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By
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February 14, 1958

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Technical Information Service Extension, Oak Ridge, Tenn.

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RADIONUCLIDES IN PLANKTON NEAR THE MARSHALL ISLANDS, 1956

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ABSTRACT

Radiochemical separations were made on plankton samples collected in and west of the Eniwetok Proving Ground in September 1956. Ion exchange resin column and precipitation techniques were used. Fission products, mainly Zr^{95}-Nb^{95} and Ce^{144}-Pr^{144}, contributed an average of 29 per cent of the total radioactivity. The remaining 71 per cent of the activity was contributed by the non-fission radioisotopes Zn^{65}, $Co^{57,58,60}$, Fe^{55} and Mn^{54}. Radioactive zinc, cobalt, and iron accounted for averages of 24, 26 and 21 per cent respectively of the total radioactivity. Mn^{54} was present in trace amounts. Variations in ratio of occurrence for the different non-fission products with change in geographical location was observed. Relatively high levels of Zn^{65} were centered in the area near Bikini and Eniwetok Atolls. The area of high levels of radioactive cobalt and iron in comparison to Zn^{65} was located approximately 480 miles west and slightly north of Eniwetok Atoll.

RADIONUCLIDES IN PLANKTON NEAR THE MARSHALL ISLANDS, 1956

Introduction

During the summer and fall of 1956 members of the Applied Fisheries Laboratory conducted two surveys in the region of the Pacific Ocean bounded by the Marshall, Caroline, and Marianas Islands. The purpose of the surveys was to measure the levels of radioactivity in the water, plankton, and fish and to determine the westward boundary of the contaminated area.

The first survey was made June 11-21, 1956, during the weapons testing period and included 53 collection stations in the ocean 11° N to 14° N and 159° E to 166° E. The results were reported in U. S. Atomic Energy Commission report UWFL-46 (6). The second survey conducted from September 1-20, 1956, about six weeks after the conclusion of the weapons tests, included 74 stations between 9° N and 15° N and approximately 145° E and 166° E (Fig. 1). The findings were published in U. S. Atomic Energy Commission report UWFL-47 (21).

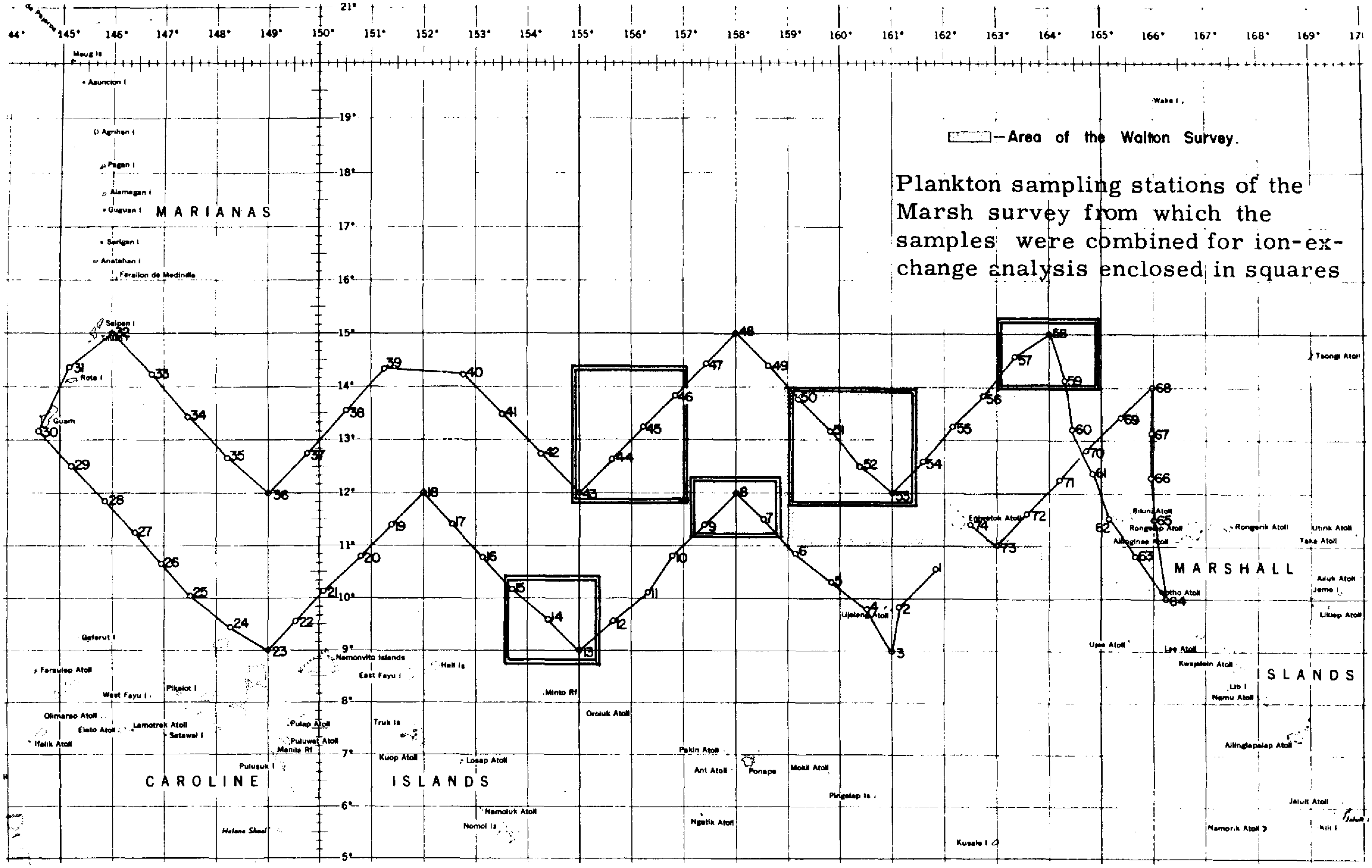
Chemical separations for fission products, cobalt, and zinc were made on a limited number of samples from both surveys, the results of which were reported in the latter report (21). According to these data fission products contributed a major portion of the total radioactivity. Gamma spectra obtained from the same samples, however, indicated that non-fission-product radioisotopes contributed more of the total radioactivity than was observed in the chemical separations, which were designed primarily for determination of fission products. Consequently a detailed study was made to determine the radioisotopic content of several plankton samples collected during the September survey. In addition, gamma spectra were determined for these and most other plankton samples collected in the latter survey. The results are reported in this paper.

Materials and Methods

The equipment and methods used for collecting the samples at sea have been described in previous reports (6, 21).

The initial counting for beta activity was done aboard ship soon after collection of the plankton samples, and the remainder of the plankton from each station was preserved in 10 per cent neutralized formalin in sea water and returned to the Seattle Laboratory.

The concentration of plankton in the tropical seas and the levels of radioactivity in the plankton are both limited. thus plankton samples from adjacent stations were combined to provide sufficient material for analysis. Figure 1 shows the track of the September survey and the groups of stations



▭ – Area of the Walton Survey.

Plankton sampling stations of the Marsh survey from which the samples were combined for ion-exchange analysis enclosed in squares

Fig. I. Track of the Marsh and area of the Walton survey.

from which samples were combined for analysis.

Five plankton samples were analysed in detail for their radioisotopic content eleven months after the date of collection. These included

Sample No.	Stations
I	7, 8, 9
II	13,14,15
III	43,44,45,46
IV	50,51,52,53
V	57,58,59

The samples were filtered through No. 1 Whatman filter papers and the plankton and filtrate fractions dried separately. Gamma spectra were made from both fractions. The plankton samples were then wet ashed with concentrated nitric acid and hydrogen peroxide and converted to the chloride form by evaporating the sample to partial dryness repeatedly with hydrochloric acid. The moist salts were dissolved in 0.2 N HCl, filtered, and the filtrate passed through a cation-exchange column (5 mm x 100 mm) of Dowex 50 (13, 24, 12, 16).

Two hundred and fifty mgm or less of the sample in 0.2 N HCl solution were passed through the column at a rate of 0.2 - 0.5 ml per minute, then the resin was washed with 50 ml of 0.2 N HCl. The eluate from the sample and the subsequent hydrochloric acid wash contained those ions not adsorbed to the resin, *i.e.* the anions. These included Zr^{95}-Nb^{95} and/or

Ru^{106}-Rh^{106}. The cations that were adsorbed onto the resin were differentially removed next by passing 40 ml of 0.5 per cent oxalic acid and 40 ml each of 5 per cent ammonium citrate solutions at pH 3.5, 4.1, 5.1, and 6.1 through the column. Oxalic acid removed cationic Zr^{95}-Nb^{95} and Fe^{55}. Ammonium citrate at pH 3.5 removed Ce^{144}-Pr^{144}, Mn^{54}, $Co^{57,58,60}$, and Zn^{65}*. Ammonium citrate at pH 4.1 removed Cs^{137}. No radioisotopes were detected in the pH 5.1 or 6.1 fractions when the fractions were counted for beta or gamma activity.

The pH 3.5 fractions were dried, wet ashed and redissolved in 5 ml of concentrated HCl (12M) and run through a column (7 mm x 250 mm) of an anionic resin, Dowex 1, to separate Mn, Co, and Zn from each other according to the method of Kraus and Moore (15). The sample was added to the column and the fractions were eluted at a flow rate of 0.1 to 0.2 ml per minute. The elutrients included 10 ml each of 12M, 6M, 4M, 2.5M, and 0.5M HCl, 20 ml of .005M HCl, and 10 ml of H_2O. Under the conditions of the experiment Mn^{54} was eluted in the 12M wash, $Co^{57,58,60}$ in the 6M fraction, and Zn^{65} in .005M HCl.

From the fractions eluted out of the Dowex 50 column one-ml aliquots of the HCl fractions were dried, treated with

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*These radioisotopes were subsequently separated from each other on a second column.

concentrated nitric acid, dried and diluted with water, then plated on one and one-half-inch stainless steel planchets. One-ml aliquots of the oxalate and citrate fractions were plated directly on the planchets and dried. The samples were counted for beta activity in a windowless methane gas-flow chamber. The oxalate plates were counted after drying, after flaming, and with a filter of 4.7 mg per cm^2 aluminum in order to identify the radiation from Fe^{55} (16).

The remainder of each fraction was dried and analysed with a gamma spectrometer equipped with a two-inch, well-type, sodium iodide crystal. The isotopes were identified by their gamma energies, maximum beta energies, half lives, and elution patterns.

Beta and gamma counts were converted to disintegrations per minute (d/m) by the use of correction factors previously described (16). Disintegration rates for the individual radioisotopes were corrected to the date of collection.

The fractions eluted from the second column (Dowex 1) were counted in solution for gamma activity and the fractions containing the peaks of radioactivity from Mn^{54}, $Co^{57,60}$, or Zn^{65} were combined into three groups (Mn, Co, Zn), dried, and the disintegration rate of each group determined.

Radiochemical separations in duplicate for Sr^{90}-Y^{90} were made on each of the five samples and their filtrates according to the method outlined in the chemical procedures of the Health and Safety Laboratory (New York Operations Office) (11).

Results

The non-fission products Co^{57}, Co^{58}, Co^{60}, Zn^{65} and Mn^{54} contributed almost all of the activity in gamma spectrum curves made from filtered plankton samples eleven months after collection, masking the weaker peaks from the fission products Zr^{95}-Nb^{95}, Rh^{106}, Ba^{137m}(Cs^{137}) and Ce^{144} (Fig. 2). The gamma peaks of the latter isotopes were detected only in the fractions eluted from the ion-exchange columns (Fig. 3).

Although the non-fission products, in each instance, contributed the major part of the gamma activity in the plankton, they were not always present at a constant ratio to each other in the different samples. Thus in some samples the amount of Zn^{65}, based on the gamma spectra, was high in comparison to radioactive cobalt, (Fig. 2, plankton stations 50, 51, 52, 53, $\frac{Co^{57}}{Zn^{65}} = \frac{0.12}{1.0}$) and in others the reverse relation existed (Fig. 2, plankton station 42, $\frac{Co^{57}}{Zn^{65}} = \frac{5.5}{1.0}$). This variation was related to the site of collection. A similar variation was observed in the levels of some other non-fission radioisotopes and will be discussed later.

Detailed descriptions of the elution patterns from ion-exchange resin columns and the identification of the separated fission and non-fission radioisotopes appear in an earlier paper (16) and are not repeated here. However, the gamma spectrum curves for plankton samples 50,51,52,53 and the separated fractions are shown in Figure 3 to illustrate the small

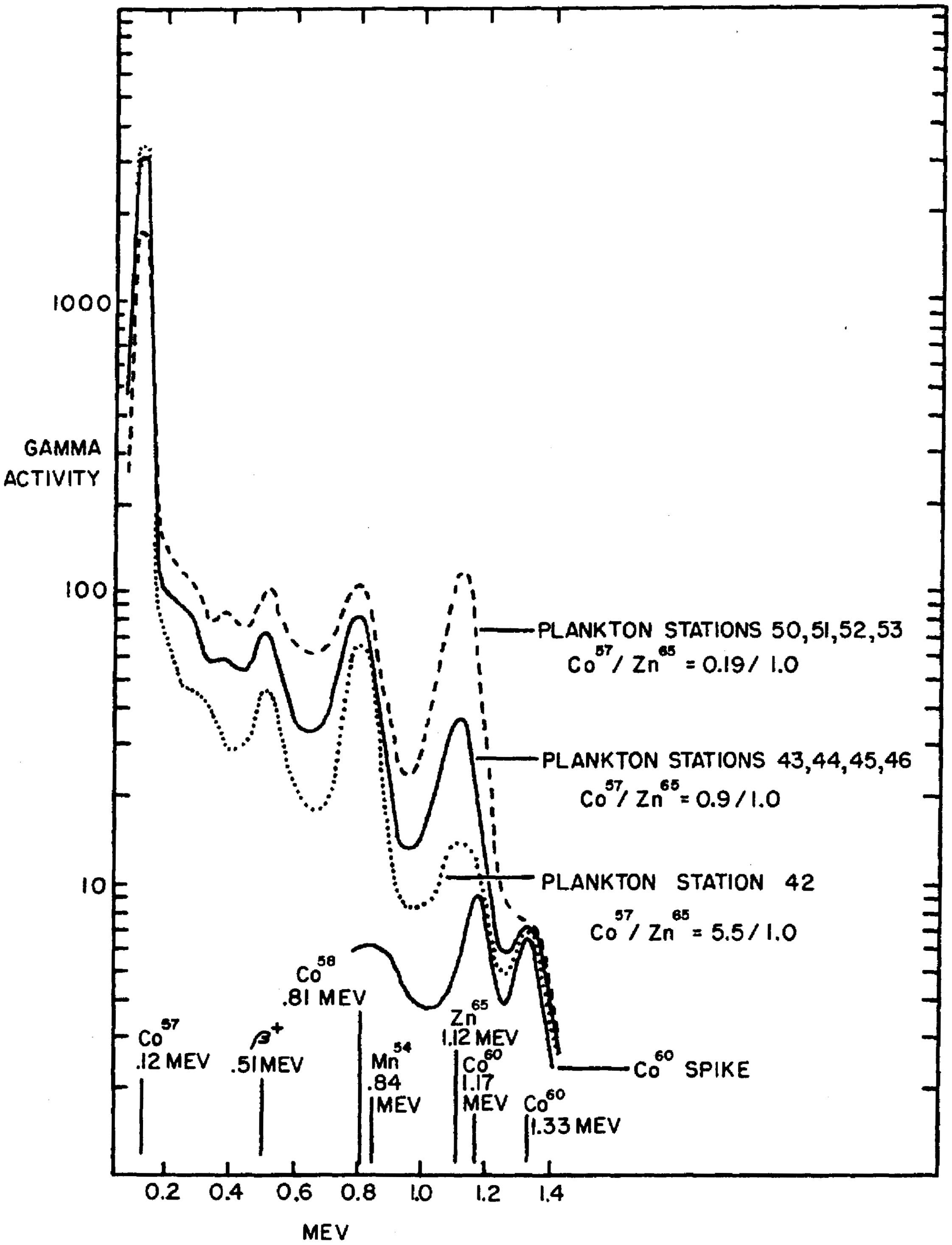

Fig. 2 Gamma spectrum curves from three areas west of Eniwetok Atoll showing the variation in ratio of Co^{57}/Zn^{65}. The gamma peaks of Mn^{54}, Co^{58}, and Co^{60} are also indicated. The gamma peaks of a Co^{60} spike are shown for comparison.

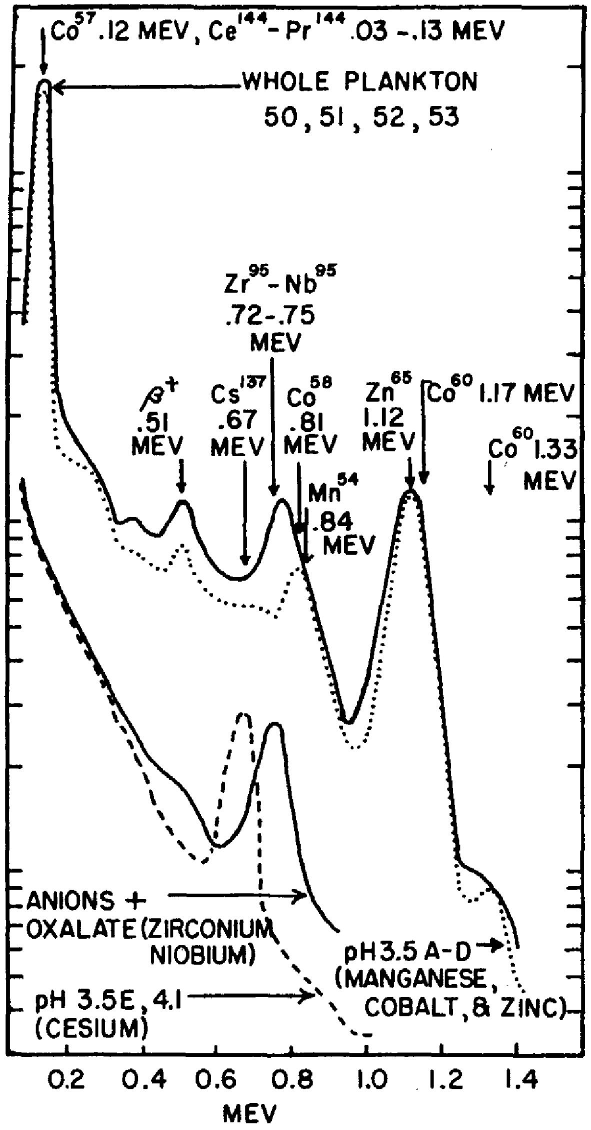

Fig. 3a Gamma curves of fractions eluted from Dowex 50. Zr^{95} was eluted by HCl (anions) and oxalate, Cs by pH 3.5-4.1 citrate, and radioactive Zn, Co and Mn by pH 3.5 a-d citrate. The separation of these elements is shown in Figure 3b.

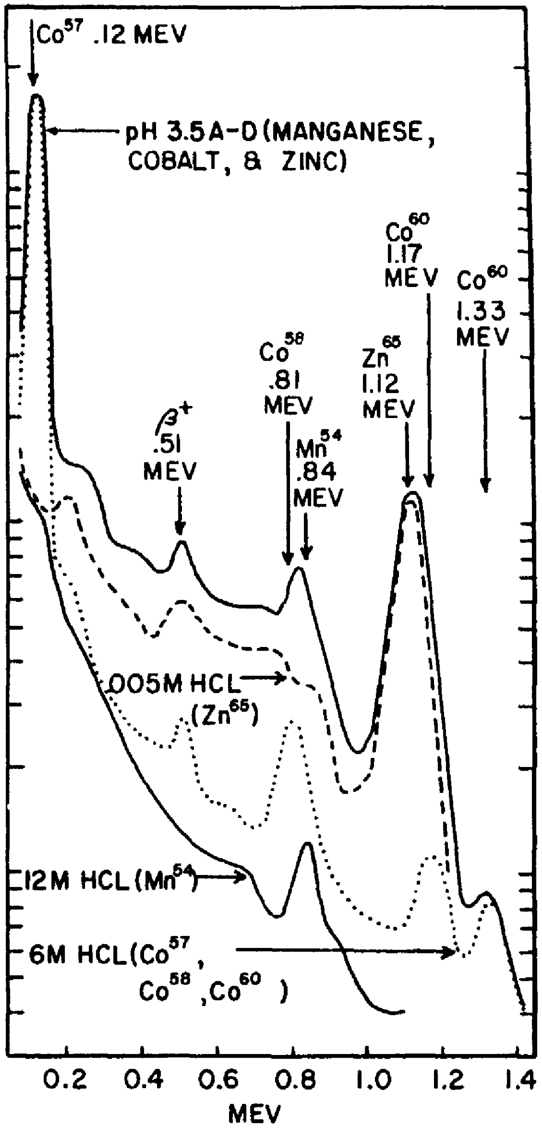

Fig. 3b Gamma curves on a second elution of the pH 3.5 Dowex 50 fraction on Dowex 1. The separations of Mn^{54}, Zn^{65} and the three Co radioisotopes are shown.

amount of cross-contamination between the radioactive elements separated by a combination of the Dowex 50 cation-exchange method and the Dowex 1 chloride complexing technique.

The results of the radioisotopic separations are summarized in Tables 1 and 2. In these samples the predominant fission-product radioisotopes were the relatively short-lived Zr^{95}-Nb^{95} (65 d), Ce^{144}-Pr^{144} (285 d), and in one sample, Ru^{106}-Rh^{106} (1 yr). The fission products accounted for 22 to 37 per cent of the total radioactivity in the three analyses from which cerium determinations were made. In the other two samples fission products contributed at least 5.7 per cent (stations 57,58,59) and 44 per cent (stations 13,14, 15) of the total radioactivity.

Sr^{90}-Y^{90} were not detected in the ion-exchange separations nor were they found in fuming nitric acid precipitations made on duplicate aliquots from both the five samples and their filtrates.

Of the non-fission radioisotopes, Fe^{55}, Co^{57}, Co^{58} and Zn^{65} contributed almost all of the activity. Although Co^{60} was present at an average level of only about one per cent, it has the longest half life (5.3 yrs) of the reported non-fission radioisotopes and is therefore important.

The variability, mentioned previously, between the ratios of Co^{57} to Zn^{65} also was evident between Co^{57} and Co^{60}, and between Co^{57} and Fe^{55}. In Figure 4a, the ratios of Co^{57}/Co^{60} in the five plankton samples are shown; also, these

Table 1. Results of radiochemical separations by ion exchange columns and precipitation techniques on five plankton samples in and near the Pacific Proving Ground.*

Isotope	I Plankton (7,8,9) 75,000 d/m/g ash Per cent of total activity	II Plankton (13,14,15) 28,700 d/m/g ash Per cent of total activity	III Plankton (43,44,45,46) 108,000 d/m/g ash Per cent of total activity	IV Plankton (50,51,52,53) 127,000 d/m/g ash Per cent of total activity	V Plankton (57,58,59) 48,000 d/m/g ash Per cent of total activity
Ru^{106}-Rh^{106}	7.1	0	0	0	0
Zr^{95}-Nb^{95}	24.9	44.0	12.5	13.7	5.7
Ce^{144}-Pr^{144}	4.7	not determined	13.2	8.0	not determined
Sr^{89-90}**	0	0	0	0	0
$\mathrm{Cs}^{137}(\mathrm{Ba}^{137m})$	0	0	0.1	0.4	0
Mn^{54}	0.1	trace	0.3	0.3	0.3
Fe^{55}	19.4	19.2	12.7	27.6	38.8
Fe^{59}	0	0	0	0	0
Co^{57}	9.9	8.5	15.5	6.1	4.6
Co^{58}	15.0	13.8	25.9	10.8	5.7
Co^{60}	0.9	0.8	1.3	0.8	1.0
Zn^{65}	18.0	13.8	17.3	32.3	43.9

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* The data are corrected to date of collection

** Chemical separations for Sr^{89-90} were made on duplicate aliquots in addition to the resin column separations. Neither Sr^{89} nor Sr^{90} was detected.

Table 2. Disintegration rates (d/m/g) of the radioisotopes separated from five plankton samples on (A) date of analysis (July 1957) and (B) corrected to date of collection (September 1956).

Isotope	I Plankton (7,8,9)		II Plankton (13,14,15)		III Plankton (43,44,45,46)		IV Plankton (50,51,52,53)		V Plankton (57,58,59)	
	A	B	A	B	A	B	A	B	A	B
Ru^{106}-Rh^{106}	1,100	1,930	0	0	0	0	0	0	0	0
Zr^{95}-Nb^{95}	266	6,800	522	13,400	460	11,800	350	8,960	82	2,100
Ce^{144}-Pr^{144}	607	1,275	*	*	5,940	12,500	2,500	5,250	*	*
Sr^{89-90}	0	0	0	0	0	0	0	0	0	0
Cs^{137}	0	0	0	0	136	139	229	234	0	0
Mn^{54}	10	20	trace	trace	137	274	109	218	51	102
Fe^{55}	4,360	5,300	4,880	5,860	10,000	12,000	15,100	18,100	11,900	14,300
Fe^{59}	0	0	0	0	0	0	0	0	0	0
Co^{57}	1,240	2,720	1,180	2,600	6,700	14,700	1,822	4,000	628	1,700
Co^{58}	242	4,100	249	4,200	1,450	24,500	421	7,110	125	2,110
Co^{60}	2,182	249	203	231	1,090	1,240	458	522	312	356
Zn^{65}	2,100	4,900	1,830	4,200	7,120	16,400	9,200	21,200	7,050	16,200

* Not determined

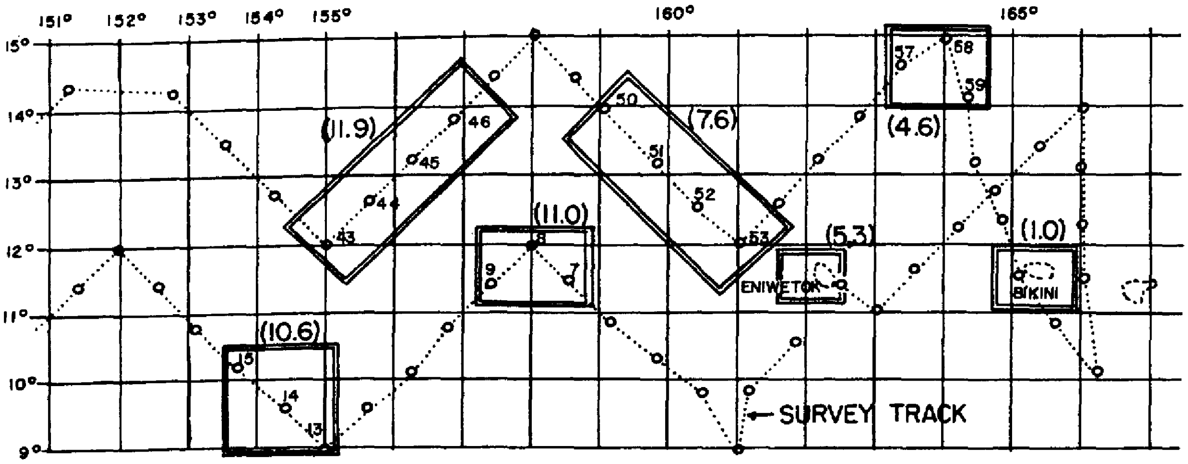

Fig. 4a Ratio Co^{57}/Co^{60} (in parentheses) at collection date from stations 7,8,9; 13,14,15; 43,44,45,46; 50,51, 52,53; 57,58,59; Bikini Atoll; and Eniwetok Atoll.

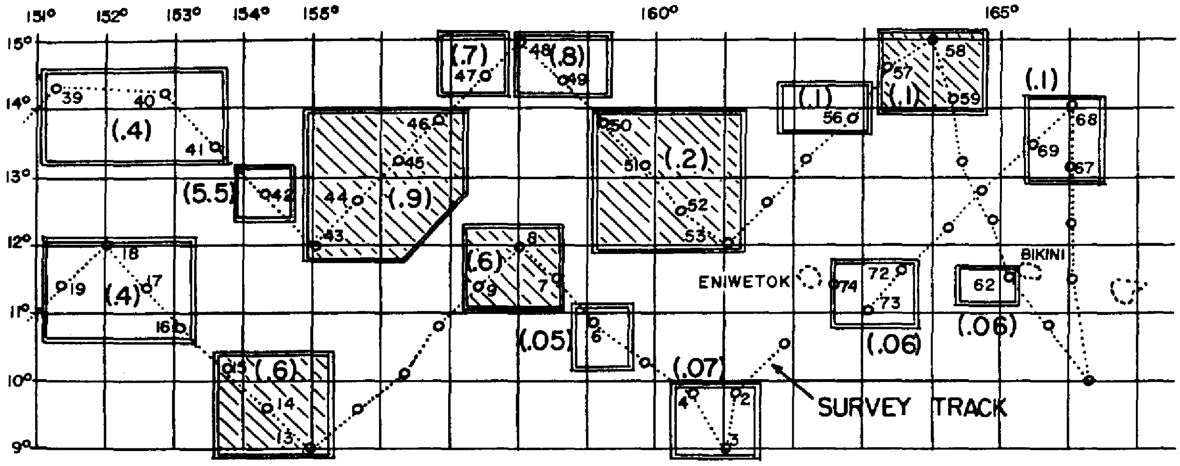

Fig. 4b Ratio Co^{57}/Zn^{65} (in parentheses) at collection date from the plankton stations; Bikini Atoll; and Eniwetok Atoll.

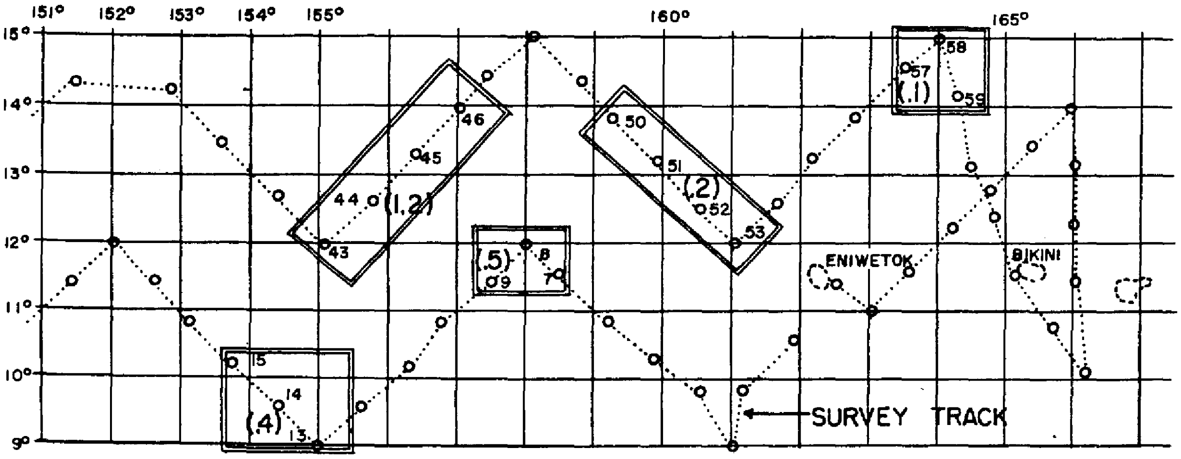

Fig. 4c Ratio Co^{57}/Fe^{55} (in parentheses) at collection date from stations 7,8,9; 13,14,15; 43,44,45,46; 50,51, 52,53; and 57,58,59.

ratios for other biological samples from within Eniwetok and Bikini Atolls, reported in an earlier paper, are given (16). Co^{57} and Co^{60} were present at Bikini Atoll in a 1:1 ratio; at Eniwetok in a 5.3:1 ratio. A similar difference in levels of the two cobalt isotopes also occurred in the plankton taken in the open sea in the vicinity of the Pacific Proving Ground. Thus, at stations 57, 58, 59, approximately 200 miles northwest of Bikini Atoll, the ratio Co^{57}/Co^{60} is 4.6:1.0 and the value 150 miles northwest of Eniwetok Atoll is 7.6:1.0. The ratio of Co^{57}/Co^{60} in an area 300 miles wide north to south and 200 to 500 miles west of Eniwetok was constant at a ratio of approximately 11:1.

The ratios of Co^{57}/Zn^{65} for the plankton stations are shown in Figure 4b. The values for the five plankton samples on which ion-exchange separations were made are shown cross-hatched; the remaining Co^{57}/Zn^{65} ratios for plankton were calculated from the gamma spectra on total samples. The ratios for Eniwetok and Bikini samples were taken from ion-exchange separation data in an earlier report (16).

The ratio of Co^{57}/Zn^{65} at both atolls was 0.06:1.0, indicating a relatively high level of Zn^{65} with respect to Co^{57}. The same condition prevailed at stations 3, 4, 5, and 6, 200 miles south and west of Eniwetok Atoll. However, the ratio north of Eniwetok and Bikini along the northern boundary of the restricted zone was slightly higher (0.1:1.0) than that of the atolls. The center of high Co^{57}/Zn^{65} ratio (5.5:1.0) was observed at station 42 approximately 480 miles west and slightly north of Eniwetok Atoll and did not coincide with the center of greatest total radioactivity which occurred approximately 110 miles due north of Eniwetok Atoll (21). The Co^{57}/Zn^{65} ratios decreased with increasing distance in all directions from station 42.

The ratios of Co^{57}/Fe^{55} for the five plankton samples are shown in Figure 4c. The ratios are similar to those of Co^{57}/Zn^{65}. The values are as follows:

Station	$\frac{Co^{57}}{Zn^{65}}$	$\frac{Co^{57}}{Fe^{55}}$
7,8,9	0.6	0.5
13,14,15	0.6	0.4
43,44,45,46	0.9	1.2
50,51,52,53	0.2	0.2
57,58,59	0.1	0.1

Discussion

The retention or rejection of a given radioactive element by the organisms in the contaminated area of the ocean is determined by several factors, many of which also function in the distribution of naturally occurring elements of the sea. The effects of some of these factors are known; others are not. A better understanding of the unknown effects may be attained by observations on the accumulation of individual radioactive elements from fallout within the trophic levels of the contaminated biosphere. By observing which radioelements are concentrated in the phytoplankton, the zooplankton, and the nekton and relating these observations to (a) the known characteristics of the fallout elements before and after entry into the sea; (b) the half lives of the radioisotopes, the amounts produced at detonation, and their distribution with respect to fallout particle size; and (c) the known biological factors involved in the uptake and retention of different elements, the eventual fate of the major fallout radioisotopes in the sea may be determined.

In the waters of the open Pacific Ocean the naturally occurring trace elements, zinc, cobalt, ruthenium, manganese, cesium, strontium, and possibly zirconium and cerium, in the water are present mainly in solution. The small amount of naturally occurring iron occurs in the colloidal and particulate form. All of the above named elements, however, except cesium and strontium, when introduced into the sea from fallout, would be present most likely in the insoluble or particulate state. Indirect evidence (8, 20) and direct observations (9) support this view. The indirect evidence is based on geochemical studies in which the potential supplies of the elements to the seas from weathering of igneous rocks are compared with the amounts of the elements present in solution in the sea. These values cannot be applied directly to fallout analyses but they do provide the basis for an estimate of the fraction of a given fallout element that would remain in solution in the sea.

The levels in the sea (ppm) of the naturally occurring forms of those elements reported in the present work are shown in Table 3. Also listed are the forms of the naturally occurring elements in sea water, the percentage of the fallout elements in solution according to direct and indirect evidence, the principal chemical state of the fallout in sea water, and the average percentages of the fallout elements observed in plankton samples collected about three months after fallout.

Of the radioactive fallout elements shown, only two, cesium and strontium, would occur normally in soluble form in the sea. The radioisotopes of these elements were found in the least amount in plankton. In contrast, the radioactive fission products with the lowest solubilities, Zr^{95} and Ce^{144}, were present in plankton in the greatest amount (20.2, 8.6 per cent)

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Table 3. Levels in the sea of naturally occurring nuclides of elements also present as radioisotopes in fallout. Also shown are the physical forms of the naturally occurring elements in sea water, per cent solution of fallout radioisotopes in sea water, and percentages of radioisotopes observed in plankton.

Isotope	Gram/ton of stable element in sea water (ppm) (8)	Form of stable element in sea water	Fallout in sea water			Average percentage of radio-element in five plankton samples
			Per cent in solution		Principal form of fallout elements in sea water	
			Indirect evidence (8)	Direct evidence (9)		
Zn^{65}	.014	mostly solution***	.02	--	particulate	25.1
$Co^{57,58,60}$	.0001	solution**	.0007	--	"	24.1
Fe^{55}	.02	particulate**	.00007	--	"	23.5
Zr^{95}-Nb^{95}	?	?	--	1.0	"	20.2
Ce^{144}-Pr^{144}	.0004	?	.001	0	"	8.6
Ru^{106}-Rh^{106}	?	?	--	0	"	1.4
Mn^{54}	.01	mostly solution**	.002	--	"	0.2
Cs^{137}	.002	mostly solution	--*	70	solution	0.1
Sr^{89-90}	.13	mostly solution	7.2	87	solution	0
Stable calcium	400.	solution				
" potassium	380.	solution				

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* A value of .05 per cent is given for cesium. The apparent insolubility is caused by a strong adsorption of the cesium ion to colloidal particles of hydrolysate sediments and not to its insolubility in sea water.

** Goldberg 1954 (7).

*** Kawabata, personal communication.

for this group of radioelements. This observation is in agreement with the findings of the Taney survey (10) in which the major fission products in plankton were found to be Ce^{144}-Pr^{144}. However, a gamma spectrum curve made on these plankton samples November 1957 by the present author revealed an appreciable amount of Co^{60}. Zr^{95}-Nb^{95} were not found in the Taney samples because of the short half life of Zr^{95} (65d).

In the case of the non-fission-product radioelements, zinc, cobalt, and iron were present in the plankton (based on an average of the five samples) in approximately equal amounts at about 24 per cent each and manganese at a level of less than one per cent of the other three elements. All of these elements, from fallout in the sea, would exist initially in the insoluble form.

Limited data from both control experiments (4) and field observations (21) suggest that the major route for uptake of fallout radioisotopes by marine animals is through the ingestion of radioactive particles. Chipman (5) noted also that, in addition to the filter feeding animals, two species of algae were able to concentrate Ce^{144} several thousand times over the levels in the water although the radioisotope was in the particulate form. The present work is in agreement with the postulate that the major source of radioactive elements from fallout to marine zooplankton is through the uptake of particulate material. The uptake may

*** Kawabata, personal

involve either adsorption of the particle onto the organism or ingestion of particulate material. The latter would include both food particles and non-living particulate detritus.

Several other factors, however, also control the uptake of specific radioelements. In the case of strontium and cesium chemical competition plays a major role. Naturally occurring potassium, the competitor to cesium, is present in sea water at a high level of about 380 ppm. In comparison to this level of occurrence, Cs^{137} would be present in very small amounts, even in an area of heavy fallout. Because of the limited ability of animals to differentiate between potassium and cesium, uptake of the latter would be low.

Natural calcium occurs at a level of about 400 ppm in sea water and exhibits chemical competition to strontium with regard to uptake by marine organisms. In the case of these elements, however, the uptake is not directly proportional to occurrence in the water; rather strontium is discriminated against with reference to and in the presence of naturally occurring calcium by factors of approximately 3 in calcareous algae and foraminiferans, 2 in arthropods, 2 to 7 in molluscs, 2.5 in bryozoans (25) and 3 to 10 in marine fishes (19).

In laboratory experiments utilizing radioactive strontium Chipman (2) observed that <u>Artemia</u> larvae reached a steady state in the level of radiostrontium about 0.7 that of sea water and Burroughs, Townsley, and Hiatt (1) reported a value of about 0.3 in a fish (Tilapia). In the case of fish bone and scales, however, the concentration factors may be higher. In the croaker (Micropogon undulatus) concentration factors for strontium in vertebrae and scales were 2.5 and 2.1 respectively those of sea water.

In addition to the above mentioned factors, isotope dilution by stable strontium would result in reduced uptake of Sr^{90} by marine organisms. Stable strontium is present in the sea at a level 6 to 1300 times that of the naturally occurring forms of the other elements represented in fallout (Table 3, column 2). Thus the discrimination against radiostrontium as a consequence of the presence of stable strontium in sea water would be 6 to 1300 times that to which the other radioelements would be subjected because of the presence of their stable counterparts.

Further discrimination against the uptake of radiostrontium in these organisms may be caused by the scavenging action of calcite formed from coral aragonite in fallout material. In a simple experiment by the author, pulverized coral was sprinkled onto and allowed to settle through sea water contaminated with $Sr^{89}Cl_2$. Approximately 11 per cent of the radionuclide was removed from solution the first hour. No reduction in activity was noted in a parallel control experiment. Suito, Takiyama and Uyeda (22) reported that the ashes from the March 1, 1954 weapons test at Bikini which fell on the No. 5 Fukuryu Maru consisted of white granules of calcite approximately

300 μ in diameter (mostly 100-400 μ). Identification of the chemical form was made by electron and X-ray diffraction techniques. Calculations of Miyoshi (18) indicate that particles of calcite 400 μ in diameter would settle from the surface to 200 meters in 26 minutes and those 100 μ in diameter in 7 hours. Thus the mechanism for rapid scavenging of Sr^{89-90} in fallout introduced into the sea would be provided by calcite settling through the thermocline.

Co-precipitation would provide another scavenging mechanism for radiostrontium in the fallout area. When calcium carbonate is precipitated in sea water strontium is co-precipitated and carried down with the calcium. This technique is commonly used in radiochemical separations on sea water. The amount of strontium removed from solution in the sea by the precipitation of calcium derived from oxidized and hydrolyzed coral is not known but probably is not low.

Strontium, co-precipitated with calcium carbonate into the particulate form, then would become available to filter feeding organisms. However, a limited fraction of the particles would settle out of reach of the biosphere into deep water. A rapid turnover of strontium has been observed in all marine invertebrates and fishes studied up to now (1, 3). Thus, ingested radioactive strontium in the particulate form would be continually recycled to the sea in solution so that in time it would come to equilibrium with the naturally occurring non-radioactive form.

Three of the radioactive non-fission-product elements, zinc, cobalt, and iron, contributed an average of 71 per cent of the total activity in the plankton samples. This observation is in contrast to those on terrestrial samples in which these isotopes were absent or, at most, present in trace amounts only (16). Thus, either a concentrating mechanism must function in the sea for the non-fission-product elements, making them available to the organisms, or an exclusion mechanism must operate on land. The extremely low levels at which these isotopes occur in dirt samples near the target area suggest that concentration occurs in the sea.

In the case of these radioelements the previously discussed factors that control uptake by marine organisms, with the possible exception of scavenging action by calcium hydroxide or calcite, would tend to cause increased uptake. Thus, these elements in fallout probably occur in the sea in particulate form, are not subject to chemical competition by similar elements or to appreciable isotope dilution by their stable counterparts, nor would co-precipitation occur in the strict sense of the word. However, negatively charged cobalt, manganese, and zinc would tend to precipitate with iron into a finely divided form, exhibiting only a limited tendency to sink below the thermocline and thus remain available to the plankton in the mixing layer.

The isotopes Mn^{54}, Fe^{55}, Co^{57}, Co^{58}, Co^{60}, and Zn^{65} are transition elements and have characteristics of variable valence, easy oxidation and reduction, and a marked tendency to form complexes. Except for the elements carbon, nitrogen, and phosphorus, the transition elements are concentrated to the greatest degree by marine plants and animals over the levels in the water (23,26). These are the elements that form the most stable complexes with organic chelating materials (17,14,27) and it is most likely by this mechanism that the heavy metals zinc, copper, nickel, cobalt, iron, and manganese are concentrated in the marine biosphere. If surface binding of these metals to organisms is a major factor for their concentration then a concurrently important factor is that of the extent of the available organic adsorptive surface. Plankton in the sea, especially phytoplankton, provide the greatest surface area with respect to protoplasmic volume and, in addition, probably provide a greater volume of living material than any other of the marine organisms. Thus, the major initial concentration of radioactive zinc, cobalt, iron, and manganese from the environment probably occurs in the phytoplankton.

In addition to the surface adsorption of non-fission-product radioisotopes onto phytoplankton and zooplankton, the process of direct uptake and assimilation may well be of importance in these organisms. The roles of these non-fission elements in the physiology of planktonic organisms are not known but in higher animal forms, at least, they are of major importance.

The probability that the non-fission radioisotopes would be present in particulate form in the sea was discussed earlier. Particles of non-fission-product material either adsorbed onto or assimilated into the phytoplankton or existing as free particulate matter in the water would be available to the invertebrate filter feeders. Thus the second concentration stage for non-fission radioisotopes would occur in the zooplankton through the filter feeders. Of course factors such as (a) average life spans of the phytoplankton and zooplankton organisms (b) average biological half lives of the different radioactive elements within the zooplankton, and (c) species differences between filter feeders regarding choice of food, the assimilation of the different radioelements and retention of the radioelements would influence the levels of the non-fission radioactive elements in the zooplankton. These factors are not known.

There is little probability, however, that the above mentioned factors or any other biological effects are responsible for the variations in ratio between Co^{57} and Co^{60} found in plankton samples from the different stations. For the heavier elements at least, organisms have little or no ability to differentiate between isotopes of a given element. Thus, the variation in ratio of Co^{57} to Co^{60} probably reflects the variation in level of occurrence of these two isotopes in the fallout material in different geographical locations.

The variation in the ratios Co^{57}/Zn^{65} and Co^{57}/Fe^{55} in plankton samples with change in collection site may also be due to differences in levels of the three isotopes in the fallout material. However, the possibilities should not be overlooked that it may be due to preferential scavenging of zinc by calcite and calcium hydroxide in the area of intense fallout or to biological effects. Data that would resolve these points are not available and an effort should be made to obtain the necessary information in any future operation.

Evidence that the differences in Co^{57}/Zn^{65} ratios are real and not due to techniques of separation and analysis is found in the gamma spectrum curves on whole plankton samples (Fig. 2). Inspection of the curves from the three samples shows clearly the differences between the levels of Zn^{65} and both of the cobalt isotopes, Co^{57} and Co^{60}, in the whole sample before separations and analyses were performed.

Summary

1. During July 1957, chemical separations by ion-exchange resin columns were made on five grouped plankton samples taken in and west of the Eniwetok Proving Ground in September 1956. Also, radiochemical separations for Sr^{90}-Y^{90} were made. The radioactive isotopes in the separated fractions were identified by decay rates, maximum beta energies and gamma energies.

2. The distribution pattern of Co^{57} and Zn^{65} in plankton samples not subjected to ion-exchange separation was determined by analysis of gamma spectrum curves.

3. The predominant fission product radioisotopes found were Zr^{95}-Nb^{95}, Ce^{144}-Pr^{144} and in one sample, Ru^{106}-Rh^{106}. Fission products accounted for 22 to 44 per cent of the total radioactivity at the collection date. Sr^{90}-Y^{90} was not found in the ion-exchange separations nor in fuming nitric acid precipitations made on duplicate aliquots.

4. The non-fission radioisotopes Fe^{55}, Co^{57}, Co^{58}, and Zn^{65} contributed 55 to 77 per cent of the total radioactivity, and Co^{60} and Mn^{54} accounted for a total of 0.8 to 1.6 per cent.

5. Co^{57} and Co^{60} were present at Bikini Atoll in a 1:1 ratio and at Eniwetok in a 5.3:1 ratio. These ratios were reflected in plankton samples collected in the vicinity of the two atolls. The ratio of Co^{57}/Co^{60} in an area 300 miles wide north to south and 200 to 500 miles west of Eniwetok was

constant at 11:1.

6. The ratio of Co^{57}/Zn^{65} at Bikini and Eniwetok Atolls was 0.06:1.0. Similar ratios occurred 200 miles south and west of Eniwetok Atoll. Slightly higher ratios occurred to the north. The center of high Co^{57}/Zn^{65} ratio (5.5:1.0) was found approximately 480 miles west and slightly north of Eniwetok Atoll and did not coincide with the center of high total radioactivity in plankton, which was located 110 miles north of Eniwetok Atoll.

7. The changes in ratio of Co^{57}/Fe^{55} with change in geographical location were similar to those of Co^{57}/Zn^{65}.

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