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THE BIOLOGICAL AND GEOGRAPHICAL DISTRIBUTION OF W^{185} IN THE VICINITY OF THE ENIWETOK TEST SITE, APRIL - SEPTEMBER, 1958

By

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ABSTRACT

Radiochemical separations were made by ion-exchange and precipitation techniques and by gamma spectrum analyses to determine the levels of tungsten-185 on selected samples from the Eniwetok and Bikini test site and from the nearby atolls of Rongelap and Ujelang The isotope was identified by its half life, maximum beta energy and gamma energy. High levels of W^{185} were found on plant-leaf and soil samples at Belle and Janet Islands (Eniwetok Atoll) and on plankton collected 155 miles northwest of Eniwetok Atoll during August 1958. Lower levels were found at Vera, Keith, and Henry Islands (Eniwetok Atoll), Nan Island (Bikini Atoll), on the plankton near Ujelang and Rongelap Atolls and on samples from the latter atolls. The W^{185} appeared to occur as surface contamination with the exception of one fish sample taken at Eniwetok Atoll in which the isotope was found in the liver and spleen.

INTRODUCTION

Following the 1954 and 1956 tests of nuclear devices at the Eniwetok Test Site, investigators from both Japan and the United States reported the occurrence of several neutron-induced radioisotopes in marine organisms from that area (Kawabata, 1954; Rinehart et al, 1955; Yamada et al, 1955; Weiss and Shipman, 1956; Amano et al, 1956; Seymour et al, 1957). In later analyses of plankton and fish samples for radioactive isotopes the non-fission products Zn^{65}, Co^{57}, Co^{58}, Co^{60}, Fe^{55}, Fe^{59}, and Mn^{54} were found to account for 50 per cent to 99 per cent of the total radioactivity of the individual samples. However, these isotopes were present in only trace amounts in soils, land plants, and terrestrial animals from the same general area. In addition to the neutron-induced radioisotopes the following fission products were present in the plankton and fish samples: Ru^{106}- Rh^{106}, Zr^{95}- Nb^{95}, and Ce^{144}- Pr^{144}.

The investigations on the uptake and retention of radioisotopes by plankton and fish at the Eniwetok and Bikini test site were continued during the 1958 atomic test series. In the period from August 8 to 14, one of three oceanic radiological surveys was made by members of the Laboratory of Radiation Biology to measure the amounts of radioactivity in sea water

and plankton and to identify the principal radioisotopes in the samples. The USS Collett (DD730) was provided by the U. S. Navy for the work and the survey included thirty-nine collecting stations spaced on a fifty-mile grid (Fig. 1) in the area bounded by 10°00'N and 13°20'N and 160°10'E and 166°00'E. In addition to the above named radioisotopes, a neutron-induced radioisotope, tungsten-185, not previously found at the test site by the present authors, was detected in high amounts in some of the samples. The gamma peak of W^{185} was first observed in gamma spectra made aboard the USS Collett on total plankton samples; however, the isotope was not positively identified until chemical separations were made. In this paper the methods used to establish the identity of the nuclides are described as well as the pattern of distribution of the isotope in plankton and other samples.

MATERIALS AND METHODS

The physical and chemical properties of W^{185} were used to identify the isotope. W^{185} decays principally by the emission of a beta particle of an energy of approximately 0.43 MEV. A gamma component (2.4 per cent) with an energy of 0.055 MEV is also emitted and can be readily identified in the gamma spectra.

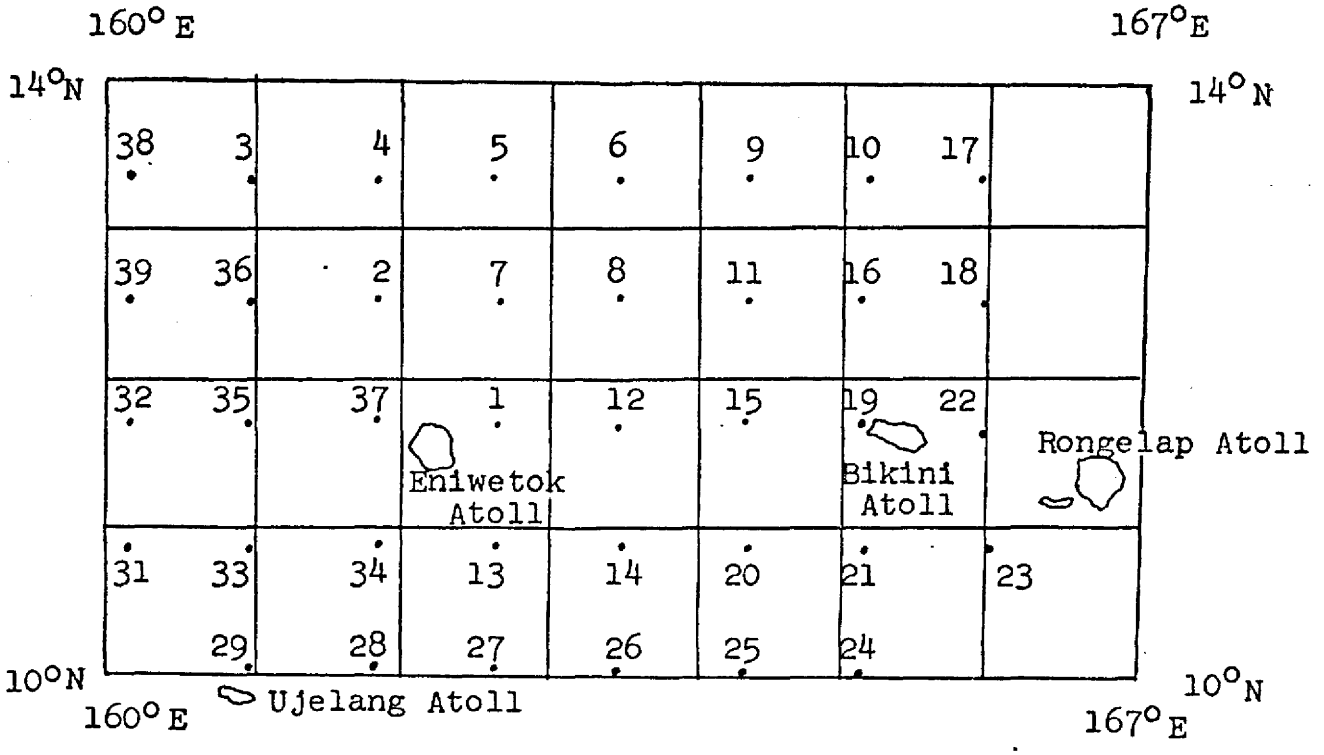

Fig. 1. Chart of the Collett plankton stations.

Gamma spectra were made from plankton samples aboard the Collett by means of a single-channel fifty-position spectrometer to determine the presence of gamma-emitting radioisotopes other than the expected fission products. Selected samples containing mixtures of gamma emitters were wet ashed, dissolved in acid solution and passed through a cation exchange column for the separation of the radionuclides. The radioisotopes were identified by beta and gamma energies and by the elution fractions in which they were found. Confirmation of the presence of W^{185} was obtained by chemical precipitation techniques, by beta and gamma energies and by comparison of sample radionuclide separations with control experiments utilizing spikes of W^{185}. Maximum beta energy was determined by mass absorption techniques.

The precipitation method of analysis (Kleinberg, 1954) for W^{185} consisted of two precipitations of tungstic acid, each followed by the dissolving of the precipitate in concentrated NH_4OH. The solution was then scavenged with $Fe(OH)_3$, and with molybdenum and bismuth sulfides in the presence of tartaric acid. Niobium was removed by extraction with chloroform and cupferron and the tungsten reprecipitated as tungstic acid. The gamma spectrum of the final precipitate showed only W^{185}, free from other gamma emitters.

The radiochemical technique for the separation of the nuclides by means of ion exchange was adapted from the procedure developed by Swartout et al, (1944) and Tompkins et al, (1947).

Preparation of samples

The dried samples were wet ashed with HNO_3 and H_2O_2, evaporated to dryness and redissolved in 0.2N HCl. To check the radiochemical procedures a "control" sample was prepared in which 0.02μc of W^{185} was added to a 300-mg portion of dry Puget Sound fish and then processed in the same manner as the test samples. The samples were filtered and the filtrates were passed through cation resin columns prepared in the following manner.

Preparation of the resin column

Dowex 50*, a sulfonated polystyrene resin of the cationic type, of mesh size 100 - 200, was washed three times with 6N HCl, rinsed three times with distilled water and twice with dilute HCl. The resin suspension was placed in a glass column 6 mm in diameter and 100 mm in length containing a glass wool plug in a constriction at the base of the tube. Three column volumes of 0.2N HCl were run through the resin until the eluate was clear and the desired flow rate was obtained.

* Available from Dow Chemical Co., Midland, Michigan

Elution procedure

The sample was allowed to pass through the column at a rate of 0.2 - 0.5 ml per minute and then the column was washed with 50 ml of 0.2N HCl. The HCl fractions, which contained ions not adsorbed to the resin, are designated as the anion fractions. The cations adsorbed to the resin were removed progressively by passing through the column 50 ml of 0.5 per cent oxalic acid followed by 5 per cent citric acid solutions adjusted to pH 3.5, 4.1, 4.6, 5.1, 5.6 and 6.1 with NH_4OH.

The eluates were collected in test tubes and dried for gamma counting. Aliquots were removed for beta counting. The resin bed was removed from the column at the termination of the elution procedure and was ignited. The ash was counted for beta and gamma activity.

The disintegration rates for the individual isotopes were determined from the beta and gamma counts.

Gamma survey procedure

After the isotope W^{185} was identified in the plankton samples, gamma spectra were made from different types of samples collected in and around the Eniwetok Test Site during the interval April 28 to September 17, 1958. The radiotungsten was identified by the 0.055 MEV gamma peak and the disintegration rates of this isotope as well as those of the other gamma

emitters present in the samples were determined from the gamma curves by use of the method of Conally (1956). A three-inch by three-inch NaI (Tl) crystal was used in conjunction with a 256-channel analyzer. Tests for the accuracy of the gamma spectrum method were made with standards and with radioactive spikes the disintegration rates of which had been determined from beta counts. Accuracy of ± 20 per cent, based on known spike mixtures, was obtained for six of the isotopes (W^{185}, Co^{57}, Cs^{137}, Zr^{95}, Co^{58} and Co^{60}) present in the samples. The lowest limit of detection of any given isotope in the samples was approximately one per cent of the total gamma activity. The determinations of the levels of Ru^{103}-Ru^{106} and Ce^{141}-Ce^{144} are not as accurate as those obtained for the other isotopes because of the overlapping gamma peaks.

The ratio of Ru^{103} to Ru^{106} was calculated in the following manner. The .624 MEV peak of Ru^{106} was determined in selected samples and the contribution of the .513 MEV peak from the same isotope was calculated. This value was subtracted from the composite .498 MEV gamma peak of Ru^{103} and the .515 MEV peak of Ru^{106} in selected samples, and the remainder was assumed to be contributed by Ru^{103}.

The ratio of Ce^{141} to Ce^{144} was calculated in a similar way. The 0.08 MEV and 0.03 MEV peaks of Ce^{144} were determined in selected samples and the contribution of the .134 MEV Ce^{144}

peak was calculated. This value was subtracted from the composite .134 MEV and .145 MEV peaks in the samples. The remainder was considered to be from Ce^{141}.

The values obtained for all isotopes were corrected to the date of collection of the sample.

DISCUSSION OF RESULTS

Ocean survey samples

During the Collett survey (August 8 to 14) gamma spectra were made on plankton samples. Pronounced peaks at 0.055 MEV and smaller peaks at 0.03 MEV, later identified as those from tungsten-185, were observed in some samples but not in others (Fig. 2). When the station positions for those samples contributing the two peaks were plotted on a chart of the area sampled, a pattern of occurrence was observed (Fig. 3). The plankton specimens containing the W^{185} were taken in three areas. The region containing the highest levels of W^{185} in the plankton (19,000,000 d/m/g dry weight) centered 155 nautical miles northwest of Eniwetok Atoll, coincided with an area of high total radioactivity, and was about 180 miles in diameter, with a tongue of activity extending to Eniwetok Atoll. The northern and western boundaries of the radioactive area were not determined. A second area, containing lower levels of W^{185}, was approximately 90 nautical miles west and south of Eniwetok and

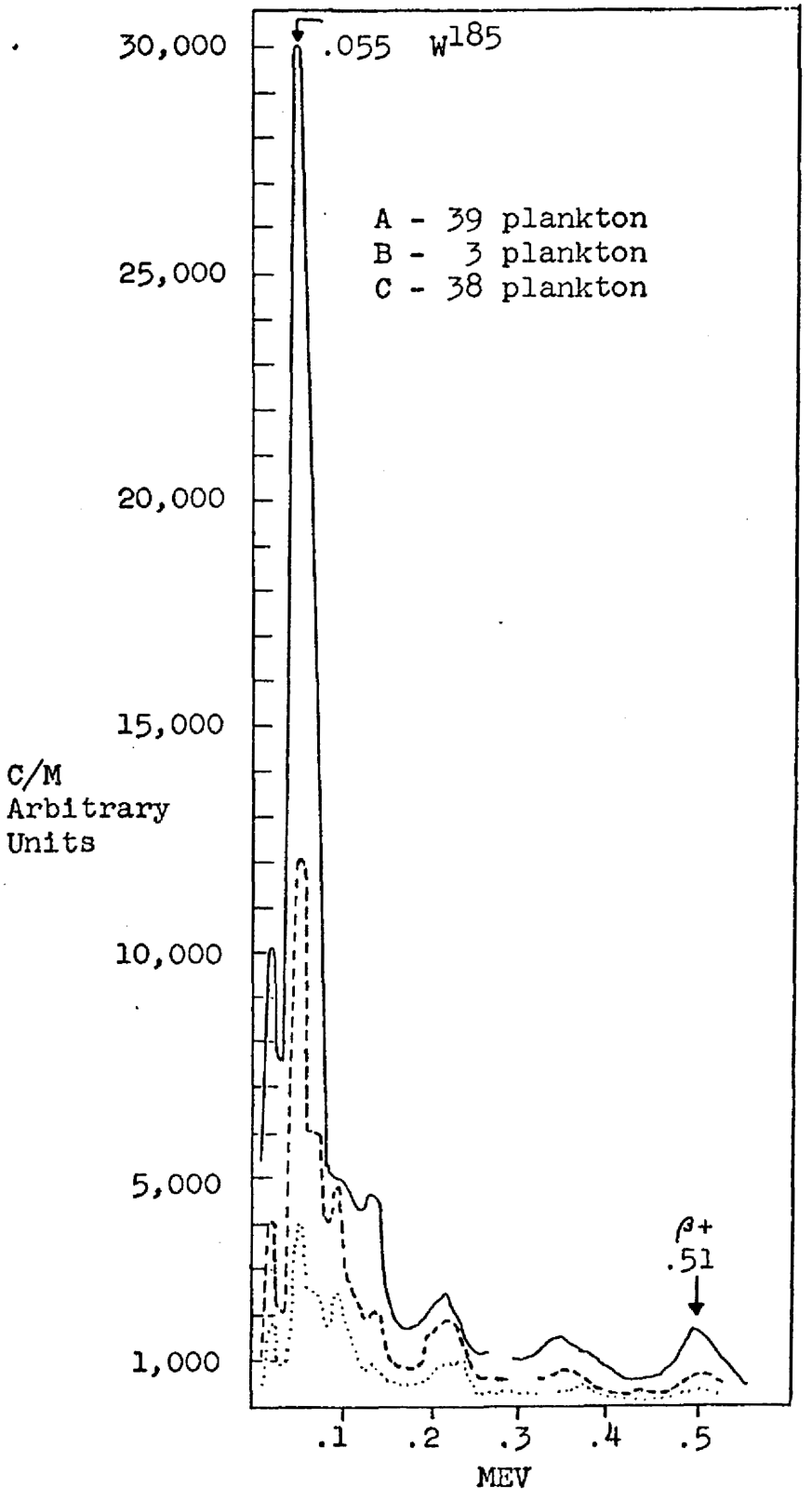

Fig. 2. Gamma spectrum curves of plankton samples collected at stations 3, 38 and 39 during the Collett survey.

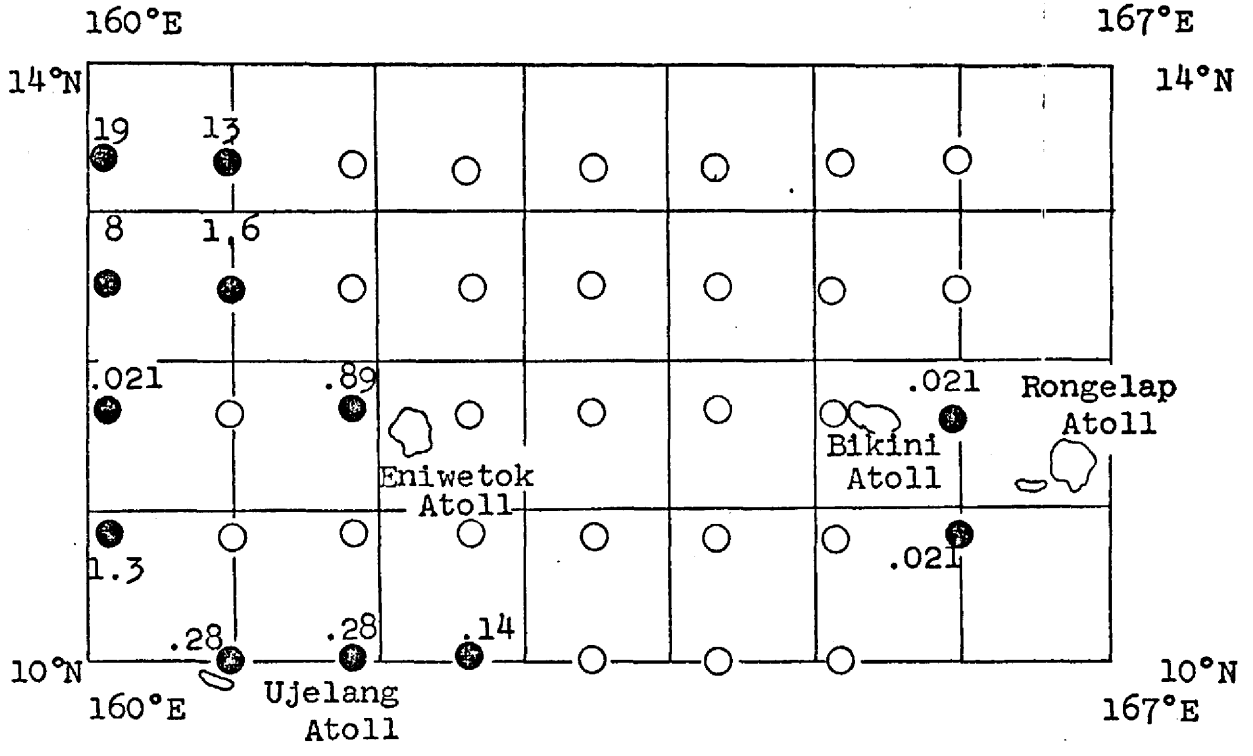

Fig. 3. Chart of the Collett survey stations. Stations at which plankton contained W^{185} are shown as solid circles. W^{185} disintegration rates are in millions per gram dry weight of plankton.

included Ujelang Atoll. Plankton from three stations between the latter contaminated body of water and Eniwetok did not exhibit the W^{185} peaks. The third area where plankton was found to contain W^{185} was east and southeast of Bikini Atoll and included Rongelap Atoll.

The results of ion-exchange chemical separations for all radioisotopes on four plankton samples, two fish tissue samples, and the gamma curve analyses for eight plankton samples are shown (Table 1 and Fig. 3). In Table 1, the disintegration rates of the plankton are given. In plankton samples from stations 36 and 3, W^{185} contributed 60 per cent and 83 per cent respectively of the total radioactivity. These samples did not contain U^{237} nor was it detected in any plankton sample containing W^{185}. In the plankton samples in which W^{185} was absent, the radioisotopes of the transition elements Co, Fe, and Zn and the fission products Ba^{140}-La^{140} contributed almost all of the radioactivity.

The gamma spectrum analysis for W^{185} in those plankton samples containing the isotope is shown at the bottom of Table 1. The highest levels of W^{185} were found in plankton samples from stations 38 and 39.

In Table 1, the disintegration rates per ml of plankton

are given. The amount of W^{185} based on volume is highest in plankton from station 3 rather than from station 38, which contained the highest level based on dry weight of plankton. The values based on dry weights are probably more accurate than those based on volume because of errors in volume determination and variation in plankton species between samples, which result in different volume to dry weight ratios.

During the three-week interval following the Collett survey, the station at which the highest values for plankton were found had shifted 150 miles westward. If this shift represents the movement of the water mass, it is a net advance of 7.3 miles per day and is a close approximation of the known westward movement of the water in this area. At the end of three weeks the center of high activity was again sampled for water and plankton. No W^{185} was found (Fig. 4).

Plankton collected at two stations just north of Ujelang Atoll contained 280,000 c/m/g dry weight of W^{185}. This isotope, which was also detected on soil, plant and animal samples taken at Ujelang Atoll, will be discussed later.

Plankton samples collected half way between Bikini Atoll and Rongelap Atoll contained 21,000 d/m/g dry weight of W^{185}. The isotope was also found in samples from Nan Island at Bikini and from Rongelap Atoll.

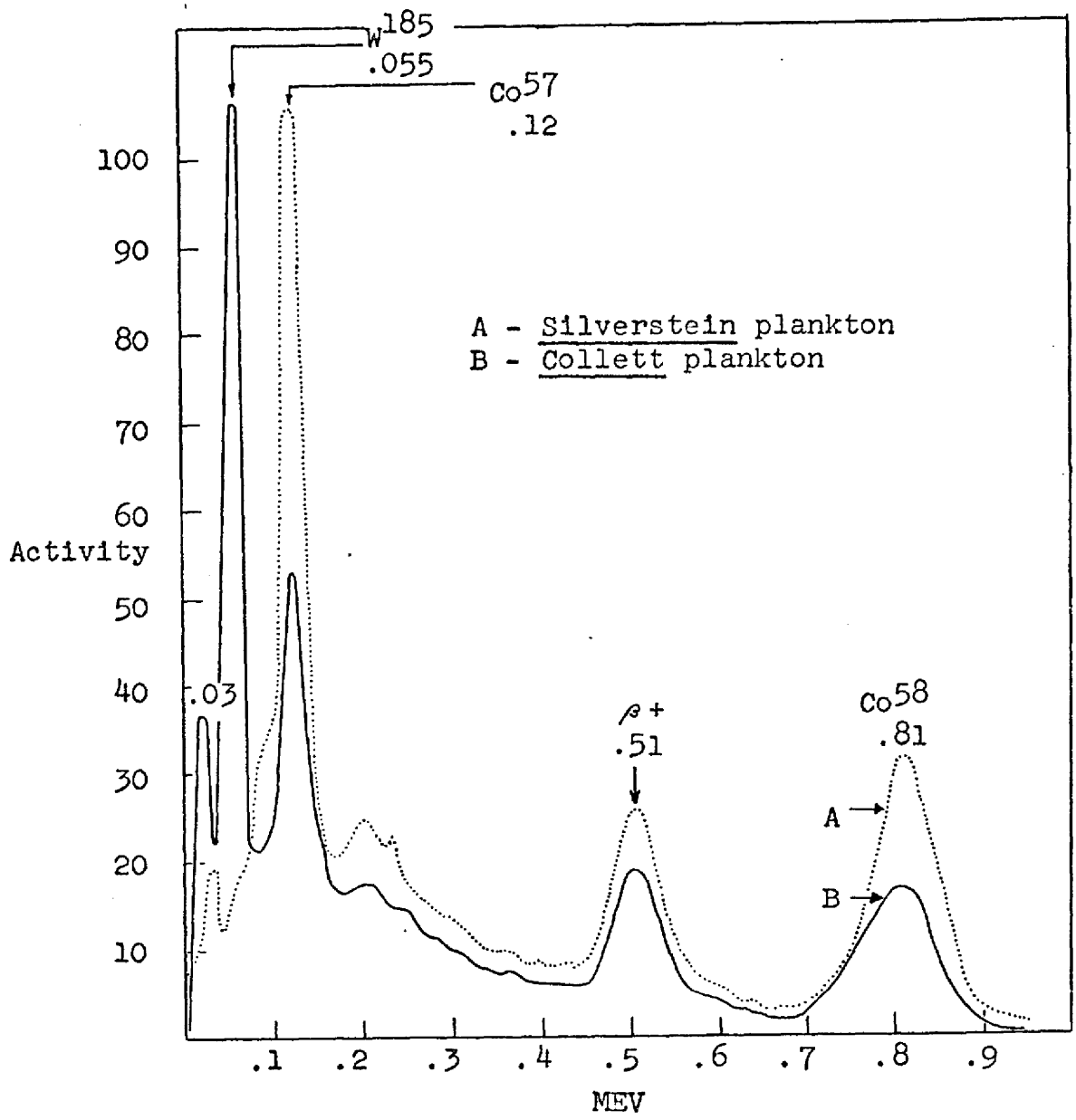

Fig. 4. Gamma spectrum curves of combined plankton samples of the Collett and of the Silverstein survey which was conducted three weeks later. W^{185} was present at a high level in the plankton from the Collett survey but was absent in the plankton from the Silverstein survey. The Collett samples contained higher levels of radioactivity than did the Silverstein samples and the gamma curves are not indicative of the relative gamma activity in the two samples.

Water samples of approximately 500 ml each taken at 0, 25, 50, 100 and 300 meter depths from five stations were pooled on January 13, 1959, filtered through millipore filters (pore size .45μ) and the filtrates dried. The filter papers contained particle sizes greater than .45μ plus the radioisotopes adsorbed to the papers. The radioisotopes with particle sizes less than .45μ plus those in solution were found in the filtrate. The results, corrected to date of collection (August 8 - 14), are as follows:

		W^{185} d/m/l	$Zr^{95}Nb^{95}$ d/m/l	W^{185} + $Zr^{95}Nb^{95}$ d/m/l	Ratio W^{185}/ $Zr^{95}Nb^{95}$
Station No. 3	>.45μ	1,200	100	1,300	12/1
	<.45μ	26,000	160	26,160	160/1
	Total	27,200	260	27,460	96/1
Station No. 31	>.45μ	0	35	35	0
	<.45μ	1,700	62	1,762	27/1
	Total	1,700	97	1,797	18/1
Station No. 37	>.45μ	0	24	24	0
	<.45μ	19,000	230	19,230	83/1
	Total	19,000	254	19,254	75/1
Station No. 38	>.45μ	0	57	57	0
	<.45μ	32,000	210	32,210	150/1
	Total	32,000	267	32,267	120/1
Station No. 39	>.45μ	1,100	210	1,310	5.2/1
	<.45μ	26,000	350	26,350	74/1
	Total	27,100	560	27,660	48/1

On the date of analysis (January 13, 1959), a major part of the radiotungsten was either in solution or in a

colloidal dispersion (<0.2μ) and was present in the filtrate fraction of the water samples. Zr^{95}-Nb^{95} was also found at higher levels in the filtrate fraction than on the filter papers but the ratio was not as high as in the case of W^{185}. In an average of the five samples approximately two per cent of the W^{185} and 30 per cent of the Zr^{95}-Nb^{95} remained on the filter papers.

The distribution of W^{185} and Zr^{95}-Nb^{95} on the filter papers and in the filtrates at the time of analysis probably does not represent the condition at the time of collection. Some leaching of radioisotopes would have occurred in planktonic organisms which died during the interval between collection and analysis. A comparison of the distribution of radioactivity in the above samples with that in aliquots of the same samples filtered at the date of collection indicates that this may be the case. The data are as follows:

Per cent of activity on filter

Water sample stations	Filtered at date of collection	Filtered January 13, 1959
No. 3	32	5
No. 31	13	2
No. 37	17	0.1
No. 38	12	0.2
No. 39	25	5

The per cent of activity remaining on the filters in those samples filtered at date of collection (August 8 - 14, 1958) is 5 to 170 times that found in the samples filtered January 13, 1959. Thus a considerable amount of leaching occurred during the intervening period.

After W^{185} was detected and identified in the samples of plankton from the Collett survey, other specimens collected at Eniwetok, Bikini, Rongelap, Ujelang, Utirik and Wotho Atolls in the Marshall Islands, Kapingamarangi Atoll in the Caroline Islands, Tarawa Atoll in the Gilbert Islands, and Kusaie and Ponape Atolls in the Senyavin Islands were examined by gamma spectrometry for the photopeak (0.055 MEV) of W^{185}. The isotope was found only in samples from Eniwetok, Bikini, Rongelap and Ujelang Atolls.

Eniwetok Atoll Samples

Chemical separations were made on Scaevola leaves from Belle Island by ion-exchange and precipitation methods. In the anions from the ion-exchange column only the peaks of energy represented by W^{185} and Zr^{95}-Nb^{95} were observed in the gamma spectra. $Ru^{103+106}$-$Rh^{103+106}$ were driven off by heat in the ashing process. In the precipitation separation, which is specific for tungsten, only the 0.055 MEV photopeak and the 0.03

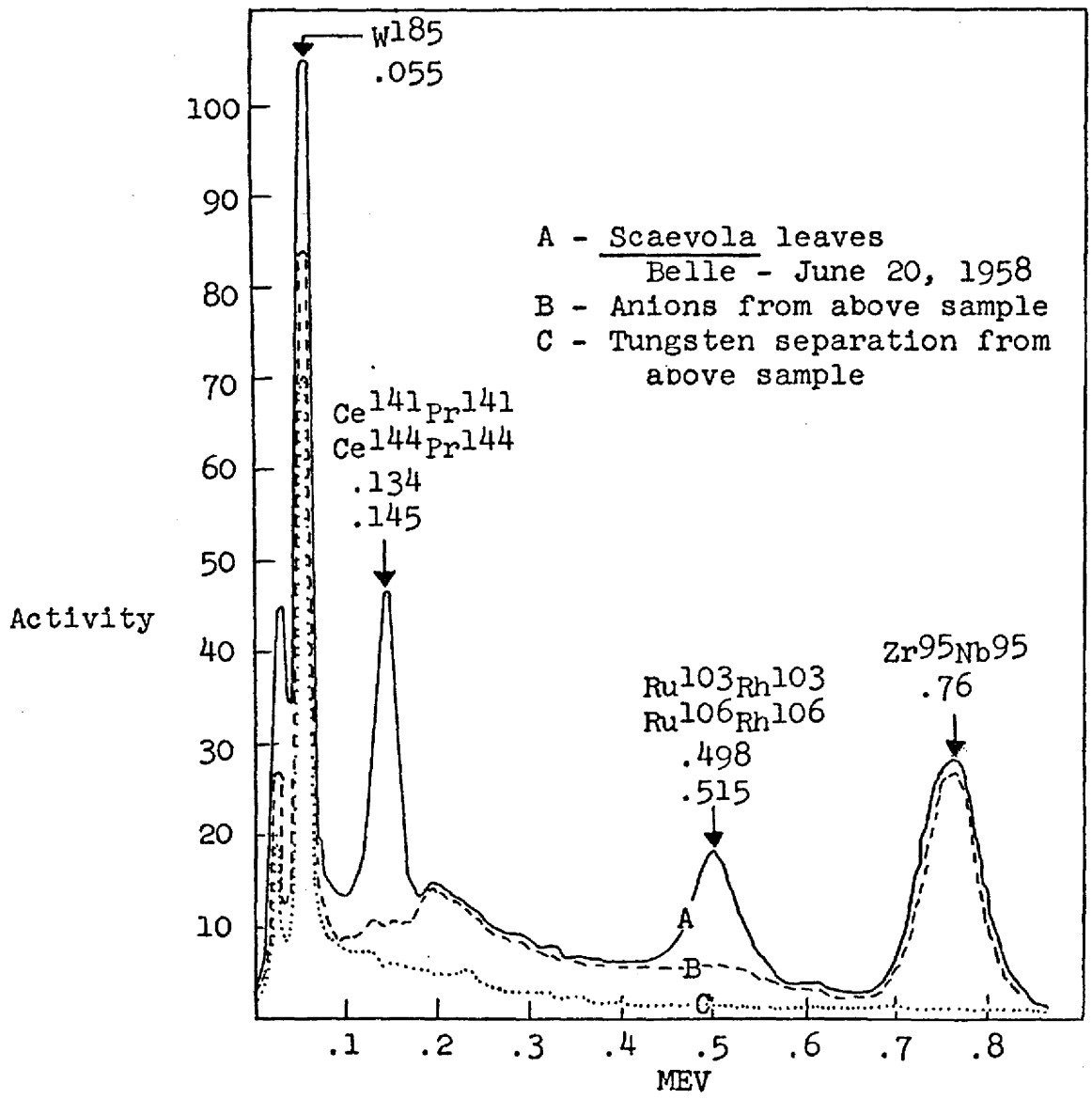

Fig. 5. Gamma spectrum curves of Scaevola leaves taken at Belle Island (Eniwetok Atoll) on June 20, 1958, and of chemical separations from the leaf samples.

MEV escape peak of W^{185} were found in the gamma spectrum (Fig. 5). A maximum beta energy of 0.43 MEV was determined by the mass absorption technique; the half life, determined by beta counting, was found to be approximately 70 days.

Plant-leaf samples collected at Janet Island on April 28, at Belle Island on May 10 (Fig. 6, Tables 2, 3) and at Henry Island on May 8, did not contain W^{185}; Cs^{137} was the only gamma emitter present. However, plant-leaf and soil samples collected at the same islands during June and August did contain radio-tungsten.

Samples of pulverized coral were taken June 5, 1958, from the crater of a nuclear device detonated near Janet Island on May 26. The results of gamma spectrum analyses on this material from the target area are shown in Tables 2 and 3. Only six gamma emitters were detected: W^{185}, Ce^{141}-Pr^{141}, Ce^{144}-Pr^{144}, Ru^{103}-Rh^{103}, Ru^{106}-Rh^{106} and Zr^{95}-Nb^{95}. The disintegration rate of W^{185} per gram of sample was 3,500,000 d/m and accounted for 67 per cent of the total radioactivity. In top soil from Belle Island collected August 20, approximately four and one-half miles from ground zero, W^{185} contributed 68 per cent of the total radioactivity (13,000,000 d/m/square inch of top soil)*.

* Two types of surface soil samples were taken -- top inch and top two inches. In the top inch samples, one cubic inch of soil was considered to represent one square inch of exposed surface. In the two-inch samples, two cubic inches were assumed to represent one square inch.

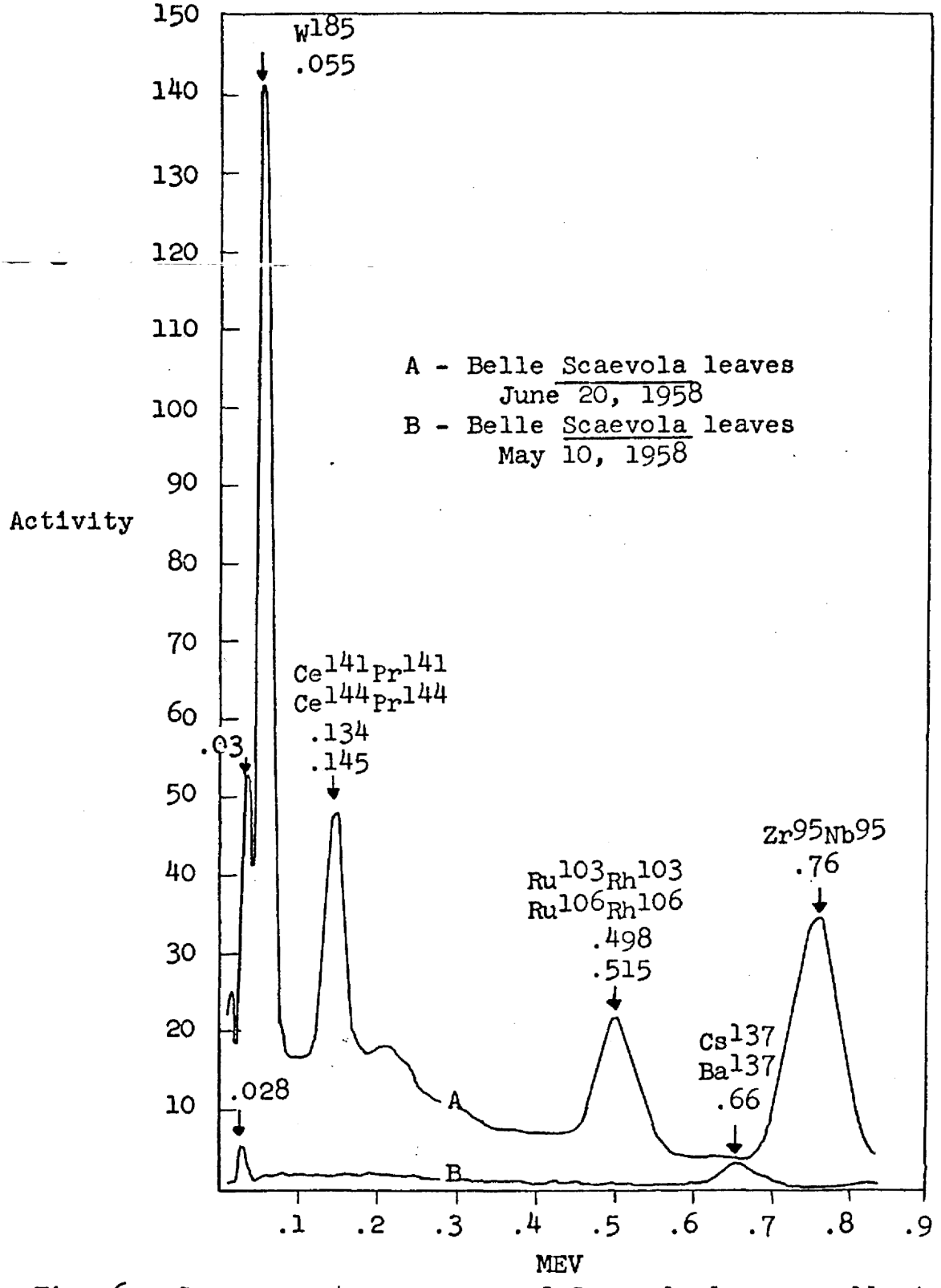

Fig. 6. Gamma spectrum curves of Scaevola leaves collected at Belle Island on May 10, 1958, prior to the W^{185} contamination and of those collected on June 20, 1958, after contamination. The earlier samples contained only Cs^{137}-Ba^{137}.

Table 2. Summary of the disintegration rates of gamma-emitting radioisotopes at date of collection Values given in d/m/g dry weight.

	W^{185}	Co^{57}	$Ce^{141-144}$	$Ru^{103-106}$	Cs^{137}	Zr^{95}	Co^{58}	Co^{60}
ENIWETOK ATOLL								
Janet I April 28, 1958								
Sida	--	--	--	--	1,200	--	--	--
August 20, 1958								
Ipomoea leaves	12,000	--	750	750	1,100	960	--	--
Fimbrystilis tops	200,000	--	5,100	6,500	--	5,800	--	--
Messerschmidia leaves	11,000	--	190	180	180	240	--	--
*Soil	2,300,000	--	740,000	650,000	--	620,000	--	--
Vera I. June 25, 1958								
Caulerpa	23,000	--	49,000	19,000	--	230,000	--	--
Ipomoea stems and flowers	15,000	--	21,000	3,000	--	2,700	--	--
Messerschmidia leaves	14,000	--	6,600	8,900	--	20,000	--	--
September 27, 1958								
Tridacna visceral mass	--	3,000	--	270,000	--	82,000	20,000	1,500
August 26, 1958								
Scaevola fruit	890	--	700	600	--	870	--	--
Belle I. May 10, 1958								
Scaevola leaves	--	--	--	--	540	--	--	--
Messerschmidia fruits and flowers	--	--	--	--	540	--	--	--
June 20, 1958								
Scaevola leaves (fair protection)	1,600,000	--	110,000	220,000	--	350,000	--	--
Scaevola leaves (poor protection)	7,900,000	--	750,000	1,300,000	--	1,900,000	--	--
August 20, 1958								
Scaevola leaves	180,000	--	19,000	21,000	1,800	26,000	--	--
Messerschmidia leaves	380,000	--	17,000	20,000	4,500	30,000	--	--
Triumfetta leaves	440,000	--	44,000	31,000	5,500	44,000	--	--
Portulaca leaves	650,000	--	20,000	24,000	12,000	29,000	--	--
*Soil - top inch	13,000,000	--	1,100,000	2,000,000	--	3,000,000	--	--
*Soil - 8 - 10 inches	270,000	--	4,600	16,000	trace	14,000	--	--
Polysiphonia	1,100,000	***	***	380,000	--	750,000	***	***
Spyridia filamentosa	1,000,000	***	***	300,000	--	480,000	***	***
Padina commersonii	910,000	***	***	580,000	--	1,500,000	***	***
Halimeda stuposa	600,000	***	***	110,000	--	170,000	***	***
Leroy I. June 28, 1958								
Scaevola leaves	**	--	420,000	440,000	--	1,100,000	--	--
Keith I. August 20, 1958								
Scaevola leaves	1,100	--	310	350	--	620	--	--
Messerschmidia leaves	--	--	--	trace	trace	trace	--	--
Henry I. May 8, 1958								
Scaevola leaves	--	--	--	--	--	--	--	--
June 23, 1958								
Scaevola leaves	2,500	--	2,200	7,500	--	11,000	--	--
Messerschmidia leaves	24,000	--	5,100	5,900	--	11,000	--	--
Coconut meat	--	--	--	--	10	38	--	--
August 20, 1958								
Scaevola leaves	510	--	840	680	--	1,000	--	--
Messerschmidia leaves	--	--	--	--	450	--	--	--
*Soil - top inch	--	--	9,500	12,000	--	7,500	--	--
Yellowwood Crater June 5, 1958								
Sludge from bottom	3,500,000	--	330,000	790,000	--	590,000	--	--
RONGELAP ATOLL August 8, 1958								
Scaevola leaves	35,000	--	1,500	1,400	--	1,300	--	--
July 27, 1958								
Breadfruit	--	--	--	--	100	160	--	--
Arrowroot	--	--	--	--	130	--	--	--
Coconut milk	--	--	--	--	29	--	--	--
UJELANG ATOLL July 19, 1958								
Scaevola leaves	2,000	--	280	430	--	440	--	--
Messerschmidia leaves	1,400	--	180	370	--	360	--	--
*Soil - top inch	1,000	--	1,500	2,800	--	810	--	--
BIKINI ATOLL								
Nan I. August 28, 1958								
Clerodendron leaves	9,800	--	310	310	--	310	--	--
Scaevola leaves	7,500	--	240	290	--	320	--	--
Pandanus - inside of fruit	--	--	--	--	210	--	--	--
Udotea indica	--	--	16,000	3,400	--	9,200	--	--
*Soil - top 2"	13,000	trace	18,000	5,200	850	--	2,300	trace
Acanthurus liver Zn^{65} 16,000	--	3,800	--	--	--	--	17,000	780

* $d/m/in^2$

** W^{185} not detected due to high compton interference

*** Present in high amount

Table 3. Per cent of total radioactivity contributed by gamma-emitting isotopes at date of collection.

	W^{185}	Co^{57}	Ce^{141} Ce^{144}	Ru^{103} Ru^{106}	Cs^{137}	Zr^{95}	Co^{58}	Co^{60}	Total d/m/g
ENIWETOK ATOLL									
Janet I. April 28, 1958									
Sida	--	--	--	--	100	--	--	--	1,200
August 20, 1958									
Ipomoea leaves	77	--	5	5	7	6	--	--	15,560
Fimbrystilis tops	92	--	2	3	--	3	--	--	217,400
Messerschmidia leaves	93	--	2	1	1	2	--	--	11,790
*Soil	53	--	17	15	--	14	--	--	4,310,000
Vera I. June 25, 1958									
Caulerpa	7	--	15	6	--	72	--	--	321,000
Ipomoea stems and flowers	36	--	51	7	--	6	--	--	41,700
Messerschmidia leaves	28	--	14	18	--	40	--	--	49,500
September 27, 1958									
Tridacna visceral mass	--	1	--	72	--	22	5	trace	376,500
August 26, 1958									
Scaevola fruit	29	--	23	20	--	28	--	--	3,060
Belle I. May 10, 1958									
Scaevola leaves	--	--	--	--	100	--	--	--	540
Messerschmidia fruits and flowers	--	--	--	--	100	--	--	--	540
June 20, 1958									
Scaevola leaves (fair protection)	70	--	5	10	--	15	--	--	2,280,000
Scaevola leaves (poor protection)	67	--	6	11	--	16	--	--	11,850,000
August 20, 1958									
Scaevola leaves	73	--	8	8	1	10	--	--	247,800
Messerschmidia leaves	84	--	4	4	1	7	--	--	451,500
Triumfetta leaves	78	--	8	5	1	8	--	--	564,500
Portulaca leaves	89	--	3	3	2	4	--	--	735,000
*Soil - top inch	68	--	6	10	--	16	--	--	19,100,000
*Soil - 8-10 inches	89	--	2	5	--	5	--	--	304,600
Leroy I. June 28, 1958									
Scaevola leaves	--	--	21	23	--	56	--	--	1,960,000
Keith I. August 30, 1958									
Scaevola leaves	46	--	13	15	--	26	--	--	2,380
Messerschmidia leaves	--	--	--	--	--	--	--	--	0
Henry I. May 8, 1958									
Scaevola leaves	--	--	--	--	--	--	--	--	0
June 23, 1958									
Scaevola leaves	11	--	10	32	--	47	--	--	23,200
Messerschmidia leaves	52	--	11	13	--	24	--	--	46,000
Coconut meat	--	--	--	--	21	79	00	00	48
August 20, 1958									
Scaevola leaves	17	--	28	22	--	34	--	--	2,440
Messerschmidia leaves	--	--	--	--	100	--	--	--	450
*Soil - top inch	--	--	33	41	--	26	--	--	29,000
Yellowwood Crater June 5, 1958									
Sludge from bottom	67	--	6	15	--	11	--	--	5,210,000
RONGELAP ATOLL August 8, 1958									
Scaevola leaves	89	--	4	4	--	3	--	--	39,200
July 27, 1958									
Breadfruit	--	--	--	--	39	61	--	--	260
Arrowroot	--	--	--	--	100	--	--	--	130
Coconut milk	--	--	--	--	100	--	--	--	29
UJELANG ATOLL July 19, 1958									
Scaevola leaves	64	--	9	14	--	14	--	--	3,150
Messerschmidia leaves	61	--	8	16	--	16	--	--	2,310
*Soil - top inch	17	--	25	46	--	13	--	--	6,110
BIKINI ATOLL									
Nan I. August 28, 1958									
Clerodendron leaves	92	--	3	3	--	3	--	--	10,730
Scaevola leaves	90	--	3	4	--	4	--	--	8,350
Pandanus - inside of fruit	--	--	--	--	100	--	--	--	210
Udotea indica	--	--	57	12	--	32	--	--	28,600
*Soil - top 2"	33	--	46	13	2	--	6	--	39,350
Acanthurus liver Zn^{65} 43	--	10	--	--	--	--	45	2	37,580

* $d/m/in^2$

At Janet Island, which was washed by the water wave from the detonation, W^{185} accounted for 53 per cent of the total radioactivity in top soil (Table 3). The contamination by this isotope is assumed to have been produced in the May 26 detonation.

Analyses were made of soil samples taken at two depths (top inch and 8-inch to 10-inch depth) at Belle Island to determine the relative rate of leaching of W^{185} through coral soil in comparison to that of Ce, Ru and Zr. Based on these samples, the radioelements leached through the soil in the following order (Fig. 7).

	Per cent of activity at 8 - 10 inches compared to surface activity
W^{185}	2.1
$Ru^{103+106}$-$Rh^{103+106}$	.8
Zr^{95}-Nb^{95}	.5
$Ce^{141+144}$-$Pr^{141+144}$	.4

W^{185} leached from the top inch of soil to a depth of 8 to 10 inches two and one-half times as fast as radioruthenium, four times the rate of radiozirconium and five times as fast as radiocerium. The above conclusions are valid only if all four of the radioelements were deposited on the ground at approximately the same time.

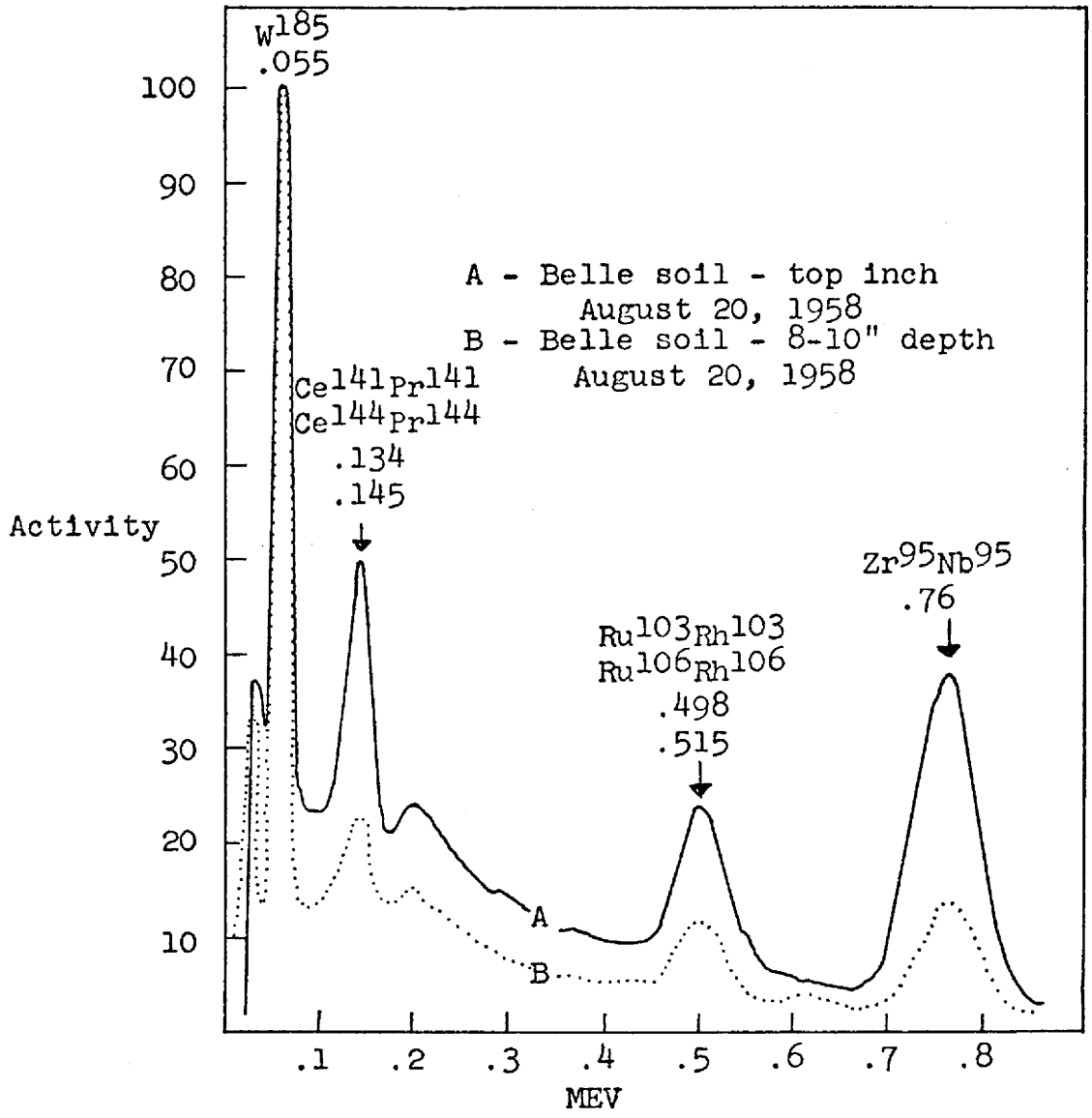

Fig. 7. Gamma spectrum curves of Belle top soil and of soil taken at a depth of 8 - 10 inches. The W^{185} peaks were arbitrarily equated to illustrate the greater relative amount of W^{185} at depth in relation to the other isotopes.

W^{185} accounted for 67 to 70 per cent of the total radioactivity in samples of Scaevola leaves taken at Belle Island on June 20, 1958 (Table 3). On August 20 the same isotope contributed 73 per cent of the total activity in a similar sample. The disintegration rates of the August 20 samples corrected to the June 20 date, however, indicate that W^{185} would have contributed 66 per cent of the total radioactivity at that time, the percentage difference being due to the presence of more radioisotopes with a short half life in the earlier sample. In Scaevola leaves taken from Henry Island on June 23, W^{185} contributed 17 per cent of the total activity. On August 20 the same isotope accounted for 11 per cent of the disintegration rate. When the August 20 disintegration rates were corrected to the June 23 date, W^{185} contributed 12 per cent of the total radioactivity. Thus the rate of removal of this isotope from the Scaevola leaves by rain and wind is not significantly different from the average of the other radioisotopes.

The disintegration rate of W^{185} on Scaevola leaves at Belle Island on June 20 was 7,900,000 d/m/g and on August 20 180,000 d/m/g, a reduction in activity by a factor of 44. The disintegration rate of W^{185} on Scaevola leaves at Henry Island on June 23 was 2,500 d/m/g and on August 20, 510 d/m/g, a

reduction by a factor of 4.9. The fact that Belle Island was washed by the water wave from at least one other test device subsequent to the contamination by W^{185} and Henry Island was not, may account for the greater reduction in activity on Scaevola leaves at Belle Island.

Four species of algae collected at Belle Island on August 20 had high levels of W^{185} as follows:

Polysiphonia	1,100,000 d/m/g
Spyridia	1,000,000 d/m/g
Padina	910,000 d/m/g
Halimeda	600,000 d/m/g

The W^{185} activity in the algae samples is related to the surface area-to-weight ratio in the four species. Polysiphonia and Spyridia are branching, finely divided forms and have the greatest surface area per unit weight. Padina is flabellate with a smaller surface area and Halimeda is a heavy-jointed calcareous form with the least surface area per unit weight. The limited data available on the levels of W^{185} in the above algae samples suggest that the contamination is present mainly on the surface, although a greater number of samples would be necessary to show with certainty that this is the case. The levels of W^{185} in the algae were higher than those in the land plants at the same island (Table 2).

The average disintegration rate of this isotope in the four algae was approximately twice that found in four species of land plants as based on disintegration rate per gram of material.

Another species of algae (Caulerpa) at Vera Island contained about one and one-half times as much W^{185} as did two species of land plants from the same island.

Because W^{185} was found in the marine algae, other samples of marine organisms from Eniwetok lagoon also were examined for this isotope. The giant clam (Tridacna gigas) is known to concentrate anions in the visceral mass (Lowman - unpublished); therefore, samples of Tridacna visceral mass were collected September 27, 1958, at Vera Island where W^{185} was known to be present. The results of ion-exchange chemical separations on this organ are shown in Tables 2 and 3. W^{185} was not present but two other anion radioelements were found -- $Ru^{103+106}$-$Rh^{103+106}$ (270,000 d/m/g dry weight, 72 per cent of total activity and Zr^{95}-Nb^{95} (82,000 d/m/g dry weight, 22 per cent of total activity). In addition to the anions, isotopes of the transition element cobalt were present in small amounts (Co^{57}, one per cent; Co^{58}, 5 per cent; Co^{60}, trace).

In a dogtooth tuna (Gymnosarda nuda) sample taken from Eniwetok lagoon on June 22, however, W^{185} was found in low level in the spleen (11,000 d/m/g dry weight, 4 per cent of

total activity) and in the liver (15,000 d/m/g dry weight, 16 per cent of total activity) (Tables 2 and 3). It was not present in the kidney, red muscle, or white muscle. This specimen is the only one in which W^{185} was detected with certainty within tissues of an organism.

In general, the levels of W^{185} in samples from the islands of Eniwetok Atoll reflected the pattern of distribution found at the plankton stations. Thus, the island (Belle) northwest of the target area (near Janet Island) had very high levels of W^{185}. The islands south (Vera) and southwest (Henry, Keith, and Leroy) of the target area had lower levels of the isotope.

Ujelang Atoll

The W^{185} contamination on the plant leaves and in the soil at Ujelang Atoll, collected July 19, 1958, was low in comparison with that at Eniwetok, with the exception of Leroy and Keith Islands, where low levels of W^{185} were observed also. The ratio of W^{185} to the other gamma-emitting isotopes in the Ujelang plant samples, however, was similar to the ratios found at Eniwetok. Thus in Scaevola and Messerschmidia leaves at Belle Island (Eniwetok Atoll) for which the disintegration rates were corrected to July 19, W^{185} contributed 66 per cent

and 82 per cent respectively of the total disintegration rate of the gamma-emitting isotopes. At Ujelang, in the same species, W^{185} accounted for 61 per cent and 64 per cent respectively of the total disintegration rate (Table 3). The presence of W^{185} on the leaves at Ujelang indicates that the radioactivity found in these samples is from recent fallout (Fig. 8).

In the Ujelang soil, on July 19, W^{185} contributed only 17 per cent of the total radioactivity in comparison with 89 per cent for the same isotope at Belle Island corrected to the same date. Janet Island soil contained W^{185} at a level of 48 per cent of the total activity corrected to July 19 (Table 3). The low value at Janet Island in comparison to that of Belle Island is due to the former island's having been washed by the water wave. The low ratio of tungsten at Ujelang Atoll in comparison to that of samples from both Belle and Janet Islands suggests that a significant part of the radioactivity in Ujelang soil was from earlier fallout, the new fallout, containing W^{185}, contributing only part of the total activity. A similar situation was observed in the soil sample from Henry Island at Eniwetok Atoll.

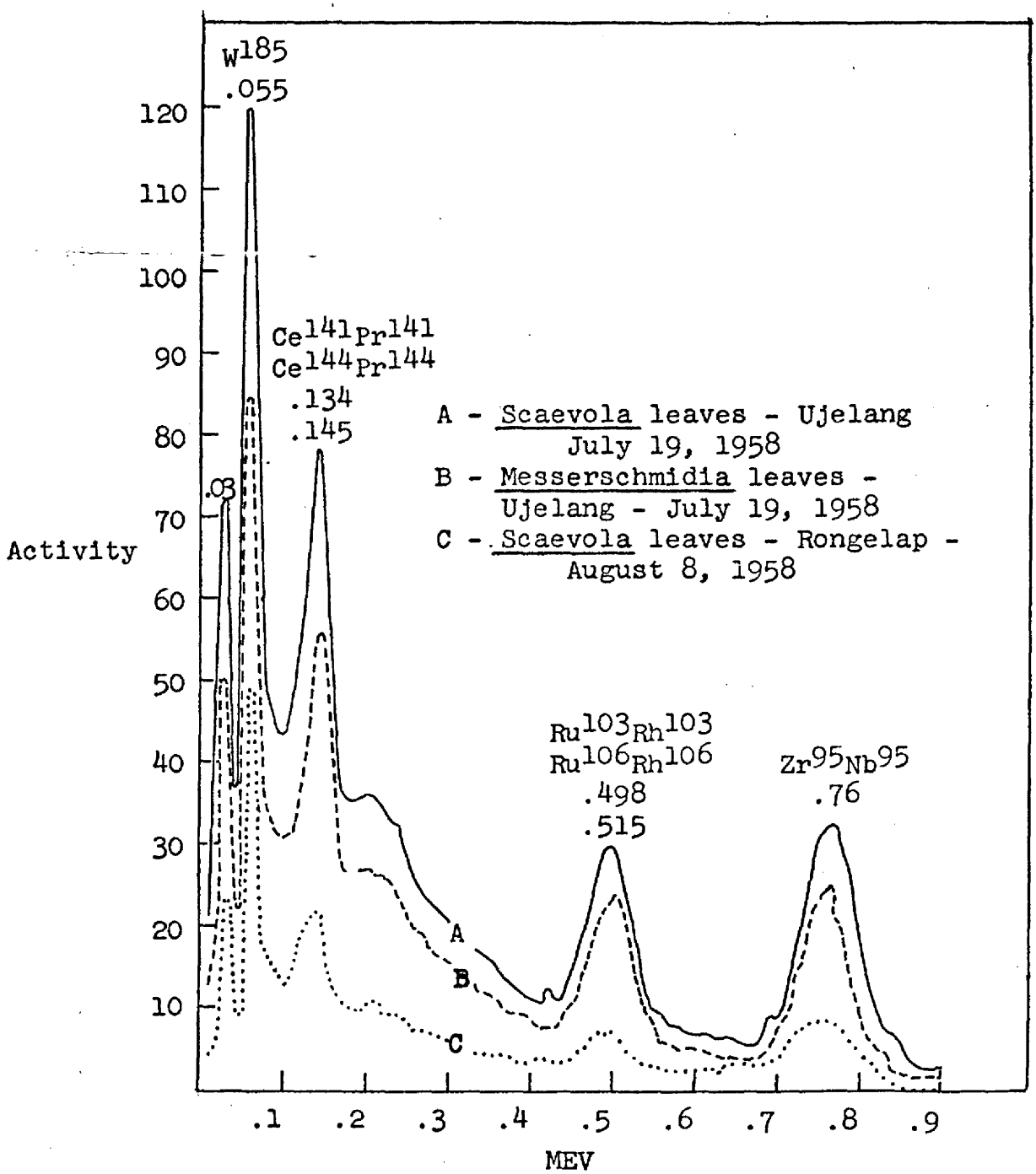

Fig. 8. Gamma spectrum curves of Scaevola leaves from Ujelang and Rongelap Atolls and of Messerschmidia leaves from Ujelang Atoll.

Rongelap and Bikini Atolls

The W^{185} content on Scaevola leaves at Rongelap Atoll on August 8, 1958, was higher than that in the same species at Nan Island (Bikini), Keith, Henry, and Leroy Islands (Eniwetok), and Ujelang Atoll (Table 3). These levels suggest that Rongelap Atoll received a greater amount of fallout than did the above islands with the exception of Leroy Island at Eniwetok Atoll. The latter island was contaminated heavily with fallout not containing W^{185} as evidenced by the comparative levels of $Ce^{141+144}$-$Pr^{141+144}$ (420,000 d/m/g), $Ru^{103+106}$-$Rh^{103+106}$ (440,000 d/m/g) and Zr^{95}-Nb^{95} (1,100,000 d/m/g) (Table 2) to those at Keith and Henry Islands.

W^{185} accounted for 89 per cent of the total radioactivity on Scaevola leaves collected at Rongelap on August 8, 1958, (Table 3, Fig. 8) and 90 per cent on leaves of the same species collected at Nan Island (Bikini) on August 28. Thus, the contamination on the plant leaves at both Bikini and Rongelap Atolls may be assumed to be due to recent fallout. In topsoil from Nan Island, W^{185} accounted for only 33 per cent of the total; the isotope was not detected in soil from Rongelap (Table 3). The ratios of W^{185} in the soil samples from Nan Island and Rongelap Atoll indicate that a significant part of the contamination was due to earlier fallout.

W^{185} was not detected in breadfruit, arrowroot, or coconut milk at Rongelap or in the internal parts of Pandanus fruit from Nan Island, Bikini Atoll. Since the above samples represent the internal parts of plants and did not contain W^{185}, although leaf samples from the same area did contain high levels of the isotope, it is probable that W^{185} is not taken up in significant amounts by plants but occurs rather as surface contamination.

SUMMARY

1. Chemical separations by ion-exchange resin column and precipitation techniques were made on selected samples collected at the Eniwetok Test Site during the summer of 1958. The chemically separated radioactive isotopes were identified by determination of maximum beta energies, gamma energies, and half life.

2. The presence of W^{185} and other gamma-emitting isotopes on or in other samples from the Eniwetok Test Site and from the nearby populated atolls of Rongelap and Ujelang was determined by means of gamma spectrum analysis of whole samples.

3. W^{185}, first observed on plankton collected approximately 150 nautical miles northwest of Eniwetok, contributed up to 83 per cent of the total activity of the gamma-emitting isotopes in the sample. Plankton specimens contaminated by W^{185} were taken in two other general areas, one approximately 90 nautical miles southwest of Eniwetok and another southeast of Bikini Atoll. Plankton taken outside these areas did not contain radiotungsten nor was it found in any plankton samples collected three weeks later in the same area.

4. Scaevola leaves from Belle Island (Eniwetok Atoll) contained a high amount of W^{185} (67 to 70 per cent of total activity). In ion-exchange separations this isotope was observed only in the anion fractions. Only the 0.055 MEV and 0.03 MEV gamma energy peaks of W^{185} were found in a precipitation separation specific for tungsten.

5. In soil samples from Belle Island collected August 20, 1958, W^{185} appeared to leach to a depth of eight to ten inches, two and one-half times as fast as radioruthenium, four times the rate of radiozirconium, and five times as fast as radiocerium.

6. The rate of removal of W^{185} from Scaevola leaves by rain and wind is not significantly different from the average of the other radioisotopes.

7. The level of W^{185} on algae samples collected at Belle Island on August 20, 1958, was higher than that on land plants, based on activity per unit weight.

8. In a dogtooth tuna specimen taken from Eniwetok lagoon on June 22, 1958, W^{185} was found in the spleen and liver. Other fish sampled failed to show the presence of this isotope.

9. W^{185} contamination on plant leaves and in the soil at Ujelang Atoll was low in comparison to that at Eniwetok Atoll

10. The level of W^{185} on Scaevola leaves at Rongelap Atoll on August 8, 1958, was higher than that at Nan Island (Bikini), Keith, Henry, and Leroy Islands (Eniwetok) and Ujelang Atoll.

11. W^{185} was not detected in the internal parts of any of the plant samples examined in this survey.

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