLARA	22	3.8-88	40	11-80	4	
DAISY			50	8-180	6	
Dense	41	22-98				
Sparse	15	3.8-33				
EDNA	18	13-24	18		1	
IRENE	11	2.4-280	13	2.3-43	21	
JANET	8.5	0.08-170	21	2.8-100	14	
KATE			28	1.8-62	3	
Dense	17	8.6-50				
Sparse	2.3	0.17-14				
LUCY	7.7	2.4-22	34	8.0-49	5	
MARY	8.0	2.0-35	18	2.0-26	3	
NANCY	9.1	2.3-28	23	9.6-35	4	
PERCY	3.5	1.5-23	11	5.5-16	2	
OLIVE			54	2.8-87	4	
Dense	7.7	2.2-30				
Sparse	2.8	1.9-4.1				
PEARL			70	4.0-400	7	
Hot spot	51	15-530				
Remainder	11	0.85-100				
RUBY	7.3	3.0-24	2.7		1	
SALLY	4.3	0.21-130	18	1.7-62	11	
TILDA			5.8	2.0-16	4	
Dense	7.6	1.4-17				
Sparse	2.5	1.1-34				
URSULA	1.3	0.26-7.3	1.5	0.6-2.7	3	
VERA	2.5	0.60-25	22	1.5-35	3	
WILMA	1.1	0.1-5.3	3.3	1.2-7.0	4	
Southern YVONNE	3.2	0.02-50	10	0.24-32	5	

Table 146. 239,240 Pu in Enewetak soils, pCi/g.

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Table 146 (continued).

	In top 1	5 cm	In top 2	cm	
Island	Median	Range	Median	Range	No. of samples
Northern					
beaches	2.7	0.34-18			
DAVID, ELM	AER,				
FRED	0.04	0.004-0.31	0.12	0.01-0.90	17
LEROY	0.63	0.02-2.0	1.7	1.1-2.6	3
All other					
southern is	lands 0.07	0.004-1 1	0.12	0.01-0.45	22

and ²⁴⁰Pu together. The small contribution from ²³⁸Pu and ²⁴¹Pu could not influence the results of the evaluation.

Predicting the Atmospheric Levels of Plutonium

It is important to provide an evaluation which considers, as far as is possible, the potential for exposure to a returned population which accounts for the population distribution on the Atoll and the patterns of living. Under conditions of habitation, large areas of soil surface will become stabilized by cultivated vegetation, coral layering in the village areas, and by buildings. These activities will tend to reduce the possibility for resuspension of soil particles. However, human activities such as construction, earth moving, agricultural activities, and children playing, tend to stir up dust. Exposure levels to individuals will depend on such local sources.

Population exposure levels due to plutonium via the inhalation pathway have been developed on the basis of a model of dust loading. Details of the model used are discussed in Appendix A of this chapter. The assumptions made, and the values for all parameters used, are chosen to be as realistic as possible but to contain an element of caution so as not to underestimate possible effects.

As shown in Appendix A, arithmetic mean values for dust loading for nonurban U.S. locations range from 9 to 79 μ g/m³; the average of all locations is 38 μ g/m³. For urban Honolulu, Hawaii the arithmetic mean mass loading is 35 μ g/m³. It seems reasonable to assume that ambient levels at Enewetak, away from sources of soil disturbance, are similar to these values for nonurban locations.

Therefore, a value of 40 μ g/m³ is taken to represent the ambient dust loading in Enewetak air under "quiet" atmospheric conditions, assumed to be approximately 60% of the time. For 35% of the time, levels are assumed to be as high as 80 μ g/m³ close to an active population. Finally, to account for extremely dusty conditions, due either to high winds or artificial agitation of the soil, levels as high as 10 times the ambient, or 400 μ g/m³, are assumed to apply 5% of the time.

With these arguments one can derive a time-weighted value for dust loading: $40(0.6) + 80(0.35) + 400(0.05) = 72 \ \mu g/m^3$. It is instructive to examine how high a value could be obtained to gain perspective on the sensitivity of the result to each component. The latter two components are components which might change with individuals who spend a good deal of time in dusty atmospheres as a result of their occupation or habits. This individual might spend 30% of the time in an atmosphere three times the ambient, and 10%of the time in an atmosphere ten times the ambient. This person would experience an average atmosphere containing $100 \ \mu g/m^3$ of particles in air of soil origin. We will use this figure for computing the expected population dosages.

Predicting Inhalation Exposure from Atmospheric Levels

Prediction of inhalation dosages has been carried out using dose conversion factors derived by Bennett^{*} from calculations using the ICRP lung dynamics model. These factors are calculated for a class-y particle, 0.4 μ m, and low in solubility. Bennett has found that ²³⁹Pu distributions in the U.S. population are most closely explained by the lung dynamics model using the characteristics of the class-y particles. Dose conversion factors used in this evaluation are developed for continuous exposure to 1 pCi/m^3 of 239 Pu in surface air. The factors are in units of cumulative rems to the lung, liver, and bone at the end of 5, 10, 30, 50, and 70 yr of exposure (see Table 147).

Dose Estimates

Using the soil data of Table 146, the dose conversion factors of Table 147, and an average dust loading of $100 \ \mu g/m^3$, cumulative dose estimates for each of the six living patterns of Table 135 have been made for the lung (Table 148), liver (Table 149), and bone (Table 150). In the living pattern analysis, it is assumed that the population spends 60% of the time on the island of residence, 20% on other islands in food gathering and agriculture, and 20% in the beach areas of the Atoll and on the water for fishing purposes.

Weighted ^{239,240}Pu concentrations in soil were developed for each living pattern for both the "modified" and "unmodified" environment. For the "unmodified" environment, the 0-2 cm concentrations of ^{239,240}Pu were used. The "modified"

*B. Bennett, <u>Fallout Plutonium-239 Dose</u> to Man, Fallout Program Quarterly Summary Report, HASL-278, Health and Safety Laboratory, U.S. Atomic Energy Commission, New York (1974).

Table 147.	Cumulative dose to the organs from continuous inhalation of
	1 pCi/m ³ of ²³⁹ Pu in air.

		E)ose, rem, aft	er		
Organ	<u>5 yr</u>	<u>10 yr</u>	<u>30 yr</u>	<u>50 yr</u>	<u>70 yr</u>	
Lung	8.6	22.0	76	130	180	
Liver	0.4	2.4	33	93	170	
Bone	0.6	4.2	61	180	360	

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Table 148. Cumula	ative rems to	organs fro	om ^{239,24}	⁰ Pu via i	nhalation	pathway, lu	ung.
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environment is represented by the 0-15 cm soil data, on the assumption that use of the land will result in a "turnover" of soil.

The southern islands are characterized by uniformly low levels of plutonium in soil. Living patterns, such as case I, which involve a predominant use of the southern part of the Atoll, can be expected to result in insignificant dosages from plutonium via inhalation. Highest dosages were computed for unmodified conditions in living pattern IV, which is an upperlimit case of living on BELLE in the northwest portion of the Atoll. For this living pattern, modifications which homogenize the topsoil would result in reducing exposures by a factor of five.

The range of plutonium concentrations in soil in the northern half of the Atoll is large, with levels of the order of 1 to 10 pCi/g in the top 15 cm of soil. 10 to 50 pCi/g in the top 2 cm of soil, and isolated high values of 100 to 500 pCi/g. The limited number of such high values does not constitute a separate, significant condition with regard to evaluation of potential population dosage. All such data has been incorporated into the development of average soil values, island by island, and are therefore accounted for in the calculations of dose. The only conditions of potential significance, unaccounted for in the evaluation, would be those conditions which

140.	Cumuranter			•	u 110 -	aradion pu	mway, 11ve
ίľ	ING PATTERN	PCI-G IN BOIL	e noe	8. FCPED 12. JPE	20 mp.S	ES MES	DB NPA
I	. MODIFIE:	0.05	0,0000	0.0000	0.0001		0.0800
	UNMODIFIED	2.12	0.0000	0.0008	0.0004	8.0C11	0.0029
II	I. MODIFIED	2.00	0.0001	0.0005	0.0065	0.0186	0.0340
	UNMOI (FIED	4.79	0.0002	0.0011	3.0155	0.0437	3.079h
III	I. NODIFIED	7.30	0.0003	2,0012	3.0241	0.0679	0.1241
	UNMOTIFIED	17.00	9.0007	0,0041	0.0561	0.1581	A.2836
IV	∕. MODI⊑:ED	15.00	0.0006	0.0036	0.0495	0.1395	0.2550
	UNNOD (FIED	77.00	0.0031	0.0185	0,2541	a.rtet	1.30%
	V. MODIFIED	7.30	0.0003	0.0018	0.0041	a.¤673	0.1241
	UNHADI IFIED	17.60	0.0007	0,0042	0.0581	n.1637	0,2993
V	I. MODIFIED	9.50	0.0004	0.0023	0.0313	0.0883	0.1615
	UNNOIIFIED	14.70	D.0005	0.0035	0.0485	0.1367	9.2499

Table 149. Cumulative rems to organs from ^{239, 240}Pu via inhalation pathway, liver,

time-weight the activities and residence of people toward areas of elevated plutonium in soil. Thus, if a large portion of the population spends a large fraction of the time in the specific areas of elevated levels of plutonium in soil, population dosages would be increased. These factors should be considered in examining the need for remedial actions, such as soil removal. An extreme example, the hypothetical occupation of the area on DAISY with up to 500 pCi/g, can be used for perspective. Under these maximum, hypothetical conditions. use of this area might lead to dosages of the order of 4 rems to the lungs in 30 yr of exposure, and a similar dosage to the bone.

YVONNE is a unique island with respect to plutonium contamination, in particular the northern part of the island (see the chapter on terrestrial soil and radiation survey for a description of the distribution of plutonium in soils). This part of the Atoll has the highest plutonium levels observed in the survey, and was the only area in which positive identification of resuspended plutonium was made in the air-sampling program. YVONNE is characterized by a nonuniformity of contamination, a large inventory of plutonium. and the existence of pure particles of <u>،</u> ، ، plutonium metal. Unrestricted land use 1 Ĩ of YVONNE, without remedial action, would produce the highest potential for H

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Table 150. Cumulative rems to organs from 239,240 Pu via inhalation pathway, bone.

population dosages from plutonium. The level of radiological significance, however, depends upon land-use plans as well as radionuclide inventory.

Bringing the radiological significance of YVONNE into line with the rest of the Atoll will require either restriction against habitation of the island, or removal of large amounts of contaminated soil.

In summary, the levels of plutonium observed in soils of the Atoll, excluding YVONNE, can be expected to lead to long-term, average air concentrations of plutonium which are the order of a millionth up to a thousandth of the MPC_a . These estimates have been made with such

assumptions that it is very doubtful that they could be underestimates of the potential population dosages. Population dosages derived from YVONNE would probably exceed ICRP guidelines if this island is used for habitation without prior soil removal.

Appendix A

Relationship Between Resuspended Plutonium in Air and Plutonium in Soil

L. R. Anspaugh Lawrence Livermore Laboratory Livermore, California

There are no general models that may be used with confidence to predict the resuspended air activity in the vicinity of an area contaminated with plutonium. However, two approximate methods may be used — the resuspension factor approach and an argument based upon ambient air particulate concentrations, with the assumption that the particulates are derived from the contaminated surface. The former method has been frequently used, but almost always in the context of a fresh surface deposit. The latter method is inappropriate to the fresh deposit situation, but should be reasonably valid after enough time has elapsed for the surface-deposited material to become fairly well mixed with a few centimeters of the soil surface.

Resuspension Factor Approach

The resuspension factor, K, is defined as

 $K = \frac{Air \text{ concentration } (Ci/m^3)}{Surface \text{ deposition } (Ci/m^2)},$

and thus has units of m^{-1} . It is almost always implied that both measurements are made at the same location. The difficulties with this approach are fairly obvious — no allowance is made for the geometrical configuration of the source, the particle-size distributions of the contaminant and the soil surface, vegetation cover, etc. Stewart¹ and $Mishima^2$ have tabulated values of K from many experiments including those involving laboratory floors as well as native soils. As would be expected, the tabulated values cover an enormous range and vary from 10^{-2} to 10^{-13} /m. Most of the high values, however, are derived from experiments with laboratory floor surfaces and/ or with artificial disturbance.

For outdoor situations, Stewart¹ suggests as a guide for planning purposes that a value for K of 10^{-6} /m be used

"under quiescent conditions, or after administrative control has been established in the case of an accident." A value of 10^{-5} /m is suggested under conditions of moderate activity. Stewart states, however, that exceptionally higher values (mean of 10^{-5} /m) were observed during the Hurricane Trial (Monte Bello Islands) and credited this to the nature of the small islands exposed to sea breezes. Values approaching 10^{-3} /m when dust is raised by pedestrians and vehicles are also reported by Stewart.

Kathren³ has also considered the resuspension factor approach and has recommended the use of 10^{-4} /m as a conservative but appropriate value for setting standards for PuO₂ surface contamination.

Langham^{4, 5} has suggested that a value of 10^{-6} /m is a reasonable average value to use in estimating the potential hazard of occupancy of a plutonium-contaminated area. At the same time, however, Langham notes that many measured values lie in the range of 10^{-5} to 10^{-7} /m and reports that his own measurements in 1956 produced a value of 7×10^{-5} /m.

These recommended values, however, are all intended for application during the time period immediately following deposition. Numerous studies^{1, 5-8} have shown that air concentrations of resuspended materials decrease with time. With the assumption that this decrease can be represented by a single exponential function, half-times of 35 to 70 days have been reported^{5, 7, 8}. This decrease in air activity is not explainable by the relatively minor loss of material from the initial site of deposition^{1, 6}, but is presumably caused by the migration of the initial surface-deposited material into the soil.

Attempts to use the resuspension factor approach to derive acceptable levels of soil surface contamination have included this "attenuation factor" as a simple exponential function with halftimes of 35 or 45 days $^{3, 4}$. There are major uncertainties in such a formulation, however. The longest study of this decrease with time extended to only 11 mo following the initial deposition⁸, which is extremely short compared to the halflife of a radionuclide such as 239 Pu. There are also published reports which indicate on experimental and theoretical bases that the decrease with time will not be adequately represented by a single exponential function, but that the rate of decrease itself will also decrease with time^{1, 6}. Fortunately, the exact nature of this time dependence is not critical in determining the integrated exposure from the time of initial deposition due to the fairly well-documented rapid decrease at early times. However, it is obviously the controlling factor for questions concerning the reoccupation of areas many years after the contaminating event.

As an illustration, the most conservative published model (Kathren³) may be used to calculate a resuspension rate for material 15 yr after deposition:

$$K = \frac{10^{-4}}{m} \exp\left(\frac{-0.693 \times 15y \times 365d}{45d}\right)$$

\$\approx 10^{-41}/m.

If, however, the resuspension rate asymptotically approached some finite value 10^{-6} of the original, then the resuspension rate 15 yr later would obviously

be 10^{-10} /m. However, the total integrated air activity (from t = 0 to ∞) for 239 Pu would be changed only by

$$A \times 10^{-4} \int_{0}^{\infty} \exp(-0.693t/45d) dt$$

+ A × 10⁻¹⁰ $\int_{0}^{\infty} \exp(-0.693t/24, 400y) dt$
= 6.5A × 10⁻³ + 1.3A × 10⁻³.

which is an increase of 20%, and more importantly, cannot be accumulated during an individual's life span.

Because the functional nature of the decrease in resuspension rate with time cannot be confidently extrapolated, previously published models should not be applied to the reoccupation of areas many years after the contaminating event.

The resuspension-factor approach can be applied in an approximate way, however, if resuspension factors are used which were derived from measurements over aged sources. Perhaps the most relevant data are unpublished results from current resuspension experiments at the GMX site in Area 5 of the Nevada Test Site. The ²³⁹Pu at this location was deposited following 22 high-explosive detonations during the period from December 1954 to February 1956. Measurements of resuspended air activity levels at this site during 1971-1973 appear to be the only available data concerning resuspension of ²³⁹Pu from a source of this age.

Data from two types of measurements are available and can be used to derive average resuspension factors. The first type of measurement⁹ was accomplished by placing five high-volume cascade impactors¹⁰ within the most highly contaminated area, and running them for 36 days, from July 7 to August 12, 1972. The collected ²³⁹, ²⁴⁰Pu activity was distributed lognormally with particle size with an activity median aerodynamic diameter (AMAD) of 3.0 µm and a geometric standard deviation of 8.2. The ^{239, 240}Pu concentration varied from 1.0×10^{-14} to $3.9 \times 10^{-14} \ \mu \text{Ci/cm}^3$, with an average of 2.3 \times 10⁻¹⁴ $\mu Ci/cm^3$ for the five samplers. At the present time only limited data are available regarding the soil activity in the area. Four soil samples of depth 0-3 cm from approximately the same location have been analyzed with results¹¹ of 2060 to 3550 dpm/g, with a mean of 2700 dpm/g. Profile data from other locations at the same general site indicate that about 90% of the total deposition is contained within the top 2.5 cm of the soil 12 . Measurements of soil density in the area average 1.8 g/cm³. The resuspension factor is therefore

$$\frac{2.3 \times 10^{-14} \ \mu \text{Ci}}{\text{cm}^3} \times \frac{\text{g}}{2700 \ \text{dpm}} \times \frac{\text{cm}^3}{1.8 \ \text{g}} \times \frac{0.9}{3 \ \text{cm}} \times \frac{10^2 \ \text{cm}}{\text{m}} \times \frac{2.22 \times 10^6 \ \text{dpm}}{\mu \text{Ci}}$$

= 3 × 10⁻¹⁰/m.

Additional air samples were taken by the Reynolds Electrical and Engineering Co. (REECo) on the edge of the contaminated area during the period of February 1971 to July 1972, with a sampling time of approximately 48 hr¹³. Measurements were made at four locations, but the most pertinent is the one which was most frequently in the direction of strong winds from the strongly contaminated area and where the highest air activities were recorded. Here, 254 individual airfilter samples were collected and detectable results reported for 236. ²³⁹, **30** Pu concentrations ranged from 3.5×10^{-17} to $6.3 \times 10^{-13} \ \mu \text{Ci/cm}^3$, with arithmetic and geometric means of 6.6×10^{-15} and $7.9 \times 10^{-16} \ \mu \text{Ci/cm}^3$, respectively. Results for four soil samples taken from approximately the same location range from 128 to 202 dpm/g, with a mean of 160 dpm/g¹¹. Because the arithmetic mean is a better representation of average lung exposure, it is used to derive a resuspension factor at this site:

$$\frac{6.6 \times 10^{-15} \ \mu \text{Ci}}{\text{cm}^3} \times \frac{\text{g}}{160 \ \text{dpm}} \times \frac{\text{cm}^3}{1.8 \ \text{g}} \times \frac{0.9}{3 \ \text{cm}} \times \frac{10^2 \ \text{cm}}{\text{m}} \times \frac{2.22 \times 10^6 \ \text{dpm}}{\mu \text{Ci}}$$

= 2 × 10⁻⁹/m.

This value is nearly an order of magnitude higher than the one previously calculated, and reflects some of the inherent difficulties in the resuspension-factor approach, i.e., that no allowance is made for the geometrical configuration of the source and that higher ground activities may be present upwind.

It is obvious that this approach is subject to major uncertainties, but does serve as an order-of-magnitude indication of the resuspended air activities that may arise from a ^{239, 240}Pu contaminated area which has weathered for 15 to 20 yr. The data discussed above also demonstrate unequivocally that resuspension of ^{239, 240}Pu does in fact occur from such aged deposits and at levels many orders of magnitude higher than would be expected if the often noted decrease with time were represented by a single exponential function with a half-time of 35 to 70 days.), 210 pu 10⁻¹⁷ imetic 5 and 5 Re-10m inge in of 2tic average a re-

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Mass-Loading Approach

The other approximate prediction method is based upon measured or ssumed levels of particulate matter in ambient air with the assumption that this material is derived from the contaminated soil. For fresh deposits this approach is not valid because the freshly deposited lebris is much more likely to be resuspended than the remainder of the weathered soil surface. After many vears of weathering since the initial deposition, however, the contaminating material should be reasonably well mixed with a centimeter or two of soil, such that the contaminant activity per gram of airborne particulate should approximate that in the upper soil. However, a major difficulty could arise if, for example, ^{239, 240}Pu were preferentially associated with the smaller particle sizes more likely to become airborne. For the Nevada Test Site, such is not the case as determined by soil analyses 14 and by the high-volume cascade impactor study. The latter study found an AMAD of 3.0 μ m for ²³⁹, ²⁴⁰Pu, whereas the total mass median aerodynamic diameter was 1.7 µm. The specific activity of the material collected on each stage can also be examined for a preferential association of plutonium with particle size. Average data from all five samplers are:

Size, µm	239,240 _{Pu, dpm/g}
>7	950
3.3 to 7	700
2.0 to 3.3	1030
1.1 to 2.0	1300
0.01 to 1.1	480
All stages	890
(Soil)	(2700)

Although there is considerable spread in these data, there is no indication of a preferential association of ²³⁹, ²⁴⁰Pu with a particular particle size; as would be expected as a result of dilution by inert aerosol, the specific activity is lower than that of the soil.

If we assume that this is generally true, a general and conservative method of predicting resuspended air concentrations of contaminants would be to simply multiply the ambient air mass loading by the contaminant concentration in soil. A factor of some uncertainty for a specific calculation is what value to use for the ambient air mass loading in the absence of specific data. This becomes even more uncertain because of the possibility that the people involved may be highly correlated with the source in the sense that children playing in sand, adults cultivating crops, etc., may generate their own "ambient air" which contains much more mass than would be recorded by a remote stationary sampler.

The lower and upper bounds of ambient air mass loading can be fixed rather easily for any site. There has been considerable interest in establishing a "background level" of mass loading, and this is generally believed to be about $10 \ \mu g/m^3$ (15). The upper bound can be established in a reasonable way by the levels found in mine atmospheres which have led to a considerable prevalence of pneumoconiosis in the affected workers¹⁶. Examination of these data indicate that current standards for occupational dust exposure (~1-10 mg/m³) have a very small, or perhaps no margin of safety, such that a reasonable upper bound can be taken as 1 mg/m^3 . British data¹⁷

indicate that if the general public were exposed to dust levels in excess of 1 mg/m^3 , the public health problem from the dust alone might be enormous. The reasonableness of the upper limit value of 1 mg/m^3 is also demonstrated by data which indicate that nonurban ambient air mass concentrations this high are usually associated with conditions described as dust storms^{18,19}.

Measurements of ambient air mass loading can be used to further define a reasonable estimate for predictive purposes. The National Air Surveillance Network (NASN) has reported such results for several years. Data 20 for 1966 show that there were 217 urban and 30 nonurban stations reporting. The annual arithmetic average for the urban stations ranged from 33 (St. Petersburg, Florida) to 254 μ g/m³ (Steubenville, Ohio), with a mean arithmetic average for all 217 stations of 102 μ g/m³. For the nonurban stations, the range was from 9 (White Pine County, Nevada) to 79 μ g/m³ (Curry County, Oregon), with a mean arithmetic average for all 30 stations of 38 μ g/m³. No data in this report are available for nonurban locations on small islands similar to the Enewetak group; perhaps the closest analog is the urban station at Honolulu, Hawaii, which had an annual arithmetic average of 35 μ g/m³.

More pertinent, but limited, data have recently been published for the island of Hawaii^{21, 22}. Data are given for three locations: Mauna Loa Observatory located at a height of 3400 m, Cape Kumukahi, and the city of Hilo. NASN data for Hilo (for an unspecified period) are given as 18 μ g/m³, and nephelometer measurements varied from 18 μ g/m³ during the day to 26 μ g/m³ at night. At Cape Kumukahi the nephelometer measure. ment was 9.2 μ g/m³. The greatest amount of data is available for Mauna Loa Observe torv. Here, the NASN measurement was $3 \ \mu g/m^3$, and the nephelometer measurements varied from 1.7 μ g/m³ at night to 6.5 μ g/m³ during the day. Additional measurements made by the USAEC Health and Safety Laboratory (HASL) were $3 \mu g/m^3$. It is of interest in the present context that $Simpson^{22}$ made the following comment concerning the HASL measurements: "The HASL filter samples contain substantial dust $(3-5 \mu g/m^3 \text{ of air sampled})$ because of the fact that the filter was located less than one meter above the ground surface near areas with substantial personnel activity at the observatory site." Thus, while this method of measurement may not have coincided with Simpson's interest, it does indicate that ambient air mass loadings may be very low on such remote islands even when considerable human activity is occurring nearby.

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On the basis of the above data, it would appear reasonable to use a value of $100 \ \mu g/m^3$ as an average ambient air mass loading for predictive purposes. Indications are that this value should be quite conservative for the Enewetak Islands, and therefore allows room for the uncertainty involved because the people themselves may generate a significant fraction of the total aerosol. Therefore, they may be exposed to higher particulate concentrations than would be measured by a stationary sampler.

Supporting evidence that 100 μ g/m³ is a reasonable value to use for predictive purposes is provided by the National Ambient Air Quality Standards²³. Here ambient air is defined as "...that portion of the atmosphere, external to buildings, to which the general public has access." The primary ambient air standards define "levels which... are necessary, with an adequate margin of safety, to protect the public health." The secondary standards define "levels which... (are)...necessary to protect the public welfare from any known or anticipated adverse effects of a pollutant." These standards for particulate matter are given below:

National ambient air quality standards for particulate matter, $\mu g/m^3$.

Annual geometric mean	Max. 24-hr comentration not to be exceeded more than once a year
Primary:	
75	260
Secondary: 60	150

Data to support these standards in terms of health effects, visibility restrictions, etc. have been provided 24 .

An arithmetic mean would be more desirable for predictive purposes. Data from 1966^{20} for nonurban locations indicate that the annual arithmetic mean is (on the average) 120% of the annual geometric mean.

Representative Calculations

Because one of the primary objects is to derive an acceptable soil level for the Enewetak Islands, the approaches developed above were used to derive such levels for both soluble and insoluble ²³⁹Pu. The derived values are given in Table 151. The two methods agree within a factor of two, at least for soil distributions like those found at the Nevada Test Site. The ambient air mass loading at

Table 151. Acceptable soil levels of ²³⁹Pu for a source which has weathered for several years. Values are approximate and are subject to uncertainty. Permissible Concentration in Air for 168-hr occupational exposure (MPC_a)25.

	Insoluble	Soluble
Acceptable air concentration, $\mu \text{Ci}/\text{cm}^3$	10 ⁻¹²	6×10^{-14}
Resuspension-factor	approach	
Assumed resuspension factor, m^{-1}	10 ⁻⁹	10 ⁻⁹
Acceptable soil deposition ^a , $\mu Ci/m^2$	10 ³	60
Acceptable soil concentration ^b , nCi/g	20	1
Mass-loading appr	oach	
Assumed mass loading, $\mu g/m^3$	10 ²	10 ²
Acceptable soil concentration, nCi/g	10	0.6

^aEquivalent to approximately $10^4 \ \mu g$ of insoluble $\frac{239}{Pu/m^2}$.

^bAssumes same distribution of ²³⁹Pu with depth and soil density as measured at the Nevada Test Site.

NTS during the cascade impactor run was measured to be 70 μ g/m³.

Such derived values must, of course, be used with a great deal of discretion. They are based on simple model systems which are believed to be generally conservative, but individual situations can be imagined which could exceed the predictions.

Other Considerations

The above calculations relate only to the resuspended air activity in ambient air, and do not consider the additional problems of resuspension of material from contaminated clothing or the resuspension of material which has been transferred to homes.

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Healy²⁶ has considered these and other problems, and has provided tables of "decision levels" for surface contamination levels and home transfer levels. A decision level is based upon National Council on Radiation Protection and Measurements (NCRP) recommended dose limitations. Because the derivations

Table 152.	Decision levels 26 for soluble 239 Pu, and their equivalent in soil mass
	based upon the "acceptable soil concentration" from Table 151.

Pathway		Decision level	Mass equivalent
Direct inhalation ^a Direct ingestion ^b Skin absorption ^c	A. Din	The contamination $2 \times 10^{-5} \text{ nCi/cm}^2$ 0.2 nCi/cm^2 $8 \times 10^{-4} \mu \text{Ci}$	$1 \times 10^{-5} \text{ g/cm}^2$ 0.2 g/cm ² 0.8 g
	B. Tr	ansfer (to homes) levels	
Resuspension ^d		$0.01 \mu \text{Ci}/\text{day}$	$10 \mathrm{g/day}$

Resuspension	$0.01 \mu \text{Ci/day}$	10 g/day
Direct inhalation	0.01 μ Ci/day	10 g/day
Direct ingestion	100 $\mu Ci/day$	10^5 g/day
Skin absorption	0.03 μ Ci/day	30 g/day

^a"The contamination level on clothing and skin that could result in inhalation of air at the MPC₂ for the public."²⁶

^b"The contamination level on skin or clothing that could result in ingestion of a quantity of radioactive material equivalent to the ingestion of water at the MPC_w for an individual in the public."²⁶

^c"The total quantity of radioactive material maintained on the skin for 24 h/day that could result in absorption of a quantity equal to that which would be absorbed from the GI tract if water at the MPC_w for "soluble" isotopes for an individual in the public were ingested. "²⁶

d"The amount transferred per day that could result in air concentrations due to resuspension in a medium-sized home averaging at the MPC_a for an individual in the public."²⁶

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it n are rather tenuous, Healy has used the phrase decision level and states that its use is to serve as a signal that further careful investigation is warranted.

Healy's decision levels for soluble 239 Pu are given in column 1 of Table 152. The values in column 2 are derived from these and an acceptable soil concentration of 1 nCi/g from Table 151 to give equivalent dirt (soil) contamination and transfer levels. The results are interpreted as indicating that the potential exists for greater dose contributions from these infrequently considered pathways than from the usually considered pathway of resuspension as calculated for ambient air. This conclusion would be the same for insoluble ²³⁹Pu. Therefore, if dose calculations based on the usual resuspension pathway should appear limiting compared to other pathways such as foodchain transfer, these pathways considered by Healy need to be carefully evaluated for the specific Enewetak situation.

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<u>Dose Estimates for the Marine Food</u> Chain

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Introduction

This analysis is designed to estimate the dose via the marine pathway. The dose assessment is based upon the marine diet discussed in the chapter on dietary and living patterns (Table 139).

Data Bank

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The data bank contains analytical results from slightly over 800 fish and approximately 50 edible invertebrates collected during the 1972 Enewetak survey (for a detailed description see the marine survey chapter). Data from the analysis of the radionuclide concentration in fish muscle have been summarized in several different ways to help in the interpretation and the assessment of the values to be used in the dose code. Figure 118 indicates the various forms of the summarized data.

Table 153^{*} lists the average radionuclide concentration — with concentrations for a nondetected nuclide set equal to the detection limit in column 4 and concentrations for nondetected nuclides set equal to zero in column 7 — for each species for samples collected at each island and in the open lagoon. Table 154 (on microfiche) ¹ presents the summary of the average radionuclide concentration for each species for the entire Atoll, regardless of location. The nuclides are identified by the

sequence of numbers in the nuclide column. The first two digits give the atomic number and the last three digits give the isotope mass number; therefore 55137 is 137 Cs. The tables also include the tissue, the number of samples in the average, the range of individual values, and, because of the skewed distribution observed in this survey and observed for trace elements and radionuclides in other populations $^{1-3}$, the lognormal median for comparison with the average value.

The reef fishes are the most plentiful around the Atoll and are the easiest to catch. Therefore they make up a considerable portion of the fresh fish intake in the diet. The most plentiful reef fishes, and also three of the preferred fish in the diet, are surgeonfish, goatfish.



Fig. 118. Summaries of marine concentration data.

Because of the sheer bulk of the data, Tables 153-155 and 157 have been reproduced on microfiche film and may be found in the envelope mounted on p. 527.

Tables 153, 154, 155, and 157. Radionuclide concentration in fish muscle.





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und mullet. Other reef fish are eaten out are not as plentiful. In addition, the larger pelagic, predator fish are eaten, but they are harder to catch and therefore supply much less of the fish diet than the reef fishes. Tridacna clams also constirute a small portion of the diet. They are considered a delicacy, are not available in large quantity, and are usually eaten raw at the time of catch. Lingusta (lobster) are also considered a delicacy out contribute a very small portion of the marine diet.

Therefore the next summary presents the average radionuclide concentration of four main fish groups — surgeonfish, goatfish, mullet and "other"— where "other" includes all species other than the three mentioned, including the tridacna clams and lingusta. The summary is shown in Table 155 (on microfiche) and is island specific.

Table 156, incorporated in the text, lists the average concentration of the radionuclides in the four fish groups for the entire Atoll. The number of samples in the average concentration, the standard deviation, and the high and low of the range are all given. The plot of the concentration of 137 Cs. 60 Co. and 90 Sr. the three main isotopes found in fish muscle, for the four fish groups is shown in Fig. 119. The standard deviations for each of the four fish groups were a factor of 2 to 3 times greater than the difference between the range of the mean values. There was therefore no statistically significant difference in the mean values of the four groups; however, the Kruskal-Wallis nonparametric test did indicate a difference in the total distribution for 60 Co and 90 Sr.



Fig. 119. Average concentration of ${}^{137}Cs$, ${}^{60}Co$, and ${}^{90}Sr$ for the four fish groups for the entire Atoll.

Because there were no statistically significant differences between mean values for the four major fish categories, the radionuclide concentration was averaged by island for all fish. These results are given in Table 157 (on microfiche). Figures 120-122 show a plot of the 137 Cs, 60 Co, and 90 Sr average nuclide concentration in all fish as a function of island location.

There appears to be a higher concentration of the three radionuclides in fish from ALICE through IRENE than from islands JANET through LEROY. Although individual samples from islands JANET through LEROY had concentrations in the

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	ORMAL AN PCI⊲5PAM 2E+00
	2E+00
40 k HUSCLE 26 8.122E+80 2.114E+80 1.104E+802 5.586E-81 8.022E+00 3.28 55 FE HUSCLE 27 1.930E+80 4.132E+80 2.189E+801 1.495E-81 1.904E+808 8.232 90 SR MUSCLE 25 8.181E-82 8.926E+80 3.041E+81 1.495E+81 1.495E+81 1.495E+81 1.495E+81 1.495E+81 1.495E+81 1.495E+81 1.904E+808 8.222E+88 4.21 90 SR MUSCLE 25 8.181E-82 8.926E+82 3.041E+81 3.459E+81 6.81E+82 4.21 106 RU MUSCLE 21 7.581E-91 3.077E+91 1.379E+80 3.618E+91 0.551E+92 7.763E+92 7.763E+92 1.655 125 SB MUSCLE 27 2.629E+91 3.732E+91 2.996E+90 7.734E+92 7.763E+92 1.87 137 CS MUSCLE 19 1.167E+81 8.185E+91 4.344E+80 2.636E+92 2.754E+91 1.40 133 BA MUSCLE 19 1.167E+81 1.603E+01 4.178E+91 2.877E+91 9.636E+92 2.754E+91	SE+00 7E-01 SE-01 7E-02 1E-01 1E-01 0E-01 7E-02 7E-02 3E-02 3E-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02 SE-01 SE-02

Table 156a. Summary of radionuclide concentrations for the entire Atoll for mullet.

*AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO DETECTION LIMIT) PCI/GRAM **AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO ZERO) PCI/GRAM

Table 156b. Summary of radionuclide concentrations for the entire Atoll for surgeon.

Table IV	JN. Dum							
NUCLIDE	TISSUE	NO. OF SAMPLES	AVERAGE PCI∞GRAM≉	STANDARD DEVIATION	RANGE PCI∕GF HIGH	RAM LŪW	AVERAGE PCI/GRAM**	LOGNORMAL MEDIAN PCI/GRAM
	MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE MUSCLE	4 25 27 28 27 27 28 28 28 28 28 28 28 28 28 28 28 28 28	3.298E-01 1.004E+01 9.892E+00 1.815E+00 2.098E-01 7.073E-01 6.618E-02 1.816E-01 5.387E-01 7.708E-02 7.345E-02 1.535E-02 1.158E-01 5.434E-02 7.724E-02 8.431E-02 CONCENTRATION	1.1666-01 2.552E+00 1.364E+01 5.789E+00 2.801E-01 4.951E-01 7.081E-02 1.243E-01 1.271E+00 8.988E+02 4.802E+02 5.551E+02 1.529E+01 3.857E+02 1.689E+01 6.707E+03 5.731E+02	4.698E-01 1.598E+01 6.757E+01 3.106E+01 1.275E+00 2.104E+00 3.719E-01 4.806E-01 6.779E+00 4.005E-01 1.924E-01 1.924E-01 1.924E-01 1.821E-01 8.874E-01 8.874E-01 1.920E-01 DETECTION LIN CP0	1.845E-81 7.194E+88 2.703E-01 6.108E-82 6.441E-83 3.017E-81 1.805E-82 7.757E-02 4.363E-82 2.445E-82 3.007E-82 3.007E-82 1.965E-82 2.271E-83 1.965E-83 1.965E-83 1.963E-83	3.298E-01 1.004E+01 9.750E+00 1.765E+00 2.029E-01 0. 0. 5.056E-01 0. 7.367E-02 0. 7.367E-02 0. 7.683E-03 0. 6.	3.128E-01 9.758E+00 4.718E+00 4.647E-01 9.282E-02 5.938E-01 4.894E-02 1.515E-01 2.222E-01 5.292E-02 6.184W-02 7.105E-02 6.825E-02 2.804E-02 5.862E-03 5.862E-03 5.863E-02
***AV	ERAGE (1F 1	NON-DETECTED.	CONCERTENTION	SEL EUGHE TO				

Table 156c. Summary of radionuclide concentrations for the entire Atoll for goatfish. RANGE PCI/GRAM STANDARD

LOGHORMAL AVERAGE

NO. OF

AVERAGE

Table 156c. Summary of radionuclide concentrations for the entire Atoll for goatfish.

NUCL IDE	TISSUE	NO. OF SAMPLES	AVERAGE PCI/GRAM*	STANDARD DEVIATION	RANGE PCI∕G HIGH	RAM LŪIJ	AVERAGE PC 126PAM***	LOGNORHAL MEDIAN PCI/GRAM
3 H	MUSCLE	3	4.108E-01	6.860E-02	4.622E-01	3.3298-01	4.103E-011	4.0076-01
40 K	NUSCLE	19	9.662E+00	3.464E+00	1.637E+01	5.9466000	14 JULY 1 1111	10, 372 4 #100
55 FE	MUSELE	20	3.0128+01	8.358E+01	3.833E+02	1.0.768.000	5.01.1.0011	1.001/0-001
60 CO	MUSCLE	24	1.5066+00	3.00/E+00	1.477E+01	1.0000E-001	1.1351.000	1 1 1 1 1 E - 1 1 I
90 SR	NUSCLE	24	2.152E-01	3.5276-01	1.541E+00	1.212E-02	2.15-004	1 . FE-56 E = FFT
106 RU	NUSCLE	10	7.548E-01	4.684E-01	1.943E+00	3.768E-01	6.	i, e ×3E÷01
102 RH	MUSCLE	21	8.293E-02	4.513E~02	1.714E~01	2.643E-02	θ.	7.204E-02
125 SB	MUSCLE	21	2.9785-01	3.943E-01	1.986E+00	9.743E-02	1.054E-01	
137 CS	NUSCLE	21	1.718E-001	1.963E-01	9.878E-01	4.201E 02	1.010E-01L	1.8.54L (011
133 BA	NUSCLE	19	1.6758-01	1.050E-01	4.118E-01	2.785E 0.1	21. DSBE - 0.2	1.114 011
144 CE	MUSCLE	1	2.739E-01	0.	2.739E-01	0.230E 01	U.	11.1130月1月1日
152 EU	MUSCLE	21	5.915E-02	4.338E-02	1.903E-01	2.279E-03	11.	は、地方でも見る
155 EU	MUSCLE	24	1.208E-01	8.445E-02	4.122E-01	BIRNDE HA	5.55.E=02	1.018E-01
207 BI	NUSCLE	21	7.464E-01	5.3208-01	1.6338+00	1.214E-01	7.379E-01	5.555E-01
235 U	MUSCLE	20	9.357E-02	4.347E+02	1.508E-01	2.482E-02	0.	8.0708-02
239,240 PU	MUSCLE	21	1.299E-02	1.513E-02	5.315E-02	1.608E-03	1.231E-02	7.288E-03
238 PU	MUSCLE	12	1.066E-02	7.512E-03	2.4325-02	3.153E-05	5.405E-03	8.090E-03
241 AM	MUSCLE	21	1.111E-01	4.604E-02	2.053E+01	2.774E+02	θ	0.907E-00

*AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO DETECTION LIMIT) PC1 SPAN **KAVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO ZEPO) PC1/SPAN

Table 156d. Summary of radionuclide concentrations for the entire Atoll for other fish.

ubic 1000.					DANGE DEL C	E .211		LOGNORMAL
		NO. OF	AVERAGE	STANDARD	RANGE PCI/G		AVERAGE	
NUCLIDE	TISSUE	SAMPLES	PCI/GRAM*	DEVIATION	HIGH	LGW	РСІ БРАМАК	MEDIAH PCI/GRAM
3 H	MUSCLE	2	5.040E-01	3.040E-01	7.189E-01	2.8906-01	5.040E-01	4.5588-01
40 K	MUSELE	46	1.593E+01	5.369E+00	2.697E+01	3.3556+00	1.593E+01	1.481E+01
55 FE	MUSCLE	50	1.717E+01	3.209E+01	1.968E+02	1.577E-01	1.704E+01	5.012E+00
60 CO	MUSCLE	52	2.347E+00	6.448E+00	3.827E+01	4.063E-02	3.282E+00	5.167E-01
90 SR	NUSCLE	52	1.403E-01	2.307E~01	8.514E-01	1.051E-03	E. BERE-RU	5.12ME 02
106 RU	MUSCLE	30	9.528E-01	4.861E-01	2.2378+00	8.236E-01	Ŋ.	81.4505E=01
102 RH	MUSELE	30 52	1.099E-01	6.257E-02	2.954E-01	2.481E-02	Ð.	9.180E-02
113 CD	MUSCLE	1	2.635E-01	Ũ.	2.635E-01	3.635E-01	2.635E-01	2.6358-01
125 SB	MUSCLE	52	2.486E-01	1.434E-01	1.025E+00	7.788E-0C	1.295E-02	2,2128-01
137 CS	MUSCLE	52	4.311E-01	5.679E~01	3.089E+00	2.689E-00	4.035E-01	2.539E+01
133 BA	MUSCLE	46	1.726E-01	1.342E-01	7.631E-01	2.632E-02	2.718E-03	1.2846-01
144 CE	MUSCLE	2	2.837E-01	1.949E-02	2.975E-01	2.6996-01	в.	2.834E-01
152 EU	MUSCLE	52	8.945E-02	7.5076-02	3.415E-01	2.933E-02	U.	6,8665-02
155 EU	MUSCLE	52	1.226E-01	9.119E-02	5.212E+01	3.570E-03	1.483E-02	1.019E-01
207 BI	NUSCLE	52	5.533E+00	3.499E+01	2.527E+02	<u>3</u> .074E-02	5.493E H IÓ	1.7766-01
235 U	MUSCLE	43	9,569E-02	5.005E-02	2.547E-01	2.3826-02	Ũ.	8,192E-02
239,240 PU	MUSCLE	49	7.000E-02	2.383E-01	1.212E+00	7.883E-04	6.368E-02	9.002E-03
238 PU	MUSCLE	31	1.805E-02	2.984E-02	1.140E-01	2.063E-03	6.089E-03	7.983E+03
241 AM	MUSELE	52	1.404E-01	1.130E-01	8.023E-01	2.700E-02	6,820E+03	1.148E-01

*AVERAGE (IF NON-DETECTED, CONCENTRATION SET COUAL TO DETECTION LIMIT) POLYGPAN **AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO ZEPO) POLYGPAN

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Fig. 120. ¹³⁷Cs concentrations in marine samples as a function of location in the Atoll. The bar above each vertical arrow indicates the maximum value for a sample included in the average. No bar or arrow indicates a single sample.

same range as individual samples for islands ALICE through IRENE, there was definitely a significant difference for the 137 Cs and 60 Co (p = 0.001 Mann-Whitney U Test) concentrations for fish from ALICE through IRENE, versus those from JANET through LEROY. There was no significant difference between these island groups for fish muscle samples for 90 Sr. If fish samples for eviscerated whole fish (which includes the bones) are included, then 90 Sr concentrations do test differently for these island groups.

However, the people living on Engebi (JANET) will fish both east and west of the island; that is, they will fish off the islands ALICE through IRENE, but will also fish off the islands KATE through WILMA. In essence, the people living on Engebi will fish the northern half of the Atoll. Therefore, in their fish diet, they will integrate the concentrations of the fish from the northern half of the Atoll, i.e., ALICE through WILMA. Again using the Mann-Whitney U Test, concentration values for the three isotopes for all fish from islands ALICE through WILMA, i.e., the northern half of the Atoll, were tested against the concentration values for all fish from islands ALVIN through LEROY, i.e., the southern



samples as a function of location in the Atoll. The bar above each vertical arrow indicates the maximum value for a sample included in the average. No bar or arrow indicates a single sample. Fig.

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Concentration - pCi/g



Fig. 122. ⁹⁰Sr concentrations in marine samples as a function of location in the Atoll. The bar above each vertical arrow indicates the maximum value for a sample included in the average. No bar or arrow indicates a single sample.

half of the Atoll. The results for fish muscle show no difference between the two halves of the Atoll for 90 Sr (p = 0.7), 137 Cs (p = 0.1), and 60 Co (p = 0.4). 137 Cs and 90 Sr tested as significantly different (p = 0.001) for eviscerated whole fish which included bone; however, the average concentration in this case for the two halves of the Atoll differs by only a factor of 3 for 137 Cs and a factor of 2 for 90 Sr, and the average for the entire Atoll is less by only a factor of 2 for 137 Cs and only 30% for 90 Sr than the average of the northern half alone. As a result of the above analysis and the fact that the Enewetak people eat only the muscle portion of the fish, the average concentration (with concentrations for nondetected radionuclides set equal to the detection limit) for fish from the <u>entire</u> Atoll was used in the dose code. Table 158 lists the average concentration, the number of samples in the average, the standard deviation, and the high and low of the range for each radionuclide for all fish.

The concentration distributions for 90 Sr, 137 Cs, and 60 Co are quite skewed (Figs. 123-125) and are consistent with other published data on radionuclide and trace-element distribution in fish, animals, and humans (1, 2, 3). The lognormal median is therefore included in the Table 158 for comparison with the average value. In general, the lognormal median is 3 to 4 times less than the average population dose for the marine pathway





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Table 158.	Average concentration, number of samples in the average, standard deviation, and
	high and low of the range for all fish in the entire Enewetak Atoll.

		NO. OF	AVERAGE	STANDARD	RANGE PCI/G	RAM	AVERAGE	LOGNORMAL
NUCL I DE	TISSUE	SAMPLES	PC IZGRAM*	DEVIATION	HIGH	LOW	PCI/GRAMH*	MEDIAN PCI/GRAM
01007	MUSSIF	2						
01003	MUSCLE	9	3.955E-01	1.517E-01	7.189E-01	1.845E-01	3.955E-01	3.712E-01
19040	MUSCLE	115	1.189E+01	5.277E+00	2.697E+01	2.982E+00	1.189E⊹01	1.075E+01
26055	MUSCLE	123	1.574E+01	4.108E+01	3.833E+02	1.577E-01	1.566E+01	5.063E+00
27060	MUSCLE	128	2.005E+00	5.377E+00	3.827E+01	4.063E-02	1.958E+00	5.9746-01
38090	MUSCLE	125	1.562E-01	2.4608-01	1.5416+00	1.051E-03	1.1778-01	6,3085-82
44106	MUSCLE	88	8.085E~01	4.558E-01-	2.237E+00	3.017E-04	Ũ.	Π.058E-01
45102	MUSCLE	128	9.044E-00	6.6018-02	3.8396-01	1.845E·0?	0.	a.10%66-012
48113	MUSCLE	1	2.635E-01	Ø.	2.635E-01	2.6356-01	2.6356-01	2.635E+01
51125	MUSCLE	128	2.449E-01	2.581E-01	2.096E+00	7.734E-02	3.910E-02	1.970E-01
55137	MUSCLE	128	3.897E-01	7.940E-01	6.779E+00	2.636E-02	3.493E-01	1.955E-01
56133	MUSCLE	104	1.431E-01	1.205E-01	7.631E-01	2.445E-02	1.598E-02	1.004E-01
58144	MUSCLE	4	2.822E-01	1.269E-02	2.975E-01	2.699E-01	Ο.	2.8205-01
63152	MUSCLE	128	7.826E-02	5.899E-02	3.415E-01	2.779E-02	0.	6.329E-02
63155	MUSCLE	128	1.107E-01	7.631E+02	5.212E-01	3.097E-03	1.411E-02	9,242E-02
83207	MUSCLE	128	2,409E+00	2.233E+01	2.5276+02	1,9656-00	2.302E+00	1.350E-01
92235	MUSCLE	122	7.932E-0C	4.7236-02	2.5-61-01	2.271E 001	υ.	6.563E-02
94000	MUSCLE	123	2.477E-01	2.0836+00	2.3066+01	4.800E-04	2.44AE+01	1,2576-02
94238	MUSCLE	64	1.390E~02	2.175E-02	1.140E-01	1.802E-03	5.241E-03	7.679E-03
95241	MUSCLE	128	1.144E-01	8.462E-02	8.023E-01	2.232E-02	2.771E-03	9.298E-02

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*AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO DETECTION LIMIT) PCI/GPAM **AVERAGE (IF NON-DETECTED, CONCENTRATION SET EQUAL TO ZERO) PCI/GRAM

Fig.

Frequency



Fig. 124. Histogram plot of the ¹³⁷Cs concentration in all fish from the entire Atoll.

we have used the average radionuclide concentration, which is conservative and leads to the higher dose estimate.

Elements other than 137 Cs, 60 Co, 90 Sr. $^{238, 239, 240}$ Pu, and 55 Fe were for the most part nondetectable. In such cases, for the purpose of dose estimates. the concentration of the radionuclide was set equal to the detection limit. The average pCi/gram value listed in column 4 in Tables 154-158 was calculated in this manner. Using this approach produces a conservative dose estimate of the contribution from these nuclides because the actual concentration of many of these nuclides may be far below the analyt-Ical detection limit. For example, detection limits for 241 Am established by wet-chemistry analysis of a few samples were found to be significantly lower than those previously established by gamma counting.

Tables 154-158 give an indication of

the isotopes whose concentrations were established by detection limits. The 8th column headed "average" (if nondetected concentration set equal to zero) means that if an element were not detected, the concentration value was then set equal to zero rather than equal to the detection limit. Therefore, if a zero appears in this column, it means that the isotope was not detected in any of the samples analyzed. If a number appears in this column but the concentration value is very low relative to the "average" column (if nondetected concentration set equal to detection limit). that indicates that the isotope was not detected in many of the analyzed samples. If the two columns have equal or approximately equal values. then all or nearly all of the samples analyzed had detectable amounts of the isotope. In any case, by setting the concentration equal to the detection limit for those isotopes which were nondetected





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maximizes the dose contribution from these radionuclides.

Table 159 shows the average concentration of the three main radionuclides found in fish. The number of samples analyzed, as well as the high and low of the range, are given. These values, corrected by a factor of 3.5 to obtain wet weight, were used along with the 600-g/day intake of fish from the predicted diet to make dose estimates. The values for $^{90}\mathrm{Sr}$ in this table deserve special comment. Most of the reef fishes, which make up a large portion of the marine diet, are small and are not easily filleted to separate meat from bone. Therefore, the eviscerated fish were homogenized in a blender to make a uniform sample and then packaged for counting. Homogenizing the whole fish (excluding viscera) includes all the bones of the fish. A significant fraction of the 90Sr in fish is, of course, lodged in the bone moiety. However, the Enewetakese do not eat the bones of the fish and are, in fact, careful to eat the meat from around the bones.

The samples where the muscle was separated from the bone showed a muscle concentration of 90 Sr of 0.075 pCi/g, which is lower by nearly a factor of 3 than that observed in the eviscerated whole fish. Therefore, the dose from 90 Sr has been calculated using the value for fish muscle of 0.075 pCi/g dry weight (or 0.021 pCi/g wet weight).

For reference, data for the ¹³⁷Cs and ⁹⁰Sr content of fish from U.S. diets, from high lakes in Colorado, from around Amchitka Island, and from around Bikini Atoll are shown in Table 160. Cesium concentrations at the Atoll are quite similar or in some cases lower than those observed in other locations, while strontium concentrations are higher in the Atoll than in the U.S. diet.

Dose Code

The doses via the marine and terrestrial food chains were estimated using the following differential equation to describe the intake and retention by man:

Table 159. Radionuclide concentrations in fish (January 1972).

			Concentra	tion, pCi	/g dry weight
Nuclide	Sample	No. of Samples	Average	High	Low
¹³⁷ Cs	All fish ^a	128	0. 39	6.8	0.026
⁶⁰ Co	All fish ^a	128	2.0	38	0.041
90 _{Sr}	All fish ^a	125	0.16	1.5	0.0010
90 _{Sr}	Evisc erat ed whole fish	74	0.21		
⁹⁰ Sr	Fish muscle only	51	0.075		
	only				

^aAll fish includes eviscerated whole fish and those fish where muscle was separated from bone and only the muscle was analyzed.

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		Concentratio mean pCi/g, dr				
Location		¹³⁷ Cs	90 _{Sr}			
Enewetak	1972	0.3	0.08			
Amchitka	1971 ^a	0.1	No data			
Chicago	1971 ^b	0.1	0.003			
Chicago	1972 ^b	0.2	0.003			
Bikini	1968 ^C	~1.0	0.7			
Colorado mountain lakes	1972 ^d	2, 5	No data			

Table 160. Comparison of cesium and strontium data for marine fish muscle.

^aAmchitka Radiobiological Program Progress Report, NVO-269-17, 1972.

^bRadiation and Data Reports 1971, 1972;

Health and Safety Laboratory Quarterly Reports 1971, 1972, 1973.

^cRadiological Report on Bikini Atoll, 1968.

^{1"}Radioecology of Some Natural Organisms and Systems in Colorado, " <u>Eleventh Annual Progress Report to Atomic Energy Commission</u>, Department of Radiology and Radiation Biology, Colorado State University, Fort Collins, Colorado, Rept. COO-1156-63.

$$\frac{dC_{man}}{dt} = \frac{I f_{man}C}{M} - \lambda_{man} C_{man},$$

where $C_{man} = concentration of nuclide in man, pCi/g,$

T

≠ food intake, g/day,

- fman = fraction of nuclide ingested reaching the organ of reference,
- C = concentration of nuclide in food product, pCi/g (i.e., fish, shellfish, coconut, land crab, etc.),
- M = mass of the organ of reference, g, and

$$\lambda_{man} = effective elimination rateof nuclide from man, day-1$$

 $\left(\lambda_{\text{man}} = \lambda_{\text{biological}} + \lambda_{\text{radioactive}}\right).$

The concentration C in the food products is calculated assuming that the nuclide disappears only by radioactive decay, i.e., that no other processes are in operation which reduce the nuclide availability in the food chain. Therefore $C = C_0 e^{-\lambda} r^t$, where C_0 is the concentration observed at the time of the survey and λ_r is the radioactive decay constant. The concentration in man at any time t after initial consumption of the food is:

$$C_{\text{man}} = \frac{I_{\text{man}} C_{\text{man}}}{M(\lambda_{\text{man}} \lambda_{r})} \left(e^{-\lambda_{r} t} - e^{-\lambda_{man} t} \right) pCi/g.$$

The dose at any time t after initial consumption is:

Dose (rem) = KE
$$\int_{0}^{t} C_{man} dt$$

= KE $\int_{0}^{t} \frac{I f_{man} C_{o}}{M (\lambda_{man} - \lambda_{r})} \left(e^{-\lambda_{r}t} - e^{-\lambda_{man}t} \right) dt$

where K is a conversion constant from pCi/g to rem and equals 5.1 × 10⁻⁵

disintegrations . g . rem, and E is the pCi · MeV · day disintegration energy of the nuclide in MeV, including a factor for relative biological effectiveness (RBE). The final dose is then determined from the integration of the equation, i.e.,

Dose =
$$\frac{\text{KE I f}_{\text{man}} C_{o}}{M(\lambda_{\text{man}} - \lambda_{r})} \left[\frac{1 - e^{-\lambda_{r}t}}{\lambda_{r}} - \frac{1 - e^{-\lambda_{man}t}}{\lambda_{man}} \right]^{(0)}$$
rem

Table 161 lists the $f_{\mbox{man}}$ (FMAN), $^{\lambda}_{
m radioactive}$ (LR), $^{\lambda}_{
m man}$ (LMAN), and disintegration energy (E) values for all of the isotopes in the dose calculations. Values for the parameters f_{man} (FMAN) (a dimensionless number) and λ_{man} (LMAN) (in days⁻¹) for the whole body, bone, and kidney are taken from ICRP 4,5 or from more recent literature reports, where such data exist. We are continually searching the literature and updating f and λ values for many isotopes when new information is available. The masses (in grams) used for the whole body and other reference organs are adopted from ICRP values. The disintegration energies, E, (in MeV), are obtained from either ICRP 4, 5 or the work of the MIRD committee 6 . The radioactive decay constants λ_r (LR) (in days ⁻¹) are calculated from isotope half-life data in the Table of Isotopes⁷.

The intake term (I) represents the average daily consumption of various dietary components. The average diet is the result of input from Jack Tobin of the Trust Territories, discussions with Dr. Mary Murai of the University of California, Berkeley and reports which she has published⁸, and direct interview and observation of the Enewetak people in their present locations (see reports by Marsh and Nelson included in the chapter on Enewetak).

Dose Estimates for the Marine Pathway

The radionuclide concentration, C_0 , is the average value for all fish from the entire Atoll determined from our survey and is listed in Tables 158 and 159 for each nuclide. The average values for radionuclide concentrations listed in the tables are in pCi/g dry weight. The data are corrected to pCi/g wet weight for use in the dose code by dividing by 3.5, the average wet-to-dry ratio for fish from the Atoll.

Integral doses calculated from the marine survey data are listed in Table 162 for the whole body and bone for 5, 10, 30. and 70 yr. The major contribution to the whole-body dose comes from 137 Cs and 60 Cc, while the bone dose comes from 90 Sr, as well as 137 Cs and 60 Co. The fourth line of the table gives the summation of the dose to each organ from the three isotopes. The bottom entry in the table lists the dose from all radionuclides which are listed in the Table 154 footnote. It is clear that almost all of the dose is contributed by 137 Cs, 60 Co, and 90 Sr. For example, the 30-yr integral wholebody dose is 47 mrem from 137 Cs and ⁶⁰Co, and only 6 mrem additional wholebody dose is contributed by other radionuclides. For bone, the total dose from all radionuclides is 840 mrem, with 94% contributed by 90 Sr, and 6% by all other nuclides.

In addition to the isotopes listed in Table 158, dose estimates for 14 C and 129 I were made and included in the summary of the marine pathway. Neither 14 C nor 129 I were detected in any of the samples, but doses were calculated on the assumption that the concentration equaled the detection limit. The $^{30-yr}$ Ē

Table 161.The disintegration energy E and the radioactive half life LR are listed for each
radionuclide.The effective biological half time LMan and the fraction of
ingested isotope reaching the organ of reference FMan are listed for three
receptor organs, bone, liver, and whole body.

1			BONE MASS=	5.000E+03	LIVER MASS=	1.800E+03	WHOLEBODY (1ASS= ?.000E+04
NUCL IDE	E	LR	-LMAN-	-FLIAN-	-LMAN-	-FNAN-	-LMAN-	-FMAN-
3 H 14 C		1.549E-04 3.314E-07	5.790E-02 1.733E-02	9.100E-02 2.500E-02	5.790E-02 6.930E-02	2.600E-02 2.600E-02	5.7908-02 6.9308-02	1 . 000E +00 1 . 000E +00
55FE 60CD 63N I	6.540E-03 7 8.740E-01 3	7.032E-04 3.609E-04 2.064E-05	1.116E-03 2.924E-02 8.869E-04	1.000E-02 2.000E-02 1.500E-01	1.954E-03 8.191E-03 1.407E-03	1.300E-02 8.310E-02 2.000E-02	1.569E-03 8.191E-03 1.060E-03	1.000E-01 3.000E-01 3.000E-01
90SR 106RU 102RH	5.500E+00 6 1.400E+00 1	5.781E-05 1.899E-03 5.544E-04	1.987E-03 3.439E-03 4.240E-02	3.000E-01 3.300E-02 1.000E-02	1.156E-01 1.180E-02 3.873E-02	7.800E-03 6.300E-02 8.000E-03	1.211E-04 7.223E-03 6.729E-02	3.000E-01 3.000E-01 2.000E-01
113CD 12558 129 I	3.600E-01 7	1.356E-04 7.032E-04 1.187E-10	5.911E-03 7.633E-03 4.950E-02	9.000E-05 3.000E-03 7.000E-02	3.601E-03 1.894E-02 9.900E-02	1.900E-03 6.000E-05 1.200E-01	1.375E-04 1.894E-02 5.022E-03	5.900E-02 3.000E-02 1.000E+00
133BA 137CS 144CE	5.900E-01 6	2.637E-04 5.329E-05 2.432E-03	1.093E-02 6.363E-03 2.894E-03	3.500E-02 9.100E-02 3.000E-05	9.745E-04 6.363E-03 4.797E-03	3.000E-05 2.600E-02 2.500E-05	1.093E-02 7.142E-03 3.662E-03	5.000E-02 1.000E+00 1.000E-04
147PM 1515M 152EU	1.523E-02 2 6.600E-01 1	7.032E-04 2.110E-05 1.531E-04	1.165E-03 4.831E-04 3.379E-04	3.500E-05 3.500E-05 3.600E-05	1.760E-03 3.727E-03 5.610E-03	6.000E-06 3.500E-05 2.500E-05	1.760E-03 1.077E-03 3.379E-04	1.000E-04 1.000E-04 1.000E-04
155EU 207BI 235 U	1.000E+00 6 4.600E+00 2	1.055E-03 5.329E-05 2.662E-12	1.240E-03 5.217E-02 8.030E-03	3.600E-05 3.000E-04 5.400E-05	6.511E-03 4.626E-02 1.899E-06	2.500E-05 1.500E-03 1.000E-02	1.240E-03 1.387E-01 8.030E-03	1.000E-04 1.000E-02 1.000E-04
238PU 239PU 240PU 241AM	5.300E+01 7 5.300E+01 2	2.134E-05 7.794E-08 2.809E-07 4.145E-06	4.032E-05 1.906E-05 1.927E-05 2.313E-05	1.350E-05 1.350E-05 1.350E-05 4.500E-05	2.323E-05 1.977E-06 2.180E-06 5.161E-05	1.200E-05 1.200E-05 1.200E-05 4.500E-05	3.083E-05 9.571E-06 9.774E-06 2.313E-05	3.000E-05 3.000E-05 3.000E-05 1.000E-05

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Table 162.	Integral dose	for 5, 10	, 3 0 ,	and 70 yr from	the marine food chain.
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	, ·	Integral dose, rem ^b								
	5 y	5 yr		10 yr		30 yr		70 yr		
Nuclide	W. B.	Bone	W. B.	Bone	W.B.	Bone	W.B.	Bone		
137 Cs	0.0061	0.0061	0.012	0.012	0.030	0.030	0.049	0.049		
⁶⁰ Co	0.0078	0.0078	0.012	0.012	0.017	0.017	0.017	0.017		
$90_{ m Sr}$		0.13		0.31		0.77		1.3		
Sum	0.014	0.14	0.024	0.33	0.047	0.82	0.066	1.4		
All nuclides	^C 0.016	0.14	0.028	0.34	0.053	0.84	0.089	1.6		

^aThe dose is based upon the average concentration for fish from the entire Atoll and upon a dietary fish intake of 600 g/day. These doses apply to all six living patterns.

^bThe concentration data were corrected to January 1974, the earliest possible return date to the Atoll; all integral doses are calculated for periods which begin on January 1974.

^CIsotopes included in the "All nuclides" calculation:

³ н	60 _{Co}	102 _{Rh}	137 Cs	$152_{\rm Eu}$	$^{235}\mathrm{U}$
¹⁴ C	⁹⁰ Sr	113 Cd	^{133}Ba	¹⁵⁵ Eu	238 _{Pu}
⁵⁵ Fe	¹⁰⁶ Ru	125 Sb	144 Ce	$^{207}\mathrm{Bi}$	²³⁹ Pu
					²⁴¹ Am

integral dose for ¹⁴C, calculated in this however, there is very good reason to believe that the actual concentration is orders of magnitude below the detection

limit reported here. Therefore, neither isotope is significant in the total dose assessment via the marine pathway.

Re 1. R. Lo Na 2. F. Siı Pr 3. A. R٤ 4. IC Ra 5. IC B (F 6. M (F

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issued by the Pacific Science Board, National Academy of Sciences-National Research Council. Evaluation of the Dosage from Terrestrial Foods

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Introduction

This chapter describes the analysis of the biota data for evaluation of the potential dose from ingestion of terrestrial foods. Because some of the items expected in the diet were not available in abundance for adequate sampling, it was necessary to examine not only data from the edible species but also those from other constituents of the ecosystem, i.e., soil, indicator plants, and indicator animals. It was necessary to take into account radiological data reported for other locations, to examine collateral information from a variety of sources, and to use all of these data to derive quantitative relationships for predicting concentrations of radionuclides in food items. Assessment of the dosage from terrestrial foods is based on the diets and living patterns discussed earlier in this section (Tables 1,4).

Sampling of Terrestrial Biota

The chapter on the terrestrial biota survey presents a detailed description of the terrestrial biota sampling program. Edible plants or animals collected include coconut, pandanus, tacca, various species of birds, bird eggs, and coconut crab. Indicator species include <u>Messerschmidia</u>, Scaevola, rice rat, and roof rat.

Table 163 lists the islands from which edible species were collected. Edible species were collected wherever they could be found but, in contrast to Messerschmidia and Scaevola, they were not available for sampling on each isited. Coconut was sampled on 16 islands distributed about the Atoll, but pandanus fruit was obtained only from BELLE and KEITH, and tacca root only from DAVID. Pandanus leaves were collected from BELLE, KEITH and eight additional islands. No breadfruit was found on the Atoll. Coconuts collected by Ken Marsh in July 1973 were the source of the coconut milk and a portion of the coconut from IRENE and MARY.

Birds were collected on 18 islands and eggs from eight islands distributed throughout the Atoll. Collection of coconut crabs was confined to islands in the south (BRUCE, GLENN, JAMES, KEITH and LEROY). Hermit crabs were collected on IRENE and on the southern islands, DAVID, REX, GLENN, HENRY and IRWIN, but they are not part of the diet. Rice rats and roof rats were collected on nine islank in the north and the south, including JANET and YVONNE. Rats are not part of the diet, but they provide useful data for assessment of the dose via poultry and livestock.

<u>Distribution of Radionuclides in</u> <u>Terrestrial Foods</u>

<u>Coconut</u> — Coconut is the edible plant for which sampling was most extensive. Table 164 lists the concentrations of radionuclides in dry coconut meat. Figure 126 is a graph of the distributions of ⁹⁰Sr and ¹³⁷Cs by island, and Fig. 127 is a graph of the distributions of the other nuclides that were above detectable limits. The term "island number" as used in Table 163 and in the figures refers to the practice of assigning consecutive numbers to each island as one proceeds around the Atoll, beginning with ALICE.

Table It Island No. Is 1. AI 2. Bl 4. D. 9. IR 10. JF 12. LI 14. M 15. N₁ 16. OI 17. PI 19. SF 20. TI 21. UI 22. VI 24. Y 29. V. 30. Al 31. BI 32. Cl 33. D. 34. RI 35. El 37. Fl 38. Gl 39. HI 40. IR 41. J/ 42. KI 43. L] ^aPandar ^bRats ar

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Tabaa										
Island No.	Island	Coconut meat	Coconut milk	Pandanus fruit	Pandanus leaves ^a	Tacca corm	Birds		Coconut c ra b	Rat ^b
and the second se	ALICE						x			
	BELLE			х	х					
4.	DAISY	х	х							
9.	IRENE	х	х				х	x		
10.	JANET	Х	Х		х		х	х		x
12.	LUCY						x			
14.	MARY	x	х				х			
15.	NANCY	x	х							
16.	OLIVE						х			
17.	PEARL						Х			x
19.	SALLY				х		х	х		x
20.	TILDA				х					
21.	URSULA	L								х
22.	VERA	x			х					
24.	YVONNE	E x					х	х		х
29.	VAN						х			
30.	ALVIN						х			
31.	BRUCE	х					х		х	х
32.	CLYDE						x	х		х
33.	DAVID	х	х		х	х	х			x
34.	REX						х	x		
35.	ELMER	х			x					x
37.	FRED	х			х					
38.	GLENN	х							х	x
39.	HENRY	x						x		
40.	IRWIN	х					х	х		
41.	JAMES								х	
42.	KEITH	x		х	x		x		х	
43.	LEROY	x			x		х		x	
-										

Table 163. Terrestrial biota survey. Edible plants and edible animals sampled.

^aPandanus leaves are not eaten but serve as indicators for pandanus fruit.

^bRats are not eaten but serve as indicators for poultry and swine.

		,			on, pCi/g di		
lsland No.	Island	³ H	⁵⁵ Fe	⁶⁰ Co	90 _{Sr}	137Cs	239, 240 _p
4	DAISY ^a	0.415		<0.059	0.200	7.17	
9	IRENE			<0.067	0.067	1.77	0.0362
			86.5	<1.70	1.61	5.11	<0.034
10	JANET	0.343		<0.069	0.207	84.7	
14	MARY		1.18	<0.055	0.136	14.3	<0.0005
			76.6	<0.017	14.1	5.58	<0.43
15	NANCY	0.333	1.95	<0.054	0.167	18.8	<0.0006
22	VERA			<0.053	0.134	9.30	0.00013
24	YVONNE	0.664		0.077	0.011	3.96	
			<0.19	<0.066	<0.054	1,99	<0.0020
31	BRUCE			<0.014		0.582	
33	DAVID	0.313		<0.060	0.014	2.59	0.0027
				<0.012	0.026	0.399	0.0034
35	ELMER	0.305	<0.63	<0.028	<0.075	3.45	<0.0052
				<0.068	0.032	2.14	0.00044
37	FRED	0.390		<0.020	0.030	2.39	
		0.302	<0.35	<0.021	0.367	0.530	<0.0058
38	GLENN		<0.27	<0.053	<0.049	1.30	<0.0013
	,			<0.029	0.020	1.01	<0.0025
39	HENRY ^b		<0.11	<0.007	<0.028	0.565	<0.0010
40	IRWIN		<0.64	0.074	<0.086	0.2~9	<0.0027
42	KEITH		<0.29	<0.064	<0.056	0.952	<0.0009
43	LEROY			<0.015	0.189	3.90	0.00073

Table 164. Radionuclides in coconut meat.

^aA concentration of 0.065 pCi 207 Bi/g was measured in the sample from DAISY. ^bA concentration of 0.098 pCi 155 Eu/g was measured in the sample from HENRY.

<u>Coconut Milk</u> — Table 165 is a comparison of the radionuclide content in fresh coconut meat and coconut milk collected from the same island. All of the milk samples represented were obtained from coconuts collected by Ken Marsh in July 1973. In Table 165 the meat samples of the bracketed pair of meat and milk samples from IRENE and MARY were also collected by Ken Marsh. Since most of

the milk was obtained from green nuts and most of the meat from ripe nuts, the bracketed meat and milk samples from IRENE and MARY are not representative of single pooled samples of coconuts.

<u>Pandanus</u> — Table 166 lists the concentrations of the radionuclides detected in fruit and leaves of pandanus. The fruit and leaves listed for BELLE and for <u>Tac</u> nuclid∉ **tacca** a

Fig. 1:

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Fig. 1

KEITH plant. islandcentrat

Concentration - pCi/g, dry

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Fig. 127. Concentrations of radionuclides in coconut meat.

KEITH are representative of the same plant. Figure 128 is a graph showing the island-by-island distribution of these concentrations.

<u>Tacca</u> — The concentrations of radionuclides detected in the one sample of tacca are listed in Table 166 and shown in Fig. 128.

<u>Birds</u> — The "edible" birds are considered to be those species that were found and collected in greatest numbers; these include the common noddy, the white-capped noddy, and the sooty tern. Table 167 lists the concentrations of radionuclides detected in muscle and liver of these species. A fourth group of pooled terns assembled from common noddies, white-capped noddies, and sooty terns is also represented in Table 167.

Figure 129 shows the distributions of 55 Fe and 60 Co in muscle and liver by island, and Fig. 130 shows the distributions of 90 Sr, 137 Cs, and 239,240 Pu in these tissues by island.

<u>Bird Eggs</u> — Table 168 presents the radionuclide concentrations detected in bird (common noddy or sooty tern) eggs, and Fig. 131 shows the distribution of the radionuclide concentrations by island.

<u>Coconut Crabs</u> — Table 169 lists the concentrations of the radionuclides detected in muscle and hepatopancreas of coconut crabs. Figure 132 is a graph of the distributions of 60 Co, 90 Sr, and 137 Cs in these tissues by island.

				(Concentratio	on, pCi/g v	vet	
Island <u>No.</u>	Island	Plant part	⁵⁵ Fe	⁶⁰ Co	90 _{Sr}	137 _{Cs}	239,240 _{Pu}	$\left(\frac{\text{Drv wt}}{\text{Wet wt}}\right)^{4}$
4	DAISY	Meat		<0.029	0.100	3.58		0.50
		Milk	<0.30	<0.051	0.068	0.084	<0.0016	0.048
9	IRENE	Meat		<0.034	0.033	0.885	0.0181	0.50
		Meat	5.60	<0.11	0.104	0.331	<0.0022	0.0656
		Milk 🖌	<2.7	<0.15	<0.077		<0.0086	0.046
10	JANE T	Meat		0.035	0.103	42.3		0.50
		Milk	<0.12	<0.030	0.084	11.2	0.0005	0.053
14	MARY	Meat	0.590	<0.027	0.068	7.14	<0.0003	0.50
		Meat	42,2	<0.009	7.79	3.07	<0.24	0.55
		Milk	<0.35	<0.016	0.042	4.52	<0.0046	0.067
15	NANCY	Meat	0,975	<0.027	0.084	9.42	<0.0003	0.50
		Milk	0,266	<0.060	0.051	6.65	<0.0010	0,045
33	DAVID	Meat		<0.030	0.0069	1.30	0.0014	0.50
		Meat		<0.0059	0.013	0.199	0.0017	0.50
		Milk	<0.13	<0.012	<0.023	1.09	<0.0015	0.047

Table 165. Radionuclides in meat and milk of coconut.

^aWhere wet and dry weights were not determined, the dry-wt/wet-wt ratio of coconut meat was assumed to be 0.50^{4,6}. ^bThis coconut sample was green and hence yielded little meat.

Statistical Analysis of Terrestrial Biota Data

Statistical Correlations Between Plants and Soil - Soil is both a logical and convenient starting point for predicting radionuclide concentrations in terrestrial foods. First, it is the source compartment from which all the terrestrial food chains derive their radioactivity. Second, it was subjected to extensive sampling and analysis on each island of the Atoll.

The uptake of radionuclides from soil to plants can be described quantitatively in terms of the concentration factor, defined in this discussion as

pCi/g dry plant ÷ pCi/g dry soil. Table 170 summarizes the concentration factors of ¹³⁷Cs and ⁹⁰Sr determined for edible and indicator plants. These two nuclides have been singled out because they were consistently detected and measured in terrestrial vegetation and they contribute most to the dosage from ingestion of terrestrial foods. The concentration factors for both ¹³⁷Cs and ⁹⁰Sr are seen to be widely distributed, with ranges varying by a factor of 100 or more. This is not really surprising since the pairing of plant and soil data for the same location is inherently lacking in precision and since soil is a complex substrate that does not exhibit

Table 1

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				C	oncentrati	on, pCi/g	dry ^a	
Island No.	Island	Plant type	³ н	⁵⁵ Fe	⁶⁰ Co	⁹⁰ Sr	¹³⁷ Cs	239, 240 _{Pu}
2	BELLE	Pandanus fruit	0.859		0.140	206	923	0.00343
		Pandanus leaves		0.438	<0.14	391	679	<0.24
10	JANET	Pandanus leav es		2.32	<0.12	4.41	0.620	0.00204
19	SALLY	Pandanus leaves		0.703	<0.11	1.97	15.0	0.0149
20	TILDA	Pandanus leaves		2.94	<0.12	1 5. 5	152	0.00698
22	VERA	P a ndanus leaves			<0.069	4.64	17.6	0.00757
3 3	DAVID	Pandanus leaves		0.127	<0.11	3.56	15.9	0.00132
		Tacca co r m	0.516	<1.31	<0.09	0.096	8.96	0.00114
35	ELMER	Pandanus leaves		0.416	<0.034	25.1	3.09	0.00 195
37	FRED	Pandanus leaves		0.851	<0.066	0.422	4.29	0.00770
42	KEITH	Pandanus fruit	1,99	12.2	<0.10	13.1	0.860	
		Pandanus leaves		0.356	<0.027	(lost)	0.569	0.00447
43	LEROY	Pandanus le aves		0.210	<0.074	1.69	9.14	0.00222

Table 166. Radionuclides in pandanus and tacca.

^aAdditional nuclides measured at levels above detection limits: (1) ¹²⁵Sb, 3.01 pCi/g in pandanus fruit from BELLE; (2) ¹⁰²Rh, 0.114 pCi/g in tacca corm (DAVID);
(3) ¹⁴⁴Ce, 0.724 pCi/g in pandanus leaves from KEITH and 0.469 pCi/g in pandanus leaves from LEROY; (4) ²⁰⁷Bi, 0.043 pCi/g in pandanus leaves from KEITH and 0.108 pCi/g in pandanus leaves from JANET.

uniform properties at any given location. Linear regression analysis 1,2 was therefore carried out to identify correlations between 137 Cs and 90 Sr concentrations in plants and those in soil that would be useful for predictive purposes.

Concentrations of 90 Sr and 137 Cs in the 0-15 cm profile samples reported in the chapter on the terrestrial soil and radiation survey were used to represent soil. <u>Messerschmidia</u> and <u>Scaevola</u>, the the dominant and most widely disseminated and extensively collected plant species, were chosen as indicator plants. The following linear regression analyses were performed to determine regressions of ⁹⁰Sr and ¹³⁷Cs in plants on those in soil: (1) coconut meat on soil, (2) pandanus



Fig. 128. Concentrations of radionuclides in pandanus and tacca.

leaves on soil, (3) <u>Messerschmidia</u> on soil, and (4) <u>Scaevola</u> on soil. The concentrations of 90 Sr and 137 Cs associated with soil locations near a given plant location were determined using the overlay figures of Appendix II. The mean of the concentrations in soil at these locations was used as the independent variable in the regression analysis.

<u>Coconut vs Soil</u> — Figure 133 shows the 137 Cs concentrations in coconut meat as a function of those in soil. Linear regression analysis¹ of the log-transformed



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				Concentration, pCi/g dry								
Island		Sample	55	Fe	60	Co	90	Sr	13'	⁷ Cs	239,2	240 _{Pu}
No.	Island	type	Muscle	Liver	Muscle	Liver	Muscle	Liver	Muscle	Liver	Muscle	Liver
1	ALICE	Common noddy	49.6	127	0.321	<0.165	0.0099	<0.0099	<0.072	<0.094		0.0622
9	IRENE	n n		49.6	0.247	0.324		0.509	<0.099	<0.057		0.0091
10	JANET	11 11	105	258	0.507	<0.165	0.0078	0.0667	<0.069	<0.097	0,0018	·0,033
	JANET	Pooled terns	59,5	172	0.159	<0.514	0.0047	0.0050	0.0621	<0.406	0,0055	0.0015
12	LUCY	Common noddy	8.78	199	<0.216	<0.398	0.0046	0.187	0,192	<0.246	0.0228	<0.073
14	MARY	11 fi	14.2	251	0.316	0.568	0.482	<0,019	0.143	<0.093	0.0022	<0.012
16	OLIVE	<u>t1</u> II	92.8	232	<0.151	<0.195	0.0016	0.0362	<0.092	<0.107	0.0919	0.0105
17	PEARL	Pooled terns		317	0.659	0.647		<0.049	<0.142	<0.127		0.0041
19	SALLY	White-capped noddy	110		0.214	0.235	0.0135	0.0283	<0.087	<0.055	0.0196	0.0110
	SALLY	Sooty tern	36.6	155	<0.114	<0.120	0.0064	0.0344	<0.081	<0.069	0.0054	<0.079
24	YVONNE	Common noddý	22.6	386	0.230	0.369	0.0073	<.0.011	0.0.086	<0.075	0.0201	0.0111
29	VAN	22 27	99.6	279	0.283	0.195	0.0034	0.0403	×0.076	~ 0 ,060	0.0014	0.0025
30	ALVIN	Pooled terns			<0.167	<0.187			0.128	<0.131		
31	BRUCE	White-capped noddy	41.3	327	0.392	<0.253	<0.0080	0. 0326	<0.079	<0.134	0.0069	0.0222
32	CLYDE	Sooty tern	20.4	146	<0.091	<0.549	0.0064	0.0149	<0.065	<0.054	<0.0015	0.0017
33	DAVID	11 11	59.0	153	<0.108	<0.345		0.0545	<0.082	<0,230	0.119	0.0420
34	REX	Common noddy	43.5	118	0,177	<0,161	0,0065	<0.0091	<0.076	<0.091	0.0056	0.0121
40	IRWIN	11 11	169	423	0.609	0.635	0.0085	0.233	0.306	~ 0. 161	0.0434	0.0242
42	KEITH	11 11			0.452	0.689	<0.0041		<0,089	<0.129	0.0010	
43	LEROY	White-capped noddy	64.4	811	2.07	2,83	<0.112	0.402	<0.134	<0.242	0.0033	0.0072

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						ration, pCi	/g d r y	
land No.	Island	Specie	s	⁵⁵ Fe	⁶⁰ Co	⁹⁰ Sr	137Cs	^{239,240} Pu
9	IRENE	Common	noddy	23.0	<0.075	0.095	<0.051	0.0015
10	JANET	11	11	57.2	<0.10	0.203	0.119	0.0148
19	SALLY	Sooty ter	n	37.6	<0.073	0.0043	<0.052	0.0154
24	YVONNE	Common	noddy	<0,59	<0.10	1.06	<0.069	0.00045
		11		56.8	<0.11	<0.0020	<0.079	0.0232
		11	11	54.5	<0.087	0.073	0 .07 9	0.00068
29	V AN	11	11	63.5	<0.10	0.0022	0.136	0.00047
32	CLYDE	Sooty ter	n		<0.82		<0.057	
34	REX	Common	n oddy	51.4	<0.10	0.0066	<0.070	0.00077
39	HENRY	11	11	54.1	0.048	0.0025	<0.015	0.00047
40	IRWIN	11		5.14	<0.12	<0.011	<0.083	0.00088

Table 168. Radionuclides in bird eggs.

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Table 169. Radionuclides in edible parts of coconut crabs.

				Conce	entration	, pCi/g	dry
Island No.	Islan d	Tissue	³ H	⁶⁰ Co	90 _{Sr}	¹³⁷ Cs	152 _{Eu} 2 39,240 Pu
31	BRUCE	Muscle Hepatopancreas	0.424 0.157	0.198 0.402	0.185 0.133	1.98 0.420	0.0012 0.0023
38	GLENN	Muscle Hepatopancreas	0.685 0.266	0.247 0.276	0.269	1.88 0.545	0.0013 <0.0066
41	JAMES	Muscle Hepatopancreas		1.05 1.56	0.079 0.0014	$1.25 \\ 0.317$	0.00076 0.0019
42	KEITH	Muscle Hepatopancreas		0.420 1.03	1.19 0.401	1.92 0.496	0.0014 0.066 0.0098
43	LEROY	Muscle Hepatopancreas	0.867 0.207	1.23 1.46	1.58 2.58	12.6 3.29	0.0031 0.0038

data reveals a significant correlation (P<0.001) between the logarithm of the 137 Cs concentration in coconut and the logarithm of the 137 Cs concentration in soil (see Table 171C). Accordingly, the mean concentration of 137 Cs in coconut

can be predicted from that in soil. Figure 134 shows the 90Sr concentrations in coconut meat as a function of those in soil. A similar analysis (see Table 172C) reveals a significant (P = 0.05) correlation between the logarithm of the 90Sr

		(pCi/g	Concent: dry plan		factors Ci/g dry so	oil)		_
		Cesium-	137			rontium	n-90	
Plant type	No. of samples	Min	Median	Max	No. of samples	Min	Median M	ax
Messerschmidia	47	0.051	5.4	270	42	0 .0 31	1.2 13	
Scaevola	45	0.059	4.7	120	39	0.023	0.74 14	
Pooled <u>Messerschm</u> and <u>Scaevola</u>	idia 92	0.051	5.2	270	81	0.023	0. 96 14	
Coconut meat	21	0.22	1.3	16	16	0.0011	0.023 1	.0
Pandanus leaves	10	0.072	4.7	42	9	0.18	1.0 57	
Pandanus fruit	2	1.3		21	2	2.5	7	.7
Tacca corm	1		16		1		0.21	

Soil-to-plant uptake of 137 Cs and 90 Sr. Table 170.

concentration in coconut and the logarithm of the ⁹⁰Sr concentration in soil. The mean ⁹⁰Sr concentration in coconut can therefore be predicted from that in soil.

Pandanus Leaves vs Soil - Figure 135 shows the concentrations of ¹³⁷Cs in pandanus leaves as a function of those in soil, and Fig. 136 shows comparable data for ⁹⁰Sr. Both the ¹³⁷Cs data and the ⁹⁰Sr data scatter widely and are relatively few. Linear regression analysis (Tables 171 and 172) reveals that the correlations of 137 Cs and 90 Sr between pandanus leaves and soil are not statistically significant.

Messerschmidia, Scaevola and Pandanus Leaves vs Soil - Figure 137 presents ¹³⁷Cs concentrations and Fig. 138. ⁹⁰Sr concentrations in Messerschmidia and Scaevola as a function of those in soil. The data points are seen to scatter widely about the regression line, but they are far more numerous than

those obtained from the edible plants. Statistical analysis reveals for both nuclides and both indicator species significant positive correlation between the



¹³⁷Cs in pandanus leaves and ¹³⁷Cs in soil.

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Table

Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y}{A} = A$	<u>+ b ln X</u> b
		A. <u>Messersch</u>	midia and Scaevo	bla	
lesserschmidia	48	0.79	<0.001	1.86	0.901
Scaevola	46	0.76	<0.001	1.52	0.823
Pooled	94	0.78	<0.001	1.69 ^b	0.864 ^a
Pandanus leaves	10	andanus leaves, M 0.52	N.S. ^g		0.682
-	48	0.79	<0.001	1.86	0.901
	46	0.76	<0.001	1.52	0.823
	104	0.76	<0.001	1.69 ^d	0.851 ^C
Pandanus leaves Messerschmidia Scaevola	48 46	0.79 0.76	<0.001 <0.001	1.52	

fable 171. Statistical correlation between 137 Cs in plants and 137 Cs in soil.

	C. Coconut	meat, pandanu	s leaves, Messers	chmidia_ and Se	caevola
Coconut meat	22	0.0	69 <0.0	01 0.847	0.620
Pandanus leave	es 10	0.	52 N.S	. ^g 1.75	0.682
Messerschmid	<u>ia</u> 48	0.	79 <0.00	01 1.86	0.901
<u>Scaevola</u>	46	0.7	76 <0.00	01 1.52	0.823
Pooled	126	0.7	74 <0.00	01 1.53 ^f	0.824 ^e

^aAn F-test for equality of the regression slopes gives F = 0.28, P = 0.597. ^bAn F-test for equality of the regression intercepts gives F = 1.50, P = 0.225. ^cAn F-test for equality of the regression slopes gives F = 0.31, P = 0.734.

^dAn F-test for equality of the regression intercepts gives F = 0.72, P = 0.489.

^eAn F-test for equality of the regression slopes gives F = 0.57, P = 0.633.

^f An F-test for equality of the regression intercepts gives F = 3.32, P = 0.22.

_Reject null hypothesis.

^gNot significant.

logarithm of the concentration in plant and the logarithm of the concentration in soil. Comparisons of the regression lines by statistical methods described in Ref. 2 support the assumption that the individual regression curves do not differ significantly and that the ¹³⁷Cs and ⁹⁰Sr data from <u>Messerschmidia</u> and <u>Scaevola</u> can each be combined and represented by a common pooled regression line (Tables 171A and 172A).

Similar analyses of the three individual plant-soil correlations for 137 Cs and 90 Sr in pandanus leaves, <u>Messerschmidia</u>, and <u>Scaevola</u> also reveal that in each case the individual correlations are statistically

Table 172. Statis	tical correlat	ion between ⁹⁰ Sr i	n plants and ⁹⁰ Sr	in soil.			13
Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y}{A} = A$	+ <u>b ln N</u> b	•	11)
		A. Messersch	midia and Scaevo	ola		•	da al
Messerschmidia	42	0.83	<0.001	0.438	0.868	4	ar
Scaevola	39	0.81	<0.001	-0.0451	0.868		re
Pooled	81	0.81	<0.001	0.208 ^b	0.866 ^a		nı
							ct fc
	В. <u>М</u>	esserschmidia, S	caevola, and pan	danus leave	S	į	10
Messerschmidia	42	0.83	<0.001	0.438	0.868		to
Scaevola	39	0.81	<0,001	-0.0451	0.866		ti
Pandanus leaves	9	0.49	N. S. ^C	1.05	0.537	•	V
Pooled	90	0.79	<0.001	0.299 ^e	0.836 ^d		ł
							3
							(
	C. <u>Messersch</u>	midia, Scaevola,	pandanus leaves	, and coconi	ut meat]
Messerschmidia	42	0,83	<0,001	0.438	0.868	•	(
Scaevola	39	0.81	<0,001	-0,0451	0.866		
Pandanus leaves	9	0.49	N.S. ^c	1,05	0.537		
Coconut meat	16	0.50	0.05	-3.01	0.482		
Pooled	106	0.62	<0.001	-0.244 ^g		•	

^aAn F-test for equality of the regression slopes gives F = 0.00, P = 0.989. ^bAn F-test for equality of the regression intercepts gives F = 3.26, P = 0.075. ^CNot significant.

^dAn F-test for equality of the regression slopes gives F = 0.80, P = 0.455. ^eAn F-test for equality of the regression intercepts gives F = 1.76, P = 0.179. ^fAn F-test for equality of the regression slopes gives F = 1.64, P = 0.185.

^gAn F-test for equality of the regression intercepts gives F = 39.41, P = 0.000.

Reject null hypothesis.

indistinguishable from the pooled correlation (Tables 171B and 172B). These results provide justification for using the pooled plant-soil regression curve for Messerschmidia, Scaevola, and pandanus leaves to predict ¹³⁷Cs and ⁹⁰Sr in pandanus leaves from the respective concentrations in soil. Analysis involving the comparison of the individual regression

lines for <u>Messerschmidia</u>, <u>Scaevola</u>, pandanus leaves, and coconut meat yielded common results for 137 Cs and 90 Sr. The individual slopes are statistically indistinguishable (Table 171C), but the null hypothesis that the four intercepts are equal has to be rejected. Thus the pooled plant-soil regression curve cannot be used to predict

r

137Cs and 90Sr in account from those in soil.

It must be remembered that the pandanus samples are few in number. Thus. although the pandanus-vs-soil correlations are not statistically significant, it is reasonable to expect that with a larger number of samples, this would not be the case. Use of the pooled regression line for predicting $\frac{137}{Cs}$ and $\frac{90}{Sr}$ in pandanus leaves is a prudent procedure that leads to prediction of somewhat higher values than those using the individual pandanus vs soil regression lines. Use of the pooled regression line implies that with a larger number of pandanus samples, the data would tend to fall above the regression lines of Figs. 135 and 136 and the slopes of the regression lines would increase. The close correspondence of the median concentration factors of 137 Cs and 90 Sr

in <u>Messerschmidia</u>, <u>Scaevola</u>, and pandanus leaves (see Table 170) provides additional justification for using the pooled regression.

Statistical Correlations Between Edible Plants and Indicator Plants — Indicator plants can also be used as the starting point for predicting radionuclide concentrations in food items. Linear regression analysis^{1,2} was performed to determine regressions of ¹³⁷Cs and ⁹⁰Sr in coconut meat on those in indicator plants. It was not possible to determine regressions of pandanus leaves on other plants because other plant species were not commonly sampled at pandanus sampling sites.

Figures 139 and 140 show respectively, 137 Cs and 90 Sr concentrations in coconut

meat as a function of those in Messerschmidia and Scaevola. Linear regression analysis 1,2 reveals significant correlations (P < 0.001) between the logarithms of the ¹³⁷Cs in coconut and those in Messerschmidia and Scaevola. Also the individual regression lines are statistically indistinguishable (see Table 173). In the case of ⁹⁰Sr, the analysis reveals a significant correlation (P = 0.05) between the logarithms of the concentration in coconut and those in Scaevola, but the correlation between the logarithms of the concentrations in coconut and those in Messerschmidia is not significant. The individual regression lines, however, are statistically indistinguishable (see Table 174). Thus, these results indicate that the concentrations of 137 Cs and 90 Sr in Messerschmidia









Table

mulcat	or plants.				
Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y = A}{A}$	+ b ln X b
Messerschmidia	19	0.80	<0.001	0.060	0.652
Scaevola	19	0.70	<0.001	0.404	0.575
Pooled	38	0.75	<0.001	0.237 ^b	0.612 ^a

Table 173. Statistical correlation between 137 Cs in coconut meat and 137 Cs in indicator plants.

^aAn F-test for equality of the regression slopes gives F = 0.17, P = 0.680.

 ^{b}An F-test for equality of the regression intercepts gives F = 0.63, P = 0.433.

and <u>Scaevola</u> may be used for prediction of the concentrations of these nuclides in coconuts growing at the same locations.





Statistical Correlations Between Rat

<u>Tissues and Indicator Plants</u> — Rats were the only mammals found on the Atoll. Previous studies by Fall, Medina, and Jackson³ indicate that although the indigenous rats of Enewetak are omnivorous, plant foods predominated in the diet. The dominant species Messerschmidia



Fig. 139. Statistical correlation between ¹³⁷Cs in coconut meat and ¹³⁷Cs in <u>Messerschmidia</u> and <u>Scaevola</u>.

Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y = A + b \ln X}{A + b}$
Messerschmidia	14	0.39	N.S. ^a	-2.91 0.438
Scaevola	14	0.54	0.05	-2.85 0 548
Pooled	28	0.46	0.02	-2.89° 0.492 ^b

Table 174. Statistical correlation between 90 Sr in coconut meat and 90 Sr in indicator plants

^aNot significant.

^bAn F-test for equality of the regression slopes gives F = 0.08, P = 0.779. ^cAn F-test for equality of the regression intercepts gives F = 0.09, P = 0.763.

and Scaevola constituted 62% of the diet of the roof rat and 78% of the diet of the Polvnesian rat. It is therefore reasonable to regard rats as "model herbivores" in which the concentrations of radionuclides in tissue could be expected to correlate with those in Messerschmidia and Scaevola.

Table 175 summarizes the transfer coefficients of 137 Cs and 90 Sr to rat muscle. The transfer coefficient is defined as pCi/g wet tissue ÷ pCi/g dry vegetation. They were calculated from the measured concentrations in rat muscle and in Messerschmidia and Scaevola growing at the same locations where the rats were captured. The water content of rat muscle was assumed to be 73% on the basis of current experience and the literature⁴. The transfer coefficients to rat muscle vary widely. The values for 137 Cs range from 0.1 to 7, and those for 90Sr range from 0.005 to 1.

Variations of the 137 Cs and 90 Sr concentrations in rat muscle as a function of those in Messerschmidia and Scaevola are presented in Figs. 141 and 142. Linear regression analysis 1 of the logtransformed data yields the results shown in Tables 176 and 177. Cesium-137 and strontium-90 concentrations in rat muscle correlate significantly with those in Messerschmidia and Scaevola $(^{137}Cs, P < 0.001, ^{90}Sr, P < 0.01)$, Further analysis² yields results that justify the conclusion that the individual regression lines have equal slopes and equal intercepts. The resultant pooled regres-





Table

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_			pCi/g w	Transf et muscle	er coeffi ÷ ÷ pCi/		getation	
		Cesi	um-137	······································	Strontium-90			
Plant type	No.	Min	Median	Max	No.	Min	Median	Max
esserschmidia	16	0.097	0.45	5.68	13	0.0053	0.040	1.28
aevola	17	0.11	1.12	6.78	13	0.0048	0.059	1.04
poled	33	0.097	0.73	6.78	26	0.0048	0.043	1.28

ble 175. Relationship between ¹³⁷Cs and ⁹⁰Sr concentrations in rat muscle and indicator plants.

'able 176. Statistical correlation between ¹³⁷Cs in rat muscle and ¹³⁷Cs in Messerschmidia and Scaevola.

Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y}{A} = A$	<u>+ b ln X</u> b
lesserschmidia	16	0.86	\0.001	0.144	0.773
c aevola	17	0.87	<0.001	0.284	0.801
'ooled	33	0.86	<0.001	0.230 ^b	0.783 ^a

An F-test for equality of the regression slopes gives F = 0.03, P = 0.869. An F-test for equality of regression intercepts gives F = 0.38, P = 0.540.

Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y = A}{A}$	+ b ln X b
lesserschmidia	13	0.76	<0.01	-2.10	0.540
caevola	13	0.70	<0.01	-2.05	0.557
ooled	26	0.73	<0.001	-2.07 ^b	0.546 ^a

able 177. Statistical correlation between ⁹⁰Sr in rat muscle and ⁹⁰Sr in <u>Messerschmidia</u> and <u>Scaevola</u>.

An F-test for equality of the regression slopes gives F = 0.01, P = 0.942.

An F-test for equality of the regression intercepts gives F = 0.03, P = 0.856.



Fig. 141. Statistical correlation between ¹³⁷Cs in rat muscle and ¹³⁷Cs in Messerschmidia and <u>Scaevola</u>.



Fig. 142. Statistical correlation between ⁹⁰Sr in rat muscle and ⁹⁰Sr in Messerschmidia and <u>Scaevola</u>.

sion lines thus can be used to predict ¹³⁷Cs and ⁹⁰Sr concentrations in rat muscle from those in <u>Messerschmidia</u> or <u>Scaevola</u>.

Since ⁹⁰Sr can be expected to concentrate in bone, transfer coefficients of 90 Sr to rat bone were calculated in the same manner. The weight of bone ash was assumed to be 38% of wet weight⁵ The values, shown in Table 178, vary from 0.2 to 4. Variation of the concentrations in bone as a function of those in Messerschmidia and Scaevola are presented in Fig. 143. Results of linear regression analysis are shown in Table 179. There is a highly significant correlation (P < 0.001) between the logarithms of the 90Sr concentrations in rat bone and those in Messerschmidia and Scaevola. Furthermore, the individual regression lines for Messerschmidia and Scaevola can be assumed to have equal slopes and equal intercepts. Thus the pooled regression line can be used to predict ⁹⁰Sr concentrations in rat bone from those in Messerschmidia or Scaevola.

Prediction of Radionuclide Concentrations in Foods

<u>Coconut</u> — Coconuts were the most extensively sampled of the edible plants; the 16 islands yielded a total of 23 samples of coconut meat. If the mean of the radionuclide concentrations in the samples from an island group were used to represent that island group, assessment of radionuclides in coconut would be as follows: Island group ALICE-IRENE would be based on three samples, JANET would be based on one sample, island group KATE-WILMA plus LEROY would be based on \mathbf{Pl}

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Concentration in bone — pCi/g, wet

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Fig. 1

	pCi/g v	Transfer coefficient, pCi/g wet bone ÷ pCi/g dry vegetation				
Plant type	No. of samples	Min	Median	Max		
Messerschmidia	13	0.20	0.79	2,53		
Scaevola	13	0.43	1.22	3.82		
Pooled	26	0.20	1.03	3.82		

Relationship between 90 Sr concentrations in rat bone and indicator rable 178.

Table 179. Statistical correlation between 90 Sr in rat bone and 90 Sr in Messerschmidia and Scaevola.

Plant type	Sample size (n)	Correlation coefficient (r)	Level of significance (P)	$\frac{\ln Y = A + b \ln X}{A b}$
Messerschmidia	13	0.94	<0.001	0.137 0.852
Scaevola	13	0.94	<0.001	0.331 0.919
Pooled	26	0.93	<0.001	0.256 ^b 0.872 ^a

^aAn F-test for equality of regression slopes gives F = 0.23, P = 0.635. ^bAn F-test for equality of regression intercepts gives F = 1.45, P = 0.241.





five samples (from four islands), and island group ALVIN-KEITH would be based on 12 samples (from eight islands). The samples from any island group are relatively small in number, and they would certainly not relate to future harvests, particularly in the case of the northern islands where coconut groves will have to be reestablished. Prediction of 137 Cs and ⁹⁰Sr concentrations in coconut for all island groups is therefore based on the statistical correlations between coconuts and soil. These take into account each of 22 coconut samples analyzed. Prediction of concentrations of the other radionuclides in coconut is necessarily based on mean values of the concentration in the samples from the island group.

The concentrations of 137 Cs and 90 Sr in coconut meat for each island group were predicted from the mean soil concentrations, determined in the following manner: Each of the islands comprising the island group was assigned the median concentration for the 0-15 cm profile as listed in Table 15 and 16 in the chapter on the terrestrial soil and radiation survey. If an island had two listed values (one for dense and one for sparse vegetation), as in the case of BELLE, DAISY, KATE, OLIVE, PEARL, and TILDA, it was assigned the lower of the median concentrations calculated for all the profiles sampled and analyzed. In weighting the mean concentration toward the areas of dense vegetation, we are in essence considering these areas as having more fertile and highly developed soils and hence as soils more suitable for agriculture. The mean for the island group was then computed from the values assigned to each island. Table 180 summarizes the data on mean concentrations of 137 Cs. and 90Sr in soil that were used as the starting point for predicting concentrations in coconut and other terrestrial plants.

For the relatively high concentrations of 137 Cs and 90 Sr in soil, the concentrations in coconut were predicted using the median value of the experimentally determined soil-to-plant concentration factor (Table 170). Use of a constant rather than varying concentration factor seems not only simple and straightforward, but also readily acceptable as a concept. Use of the median value is consistent with the observed data (Figs. 133 and 134), and when the concentrations in soil are relatively high, it leads to predicted concentrations in coconut that are greater than those derived from the linear regression expression. For the relatively low concentrations of 137 Cs and 90 Sr in soil, concentrations in coconut were predicted from the linear regression expressions (Tables 171 and 172).

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The above procedure for predicting 137 Cs and 90 Sr concentrations in coconut meat can be summarized as follows: If Y = concentration (pCi/g) in dry coconut meat and X = concentration (pCi/g) in dry soil,

¹³⁷Cs:

(1) X< 4.7 pCi/g	
lnY = 0.847 + 0.620 lnX	$Y = 2.33X^{0.620}$
(2) X≥4.7 pCi/g	Y = 1.3X

⁹⁰Sr:

(1) X<4.3 pCi/g	
ln Y = -3.01 +0.482ln X	$Y = 0.049 X^{0.482}$
(2) X≥4.3 pCi/g	Y = 0.023X .
These relationships are s	hown as the
solid lines in Figs. 133 a	nd 134.

<u>Coconut Milk</u> — In the absence of definitive data it is reasonable to expect that the distributions of 137 Cs and 90 Sr in meat and milk of coconut would be similar to those of their stable element counterparts potassium and calcium. Thus, the amount of 90 Sr in land plants from Bikini Atoll appeared to depend on the amount of calcium present⁴. Table 181 lists stable potassium and stable calcium concentrations in coconut and other edible and indicator plants. Table 181 indicates that for fresh coconut the potassium concentration is somewhat greater in meat,

	pC	on in top 15 cn ï/g
	90 _{Sr}	¹³⁷ Cs
Island group ALICE-IRENE		
1. ALICE	80	36
2. BELLE (dense median)	123	48
3. CLARA	6 5	26
4. DAISY (mean)	108	11
5. EDNA	46	4.2
9. IRENE	30	3.2
Mean	75.3	21.4
Island group BELLE		
2. BELLE (dense median)	123	48
Island group JANET		
10. JANET	44	16
Island group KATE-WILMA + LEROY		
11. KATE (mean)	43.5	13.1
12. LUCY	32	11
13. PERCY	13	0.94
14. MARY	29	9.9
15. NANCY	36	12
16. OLIVE (90 Sr dense median, 137 Cs mean)	22	7.65
17. PEARL (mean)	28.2	12.4
18. RUBY	12	1.4
19. SALLY	8.4	3.0
20. TILDA (mean)	19.2	4.2
21. URSULA	6.8	1.7
22. VERA	6.3	2.0
23. WILMA	3.3	1.3
43. LEROY	11	3.2
Mean	19.3	6.00
· · · · · · · · · · · · · · · · · · ·		
Island group ALVIN-KEITH	0.11	A 91
33. DAVID, 35. ELMER, 37. FRED	0.41 0.52	0.21 0.14
All others (14 islands)	0.52	0.14
Weighted	mean 0.50	0.15

Table 180. Mean concentrations of 90 Sr and 137 Cs in Enewetak soils.

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indical	tor plants.			
Observa	tion	Concer mg/g	ntration ^a , dry R	leference
Plant type	Source	K	Ca	
Breadfruit	Marshall Is. 1951	-	0.85	19
Breadfruit	Caroline Is. 1951	-	0.70	19
Breadfruit	Panama 1966-67	32	2.2	22
Breadfruit	Colombia 1966-67	26	2.1	22
Breadfruit	Handbook data	15	1.1	6
Coconut meat	Bikini I 1964	-	0.71	4
Coconut meat	Handbook data	5.9	0.26	6
Coconut milk	Handbook data	25 [°]	3.4 [°]	6
Coconut meat	Enewetak 1972-73	$7.5(23)^{1}$	^o 0.24(10)	This study
Pandanus fruit	Rongelap I 1958–63	-	6.1(23)	10
Pandanus leaves	Rongelap I 1958-63	-	13 (66)	10
Pandanus fruit	Eniaetok I, Rongelap 1958-63	-	3.3(9)	10
Pandanus leaves	Eniaetok I, Rongelap 1958-63	-	12 (12)	10
Pandanus fruit	Kabelle I, Rongelap 1958-63	-	9,2(6)	10
Pandanus leaves	Kabelle I, Rongelap 1958-63	-	12 (8)	10
Pandanus fruit	Bikini I 1964	-	17	4
Pandanus leaves	Bikini I 1964	-	17	4
Pandanus fruit	Enewetak 1972-73	16(2) ^b	4.4(1)	This study
Pandanus leaves	Enewetak 1972-73	11(9) ^b	12.6(9)	This study
Tacca whole corm	Rongelap 1958–61 ^d	6.8(3)	6.0(6)	10
Tacca peeled	_			
corm	Rongelap 1958-61 ^d	4.9(3)	1.1(3)	10
Tacca peels	Rongelap 1958-61 ^d	19 (3)	5.4(3)	10
Tacca processed	Rongelap 1958–61 ^d	0.14(1)	0.41(2)	10
Tacca whole corm	Bikini 1964	-	5.0	4
Tacca whole corm	Enewetak 1972-73	8.0(1) ^b	9.8(1)	This study
Cassava root	Panama 1966-67	11	1.5	22
Cassava root	Colombia 1966-67	12	1.0	22
Tapioca	Handbook data	0.18	0.10	6

Table 181. Stable potassium and stable calcium content of selected edible and indicator plants.

^aThe number shown within parentheses is the number of samples. ^bThe stable K concentration was estimated from the concentrations of ⁴⁰K. ^cThe concentrations of stable K and stable Ca in fresh coconut meat are 2.56 mg/g and 0.13 mg/g, respectively. In fresh coconut milk they are 1.47 mg/g and 0.20 mg/g, respectively.⁶ ^dThe tacca samples from Rongelap Atoll were collected from the islands of Rongelap,

Eniaetok, and Kabelle.

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P \underline{N} Table 181 (continued).

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Observat	tion		tration ^a , g dry	Reference
Plant type	Source	K	Ca	
Messerschmidia				
basal leaves	Bikini 1964	-	52	4
Messerschmidia				
terminal leaves	Bikini 1964	-	35	4
Messerschmidia				
leaves	Enewetak and Bikini 1964	9 . 9(3 1)	23(31)	24
Messerschmidia				
leaves	Enewetak 1972-73	14(58) ^b	-	This study
Scaevola basal				
leaves	Bikini 1964	-	23	4
Scaevola				
terminal leaves	Bikini 1964	-	19	4
Scaevola leaves	Enewetak 1972-73	16(56) ^b	39(6)	This study

^aThe number shown within parentheses is the number of samples.

^bThe stable K concentration was estimated from the concentrations of 40 K.

^cThe concentrations of stable K and stable Ca in fresh coconut meat are 2.56 mg/g and 0.13 mg/g, respectively. In fresh coconut milk they are 1.47 mg/g and 0.20 mg/g, respectively.⁶ dThe tacca samples from Rongelap Atoll were collected from the islands of Rongelap,

Eniaetok, and Kabelle.

while the calcium concentration is somewhat greater in milk 6 , but the concentrations in meat and milk differ by less than a factor of two.

Table 165 lists the concentrations of the radionuclides measured in milk and meat of coconuts in this survey. The data are too few to allow for statistical analysis, and they show an inconsistent pattern of ¹³⁷Cs distribution. Concentrations of 137 Cs are greater in meat in samples from DAISY, JANET, and NANCY, but they are greater in milk in the samples from MARY and DAVID. Strontium-90 concentrations are lower rather than higher in milk in all samples, except possibly those from DAVID. In view of these considera-

tions, it seems reasonable and expedient to assume for purposes of prediction that the concentrations of $^{137}\mathrm{Cs}$ and $^{90}\mathrm{Sr}$ in fresh meat and fresh milk are the same. It is interesting to note that the average ¹³⁷Cs concentrations in meat and milk from coconuts sampled at Bikini Atoll in 1969 were comparable to within 10-15%⁷. Wet-weight concentrations of radionuclides were calculated from the dry-weight concentrations, assuming 50 and 95% as the water contents of fresh coconut meat and fresh coconut milk. Both of these values are consistent with literature values 4,6 and with laboratory records.

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Pandanus Fruit - Although pandanus was found on 10 islands, samples of fruit could only be obtained from BELLE and KEITH. These fruit samples seem to display soil uptake patterns for 137 Cs and 90 Sr that are similar to those displayed by the leaves. Figure 144 shows the variation of the ¹³⁷Cs concentrations in fruit and leaves as a function of that in soil. and Fig. 145 shows comparable data for ⁹⁰Sr. Fruit and leaves of pandanus seem to display similar soil uptake patterns for 137 Cs and 90 Sr, and the concentration factors of fruit are within the range of the concentration factors of leaves (see Table 170).

As Table 182 indicates, data on radionuclide content in fruit or leaves of pandanus from previous radiological surveys



Fig. 144. Correlation of ¹³⁷Cs in edible plants with ¹³⁷Cs in soil.



Fig. 145. Correlation of 90 Sr in edible plants with 90 Sr in soil.

of Bikini and Enewetak were limited to just a few samples^{4, 7-9}. It was our good fortune to be provided with unpublished data on the radionuclide content in fruit and leaves of pandanus sampled during radiological surveys conducted by the University of Washington on Rongelap Atoll¹⁰. Table 183 summarizes the available data on concentrations of ¹³⁷Cs and ⁹⁰Sr in fruit and leaves of pandanus from the same site on Rongelap¹⁰ and elsewhere⁴. The two sets of pandanus fruit and leaves from the current survey are included in the table.

Inspection of Table 183 leads to the

			Concentration,	pCi/g wet					
		Pand	anus	Tacca		Tacca		Reference	
		Fruit	Leaves	Corm	Processed arrowroot				
Year	Nuclide	Bikini Enewetak	Bikini Enewetak	Bikini Enewetak	Bikini Enewetak				
1964	60 _{Co}			0. 12 ^a		1			
		ac	ас			4			
	90 _{Sr}	32 ^{a, c}	24 ^{a, c}	· 0. 068		4			
	90 Sr	9300 SU ^d	7200 su^{d}	6900 SU ^d		4,8			
	106Ru			1.8 ^a		4			
	137 _{Cs}	3.1 ^a 180 ^b		50 ^a		4			
1967	⁹⁰ Sr								
		19 (4-45)		0.17		9			
	137 Cs	52 (14-90)		92 (15-170)		9			
1969	$90_{ m Sr}$	28 ^a			2. 4^{a} 0. 4^{b}	7			
	137Cs	$130(26-400)^{a}$			4. t U. t	1			
	65	87 ^b			0.6(0.4-1.1) ^{a or b}	7			

Table 182.	Average radionuclide	content of panda	nus and tacca	collected on	previous su	urveys at	Bikini and E	Enewetak.

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a Bikini Island. ^bEnyu Island. ^cWater content of pandanus and tacca corm is assumed to be 80%. ^d1 SU (strontium unit) is equal to 1 pCi ⁹⁰Sr/g Ca. ^ePrepared according to the Marshallese method of preparation by grinding, rinsing three times with salt water and once with fresh water.⁷

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			Concentra	tion, pCi/g	dry	
		9 0	Sr	137	Cs	
Date	Location	Fruit	Leaves	Fruit	Leaves	Reference
Aug. 1958	Rongelap Atoll					
	Eniaetok	6.0	25	230	78	10
	Kabelle	6.0	34	309	346	10
Sept. 1959	Rongelap Atoll					
	Rongelap	12.0	14	160	100	10
	Rongelap			152	48	10
	Eniaetok	7.1	19	360	82	10
	Eniaetok	9 .0	38	226	105	10
	Kieshiechi	30.0	92	422	97	10
	Mellu	7.1	29	298	50	10
	Gejen	30.0	54	991	111	10
	Aerik	21.0	65	620	496	10
	Tufa	5.8	11	126	42	10
Aug. 1963	Rongelap Atoll					
	Rongelap			170	58	10
	Rongelap	6.6	11	44	21^{a}	10
	Rongelap			140	62^{a}	10
	Eniaetok	15.0	45	260	67	10
	Kabelle			220	170	10
Aug. 1964	Bikini Atoll					
	Bikini	160.0	120			4
Oct. 1972-	Enewetak Atoll					
Feb. 1973	Bogombogo (BELLE)	206.0	391	923	679	This study
	Giriinian (KEITH)			0.86	0.57	This study

Table 183. 90Sr and 137Cs in fruit and leaves of pandanus.

^aMean of concentrations in terminal and basal leaves.

conclusion that the concentration of ¹³⁷Cs in pandanus fruit can be expected to exceed that in leaves, while the concentration of ⁹⁰Sr in pandanus leaves can be expected to exceed that in fruit. Thus, if a quantitative relationship can be established between the concentrations in fruit and leaves, it would be possible to predict future concentrations of radionuclides in pandanus fruit using the data obtained from pandanus leaves.

The Wilcoxen matched-pairs, signedranks test¹¹ was used to determine the appropriate conversion factors to be used with the 90 Sr and 137 Cs concentrations in pandanus leaves to determine those in fruit. In this nonparametric test, signed differences between concentrations in fruit and leaves are determined, and the differences are ranked according to absolute value. The ranks of like sign are then summed. and the lower sum of the like-signed ranks is compared with an appropriate critical value from a special table. If the observed sum is equal to or less than this critical value for a particular significance level, the null hypothesis may be rejected at that level of significance.

Tables 184 and 185 show the results of Wilcoxen test carried out on the matched pairs of 137 Cs and 90 Sr concentrations listed in Table 183. A series of values was tested to determine the appropriate conversion factors for the two nuclides. Table 184 indicates that if the concentrations of 137 Cs measured in pandanus leaves were increased by any factor from 2 to 3.5, the resulting concentrations would be statistically indistinguishable from those in fruit. A conversion factor of 2.5, which gives essentially equal sums of minus and plus ranks has been adopted in this evaluation; i.e., the 137 Cs concentration in pandanus fruit is assumed to be 2.5 times that in pandanus leaves. In the case of 90 Sr, Table 185 indicates that if the concentrations measured in pandanus fruit were increased by any factor between 2 and 3, the resulting concentrations would be statistically indistinguishable from those in leaves. A conversion factor of 2.5, which gives essentially equal sums of minus and plus ranks, has been selected. Thus the concentration of 90 Sr in pandanus fruit is assumed to be 40% of that in pandanus leaves.

The concentrations of 137 Cs and 90 Sr in pandanus leaves for each island group were predicted from the mean soil concentrations in essentially the same manner described for coconut. The mean concentrations in soil listed in Table 180 were used as the starting point. When the soil concentrations were relatively low. the statistical correlation between pooled pandanus leaves, Messerschmidia, and Scaevola and soil (Tables 171B and 172B) was used for prediction. When the soil concentrations were relatively high, the median of the experimentally determined soil-to-plant concentration factors (Table 170) was used. The median values chosen for the concentration factor in the pooled plants were 5.2 for 137 Cs and 1.0 for ⁹⁰Sr. The relationship used for predicting concentrations in pandanus leaves from those in soil are represented as solid curves on the graphs showing Messerschmidia and Scaevola vs soil (Figs. 137 and 138).

The concentrations of ¹³⁷Cs and ⁹⁰Sr in pandanus fruit were subsequently pre-

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			ed-ranks test (Ref. 11) ^a
	X = pC1	/g dry iruit	Y = pCi/g dry leaves
Samples tested	Sum of minus ranks $\left(\sum_{-}\right)$	Sum of plus ranks $\left(\sum^{+}\right)$	Conclusion about null hypothesis (H _o)
X - Y	3	168	Reject H_0 , $P < 0.005$; $X > Y$
X - 1.5Y	40	131	Reject H_{0} , P = 0.025; X > 1.5 Y
X - 2 Y	64	107	Not rejected; $X = 2 Y$
X - 2.5Y	86	85	Not rejected; $X = 2.5 Y$
X - 3 Y	97	56	Not rejected; $X = 3 Y$
X - 3.5Y	127	44	Not rejected; $X = 3.5 Y$
X - 4 Y	139	32	Reject H_0 , P < 0.01; X < 4 Y

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Table 184. Relationship between ¹³⁷Cs concentrations in fruit and leaves of pandanus.

^aThis table summarizes the results from the Wilcoxen test using the 18 pairs of data listed in Table 183.

	11 PO	ing dry man	r porfgary reaves
Samples tested	Sum of minus ranks $\left(\sum_{i=1}^{i} -\right)$	Sum of plus ranks $\left(\sum_{+}^{+}\right)$	Conclusion about null hypothesis (H _O)
Y - X	11	94	Reject H_{0} , $P < 0.005; Y > X$
Y ~ 1.5X	17	88	Reject H_0 , $P < 0.025$; $Y > 1.5X$
Y - 2X	36	69	Not rejected; $Y = 2X$
Y - 2.5X	53	52	Not rejected; $Y = 2.5 X$
Y - 3X	60	31	Not rejected; $Y = 3 X$
Y - 3.5X	89	17	Reject H_0 , P < 0.025; Y < 3.5X

Table 185. Relationship between 90 Sr concentrations in fruit and leaves of pandanus.

Wilcoxen matched-pairs, signed-ranks test (Ref. 11)^a X = pCi/g dry fruit Y = pCi/g dry leaves

^aThis table summarizes the results from the Wilcoxen test using the 14 pairs of data listed in Table 183.

dicted from those in leaves using respective conversion factors of 2.5 and 0.4. The overall procedure for predicting concentrations in pandanus fruit from those in soil can be summarized as follows: If Y = concentration (pCi·g) in dry pandanus fruit and X = concentration (pCi/g) in dry soil,

 $^{137}Cs:$

(1) $X \le 1.30 \text{ pCi/g}$ $\ell_n Y = 2.60 \pm 0.851 \text{ (nX)}$	Y = 13.5X ^{0.851}
(2) X≥1.30 pCi/g	Y = 13X

⁹⁰Sr:

(1) $X \le 6.1 \text{ pCi/g}$	0.000
$\ln Y = -0.617 + 0.836 \ln X$	Y = 0.539X ^{0.630}
(2) X≥6.1 pCi/g	Y = 0.4X

Tacca (Arrowroot) - Figure 144 shows graphically the distribution of ¹³⁷Cs in edible plants as a function of that in soil. Although only one sample of tacca corm was collected in the current survey, it seems to fall naturally within the overall distribution. The concentration factor of 137 Cs in this sample is 16, which is about a factor of three greater than the median concentration factor in pandanus eaves and ten times greater than the median in coconut (see Table 170). This sample of tacca also seems to be similar to the other edible plants as far as uptake of ⁹⁰Sr from soil is concerned (see Fig. 145). The concentration factor of 90 Sr is 0.21, a value intermediate between 0.023, the median concentration factor in coconut meat, and 1.0, the median concentration factor in pandanus leaves (see Table 170). Data on tacca collected in previous radiological surveys of Bikini and Enewetak are shown in Table 182 and are relatively few^{4,7-9}. Although the data suggest that the ⁹⁰Sr concentration in pandanus fruit would exceed that in tacca corm by more than an order of magnitude and that the ¹³⁷Cs concentrations in the two plant types would be comparable, it cannot be ascertained that the two plant types were sampled at the same sites. If comparison of the 1967 data on tacca corm from Bikini and the 1969 data on processed arrowroot from Bikini is valid, it can be concluded that most of the ¹³⁷Cs content of tacca is lost during processing.

Table 186 shows unpublished data on the concentrations of 137 Cs in tacca and pandanus leaves from the same sites on Rongelap Atoll¹⁰. The concentrations are greater in pandanus leaves, but they do not exceed those in tacca corm by more than a factor of two. Comparable data on 90 Sr in Table 187 indicate that the 90 Sr concentration in pandanus leaves can be expected to exceed that in tacca corm by a substantial amount. The concentrations in pandanus fruit and tacca would be more comparable; perhaps the concentration in tacca would be somewhat less. If we assume that the stable K concentration provides a measure of the relative uptake of 137 Cs, the stable K data of Table 181 suggest that the uptake of 137 Cs into tacca from soil would not exceed that to pandanus leaves, Messerschmidia, Scaevola, or coconut. Accordingly, on the basis of the stable K data and the Rongelap data, we have assumed that pandanus leaves serve as direct indicators for the uptake of ¹³⁷Cs to tacca corms, and that the 137 Cs in tacca can be predicted from that

			Concentrati	on, pCi/g dry
Locat	ion	Date	Tacca corms	Pandanus leaves
Rongelap I	Pit 1	Mar. 1958	47	110
Eniaetok I	Pit 11	Mar. 1958	140	203
Kabelle I	Pit 7	Mar. 1958	182	270

Table 186. Comparison of ¹³⁷Cs in tacca and pandanus from the same sites ^a

^aThese unpublished data from University of Washington radiological survey at Rongelap were provided by Drs. A. H. Seymour and E. E. Held¹⁰.

in soil using the plant-vs-soil correlation derived for pooled <u>Messerschmidia</u>, <u>Scaevola</u>, and pandanus leaves (Table 171B). Similarly, if the stable calcium content is a measure of the potential 90 Sr uptake, one may conclude that the uptake of 90 Sr to tacca corm and pandanus fruit would be comparable and exceed that to coconut. On the basis of the stable calcium data and the Rongelap data, we have predicted the 90 Sr concentrations in tacca corm from those in soil, assuming that concentration in tacca and pandanus fruit would be the same.

Unpublished data on tacca from the University of Washington radiological surveys on Rongelap¹⁰ have also provided us with information on expected reductions in the ¹³⁷Cs and ⁹⁰Sr dry-weight concentrations from the processing of tacca into arrowroot starch. The ¹³⁷Cs concentration would be reduced by a factor of 50 or greater, and the ⁹⁰Sr concentration would be reduced by a factor of 20 or greater. Stable potassium and stable calcium experience similar reductions (see Table 181). On this basis the concentrations of ¹³⁷Cs and ⁹⁰Sr predicted in tacca corm are reduced by factors of 50 and 20 to give the final concentrations of 137 Cs and 90 Sr in dry processed arrowroot starch.

The overall procedure for predicting the concentrations of 137 Cs and 90 Sr in arrowroot starch from those in soil can be summarized as follows: If Y = concentration (pCi/g) in dry arrowroot starch and X = concentration (pCi/g) in dry soil,

 $137_{Cs}:$ (1) X< 1.3 pCi/g ln Y = -2.22 + 0.851 lnX Y = 0.108X^{0.851} (2) X = 1.3 pCi/g Y = 0.10X

⁹⁰Sr:

(1) X<	6.1 pCi/g	0.000
ln	$Y = -3.61 + 0.836 \ln X$	$Y = 0.027 X^{0.836}$
	=6.1 pCi/g	Y = 0.020X

<u>Breadfruit</u> — Breadfruit was not obtainable on this survey. The data of Table 181 indicate that stable potassium concentration in breadfruit is relatively high, greater than that in coconut or tacca and comparable to or even greater than that in <u>Messerschmidia</u>, <u>Scaevola</u>, or pandanus fruit. The data of Table 181 also indicate that the stable calcium con-

				C	oncentration, pCi/	g dry		
Location		Date	Tacca corms	Date	Pandanus leaves	Date	Pandanus fruit	
Rongelap I	Pit 1	Mar. 1958	3.2	Aug. 1958	47			
Eniaetok I	Pit 11	Mar. 1958	3.1	Mar. 1958	42	Mar. 1958	10	
				Aug. 1958	41			
						Sept. 1959	9	
		Sept. 1961	6.9					
				Aug. 1963	45	Aug. 1963	15	
Eniaetok I	Pit 10					Sept. 1959	9	
		Sept. 1961	16.7					
Kabelle I	Pit 7	Mar. 1958	4.9	Mar. 1958	36	Mar. 1958	17	
				Sept. 1959	26			
		Sept. 1961	20	Sept. 1961	40 ^b			
						Mar. 1963	26	
				Aug. 1963	26	Aug. 1963	1.5	

Table 187. Comparison of 90 Sr in tacca and pandanus from the same sites.^a

^aThese unpublished data from University of Washington radiological surveys at Rongelap Atoll were provided by Drs. A. H. Seymour and E. E. Held¹⁰. ^bMean of concentrations in terminal and basal leaves.

centration in breadfruit is relatively low; lower concentrations are listed only for coconut. In the absence of additional data, we have assumed that uptake of 137 Cs and 90 Sr will be proportional to the concentrations of stable potassium and stable calcium. Therefore, for predicting 137 Cs and 90 Sr concentrations in breadfruit, we have assumed that breadfruit and pandanus fruit will experience the same uptake from soil and have simply adopted the same procedure described previously for predicting 137 Cs and 90 Sr concentrations in pandanus fruit from those in soil.

<u>Birds</u> — Birds were captured in numbers on 18 islands distributed over all sections of the Atoll. The mean of the radionuclide concentrations in the samples from each island group were used to represent the island group. Since both muscle and liver are consumed as food, radionuclide concentrations in both muscle and liver are presented in Table 167. The average concentration in edible bird flesh was computed from these data, assuming that the weight of muscle consumed is six times that of liver, a relationship derived from laboratory records.

<u>Bird Eggs</u> — Common noddy or sooty tern eggs were collected on eight islands distributed more or less throughout the Atoll. The mean of the concentrations in the samples from each island group (see Table 168) was used to represent the group.

<u>Coconut Crabs</u> — Coconut crabs were captured on only five of the southern islands. The means of the concentrations in the four samples from BRUCE, GLENN JAMES, and KEITH were used to represent the southern island group (Group E. ALVIN-KEITH), and the concentrations in the samples from LEROY was used to represent island group KATE-WILMA and LEROY. Since coconut crabs could not be captured elsewhere on the Atoll, we conclude that only the southern islands would yield coconut crabs in numbers sufficient to contribute substantially to the diet. Both muscle and hepatopancreas are consumed as food. Laboratory records indicate that the dry weights of hepatopancreas (and associated tissues) and muscle are about the same. The concentrations of radionuclides in coconut crab were therefore computed from the concentrations listed in Table 169, assuming equal contributions from hepatopancreas and muscle.

 $\frac{\text{Livestock and Poultry}}{137}$ — Although most effectively transferred to man via meat and poultry, this pathway would still contribute significant quantities of ⁹⁰Sr to the diet. Prediction of 137 Cs and 90 Sr concentrations in pork and chicken has been based on data obtained from rats. Table 188 summarizes data on the transfer coefficient of ¹³⁷Cs in muscle of herbivores that provide meat for human consumption, calculated from environmental data reported for cattle¹², sheep, and deer¹³. The transfer coefficient is defined as pCi/g wet tissue $\div pCi/g dry$ feed. Included in Table 188 are the transfer coefficients for rock ptarmigan and willow grouse 14 , two herbivorous game

		pCi/kg	<u> </u>			
			Ra	nge		
Location	Observation	No.	Min	Max	Mean Referer	
Fort Collins, Colo.	Dry-lot and pasture-fed cattle	17	0.049	0.57	0,25±0,15 ⁸	a 12
Mendocino County, Calif.	Sheep, age 4 mo	4	0.13^{b}	0.29^{b}	0.19 ^b	13
Mendoemo county, cerri	Sheep, age 16-17 mo	4	0.10^{b}	0.15^{b}	0.12^{b}	
	Sheep, age 40-41 mo	4	0.23^{b}	0.52^{b}	0.27^{b}	
	Deer (Columbia black-tailed), age 4 mo	4	0.21^{b}	0.36 ^b	0.26 ^b	
	Deer, age 16-17 mo	5	0.17 ^b	0.41^{b}	$0.26^{\rm b}_{-}$	
	Deer, age 40-41 mo	5	0.087^{b}	0.32^{b}	0.17^{b}	
Lapland, Finland	Rock ptarmigan	2°	0.15 ^d	0.24^{d}		14
Lapland, Limand	Willow grouse	2°	0.29 ^d	0.35 ^d		

Rehavior of 137 Ca in the plant-berbivore-meat pathway. m. L.L. 100

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^aMean value ± σ.
 ^bTransfer coefficients were calculated assuming that dry weight of forage equals dry weight of ruminal contents and that wet weight of muscle equals 3.5 times dry weight of muscle.
 ^cPooled samples each composed of 6-15 specimens.
 ^dThe concentration in dry crop contents was assumed to be the concentration in dry forage.

birds. Table 189 summarizes transfer coefficients of ¹³⁷Cs in rat organs calculated from data on rats and indicator plants reported for previous studies on Enewetak and Bikini by the University of Washington⁴ and Bowling Green University¹⁵. It was assumed that the rats and plants were collected at the same locations. Except for the transfer coefficient of 1.39 reported for Japtan, the concentration factors listed in Table 189 for rats fell within the range of those listed in Table 188 for cattle, sheep, deer, and game birds.

Tracer experiments have demonstrated a similar pattern of deposition of radiocesium in muscle of livestock and poultry. Hood and Comar¹⁶ noted that the relative concentrations of ¹³⁷Cs in various tissues of farm animals 7 days after a single oral dose were quite similar. When normalized to a common body weight, the relative concentrations in muscle of cow. sheep, pig, and hen were 30, 41, 23, and 24, respectively. Although equilibrium between intake and accumulation in organs requires a period of time that varies with species, the equilibrium content in organs following chronic feeding differs very little. The equilibrium content of 137 Cs in muscles of rat, rabbits, dogs, and pigs was found to be 14.5 to 28.5 times the daily dose 17 .

The basic parameters that influence the transfer of radionuclides from vegetation to muscle of herbivores can be conveniently described in terms of a simple model. The radionuclide concentration in muscle can be described by the equation:

$$Q(t) = \frac{f_B I}{\lambda_E} (1 - e^{-\lambda_E t}),$$
 (1)

where

- Q(t) = quantity of radionuclide in muscle at time t, day,
- f_B = fraction of ingested nuclide deposited in muscle,
- I = quantity of radionuclide ingested daily, pCi/day, and
- $\lambda_{\rm E}$ = effective elimination constant, day⁻¹.

The quantity of radionuclide in muscle at equilibrium $\boldsymbol{Q}_{e\boldsymbol{\alpha}}$ is:

$$Q_{eq} = \frac{f_B I^*}{\lambda_E}$$
, (2)

since

$$Q_{eq} = mC_B^*$$
 and $I^* = JC_P^*$, (3a, 3b)

where

m = mass of muscle, g,

- C^{**}_B = concentration of radionuclide in muscle, pCi/g,
- J = quantity of vegetation ingested daily, pCi/day, and
- C_P^* = concentration of radionuclide in vegetation, pCi/g.

One can substitute for Q_{eq} and I^* and bbtain the following expression for the transfer coefficient

$$\frac{C_{\rm B}^{*}}{C_{\rm P}^{*}} = \frac{Jf_{\rm B}}{m\lambda_{\rm E}} \,. \tag{4}$$

Table 190 presents muscle weight and daily intake of dry feed in livestock and rats. Table 191 presents effective halflives or accumulation factors of 137 Cs in muscle. The accumulation factor is obtained from chronic-administration experiments and is the ratio of the quantity of radionuclide in an organ to the daily

Date	Location	Rat species	Organ	Plant species	Transfer coefficient	Refer- ence
1964	Enewetak Atoll					
	Engebi (JANET)	Roof rat	Muscle	Messerschmidia and Scaevola	0.28	15
	Engebi (JANET)	Roof rat	All	A11	0.12	4
	N. Runit (YVONNE)	Roof rat	Muscle	<u>Messerschmidia</u> and <u>Scaevola</u>	0.58 [°]	15
	Japtan (DAVID)	Polynesian rat	All	All	1.39 ^b	4
	Bikini Atoll					
	Enyu (NAN)	Polynesian rat	A11	All	0.32	4
	Bikini (HOW)	Roof and Polynesian rat	All	All	0.29	4
1965	Enewetak Atoll					
	Engebi (JANET)	Roof rat	Muscle	Messerschmidia and <u>Scaevola</u>	0.11	15
	Biijiri (TILDA)	Polynesian rat	Muscle	Messerschmidia and Scaevola	0.46 [°]	15
	Runit (YVONNE)	Roof rat	A11	A11	0.26	4

Table 189. Transfer coefficients of ¹³⁷Cs in rat organs^a.

^aTransfer coefficient = pCi/g wet tissue ÷ pCi/g dry forage. ^bBoth tissue and plants were low in ¹³⁷Cs content. ^c<u>Messerschmidia</u> and <u>Scaevola</u> differed widely in ¹³⁷Cs content.

dose. From Eq. (1) the accumulation factor is

$$AF = Q_{eq} / I^* = f_B / \lambda_E$$
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The effective half-life $T_{F_{i}}$ is related to the elimination rate $\lambda^{}_{\rm E}$ through the relationship λ_E = ln 2/T_E. Half-lives were generally obtained from experiments involving single administration of 137 Cs.

Table 191 also lists the resultant transfer coefficient to muscle when the data listed in Tables 190 and 191 are

combined according to Eq. (4). The f_B to muscle has been set equal to 0.5 for the calculation. Table 191 includes wholebody values for half-life and accumulation factor. In using the exponential retention for the whole body to represent muscle, we are following accepted practice.

The transfer coefficients calculated for Table 191 are seen to exceed those of Table 188. This difference is explainable by the difference in character of the two sets of data. Table 188 is representative

Animal	Muscle weight (m), kg	Daily intake of dry feed (J), kg/day	Ratio of daily intake to muscle weight (J/m), day ⁻¹	References
	•			
Rat ^a	0.12	0.017-0.025 ^a	0.18	5, 25
Beef cattle	180	8-15	0.064	26, 27
Dairy cattle	160	10-20	0.094	26, 27
Sheep	24	1.2-2	0.067	26, 27
Swine	85	3.1-4	0.042	26, 27
Chicken ^b	0.7	0.08	0.11	28

Table 190. Muscle weight and daily intake of dry feed in livestock and rats.

^aThe entries for rat assume a total body weight of 260 g. The 25-g/day daily intake is based upon the personal experiences of A. J. Silva in the Bio-Medical Division laboratories at LLL.

^bThe entries for chicken assume that about 50% of the total body weight is muscle and that feeding practice is as described in Ref. 28.

and rats.					
Animal	Half-life (T _E), day	Accumulation factor (AF), day	Transfer coefficient (C_{B}^{*}/C_{P}^{*})	Reference	
Rat	13		1.7	29	
	8.6 ^a		1.1	30	
		8.9 ^a	1.8	25	
		16	2.9	17	
Dairy cattle	17 ^a	•	1.2	31	
	15 ^{a,b}		1.0	31	i
Sheep	12 ^{a,b}		0.58	31	ĺ
•	17 ^a		0.82	32	
Swine	29 ^{a,b}		0.88	31	
	23 ^C		0.70	33	
		16	0.67	17	
Hen	27 ^a		2.1	33	

Table 191. Half-lives and accumulation factors of 137 Cs in muscle of livestock and rats

^aWhole-body value.

^bIsotope administered intravenously.

^cSlow component.

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of field data. Since not all of the 137 Cs associated with vegetation is absorbed in the gastrointestinal tract, fractional biological-availability factors are implicit in the data. The data of Table 191, on the other hand, were derived from tracer experiments; in these situations the biological availability of ¹³⁷Cs is characteristically near 100° . In Table 191 the transfer coefficients of 137 Cs to muscle are two to three times greater in rats than in swine. Table 175 lists the median and range of the transfer coefficients of 137 Cs in rat muscle calculated for this study, using data from rats and Messerschmidia and Scaevola sampled at the same locations. The median transfer coefficient exceeds by a factor of two or greater the transfer coefficients listed in Table 188 for cattle, sheep, and deer. The considerations enumerated above lead to the conclusion that the transfer coefficient of 137 Cs to pork is half as great as the transfer coefficient to rat muscle.

The transfer coefficients of ¹³⁷Cs in Table 188 for rock ptarmigan and willow grouse muscle are comparable to those for muscle of cattle, sheep, and deer. On the other hand, the transfer coefficient calculated for poultry muscle in Table 191 is about equal to that for rat muscle and about two times that for cattle and swine. Surveillance data on ¹³⁷Cs in the Chicago diet¹⁸ indicate that the ¹³⁷Cs concentrations in poultry were substantially less than those in meat when the fallout rate was relatively high. During the recent periods of relatively low fallout rate, ¹³⁷Cs concentrations in poultry and meat

have been more or less comparable (see

Table 192). It must be remembered, however, that poultry raised for commerce do not forage but are kept under shelter and given stored feed. We have assumed for present purposes that the transfer coefficients of 137 Cs to rat muscle and poultry muscle are equal.

Strontium accumulates in bone rather than in soft tissues. In repeated oral administration of 90Sr the accumulation patterns of 90Sr in skeleton of rats and swine were similar, with accumulation factors intermediate between those of calves and dogs and those of ewes¹⁷. The maximum accumulation factor varied from 7.2 to 17.5 in the skeleton of rats and from 10.7 to 17.5 in that of pigs¹⁷. In the establishment of the equilibrium state between intake and elimination in young rats during chronic feeding, the 90Sr content in the skeleton was 200

times and the concentration 2000 times greater than in muscle. In old animals these relationships were 99 and 665,respectively, and in rats on high calcium diets, 55 and 333^{17} . In pigs the concentration in skeleton was 140 times greater than in muscle¹⁷. In the present study the ratio of the ⁹⁰Sr concentration in rat bone (pCi/g wet) and that in rat muscle (pCi/g wet) varied from 3.0 to 150, with a median value of 41 (n = 11).

The quotient of the daily intake of feed and the mass of bone in rat could be expected to exceed that in swine in much the same way that the quotient of the daily intake of feed and the mass of muscle in rat exceeds that in swine (see Table 190). If the accumulation factors to bone in rat and swine are as Ref. 17 indicates, then by Eq. (4) the transfer coefficient from

	Concentration, pCi/kg			
Date	Poultry	Meat		
January 1970	11	28		
April	5	12		
October	0	14		
January 1971	0	24		
April	8	25		
July	6	33		
October	10	19		
January 1972	10	12		
April	13	18		
July	22	20		
October	9	19		
April 1973	4	10		

Table 192. ¹³⁷Cs in poultry and meat from Chicago.^a

^aThe data in this table were abstracted from Ref. 18.

indicator plant to bone for 90 Sr would be greater in rats than in pigs by about a factor of four. On the other hand, the data of Ref. 17 also indicate that for the same 90 Sr concentration in bone, the 90 Sr concentration in muscle of pigs could be expected to exceed that in muscle of rats by about the same factor. These relationships thus provide a basis for using the 90 Sr concentrations in rat muscle as a direct indicator for that in pork, and in the absence of data on the behavior of 90 Sr in poultry, as a direct indicator for meat from chicken.

Comparison of Figs. 142 and 143 and Tables 177 and 179 reveals that the correlation between the ⁹⁰Sr concentrations in rat bone and indicator plants is stronger than the correlation between rat muscle and indicator plants. Thus the bone versus plant correlation, together with the bone-to-muscle concentration ratio, could be used to predict ⁹⁰Sr concentration in rat muscle from that in <u>Messerschmidia</u> or <u>Scaevola</u>. It was not possible to follow such an approach and develop a simple, straightforward scheme for predicting ⁹⁰Sr in rat muscle and obtain results consistent with the observed concentrations in muscle.

The concentrations of 137 Cs and 90 Sr in rat muscle were both predicted from those in <u>Messerschmidia</u> and <u>Scaevola</u>. The median transfer coefficients, which are listed in Table 175, were used with the higher concentrations in the indicator plants, and the statistical correlations between rat muscle and pooled <u>Messer-</u> <u>schmidia</u> and <u>Scaevola</u> (see Tables 176 and 177) were used with the lower concen-

tration: 137 Csschmid predict correla Tables and 138 Messei in soil. soil for in Tabl tions tl and poi arbitra much 1 The 137Cs those i follows fresh 1 tion (p ¹³⁷Cs: (1) X< 1 ln Y (2) 1.3 ln Y (3) X≥2 ⁹⁰Sr: (1) X< (ln y (2) 6.1 ln } (3) X > As:tria Me nuclic was c predirations in the indicator plants. The ³⁷Cs and ⁹⁰Sr concentrations in <u>Messerchmidia</u> and <u>Scaevola</u> initially were redicted from those in soil using the orrelations in Tables 171A and 172A or lables 171B and 172B (also see Figs. 137 nd 138) between the concentrations in <u>Messerschmidia</u> and <u>Scaevola</u> and those n soil. The mean concentrations in soil for the islands of interest are listed n Table 180. To simplify the calculaions the average concentrations in meat and poultry were computed assuming arbitrarily that pork contributes twice as nuch meat to the diet as chicken.

The overall procedure for predicting ${}^{37}Cs$ and ${}^{90}Sr$ in meat and poultry from hose in soil can be summarized as follows: If Y = concentration (pCi/g) in fresh meat and poultry and X = concentration (pCi/g) in dry soil,

 ${}^{137}Cs:$ (1) X< 1.3 pCi/g ln Y = 1.15 + 0.666 ln X Y = 3.15X^{0.666} (2) 1.3 pCi/g \leq X< 2.4 pCi/g ln Y = 1.12 + 0.783 ln X Y = 3.05X^{0.783} (3) X \geq 2.4 pCi/g Y = 2.53X

⁹⁰Sr:

1) X< 6.1 pCi/g ln Y = -1.91 + 0.456 ln X Y = 0.148X^{0.456} (2) 6.1 pCi/g \leq X< 10.7 pCi/g ln Y = -2.07 + 0.546 ln X Y = 0.126X^{0.546} (3) X> 10.7 pCi/g Y = 0.043X

Assessment of the Dosage from Terrestrial Foods

<u>Methodology</u> — The quantity of radionuclides ingested via terrestrial foods was computed from the measured and predicted concentrations according to the expected daily diets listed in Table 139 of the chapter on dietary and living patterns. Except for coconut and arrowroot, the daily intake of the food items listed in this table refers to the grams per day of fresh food. The gramper-dav intakes listed for coconut and arrowroot refer to the dry-weight intake of coconut meat (copra) and processed arrowroot starch. Water content of food items used to compute fresh-weight concentrations from dry-weight concentrations were determined from laboratory experience or estimated from the literature. The water content was assumed to be 50% in fresh coconut meat $^{4, 6, 19}$, 95% in coconut milk $^{6, 19}$ and 70% in breadfruit^{6, 19}. Pandanus was initially assumed to be similar to other tropical fruits and have a water content of $80\%^6$, which was subsequently confirmed by Ref. 19. The water content was assumed to be 70% in bird muscle and liver on the basis of poultry data⁶; it was assumed to be 75%in $eggs^6$. In the case of coconut crabs. the water content was assumed to be 81%in liver²⁰ and 62% in hepatopancreas²¹.

Evaluation of the potential dose to the returning population has been structured on the basis of basic living patterns (see Table 135) and involves assessment of the contributions of terrestrial food from certain islands or island groups: (A) ALICE-IRENE, (B) BELLE, (C) JANET, (D) KATE-WILMA + LEROY, and (E) ALVIN-KEITH. Table 193 lists the initial concentrations of the radionuclides in the terrestrial foods from these islands or island groups. Two reference dates are shown on Table 193. The concentrations based on values in