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Radionuclides in Plankton and Tuna from the Central Pacific.

Radioactivity of Marine Organisms from Guam, Palau and the Gulf of Siam.

Qualitative Distribution of Radionuclides at Rongelap Atoll.

Increase in Radioresistance of Fish to Lethal Doses with Advancing Embryonic Development.

Factors Controlling the Distribution of the Rare Earths in the Environment and in Living Organisms.

Iron and Cobalt in Ecology.

Some Aspects of the Biology of Zirconium.

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RADIONUCLIDES IN PLANKTON AND TUNA FROM THE CENTRAL PACIFIC

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INTRODUCTION

The uptake and retention of radionuclides by marine organisms are of practical interest to man insofar as the radionuclides constitute a potential hazard in his food materials obtained from the sea. In addition to the practical considerations, information may be derived concerning the processes which control the cycling of stable elements from sea water through the various levels of food chains. If the amounts of radionuclides in organisms comprising a food chain are determined at intervals of time following contamination of the environment, the interrelations between a trophic level and the physical environment and with other trophic levels may be delineated, at least in regard to the movements of mineral components through the system.

Except for the light elements, marine organisms do not discriminate between different isotopes of a given element; thus, if the introduced radionuclide has the same chemical and physical form as the corresponding stable element, the two are taken up according to their ratio of abundance. However, radioisotopes do differ from their stable counterparts in that they are subject to radioactive decay. If the biological half-lives of a given element, in several organisms comprising a food chain, are not taken into account, erroneous interpretations may be made, especially if the radioisotope has a relatively short physical half-life. These errors may be corrected if the average ratio of radioactive to stable isotope of an element is determined for each level within the food chain.

The principal factors which control the uptake and retention of radionuclides by marine organisms are: (1) the amount of radionuclide introduced into the sea, (2) the site of introduction into the sea in relation to position and depth, (3) the degree of physical dispersion by currents and gravity, (4) the chemical and physical characteristics of the radioelement, (5) the chemical and physical forms of non-radioactive materials associated with the radionuclide, (6) the degree of isotope dilution of the radioelement by the corresponding stable element (or chemically similar elements) in sea water, (7) the degree to which the radionuclide is adsorbed to the organisms, (8) the degree of selective uptake, and (9) the biological half-life of the element in the organisms.

Fallout areas near the sites of nuclear weapons tests are of value for investigations of the fate of radionuclides in the marine environment in that large amounts of radiomaterials may be introduced into a given area in a short time and at adequate levels to be detected for a relatively long time following contamination. Because the fallout is usually deposited over large areas, most of the animals collected over a period of time in the area may be considered to have been exposed to a chronic rather than to an acute exposure of radioactive materials.

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MATERIALS AND METHODS

During the nuclear weapons test series in 1958 (Operation HARDTACK I) three oceanographic surveys were made by members of the Laboratory of Radiation Biology (Lowman, 1960; Lowman et al., 1958; Palumbo et al., 1959). In the first survey, measurements were made and samples were collected in a contaminated body of water for an interval of time starting less than one hour after detonation of the nuclear device and extending over a period of 48 hours. The samples included plankton and water samples taken at depths to 300 meters. The remaining two surveys included areas which had been contaminated for more than one week and for an average time of six weeks, respectively. During the time of the above described surveys, samples of tuna (Thunnus spp.), albacore (Thunnus alalunga), skipjack (Euthynnus pelornis), dolphin (Coryphaena sippurus), barracuda (Sphyraena argentea), and shark caught in the vicinity of the Eniwetok Proving Ground were collected at ports-of-landing in Japan by Dr. Toshiharu Kawabata.[2] Samples of the organs and tissues of tunas comprised the major part of the collection of more than 1,000 samples. Wet weights were taken, the samples were dried, and then sent to the Laboratory of Radiation Biology by Dr. Kawabata.

The samples of sea water, plankton, and fish were counted for total beta radioactivity and selected samples were subjected to radiochemical separation and analysis. These techniques and methods have been described elsewhere (Donaldson et al., 1956; Kawabata and Held, 1959; Lowman, 1958; Lowman et al., 1957; Palumbo and Lowman, 1958).

RESULTS

In the present paper the ratios of total radioactivity and the percentage of total radioactivity contributed by individual radionuclides are considered with respect to sea water and in the food chain composed of plankton, omnivorous fish, and carnivorous fish. In most cases the averages of several samples collected within a contaminated area are used in order to reduce the effect of individual variability, a factor which has been considered elsewhere (Bonham, 1958; Donaldson et al., 1956; Lowman 1958, 1960; Welander, 1958).

In studies on the uptake of radioactivity from sea water by plankton the fractionation of the radioactivity between the water and plankton is usually compared on an equal volume or equal weight basis and a concentration factor for the radioactivity by the plankton may thus be derived. However, uptake of radioactivity by plankton from sea water may also be compared on the basis of the fraction of the total radioactivity in the water that is accumulated by the contained organisms.

The importance of marine organisms in affecting the movement and distribution of radioactive contamination in the sea has been emphasized by several authors (Ketchum, 1957; Lowman, 1958; Revelle and Shaefer, 1957). In areas of the sea where relatively large populations of organisms exist in relation to the total water volume the organisms may exert an effect of this type (Ketchum, 1957).

[2]Scientific names from: List of common and scientific names of fishes from the United States and Canada, 2nd edition. Am. Fish. Soc. Spec. Publ. 2. 102 pp. (1960).

Table 1. Percentage of total radioactivity in sea water and plankton contributed by fission products and neutron-induced radioisotopes at approximate times of 48 hours, one week, and six weeks after detonation.

	Time after detonation					
	Less than 48 hours		Greater than one week		Six weeks	
	Water	Plankton	Water	Plankton	Water	Plankton
Molybdenum-99; technetium-99m	5.9	12.0	5.0	0.0	0.02	0.0
Cerium-141, 144; praesodymium-141, 144	0.83	3.0	2.1	0.0	20.0	5.0
Ruthenium-103, 105, 106; rhodium-103, 105, 106	2.1	3.0	2.2	5.0	16.0	1.4
Barium-140; lanthanum-140	2.6	2.0	6.8	23.0	18.0	0.0
Tellurium-132; iodine-132	6.2	8.0	5.6	0.0	0.05	0.0
Zirconium-95; niobium-95	0.38	1.0	1.0	6.0	18.0	20.0
Strontium-89, 90	0.30	0.0	0.76	0.0	8.2	0.0
Cesium-137	0.01[1]	0.0	0.05	0.0	0.08	0.0
Neptunium-239	54.0	69.0	33.0	2.0	0.02	0.0
Uranium-237	24.0	3.0	43.0	2.0	18.0	0.0
Cobalt-47, 58, 60	0.02	0.0	0.05	43.0	0.67	24.0
Zinc-65	0.02	0.0	0.05	3.0	0.84	25.0
Iron-55, 59	0.02	0.0	0.05	16.0	0.93	24.0
Manganese-54	trace	0.0	0.01	0.0	0.12	0.6

[1] Less than 0.01 per cent.

However, in some tropical areas, where the total mass of organisms is very small in comparison to the volume of the water, the influence on the distribution of radionuclides may be small and in the open seas surrounding the Marshall Islands, at least, appears to be insignificant.

In the sea at Eniwetok Proving Ground the ratio of the volume of the sea water to contained plankton is approximately 100,000,000 to 1 (Anonymous, 1958). The ratio of total radioactivity in the water to radioactivity in the plankton in the same area was approximately 10,000 to 1 at one week and 30,000 to 1 at six weeks after contamination. Thus at one week the plankton showed an average concentration factor of 10,000 and at six weeks 3,000 for the introduced radioactivity. The cause of the apparent drop in concentration factor of the plankton with increased time is not known but has been observed to occur consistently in samples collected in the open sea at the Eniwetok Proving Ground (Lowman, 1960; Lowman et al., 1957; Seymour et al., 1957).

In view of the observations that the plankton contained only 1/10,000 and 1/30,000 of the total radioactivity in the contaminated area at one week and six weeks, respectively, the organisms could not have exerted a significant influence upon overall movement and distribution of the introduced radioactivity. However, the plankton might be expected to affect the distribution patterns of those radionuclides concentrated to the greatest degree (iron, zinc, and cobalt). At six weeks the ratios of total calculated amounts of radioactive iron, zinc, and cobalt in sea water to those in the contained plankton were 1,000, 900, and 700, respectively. Thus, even for those radionuclides for which the plankton exhibited high concentration factors, the physical factors of the environment such as gravity and water currents probably exerted the major influence upon their total movement and distribution.

In Table 1 the percentages of radioactivity contributed by the different radionuclides in sea water and in plankton are shown at times of less than 48 hours, greater than one week, and greater than six weeks after fallout. The values for water are based on the theoretical radionuclide composition of fallout[3] at the given times and are subject to error because they are not corrected for fractionation and precipitation in the sea water. Because of the resulting uncertainty, the minor differences between the percentages of radionuclides in plankton and water less than 48 hours cannot be considered to be significant. At less than 48 hours the radionuclides probably were taken up by the plankton in an approximately direct ratio to their occurrence in the water with the exception of strontium and possibly cesium. Of all the radionuclides listed in Table 1 only radioactive stontium and cesium are initally present in solution in the water. Cesium-137 has not been found in any significant amount in any plankton sample.

Experimental data taken in 1958 at the Eniwetok Proving Ground showed that radioactive strontium was actively discriminated against by plankton relative to other radionuclides (Lowman, 1960). Samples of plankton and water were taken and the amounts molybdenum-99, technetium-99m; cerium, promethium-141; barium, lanthanum-140; telluriumu, iodine-132; zirconium-95 and strontium-90 were determined by radiochemical analysis. The ratio of observed strontium-90 to expected strontium-90, based on the observed levels of the other radionuclides, was calculated for various times after detonation. The results were as follows:

Time	Observed to expected
3 hours	.43
21 hours	.06
36 hours	.03

At three hours approximately ½ as much strontium-90 was associated with the plankton as was expected. At 21 hours and 36 hours the observed levels were approximately 1/17 and 1/30 of those expected. The discrimination against strontium-90 in relation to the uptake of the other radionuclides, therefore, increased with time up to at least 36 hours. In samples of plankton collected at later times the levels of strontium-90 in the plankton were too low to be measured.

Although the radionuclides were taken up by the plankton less than 48 hours in approximately the same ratio as they were present in the water, selective uptake at greater than one and six weeks was evident. At approximately one week the three radioelements, cobalt, zinc, and iron contributed only .15 per cent of the total radioactivity in the water but accounted for 62 per cent of the radioactivity in the plankton. At approximately six weeks the values were 2.44 per cent and 73 per cent respectively. The accumulation of fission products by the plankton at one and six weeks was variable and no marked pattern of active concen-

[3] Ratio of cobalt-60 to strontium-90 (Strom et al., 1958), ratios of cobalt-60 to manganese-54, iron-59, cobalt-58, uranium-237, and neptunium-239 (Knapp, 1960), ratio of iron-55 to iron-59 (Lowman, 1960).

tration of these radionuclides by the organisms was evident.

In the food chain under consideration, the plankton are fed upon by omnivorous fish. Carnivorous fish, in turn, feed upon the omnivores and probably, to a lesser degree, on the plankton. During the 1958 surveys samples of plankton and omnivorous flying fish were taken at the same sampling station in an area which had been contaminated by fallout about one week earlier. The tuna (carnivores) samples were collected south and west of the sampling station, described above, about five weeks later.

The plankton and omnivorous fish may be assumed to have remained in the contaminated body of water during the week following fallout and the total radioactivity and the levels of individual radionuclides contained in these organisms may be directly compared. However, the total radioactivity in the tissues and organs of the tunas cannot be directly compared with that in the plankton or omnivorous fish. Tunas are pelagic fishes and are capable of migrating great distances, in comparison to the size of the main mass of the contaminated area, in short periods of time. Thus the length of time that the fish remained in the contaminated area prior to being taken cannot be determined. The levels of individual elements in the organs and tissues of the tunas can be compared with those in the other organisms, however, if the total time spent in the contaminated area by the tunas is relatively long in comparison to the biological half-life of the elements in these fish. The tunas were taken in a mass of water that had been contaminated for about six weeks. During this time the contaminated body of water had been dispersed over an area with a diameter of several hundred miles although a central area of higher radioactivity still persisted. If one assumes a more or less random movement of the tunas and if a similar distribution of radionuclides is found in the same organs of different fish, it is probable that the tunas had remained in some part of the contaminated area sufficiently long for the individual radionuclides within the tissue and organs to have reached a state of equilibrium with the radionuclides in the food items. The average level of radioactivity in the food items would not necessarily be the same, however, as those of the food items taken at the above described sampling station.

In Table 2 is shown the percentage of total radioactivity contributed by individual radionuclides in plankton and flying fish at one week and in tunas and water at six weeks after contamination. The total disintegration rates for the radioactivity in the biological samples are also given. Although the fission products plus uranium-237 accounted for 36 per cent of the radioactivity associated with the plankton, they were present only in trace amounts in the white muscle and liver samples of the omnivorous flying fish. In the tuna tissues and organs the only fission product that was detected was cesium-137 which was present in one sample of white muscle. The cesium-137 in this sample accounted for only 1.1 per cent (36 disintegrations per minute per gram of dry weight) of the total radioactivity and was present at about the same percentage value of total radioactivity as that in the water. Cesium-137 is present in solution in sea water and would be expected to follow the u[illegible] of naturally occurring potassium which is concentrated in muscle tissue.

Radioisotopes of manganese, iron, cobalt, and zinc contributed 62 per cent of the total radioactivity in the plankton and almost 100 per cent in the omnivorous and carnivorous fishes.

Metabolism is similar in all forms of life at the cellular level in both plants and animals although they may differ significantly in form and complexity. Marine plants and animals tend to concentrate the stable transition elements manganese, iron, copper, cobalt, and zinc which become tightly bound to the organisms (Krumholz et al., 1957). The details of the processes involved in the uptake of these elements have been discussed elsewhere (Lowman, 1960) and will not be reviewed again. However, observations by Korringa (1952) and Lehninger (1951) indicate that marine organisms tend to concentrate positive polyvalent ions but not positive monovalent ions although the latter may be present in the environment in high amounts.

The ability of the transition elements to form complexes with biological materials is usually not affected by the chemical composition of the biological sustrate, and the stability of the metal-biological complex is usually in the following biological order.

$$Mn^{++} < Fe^{++} < Co^{++} < Ni^{++} < Cu^{++} \;|\; > Zn^{++}$$

Uptake of the radioisotopes of these elements from sea water by the plankton probably reflects the stability of the metal-biological substrate described above. In plankton samples taken at approximately one and six weeks (Table 1) the order of uptake in relation to the levels of radioisotopes in

Table 2. Percentage of total radioactivity contributed by fission products and by the neutron-induced isotopes uranium-237, cobalt-57, 58, 60; iron-55, 59; zinc-65, and manganese-54 determined in plankton and fish samples collected at the Eniwetok Proving Ground in 1958. The total radioactivity per gram dry weight is also shown.

	Water	Plankton[1]	Flying Fish		Tuna					
			White muscle	Liver	White muscle A	White muscle B	Liver C	Liver D	Dark muscle A	Dark muscle B[2]
Ruthenium, zirconium and uranium	52.0	13.0	trace	trace	0.0	0.0	0.0	0.0	0.0	0.0
Barium-140; lanthanum-140	18.0	23.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cesium-137; barium-137m	0.08	0.0	0.0	0.0	1.1	0.0	0.0	0.0	0.0	0.0
Cobalt-57, 58, 60	0.67	43.0	10.0	8.7	0.9	2.5	0.0	2.3	1.1	2.1
Iron-55, 59	0.93	16.0	31.3	81.3	5.8	8.1	25.5	15.0	12.4	9.7
Zinc-65	0.84	3.0	58.8	9.9	91.9	89.0	74.5	82.6	86.3	88.0
Manganese-54	0.12	0.0	0.0	0.0	0.2	0.4	0.0	0.1	0.2	0.2
Total radioactivity (disintegrations per minute per gram dry weight)	-	2.3 $\times 10^6$	2.2 $\times 10^4$	1.1 $\times 10^6$	3.3 $\times 10^3$	1.2 $\times 10^4$	3.6 $\times 10^4$	2.0 $\times 10^5$	5.3 $\times 10^3$	2.9 $\times 10^4$

[1] At six weeks.

[2] Samples from four different fish: A = big eye tuna (Thunnus obeus); B = yellow fin tuna (Thunnus albacares); C = yellow fin tuna; D = big eye tuna.

the water was

manganese < iron < cobalt < zinc.

The order of accumulation is therefore in agreement with the order of stability of the metal-biological sustrate complex and suggests that the primary process of accumulation of the radioisotopes of these elements by plankton is probably that of surface adsorption although the process of direct uptake and assimilation may also occur. In higher animals, at least, these elements are intimately associated with oxygen transport, electron transport, enzyme function, and the structure of vitamin B_{12}.

Although surface adsorption may be an important mechanism in the uptake of radionuclides by plankton, this process is of minor importance in the accumulation of radionuclides by omnivorous and carnivorous marine fishes. In the fishes the radionuclides considered here are primarily accumulated by metabolic processes and the route of uptake is mainly through the food supply. Only those radionuclides which are biologically important and are subjected to only limited isotope dilution in the sea are concentrated by the fishes. The radioisotopes of cobalt were concentrated more than the other radionuclides by the plankton and contributed 43 per cent of the total radioactivity. In the omnivorous fish radioactive cobalt was discriminated against, probably as a result of limited biological demand, and accounted for only 10 per cent of the total radioactivity in white muscle and 8.7 per cent in the liver. In the tunas the percentages of radioactivity contributed by the radioisotopes of cobalt in the tissues and organs were reduced further to values of 0.9 per cent to 2.5 per cent in the liver. Thus, the percentage of radioactive cobalt in the tissues and organs decreased progressively through the two higher trophic levels of the food chain. This decrease may be attributed, in part, to higher turnover rates for cobalt in marine fishes than for iron, zinc, and manganese.

The elements iron, zinc, and manganese are biologically important in enzyme systems and in oxygen and electron transfer. Iron is a component of hemoglobin, myoglobin, cytochromes, and cytochrome oxidases and other enzymes associated with oxidation. Zinc is found in the respiratory protein of the blood in some snails and in the enzyme carbonic anhydrase. Manganese is associated with peptidases, transphorases, dehydrogenases, decarboxylases, and oxygen disposal system in plants (Eyster et al., 1958; Steward and Pollard, 1957).

Of the three elements, the radioisotopes of iron and zinc were concentrated in the liver of the omnivorous fish to a level in which the disintegration rate per unit weight of tissue was approximately double that in the plankton. On a percentage basis radioactive iron contributed 16 per cent of the total activity in the plankton and approximately 31 per cent and 81 per cent respectlvely in the white muscle and liver of the omnivorous flying fish. In the carnivorous tunas iron-55,59 accounted only for an average of 7 per cent and 20 per cent respectively in the same organs. Thus the percentage of total radioactivity contributed by iron-55,59 in plankton was about $\frac{1}{2}$ and 1/5 respectively of the percentage accounted for in the white muscle and liver of the omnivore and radioactive iron contributed only 1/4 as much of the total percentage or radioactivity in the muscle and liver of the carnivorous tunas as it did in the corresponding tissues of the omnivore.

The reduction in percentage composition of radioactive iron in the tissues of the tunas compared with those in the flying fish is compensated by the increased percentages of zinc-65. Whereas radioactive zinc contributed only 3 per cent of the total radioactivity in the plankton it accounted for about 59 per cent in the white muscle and 10 per cent in the liver of the omnivore. In the tunas the percentages were greater than those in the flying fish, accounting for approximately 90 per cent and 78 per cent respectively in the muscle and liver of these carnivorous fishes. The high levels of zinc-65 in the tissues of the tunas are probably related to the biological requirements for metabolism in these active fish.

Radioactive manganese was not detected in the plankton or omnivorous fish. In the tuna samples manganese-54 accounted for up to 0.4 per cent of the total radioactivity with an average percentage in all organs approximately equal to the percentage of total radioactivity in the water contributed by that radioelement.

SUMMARY

Three factors appeared to control the selective uptake of radionuclides from sea water by the plankton, omnivorous fish, and carnivorous fish studied. These were isotope dilution by the corresponding stable element or chemically similar elment in the sea water, the tendency of divalent cations to complex strongly with biological substrates, and the biological requirements for certain elements in metabolic processes.

The uptake patterns in the three trophic levels were as follows:

During the first 48 hours following fallout the plankton in the contaminated area accumulated radionuclides in approximately the same ratio as they occurred in sea water. After one week the radioisotopes of the three elements cobalt, iron and zinc were actively taken up by the plankton in comparison to the other radionuclides which were associated with the plankton at percentages similar to those in sea water. Omnivorous fish which feed on the plankton, almost completely excluded the fission products and, on a comparative basis, concentrated zinc-65 and iron 55,59, but discriminated against cobalt-57,58,60. Carnivorous tunas, which feed primarily on omnivorous fishes, discriminated in favor of zinc and manganese and against iron and cobalt in comparison to the relative percentages of these radionuclides in their food supply.

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RADIOACTIVITY OF MARINE ORGANISMS FROM GUAM, PALAU AND THE GULF OF SIAM, 1958-1959

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INTRODUCTION

Following the Operation Hardtack (Phase I) nuclear test series at Bikini and Eniwetok Atolls in 1958, a study was made to measure changes with time in the radioactivity in marine organisms collected at stations located at distances of over one thousand miles from the test site. The radionuclides of principal interest were those from local Hardtack fallout that were transported by ocean currents from near the test site to the collecting areas, rather than the fallout nuclides transported in the stratosphere. The study following Hardtack supplemented a somewhat similar program that was begun after the Redwing test series in 1956 (Thomas et al., 1958).

During the Bikini-Eniwetok phase of Hardtack there were 24 barge, 2 surface, and 2 underwater detonations between May 5 and July 26, 1958. A large portion of the 19 megatons of total fission yield credited to nuclear tests by the United States and the United Kingdom for 1957 and 1958 (Dunham, 1959) was produced by these detonations. Thirty to eighty per cent of the radioactivity produced by the barge and surface detonations could be expected to occur as local fallout (Libby, 1959) and to enter the ocean within a day or two after detonation and within a few hundred miles of the test site. Practically all of the radioactivity from the underwater detonations would remain in the ocean.

One objective of the study was to determine if local fallout from Hardtack could be detected in ocean waters at distances of 1,000 miles or more from the test site (Guam and Palau are 1,200 and 1,950 miles, respectively, west of Eniwetok). If so, then an estimate could be made of the rate of movement of local fallout by ocean currents, in this instance the North Equatorial Current, since both the test site (Eniwetok and Bikini) and two of the collecting areas (Guam and Palau) are within the path of the westward-flowing part of this current system (see Figure 1). The collecting area in the Gulf of Siam is 4,250 miles west of Eniwetok but outside the North Equatorial Current.

The arrival of Hardtack fallout at the collecting areas was determined indirectly rather than by a direct measurement of the radioactivity in sea water. The criterion was a significant increase in the gross beta activity of marine organisms. There are two reasons for using this criterion: first, small amounts of radionuclides in sea water are difficult to detect in the presence of the salts that are normally present; and, secondly, biological organisms concentrate certain fallout nuclides and thus the specific activity for them is higher than for sea water. For these reasons biological samples are easier to prepare for counting and require less counting time than sea water samples.

Other objectives of the program were to document the level of radioactivity in marine organisms from the western Pacific, to provide additional information about the biological distribution of fallout nuclides, and to compare the results of this study with the study following the Redwing test series of 1956.

SAMPLE COLLECTION, PREPARATION AND COUNTING

Samples were collected at approximately three-month intervals beginning in June 1958 and continuing until November 1959, except in the Gulf of Siam. There were four collections in the Gulf of Siam and six each at Palau and Guam. Samples included fish, crabs, lobsters, snails, clams, algae, and plankton. In addition, 72 weekly plankton samples were collected at Palau, site of field headquarters for the George Vanderbilt Foundation.

Plankton samples were preserved in ten per cent formalin, but all other samples were sent fresh-frozen from the field stations to the laboratory. The samples were held in a freezer until they were processed, at which time they were thawed, dissected, weighed, dried at 95° centigrade, reweighed, ashed at temperatures up to 540° centigrade, and again reweighed prior to counting.

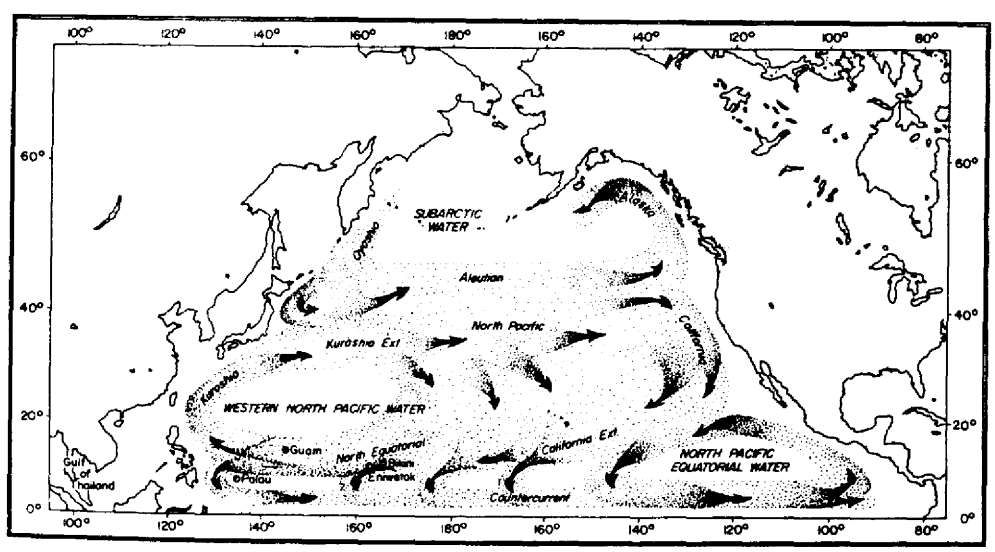

Figure 1. The major surface current systems in the north Pacific Ocean (after Seckel and Waldron, 1960).

All of the samples were counted for gross beta activity and some for gamma-emitting nuclides. Generally, gross beta counts are of limited value because they are neither quantitative nor qualitative but have been useful here to indicate the arrival at the collecting areas of water-borne fallout nuclides. In other instances beta counts are useful for purposes of comparison when it can be assumed that the isotopic composition of the samples being compared is approximately the same. In addition to the determination of gross beta activity, specific nuclides were identified, qualitatively, in a limited number of samples by gamma spectrum analysis.

The beta-counting system was composed of the following units: a thin-window 1 3/4-inch, pancake-type Geiger-Mueller detector, an automatic sample changer with the lead shield modified to accommodate the detector, a decade scaler, and a printing timer. The cups for the automatic sample changer were adapted to hold 1¼-inch stainless steel planchets.

The gamma-counting system was a model of a system described by Heath (1957). The copper and cadmium-lined shield weighed five tons and was built with lead bricks into the form of a cube, 36 inches on a side and four inches thick. The detector head consisted of a three-inch by three-inch sodium iodide crystal and a 12-stage, three-inch photo cathode that was placed in the center of the shield, a minimum distance of 12½ inches from the sides. The impulses from the detector head were transmitted to a 256-channel analyzer.

RESULTS

The results of gross beta counting are given in Appendix Table A as average values. With few exceptions the average is computed from the counts of three samples, each from a different specimen. The total number of samples counted was 825, but in addition there were 495 recounts either to establish decay curves or to confirm the original counts. The common and scientific names of the specimens are listed in Appendix Table B.

Some of the data from Appendix Table A have been selected for graphical presentation in Figure 2. Since all of the data could not be presented conveniently in Figure 2, data representative of general results were selected.

Seventy-two plankton samples, collected weekly, were obtained from Palau in addition to six trimonthly samples from Guam and four trimonthly samples from the Gulf of Siam. The data for these samples are presented in Appendix Table C and Figure 3.

Gamma spectrum analyses were made of 17 samples to determine the presence of specific gamma-emitting nuclides. The samples were selected for geographical location and high gross beta counts. Results of analyses corrected to date of collection are given in Table 1.

One of the limitations in the interpretation of the results is the fact that the gross beta counts have not been corrected for decay during the interval between times of collecting and of count-

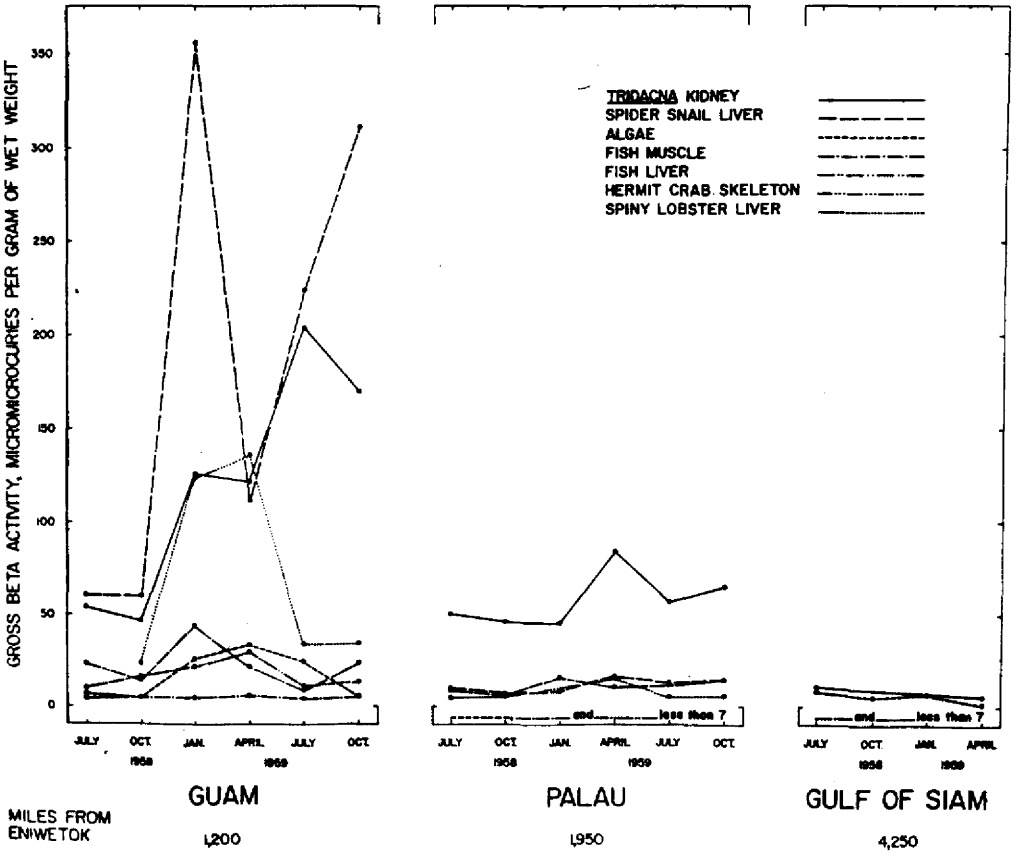

Figure 2. Average gross beta activity of marine organisms from Guam, Palau, and the Gulf of Siam, 1958-1959. Average based on two to thirteen observations.

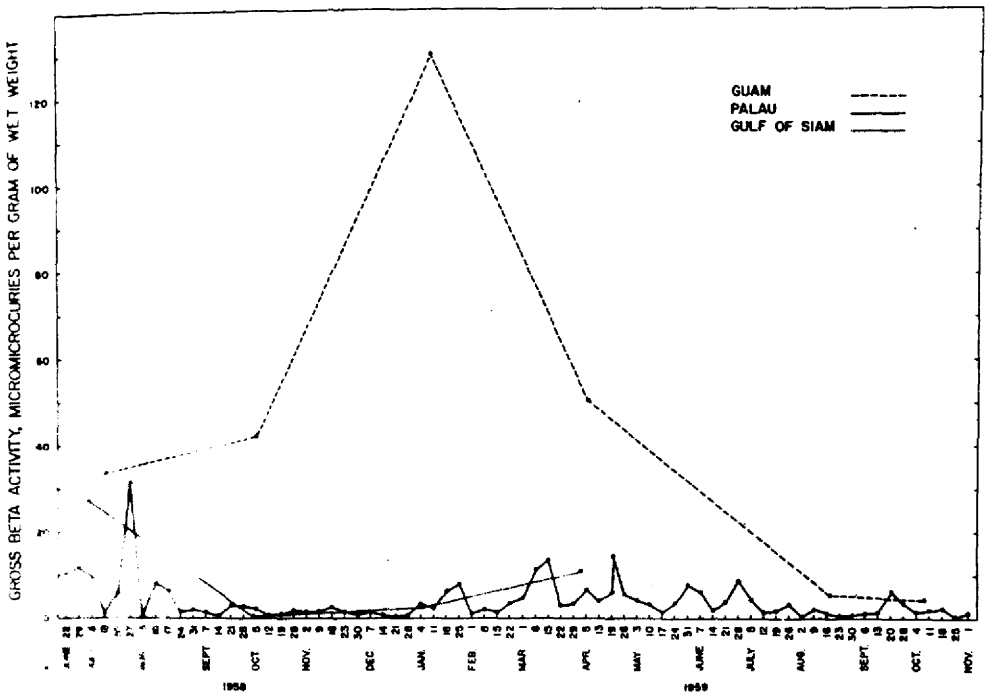

Figure 3. Gross beta activity of plankton from Guam, Palau, and the Gulf of Siam, 1958-1959, based on single observations.

ing the sample. This interval averaged 247 days for the first collection, 161 days for collection II, and 38 to 78 days for the other four collections. Therefore, all reported values are low and by an amount that is estimated to range from 12 to 35 per cent. The percentage error was estimated from decay curves that were arbitrarily selected after inspection of 19 decay curves for these data and also decay curves of Bonham (1959). On a log-log plot the slopes of the selected curves were -1.5 for samples collected prior to the arrival of the Hardtack fallout and -1.4 for later samples.

A reliable correction factor for decay was difficult to derive either empirically from other data or theoretically from consideration of the composition of fallout because the nuclides present in the samples were a mixture of unknown nuclides with different, unknown dates of origin. The best source of information about decay rates that are most like the Guam and Palau samples is Bonham (1959), who has determined the gross beta decay rates for 106 types of samples from the Eniwetok area for periods up to seven years. However, because of within-sample and between-sample variation,

Table 1. Nuclides identified by gamma spectrometry in single samples from Guam, Palau and Gulf of Siam, 1958-1959. (Values in terms of micromicrocuries per gram of wet weight and corrected to date of collection.)

Area	Sample type	Date collected	Number of specimens	Potassium -40	Manganese -54	Cobalt -57	Cobalt -60	Zinc -65	Zirconium -95	Silver -110m
Guam	(surgeonfish)									
	muscle	October 6, 1959	6	4 ± 1[1]						
	liver	April 4, 1959	3	28 ± 17		20 ± 12	2 ± 1	137 ± 12		
	(spider snail)									
	liver	January 8, 1959	4		180 ± 8	59 ± 3	4 ± 2			
	liver	July 29, 1959	6	79 ± 11	357 ± 5	231 ± 3	19 ± 1			
	liver	October 7, 1959	7	54 ± 13	207 ± 3	109 ± 2	10 ± 1	63 ± 5		
	(spiny lobster)									
	liver	November 3, 1959	6	91 ± 2	?	25	?	40	30	50[2]
	(Tridacna clam)									
	kidney	October 21, 1959	10		1003 ± 31	2340 ± 30	275 ± 19	22 ± 1		
Palau	(goatfish)									
	muscle	September 29, 1959	10	3 ± 1						
	(Tridacna clam)									
	kidney	September 29, 1958	7		12 ± 2	16 ± 1	57 ± 1			
	kidney	April 14, 1959	1		5 ± 1	11 ± 1	20 ± 1	30 ± 6		
	kidney	September 28, 1959	3		85 ± 20	294 ± 20	50 ± 7			
Gulf of Siam										

No gamma emitting nuclides other than potassium-40 identified in the six samples examined which included: fish muscle (Epinephelus), fish liver (Katsuwonus), clam kidney (Tridacna), clam soft parts (Arca), algae (Dictyota), and plankton.

[1] 95 per cent error in net count; $E_{95} = 1.96\ (Ns/t_s + Nb/t_b)^{1/2}$; from U. S. Public Health Service Radiological Health Handbook, 1960.

[2] Approximate value based upon decay scheme for silver-110m and efficiency factor for cesium-137. Without a standard for silver-110m Compton correction factors not calculated and therefore estimates of other radionuclides in sample also are approximate.

Bonham's decay rates were of limited usefulness. The theoretical decay curve for mixed fission products could not be used because the composition of the nuclides in the samples was not that of mixed fission products. Probably the best estimate of decay can be made from a decay curve that is determined for each sample, but even then there will be some degree of uncertainty because of the need to extrapolate from a decay curve that is not necessarily linear on either a semi-log or a log-log plot.

A limitation on the interpretation of the plankton data resulted from the use of whole samples including many species of organisms rather than of samples which were sorted by species. The composition of plankton catches varied a good deal from catch to catch and the uptake of specific nuclides by various species of plankton organisms could be expected to differ markedly; therefore, the variation in radioactivity of whole plankton samples may reflect species differences only. However, separation of these plankton catches into species is entirely impractical because, in addition to the great effort required, the quantity obtained of each species would provide only a minute sample.

Accuracy of the estimate of the rate of transport of Hardtack fallout by the North Equatorial Current was limited by lack of information as to the exact time and place of entry of the fallout into the ocean. There may be an error up to 50 days in estimating time and one to two hundred miles in estimating place of entry. Also, it was assumed that the fallout was transported in a direct line between the two points of reference, but Miyake et al. (1955) have shown that there is considerable deviation in the flow of the North Equatorial Current near Bikini and Eniwetok. It is obvious that the percentage error in calculating rate of transport becomes less as time and distance increase.

CONCLUSIONS

The following conclusions are based upon data presented in Appendix Tables A and C and Figures 2 and 3.

(1) It appears reasonable to assume that significant increases in gross beta counts of some marine biological samples were a valid criterion of the arrival of water-transported, local fallout from Hardtack.

(2) One type of sample in which the presence of Hardtack fallout was not evident was fish muscle, for which the counts remained at essentially background level for all collections regardless of date or area of collection. For Guam the values ranged fro 3.9 to 6.1 micromicrocuries per gram of wet weight; for Palau, from 3.5 to 5.6; and for the Gulf of Siam, from 3.5 to 6.6.

(3) The counts of gross beta activity differed greatly for the three collecting areas. The counts of Guam samples were considerably higher than those from Palau, which in turn were very much higher than the counts of the Gulf of Siam samples. The levels of radioactivity in the Gulf of Siam samples were approximately background and did not change significantly with date of collection.

(4) The arrival at Guam of Hardtack fallout transported by the North Equatorial Current occurred between October, 1958, and January, 1959. The arrival at Palau was approximately three months later, between January and April, 1959, but was not as sharply indicated because the levels of radioactivity were considerably lower than at Guam. Variation in the counts of the weekly plankton samples from Palau partially masks the upward trend of the counts during the first part of 1959. This variation is believed to be due to differences in the species composition of the plankton catches.

(5) The samples with the highest gross beta counts were clam kidney (Tridacna sp.) and spider snail liver (Lambis lambis). The relatively low values for liver samples of Palau spider snails were an anomaly. The highest values for clam kidney and for spider snail liver were, respectively, 204 and 356 micromicrocuries per gram of wet weight.

(6) Gross beta counts of some Guam and Palau samples prior to the arrival of Hardtack fallout indicated the presence of radionuclides from prior test series. Values were higher at Guam than at Palau.

Two estimates of the rate of westerly advance of Hardtack fallout were made: one, the rate from Eniwetok to Guam; the other, from Eniwetok to Palau. The estimates were based on the assumption that the fallout occurred at Eniwetok on June 15 (mid-point of test series), traveled in a straight line to Guam and to Palau and arrived at Guam on November 15, 1960 (mid-point between the October, 1958, and January, 1959, collections) and at Palau on Febuary 15, 1959 (mid-point between the January and April, 1959, collections). The calculated rates were 8.0 and 8.1 nautical miles per day, respectively. The closeness of the values is fortuitous but they are in reasonable agreement with previous estimates of the rate of transport by the North Equatorial Current of fallout from other test series. From data published by Miyake et al. (1955), Harley (1956), Seymour et al. (1957), and Lowman (1960) the calculated rate for periods from one month to one year ranged from seven to ten nautical miles per day.

In Table 1 the amounts of specific gamma-emitting nuclides as determined by gamma spectrometry are tabulated. Samples were selected for type, area of collection, and high gross beta count. In addition to naturally occurring potassium-40, the radionuclides present, in order of abundance, were cobalt-57, cobalt-60, manganese-54, cerium-144, zinc-65, zirconium-95, and silver-110m. The greatest value was 2,300 micromicrocuries of cobalt-57 per gram of wet weight of clam kidney (Tridacna).

It is to be noted that silver-110m was reported as cesium-137 and manganese-54 at the time this paper was presented at the Symposium. The nuclides had been identified by gamma spectrometry and the two principal gamma peaks for silver-110m, 0.67 Mev (million electron volts) and 0.89 Mev, were mistaken for the 0.66 Mev gamma peak of cesium-137 and the 0.84 Mev gamma peak of manganese-54. Further investigations by Palumbo and other co-workers in our Laboratory suggested the presence of silver-110m, which was verified by chemical separation of silver from the sample and by gamma spectrometry of the separated component. The identification of silver-110m is of special interest because it is a fallout nuclide which has not been reported previously, and so far it has been detected only in the "liver" of the spiny lobster. At the time that silver-110m was identified in the sample of spiny lobster, collected at Guam in November 1959, Palumbo also identified as silver-110m an "unknown" nuclide in spiny lobster liver collected at Eniwetok in March 1961.

Gamma-emitting nuclides other than those listed in Table 1 may have been present in the samples but were not identified because their radioactivity was below the level of detectability, or their photopeak

hidden by the photopeak of a more abundant nuclide with gamma rays of approximately the same energy. Nuclides in the latter category that might have been expected in the samples analyzed included cobalt-57 and cerium-144 as well as cobalt-60 and zinc-65. When two nuclides that emit gamma rays of approximately the same energy are present in a sample, often one nuclide can be identified by scanning the gamma spectrum for other photopeaks from the same nuclide or from assocated nuclides, without resorting to radiochemical methods. The nuclides listed in Table 1 were identified solely by scanning the gamma spectrum except for silver-110m, as mentioned above.

Further analysis of the samples from Guam, Palau, and the Gulf of Siam for specific radionuclides was not attempted because of the extensive work done by the Hanford Laboratories on radionuclide composition of similar samples collected after the Redwing test series in 1956 (Thomas et al., 1958). To the extent that the 1956-1957 samples could be compared with the 1958-1959 samples there were no gross differences in results between the two groups of data.

SUMMARY

Following the Hardtack weapons test series at Bikini and Eniwetok in 1958, samples of fish, crabs, lobsters, snails, clams, algae, and plankton were collected at Guam, Palau, and in the Gulf of Siam by the George Vanderbilt Foundation for radiological analyses at the Laboratory of Radiation Biology. The collecting areas were 1,200, 1,950, and 4,250 miles, respectively, west of the test site. The gross beta activity was determined for all samples and the gamma-emitting nuclides were identified in selected samples.

The rate of westward transport of local fallout from the Hardtack series by the North Equatorial Current was estimated at eight miles per day between the test site and Guam and Palau. The criterion for the arrival of the fallout at the collecting area was a significant increase in the gross beta count of certain biological samples. The levels of radioactivity were considerably different for samples from the three collecting areas: the counts of samples from Guam were notably higher than those from Palau, which in turn were very much higher than those from the Gulf of Siam, which were essentially at background level for all collections The gross beta counts of fish muscle from all areas from all collections were constant and less than seven micromicrocuries per gram of wet weight. The samples with the highest gross beta counts were clam kidney and spider snail liver, with maximums of 204 and 356 micromicrocuries per gram, respectively. Gross beta counts of some Guam and Palau samples prior to the arrival of the Hardtack fallout indicate the presence of radionuclides from prior test series. Gamma-emitting nuclides other than naturally occurring potassium-40 included, in order of abundance, cobalt-57, cobalt-60, manganese-54, cerium-144, zinc-65, and silver-110m. The greatest value was 2,300 micromicrocuries per gram of wet weight for clam kidney (Tridacna). The occurrence of silver-110m in the liver of the spiny lobster was of special interest because it is a previously unreported fallout nuclide and so far has been detected only in spiny lobster "liver." Additional information on radionuclides to be expected in marine organisms from the western Pacific can be found in the report of the Hansford Laboratories on the radionuclide analyses of samples collected after the 1956 Redwing test series.

ACKNOWLEDGMENTS

The western Pacific samples were collected by the George Vanderbilt Foundation, Stanford University, and sent to the Laboratory of Radiation Biology, University of Washington, for radiological analyses.

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APPENDIX TABLE A. Average gross beta values, micromicrocuries per gram of wet tissue, of marine organisms collected at Guam, Palau, and Gulf of Siam, 1958 and 1959. Average based on three observations unless noted.

			Guam						Palau						Gulf of Siam			
			1958		1959				1958		1959				1958		1959	
Group	Name	Tissue	June	October	January	April	June	October	June	October	January	April	June	October	June	October	January	April
Fish	Grouper	muscle	4±.46	4±.44	4±.22	5±.23	4±.51	5±.26	4±.21	4±.32	4±.21	4±.65	4±.22	4±.20	5±.43	4±.40	5±.15	3±.33
		liver	10±.65	17±4.8	16±3.8	43±13.	5±1.8	14±7.3	7±1.7	7±2.6	3±.52	5±1.4	3±.44	4±.23	6±.75	2±1.1	2±.25	3±.33
	Surgeonfish	muscle	4±.04	6±.79	5±.24	6±.34	4±.03	5±.21	5±.28	5±.11	5±.62	4±.45	4±.20	5±.41				
		liver	8±.67	20±1.6	26±1.7	31±3.3	19±1.4	16±1.1	5±.73	6±1.2	8± 1.6	6±.89	5±.58	8±.59				
	Goatfish	muscle	5±.13	6±.58	5±.33	6±.52	4±.41	4±.22	5±.51	6±.32	4±.34	4±.07	5±.66	5±.37				
		liver	11±2.5	11±.45	24±3.1	15±2.2	9±2.8	10±3.3	5±.77	4±1.4	5±.94	6±1.1	7±.80	5±.82				
	Tuna	muscle,dark													4±.05	4±.41	3±.39	3±.05
		muscle,light													5±.25	5±.73	4±.39	4±.30
		liver													5±.23	4±.07	4±.20	4±.03
	Anchovy	muscle													3±.35	7±1.6	4±.29	
		liver														5±3.0	3±.29	
		viscera													4±.44			
		midsection																3±.11
	Leiognathus	muscle													4±.34	6±.24	4±.48	2±.08
		liver														14±.14	2±1.1	2±.41
		viscera													4±.62			
Molluscs	Spider snail	muscle	5±.90	5±.45	14±2.4	28±1.4	21±6.0	23±11.	4±.56	7±2.0	4±.21	4±.61	5±.21	4±.08				
		liver	61±13.	60±14.	356±54.	112±12.	224±27.	312±50.	9±1.5	7±.28	8±1.1	16±6.7	13±2.3	14±1.1				
	Tridacna clam	muscle	4±.36	3±.24	6±.86	5±.64	4±.28	4±.46	2±.21	2±.42	3±.16	3±.21	3±.21	3±.10	3±.48			3±.31
		kidney	54±8.9	47±4.2	126±22.	122±3.7	204±29	170±56.	50±13.	46±10.	45±7.0	84±13.	57±3.3	65±5.5	11±1.41			6±.55
	Arca clam	foot															1±.66	2±.40
		soft parts													3±.43	2±.32	2±.29	3±.31
	Pinna pen shell	muscle															2±.20	
		viscera															4±.41	
	Pteria oyster	muscle															2[2]	
		viscera															3[2]	
Crustacean	Land hermit crab	muscle	4±.05	4±.38	3±.16	5±.37	2±.43	3±.19	3±.02	3±.23	2±.36	2±.16	3±.05	3±.36				
		skeleton	23±2.4	14±2.1	44±29.	22±5.8	9±2.2	23±1.3	8±.52	6±1.2	16±3.6	10±1.7	12±3.1	14±3.7				
		skeleton and muscle	12[2]															
		viscera	8[2]															
	Swimming crab	muscle													3±.11	2±.17	2±.26	2±.37
		liver													4±.85	1±.21	3±.61	1±.18
	Coconut crab	muscle							2[2]									
		skeleton							10[2]									
	spiny lobster	muscle		6±.88	8±.65	15±2.4	7±.93	10±1.7	3±.44	4±.66		6±.30	4±.45	4±.30				
		liver		23±7.4	124±21.	136±67.	34±2.4	34±7.5	4±.27	5±1.3		15±3.3	6±.38	6±.38				
Algae	Caulerpa	whole	10±.22	7±.03	36±7.9	61±4.2	44±3.8	6±50	1±.16	2±.20	2±.08	3±.18	1±.13	1±.12				
	Halimeda	whole	4±.46	2±.10	16±.53	5±.87	5±.24	5±.78	0.7±.68	1±.27	2±.08	3±.37	2±.45	4±.37				
	Sargassum	whole		5±.15													7±.10	2±1.0
	Dictyota	whole													11±.30			
	Gracilaria	whole																2±.05
	Laurencia	whole													5±.60			

[1] Average ± one standard error of the mean.

[2] One sample only.

Appendix Table B. Common and scientific names and collecting areas of samples as recorded in "Field Data Record" of the George Vanderbilt Foundation.

Group	Common name	Scientific name	Guam	Palau	Gulf of Thailand[1]
Fish	surgeonfish	Acanthurus lineatus	x	x	
	goatfish	Mulloidichthys samoensis	x	x	
	grouper, sea bass	Cephalopholis argus	x	x	
	grouper, sea bass	Epinephelus merra	x	x	
	grouper, sea bass	Epinephelus hexagonatus	x	x	x
	tuna, bonito	Katsuwonus pelamys			x
	anchovy	Engraulis spp.			x
	anchovy	Stolephorus spp.			x
	leiognathid	Gerres argyreus			x
Crabs	hermit crab	*	x	x	
	coconut crab	*		x	
	swimming crab	*			x
Spiny lobster	lobsters, "langoustre," langusta	*	x	x	
				x	
Snails	sea snails	Lambis lambis	x	x	
Clams	clam	Tridacna		x	x
	clam	Tridacna elongata	x	x	
	clam	Area sp.			x
Oyster	oyster	Pinna			x
Plankton	plankton		x	x	x
Algae	alga	Caulerpa	x	x	
	alga	Halimeda			x

[1] Referred to as Gulf of Siam in text.
*Not given.

Appendix Table C. Gross beta activity of plankton from Guam, Palau, and the Gulf of Siam, 1958–1959. Single determinations (micromicrocuries per gram of wet weight).

Guam		Palau						Gulf of Siam	
Sample date	Activity	Sample date	Activity	Sample date	Activity	Sample date	Activity	Sample date	Activity
1958		1958		1959		1959		1958	
July		June		January		July		July	
13	34±3.7	22	11±2.0	4	3±1.0	5	4±1.1	4	27±3.1
October		29	12±3.6	11	2±0.7	12	1±0.7	October	
5	42±3.4	July		18	6±1.0	19	2±1.0	3	0.5±.35
1959		6	10±1.5	25	8±0.6	26	3±0.8	1959	
January		13	1±0.8	February		August		January	
9	130±4.3	20	6±0.9	1	1±0.8	2	0.5±0.8	5	3±0.8
April		27	31±3.0	8	2±1.5	9	2±0.7	April	
6	50±4.0	August		15	1±0.9	16	1±0.6	2	11±2.3
August		3	0.6±0.4	22	4±0.9	23	0.6±0.9		
18	6±0.8	10	8±1.5	March		30	0.9±0.7		
October		17	6±1.5	1	5±1.2	September			
8	4±0.8	24	1±0.6	8	11±1.6	6	1±0.6		
		31	2±0.6	15	14±2.2	13	1±0.7		
		September		22	3±0.9	20	6±1.0		
		7	1±0.5	29	3±1.0	28	4±1.2		
		14	0.5±0.6	April		October			
		21	3±0.9	5	7±1.2	4	2±0.7		
		28	3±1.0	13	4±1.0	11	2±0.8		
		October		19	6±1.4	18	2±1.0		
		5	2±1.0	20	14±1.7	25	0.5±0.4		
		12	0.8±1.0	26	5±2.0	November			
		19	1±1.3	May		1	1±0.7		
		28	2±2.5	3	4±1.5				
		November		10	3±1.3				
		2	1±1.0	17	2±1.3				
		9	2±2.1	24	3±1.4				
		16	3±1.8	31	8±2.1				
		23	1±0.6	June					
		30	0.9±0.4	7	6±2.2				
		December		14	2±1.5				
		7	2±0.7	21	4±1.2				
		14	0.8±1.0	28	9±2.7				
		21	0.6±0.8						
		28	0.8±0.6						

[1] Values are the 95 per cent errors in the net count; $E_{95} = 1.96\ (Ns/t_s + Nb/t_b)^{1/2}$; from U.S. Public Health Service Radiological Health Handbook, 1960.

QUALITATIVE DISTRIBUTION OF RADIONUCLIDES AT RONGELAP ATOLL

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In March, 1958, a radioecological study of Rongelap Atoll was instituted at the request of the Division of Biology and Medicine, U. S. Atomic Energy Commission (AEC). This report will be concerned with generalizations regarding the distribution of radionuclides at the atoll in the fall of 1959, some five years after contamination with radioactive fallout.

Rongelap Atoll was accidentally contaminated on March 1, 1954, with radioactive fallout from a thermonuclear device detonated at Bikini Atoll some 80 miles to the west. Gamma radiation dose rates at Rongelap on D + 1 (detonation + one day) ranged from 3.5 roentgens per hour at the southern islets of the atoll to 35 roentgens per hour at the northern islets (Dunning, 1957). Eighty-two natives residing on Rongelap Island, in the south, were evacuated and did not return until June, 1957. At that time the returning population approached 300 in number but since appears to have stabilized at 230.

Several radiological and biological surveys, primarily of a monitoring nature, were conducted from the time of the first contamination until 1958 (Dunning, 1957). During this time the gamma radiation dose rates over land areas declined at approximately the rate predicted for mixed fission products by Miller and Loeb (1958). Slight rises in gamma dose rate were observed in 1956 and 1958, resulting from tests conducted during these years. However, the total contribution of radionuclides from these subsequent fallouts amounted to a fraction of one per cent of the amount from the 1954 fallout.

Rongelap Atoll is located in the Marshall Islands, in the Central Pacific Ocean, at about 11° North. It is a typical atoll with a lagoon area of 388 square miles and about 180-foot average depth. The emergent land area is about three square miles and is made up of 61 small islets ranging in size from a fraction of an acre to the largest island, Rongelap, which is about four miles long and one-half mile across at its widest point.

There is one small islet on the western reef and the remainder are strung along the northern, eastern, and southern reefs. The islets on the northern reef are not as well developed as those to the east and south. The waters of the lagoon are essentially isothermal (Robinson, 1954). The circulation, generated by the northeast trade winds, is from east to west at the surface with a returning bottom current (Von Arx, 1954). The estimated time for renewal of water in the lagoon is about 30 days.

The parent material of both soils and the lagoon bottom is primarily calcium carbonate originating mainly from coralline algae, corals and foraminifera. There is also some accumulation of [illegible] in the soils.

The natives of the area are Micronesians. Their agriculture is limited in variety of products and the only significant export is copra. About half of the food consumed at the present time is imported. Fish and other marine organisms are eaten but these sources are not exploited as much as they could be. The Rongelapese are almost entirely dependent on cisterns as a source of water. Rainfall in this area is comparatively low and the islets small, so that there is not a well-developed fresh water lens. There is, however, some potable water in wells at Rongelap and Eniwetok Islets.

The native style wattle and palm frond buildings have been replaced by plywood and aluminum structures built to Rongelapese specifications by the AEC. Sanitation habits have been altered by the advent of pit toilets.

The terrestrial fauna is limited in variety. The only mammal present is the small field rat, Rattus exulans. The most common birds are the fairy tern, Gygis alba, and the noddy terns, Anous stolidus and A. tenuirostris, which nest in large numbers on some of the uninhabited islets. The reptiles are represented by skinks, geckos, and a blind snake. Land crabs are common, the most spectacular being Birgus latro, the coconut or robber crab. Insects are few, both in number of species and individuals. The most severe pest appears to be the beetle, Brontispa sp., which attacks the coconut palm.

In contrast to the land areas there is a tremendous proliferation of both numbers and variety of organisms on the reefs and in the lagoon. For example, there are over 700 species of fish. Plankton, however, is extremely sparse and as a consequence the water is so clear that green algae are found growing at depths of 180 feet.

Since the question of the effects of radiation on the organisms inevitably arises, it might be well to consider it briefly before going on to the main subject. There is no doubt that the levels of radiation were of sufficient intensity to affect living organisms, However, under actual field conditions and without benefit of study before the addition of radiation as an ecological factor, it is difficult to do more than speculate concerning the cause of the specific anomalies observed. Fosberg (1959) has accurately described the poor condition of the plants at the northern islets of Rongelap Atoll and has suggested that the primary cause of this condition is radiation. In our opinion, however, other factors, particularly edaphic factors, have probably been more important than radiation. The fact that the nitrogen content of the soils of the northern islets is lower than that of the rest of the atoll is at least circumstantial evidence that for some time there have been differences between these areas with respect to plant growth. Stone et al. (1957) have concluded from studies of Drosophila populations at Bikini, Eniwetok, Rongelap, and uncontaminated atolls that while there is evidence of genetic changes caused by radiation other factors mask the radiation effects. In short, it is not likely that such questions will be resolved without controlled experimentation with the species involved, under varying conditions, and with an eye toward the possibility of synergistic effects.

Approximately five years after fallout the long-lived fission products cesium-137 and strontium-90 are the principal radionuclides found in the land organisms, while the neutron-induced radionuclides zinc-65, cobalt-60 and manganese-54 are found primarily in the marine organisms.

Still detectable in the soil are manganese-54, iron-55, cobalt-57, cobalt-60, zinc-65, strontium-90, zirconium-95, ruthenium-106, antimony-125, cesium-137, cerium-144, and europium-155, which remain concentrated in the upper one to two inches. Where higher levels of radionuclides have been present these nuclides have been reported in a wide variety or organisms. It is likely that most of these radionuclides are actually present in most if

not all organisms at Rongelap but that the levels at which they occur are extremely low and so escape detection.

Passing from the soil to the soil solution, the term being used here to mean leachates collected in the field from lysimeters, strontium-90, cesium-137, and antimony-125 are the principal nuclides found, although Ru^{106}-Rh^{106}, cerium-144, and europium-155 are also detectable (Cole et al., 1961). Here differences exist with respect to soil type in that the leachates from immature soil, consisting almost exclusively of parent material, contained only antimony-125 and strontium-90. There is to us no evident explanation for this difference.

The ground water probably contains these nuclides since their movement has been detected in leachates to depths of 30 inches, but the levels are so low in ground water that special techniques would have to be developed to detect them.

The land plants contain principally cesium-137 and strontium-90. Manganese-54 and zinc-65 have been found in plants from the more heavily contaminated islets but are present in relatively insignificant amounts. In general, cesium-137 accounts for 90 per cent or more of the radioactivity in the land plants and strontium-90 for the remainder. This is unlike the situation usually found on continental soils and is a consequence of the low potassium content of Rongelap soil. Amendments of potassium to Rongelap soil reduce the uptake of cesium-137 by plants (Walker et al., 1961), and affect the distribution of cesium-137 within the plant. There are, of course, differences between plant species and plant parts with respect to the relative amounts of cesium-137 and strontium. For example, copra contains very little strontium-90 as compared with Pandanus fruit, and the basal leaves of various plants contain more strontium-90 relative to cesium-137 than do the terminal leaves. This variation is related to differences in mobility between cesium and potassium, and strontium and calcium.

The rats contain cesium-137 and strontium-90, reflecting the radionuclides present in the plants on which they feed. The coconut crab and the land hermit crab (Coenobita perlatus) contain the same nuclides but concentrate strontium-90, as has been reported for Coenobita from Eniwetok Atoll (Held, 1960).

The occurrence of radionuclides in man at Rongelap has been summarized by Cohn et al. (1960). In 1958 these nuclides were cesium-137 and strontium-90 coming form the food plants, and zinc-65 coming to man from marine products.

The birds, which feed almost exclusively on marine organisms, contain primarily zinc-65 and occasionally small amounts of manganese-54 and cobalt-60. Strontium-90 is also found in small amounts in bird bone and may reflect direct uptake from the ingestion of soil, although there is no direct evidence that this occurs.

Radionuclides in fish are limited to manganese-54, cobalt-60, and zinc-65, the latter being predominant. On a dry-weight basis for a sample of goatfish (Mulloidichthys samoensis) testes have the highest levels, the liver, gastrointestinal tract, and eyes are lower by about an order of magnitude, and the muscle and bone lower by still another order of magnitude. If the total amount of radioactivity by tissue is considered, then bone is the principal depository of zinc-65 (Joyner, 1961, personal communication). The sources of zinc-65 for fish are open to question. In some instances invertebrates containing zinc-65 are known to be consumed by fish found to contain zinc-65, but, in general, no definite sources of zinc-65 are known to exist five years after fallout. It is possible that there is concentration of undetectable levels from the sea water or algae. The possibility that most of the zinc-65 radioactivity in fish is residual appears to be ruled out by the fact that young fish contain relatively high levels.

The marine invertebrates taken as a whole contain a wider spectrum of radionuclides than do the fish. These are manganese-54, cobalt-57,60, zinc-65, strontium-90, cerium-144, and probably europium-155. The corals contain cobalt-60 and are the only invertebrates in which strontium-90 has been consistently detected. From limited data available thus far it appears that these nuclides were deposited in the skeletal material soon after fallout and have remained localized in portions of the coral colony actively growing at that time. The clams contain mostly zinc-65, cobalt-57 and cobalt-60. Weiss and Shipman (1957) originally reported the concentration of cobalt-60 in the kidney of Tridacnid clams collected at Rongelap in 1956. Animals such as the sea cucumber (Holothuria, Stichopus) and spider snail (Lambis, Strombus), which ingest large amounts of bottom sediments, contain ruthenium-106, cerium-144, and probably europium-155.

Of several species of algae sampled in 1959 the only radionuclides detected were ruthenium-106, cerium-144 and europium-155. In general, the levels of radioactivity in the algae are lower than in the fish or invertebrates.

The plankton contain manganese-54, cobalt-57,60, zinc-65, zirconium-95, ruthenium-106, and cerium-144, but all in minute amounts. In 1959 plankton samples collected by pumping a total of two and a half million gallons of water were pooled for gamma-ray spectrum analysis and were found to contain only enough of these nuclides for qualitative analysis without resorting to chemical separations. Further analysis has been deferred until other studies with the individual samples can be completed.

The lagoon sediments contain strontium-90, ruthenium-106, cerium-144, and europium-155. The radioactivity is associated mainly with the fines and is concentrated in the top two to four inches, dropping off rapidly with depth.

Radionuclides other than naturally occurring potassium-40 were not detected in sea water although larger samples and more sensitive techniques undoubtedly would have revealed their presence.

In sum, on land the present distribution of long-lived fission products, strontium-90 and cesium-137, can be expected to remain very much as it is now. The levels of radioactivity will be reduced primarily by physical decay of the radionuclides so long as other factors such as changed agricultural practices or a catastrophic storm do not occur. In the lagoon, the levels of radioactivity will decline more rapidly than on land because of the presence of shorter-lived radionuclides, with the exception of strontium-90. The latter does not enter the marine food web to any significant extent and may remain as a label useful in evaluating the long-term effects of physical forces in the lagoon.

SUMMARY

The qualitative distribution of radionuclides at Rongelap Atoll as determined approximately five

ears after contamination by fallout from a thermonuclear device indicates distinct differences between the terrestrial and marine environments. The levels of radioactivity are low, the concentrations being less than the maximum permissible concentration for radionuclides in food or drinking water of man. Of the wide spectrum of radionuclides concentrated in the surface layers of the soil, strontium-90, antimony-125, and cesium-137 are the principal nuclides entering into the soil solution. The principal nuclides in the land plants and plant-eating animals such as coconut crabs and the indigenous rats are cesium-137 and, to a lesser degree, strontium-90. Bottom sediments contain mainly strontium-90 and europium-155. The radionuclides in the lagoon water have not been detected but are probably present in minute amounts. Planktonic organisms contain traces of manganese-54, cobalt-57,60, zinc-65, zirconium-95, ruthenium-106 and cerium-144. The principal nuclide found in the marine algae is cerium-144. In the marine invertebrates cobalt-60 and zinc-65 occur most commonly. Corals and coralline algae contain some strontium-90, while the fish and sea birds are found to contain mostly zinc-65. The presence of zinc-65, cesium-137, and strontium-90 in the body of the natives reflects a diet of both marine and terrestrial origin.

ACKNOWLEDGMENTS

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INCREASE IN RADIORESISTANCE OF FISH TO LETHAL DOSES WITH ADVANCING EMBRYONIC DEVELOPMENT

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INTRODUCTION

Few papers exist on the radioresistance of the eggs of fishes at various developmental stages. Welander (1954) showed the resistance of rainbow trout eggs to increase with advancing age of embryo, as did Rugh and Clugston (1955) for Fundulus heteroclitus. Belyaeva and Pokrovskaya (1957, 1958) reported on several stages of development of the eggs of the loach, Misgurnus fossilis through late blastula. Golvinskaia and Romashov's useful review of the literature appeared in 1958. Rugh (1954) gave pertinent data for amphibian development. Interpretation of the findings of all of these authors discloses a relationship of radioresistance to stages of embryonic development, that is common to fish and to at least some other organisms. Thus it is our objective to determine the rate of increase in radioresistance of fish eggs with advancing embryological development, to compare this rate with some other type of organism, and to quantify the relationship if possible. We present one block of Welander's heretofore unpublished data on irradiated silver salmon eggs and refer to published literature for the rest of the data.

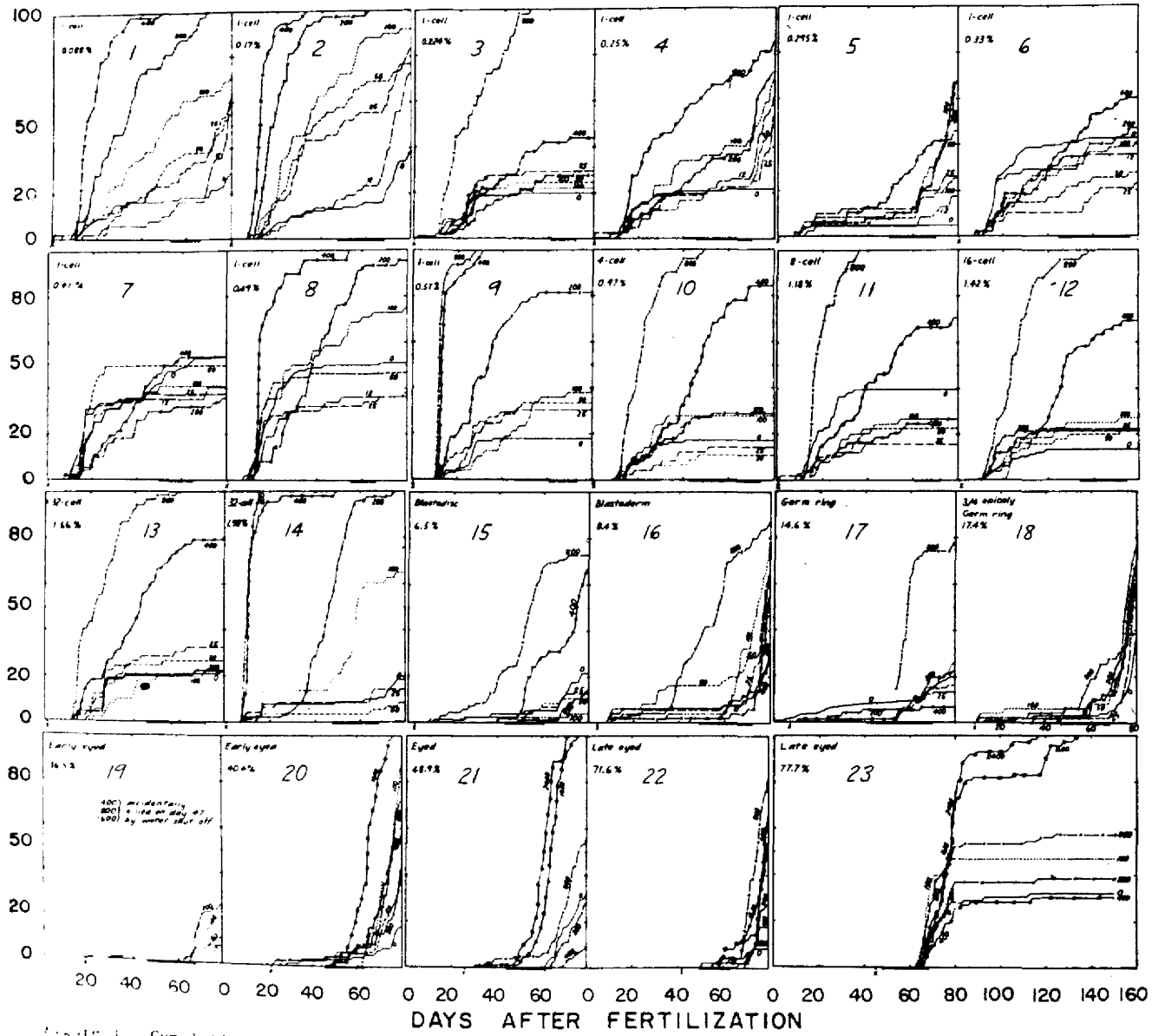

FIGURE 1. Cumulative percentage mortality of eggs and young of silver salmon in 23 experiments involving irradiation at different stages of development. Numbers along the curves are doses in roentgens. Mortalities are shown up to 80 days, and to 150 days in experiment 23. Below the abscissa are indicated, by an X, the day of irradiation, and, by a horizontal line, the maximum duration of the hatching period. At the upper left of each rectangle are given the stage of development and the percentage of incubation completed at the time of irradiation.

MATERIALS AND METHODS

In November and December 1951 the silver salmon, Oncorhynchus kisutch which had homed to the rearing ponds at the University of Washington in Seattle were spawned artificially and incubated in screen baskets in hatchery troughs. Methods of irradiating eggs and of handling eggs and larvae were as described by Welander (1954, p. 229), and were briefly as follows.

Fertilized eggs were irradiated at each of 23 stages of development from zygote to late-eyed stage. About 50 to 100 eggs, barely covered by water, rested in a single layer on a flat screen, and were confined by a low plastic collar. A Picker deep therapy x-ray machine operating at 200 kilovolt peak and 20 milliamps, with tungsten target and filters of one millimeter aluminum and 1 millimeter copper plus an approximately equal inherent filtration, delivered about 100 roentgens per minute at a distance to the eggs of 39 to 40 centimeters. Dose rate was determined with a Victoreen 250-roentgen medium energy chamber and meter. Doses ranged from 12 to 2,400 roentgens, incremented by a factor of approximately two. Seven doses distributed to embrace the anticipated lethal dose-50 were used at each embryological stage. Eggs were returned to the incubating baskets in the hatchery troughs immediately after irradiation. Dead eggs (at least partially opaque) were removed once or twice daily and preserved in formalin.

RESULTS

Figure 1 shows post-irradiation mortality related to time for each of 23 experiments. The stage of development and the percentage of the incubation period attained at the time of irradiation are inserted at the upper left in each graph. Below the abscissa the day of x-raying is indicated by an X, and the maximum range of the hatching period, by a solid horizontal line at about 60 days. After about 80 days most of the mortalities remained fairly constant to the end of the observation period at 150 days, as seen in the last graph (23) where this later period is shown for an example.

The lethal dose-50's at hatching were calculated for each experiment except numbers 7, 19, and 23 using the method of Kärber (1931; p. 481) as modified by Irwin and Cheeseman (1939; p. 574), after adjusting for control mortalities as shown by Finney's (1944; p. 68) use of Abbott's formula. Lethal dose-50's at 150 days were similarly calculated omitting experiments 7 and 19. The few (less than 0.1 per cent) accidental deaths were simply subtracted from the original totals when computing percentage mortalities.

In order to utilize the data of the experiments omitted in calculating lethal dose-50's, and to serve as a uniform basis for comparison with other organisms reported in the literature, subjective estimates were made from the graphs of Figure 1 at hatching, and from extensions of these graphs to 150 days as exemplified in Figure 1 by experiment 23. Graphs for experiments 1 through 22 were arbitrarily shown to only 80 days, although their extensions to 150 days were available for reference. Table 1 gives the resulting evaluations of lethal dose-50. Graphs of these relationships are shown in Figure 2.

For relating lethal dose-50 to stage of embryonic or larval development at the time of irradiation, the stage of development was expressed as percentage of developmental period from ferti-

Table 1. Lethal dose-50 of silver salmon irradiated at 23 stages of incubation, 50 to 100 eggs per calculation.

Per cent of incubation when irradiated	Lethal dose-50 in roentgens			
	At hatching		At 150 days	
	calculated[1]	estimated[2]	calculated[1]	estimated[2]
.088	66	80	25	20
.170	30	50	16	20
.224	373	450	277	300
.250	177	400	26	80
.295	249	600	84	150
.331	471	600	309	400
.413		800		400
.493	114	400	97	200
.57	108	170	80	120
.97	268	350	252	300
1.18	350	400	300	400
1.42	264	400	211	300
1.66	298	350	296	300
1.98	98	120	93	100
6.5	586	800	465	400
8.4	669	800	88	800
14.6	702	800	635	700
17.4	757	1,200	138	800
36.5		1,600		800
40.4	921	1,600	115	600
48.8	1,423	2,400	607	700
71.6	1,871	3,000	488	700
77.7		3,000	874	1,000

[1]Calculated lethal dose-50's.

[2]Lethal dose-50's estimated from graphs.

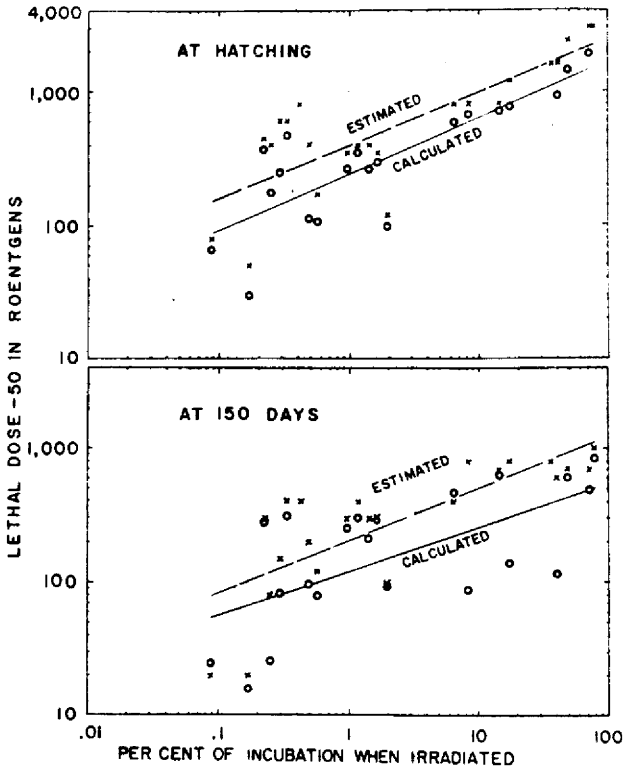

Figure 2. Relationship of lethal dose-50 to per cent of incubation attained at time of irradiation of eggs of silver salmon. Upper, lethal dose-50 at hatching. Lower, lethal dose-50 at 150 days. Circles and solid lines, calculated lethal dose-50's; crosses and broken lines, estimated lethal dose-50's from graphs.

lization to hatching for fish, or to metamorphosis for amphibia. Regressions were calculated (Snedecor, 1956) using logarithmic transformation. The number of observations, the regression coefficients, and the probabilities determined from the value of "t" appear in a summarizing table (Table 6).

ANALYSIS OF PUBLISHED RESULTS

Welander (1954) irradiated the eggs of rainbow trout (Salmo gairdnerii) in various stages from one-cell to late-eyed stage. From his Table 1 and 2 it is possible to relate lethal dose-50 as determined both at hatching and about two months later at the completion of yolk absorption to percentage of incubation reached at the time of irradiation. Lethal dose-50's were also estimated from Welander's (1954, page 235) graphs, and both calculated and estimated lethal dose-50's are used in Table 2 and Figure 3.

Table 2. Lethal dose-50 of rainbow trout at six stages of incubation.

Per cent of incubation when irradiated	Lethal dose-50 in roentgens			
	At hatching		At end of yolk stage	
	calculated[1]	estimated	calculated[1]	estimated
.28	78	80	58	70
2.54	468	600	313	400
17.3	524	700	461	600
25.	735	1,000	454	500
67.	-	3,000	415	600
89.	-	4,000	904	1,000

[1]Lethal dose-50's calculated by Welander (1954, page 233).

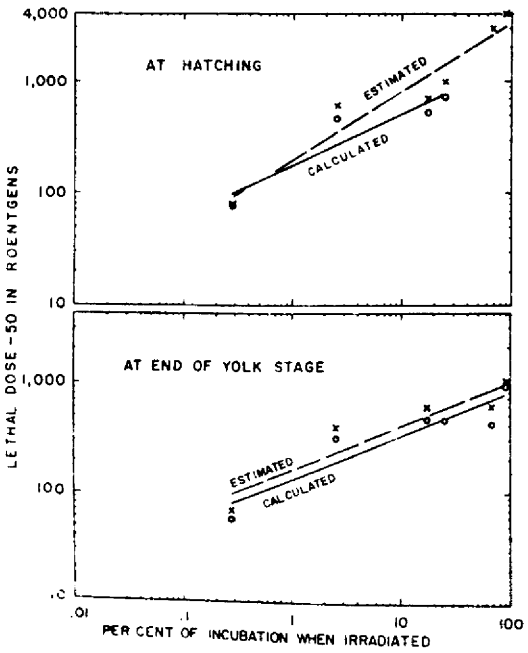

Figure 3. Relationship of lethal dose-50 to per cent of incubation of rainbow trout eggs when irradiated (Welander, 1954). Lethal dose-50's determined at hatching (upper), and at end of yolk stage (lower). Circles and solid lines, based on calculated lethal dose-50's, crosses and broken lines, based on estimated lethal dose-50's.

Rugh and Clugston (1955) described the effects of x-radiation upon various stages in the embryonic development of Fundulus heteroclitus. Although the lethal dose-50 is not stated, their descriptions of results permitted our estimating the lethal dose-50 (Table 3) at four stages which can be converted to percentage of the total incubation period (11 days) using the time-table of embryonic development contributed by Solberg (1938). Table 3 and Figure 4 give the trend of embryonic radioresistance of Fundulus which closely resembles the preceding trends.

Table 3. Responses of embryos of Fundulus heteroclitus to x-irradiation, from Rugh and Clugston, (1955), with subjective estimates of lethal dose-50 based upon their descriptions.

Stage irradiated			
Name or number	Per cent of incubation	Effect	Estimated lethal dose-50 at hatching (roentgens)
1- to 2-cell	1.14	-300 to 400 roentgens resulted in stunted, but otherwise normal development;600 roentgens,25 per cent hatched, stunted; reached stage 15 when controls at stage 22....	400
2- to 8-cell	1.90	-Only slightly less sensitive than 1- to 2-cell; 500 roentgens permitted quite normal development.....	600
11;expanding blastula	5.30	-1,000 roentgens permitted only a few to hatch.....	800
8-day embryo	72.8	-Tolerated up to 3,000 roentgens without a marked decrease in the number developing normally.....	3,000

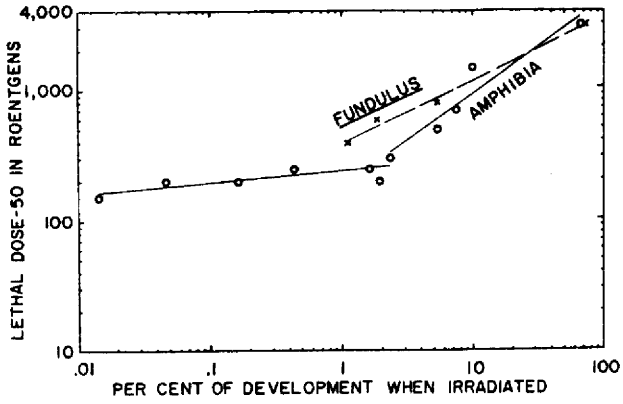

Figure 4. Relationships of lethal dose-50 to per cent of incubation of the eggs of Fundulus (Rugh and Clugston, 1955), crosses and broken line; and of lethal dose-50 to per cent of time from fertilization to metamorphosis of amphibia (Rugh, 1954), circles and solid lines.

Belyaeva and Pokrouskaya (1957, 1958) reported for the loach, Misgurnus fossilis, that anaphase-telophase was the most sensitve mitotic phase, 20 times as sensitive as interphase, and that radioresistance rose with advancing embryological development. From their description of results, lethal dose 50's may be roughly estimated for four stages (Table 4), but, because of the high resistance at last blastula, variability is too great for statistically significant regression (Table 6).

Rugh (1954) described the results of irradiating amphibia at various stages from the uterine egg

Table 4. Estimates of lethal dose-50's of Misgurnus fossilis embryos based upon descriptions of x-irradiation experiments by Belyaeva and Pokrovskaya (1957, 1958) at four stages of development (referred to Solberg's 1938 time table for Fundulus for quantifying).

Stage irradiated					Estimated lethal dose-50 at hatching (roentgens)
Stated	Per cent incubation	Year	Source	Description	
Anaphase 1st cleavage	0.37	1958	Figures 1a and 1b		100
Anaphase 2-3 cleavages	0.85	1958	Figure 2		300
Early blastula	2.3	1957	Table 1	200 roentgens, hatching, but abnormal forms found; 500 roentgens, some larvae hatch, abnormal.	300
Late blastula	5.3	1957	Table 1	500 roentgens, all hatch; 1,000 roentgens, majority hatch.	1,200

to metamorphosis, a period of 90 days. Some of his descriptions pertaining to the lower doses, and our own estimates from these descriptions of the resulting lethal dose-50's at metamorphosis appear in Table 5. The relationship of lethal dose-50's to percentage of the period from fertilization to metamorphosis at the time or irradiation is plotted in Figure 4 along with the relationship for Fundulus. For amphibia two trends are evident, a slow increase in radioresistance in the early stages up to the time of neurulation (two per cent of the premetamorphic period) expressed by the logarithmic equation $Y = 229\ X^{0.0712}$ followed after neurulation by a more rapid increase in resistance, $Y = 185\ X^{0.698}$. Hatching, at 5.4 per cent of the premetamorphic period, appears not to affect the rate of increase in radioresistance.

Table 5. Responses of amphibian embryos and larvae to X-rays, modified from data of Rugh (1954). Subjective estimates of lethal dose-50 are based on Rugh's description of effects.

No.	Days of age	Per cent of 90 days	Description and results	Estimated lethal dose-50 at metamorphosis (roentgens)
0	0	0	-Newly fertilized egg(sperm enters egg):150 roentgens,50 per cent developed normally	150
1	.013	.0144	-Polar body extrusion(19 minutes):300 roentgens,developed only to yolk plug stage	150
2	.042	.047	-Zygote: 300 roentgens, 16 per cent gastrulated, then died	200
3	.15	.167	-2-cell, 2½ hours:300 roentgens, reached late gastrula 54 per cent neurulated	200
6-8	.4	.44	-Blastula, 16-many-cell:180 roentgens, lived indefinitely; 360 roentgens, 70 per cent had nerve cord	250
11	1.47	1.63	-Gastrula:300 roentgens, developed to external gill larvae,but with ventral tail flexure	250
12	1.75	1.94	-Yolk plug stage: 300 roentgens, died at 9 days	200
13	2.1	2.33	-Neurula:300 roentgens, some outlived duration of experiment	300
19	4.9	5.4	-Hatching: 300 roentgens, no effect; 600 roentgens, 54 per cent lived 5 days	500
21	6.8	7.6	-Early external gill stage:500 roentgens,survived 3 months to metamorphoris;1,000 roentgens, survived 7 days	700
23	9.0	10.0	Absorbing external, and growing internal, gills:1,000 roentgens, indefinite survival; 2,000 roentgens,20 per cent died in 8 days; 15,000 roentgens, 50 per cent died in 9 days	1,500
40	60	67	-Premetamorphosis	3,000

Table 6. Regression of lethal-dose-50 upon per cent of development from fertilization of the egg to hatching, for fish, or to metamorphosis, for amphibia.

			Lethal dose-50 (y) in terms of per cent development (x) when irradiated											
			Using calculated lethal dose-50						Using estimated lethal dose-50					
Organism	Authority	Stage when lethal dose-50 was determined	Number of observations	Roentgens at one per cent incubation(a)[1]	Regression coefficient or slope (b)[1]	$t_{.05}^2 s_b$	t	P	Number of observations	Roentgens at one per cent incubation (a)[1]	Regression coefficient or slope (b)[1]	$t_{.05}^2 s_b$	t	P
Silver salmon	(present)	hatching	20	240	.421	.194	4.56	<.001	23	393	.398	.137	5.83	<.001
Rainbow trout	Welander	150 days hatching	21	125	.331	.188	3.68	<.005	23	187	.390	.135	6.46	<.001
Fundulus	Rugh and Clugston	end of yolk hatching	4	179	.457	.547	3.59	<.08	6	198	.612	.245	6.92	<.005
			6	131	.390	.231	4.68	<.01	6	163	.390	.214	5.04	<.01
									4	394	.469	.149	13.7	<.01
Misgurnus	Belyaeva and Pokrovskaya	hatching							4	214	.925	1.62	2.33	<.2
Amphibia	Rugh	metamorphosis												
		(preneurulation)							7	241	.0896	.0794	2.90	<.05
		(postneurulation)							5	185	.698	.415	5.35	<.02

[1] Values in the fitted logarithmic equation, $y = ax^b$.

[2] Addition and subtraction of $t_{.05}^2 s_b$ from b give the .05 confidence interval.

DISCUSSION

The patterns of embryonic radioresistance of fishes and amphibia under consideration may be expressed mathematically with fair correspondence between species. Table 6 shows the available regressions of lethal dose-50 determined either at hatching or somewhat later upon percentage development to hatching for fish, or to metamorphosis for amphibia when irradiated.

Disregarding the data for Misgurnus which lacks statistical significance, and considering Fundulus and the four pairs of values for salmon and trout based on both calculated and estimated lethal dose-50's, the data of Table 6 show a mean and standard error lethal dose-50 for the one per cent (logarithmic Y-intercept) level of incubation, of 223 ± 34 roentgens. The nine corresponding coefficients of regression averaged 0.431 and ranged from 0.331 to 0.611, with probability values at or beyond the 0.08 level when tested with "t" The preneurulation and postneurulation regressions for amphibia are of comparable statistical significance.

For salmon and trout a comparison was made between calculated and estimated values of the lethal dose-50 given by the regressions. Figures 2 and 3 and Table 6 show the value of the estimated, tending to exceed that of the calculated, lethal dose-50. The slopes of the lines do not differ significantly, however, when regression coefficients are compared by the method of Fisher (1946, p. 140).

Since time is required for radiation to take effect, the apparent increase in radioresistance with advancing stage of development of an organism, is in part a function of the decreasing time interval between irradiation and the stipulated time of determination of the lethal dose-50. The shortening of this interval loses importance, however, when the lethal dose-50 is determined at an appreciably later stage for doses administered before hatching. Because of the decreased mortality rate between hatching and either yolk sac absorption (trout) or 150 days post-fertilization (salmon), the lethal dose-50's at the later stages were only slightly lower than at hatching (Figure 1, experiment 23). Thus, the increased resistance in the later stages is real rather than apparent.

Although variability in early radioresistance is apparently only slightly species dependent, variation in radioresistance is nevetheless believed to occur as a result of difference due to stage of development, especially in the early embryonic stages. Fitting of straight lines to our observations representing random sampling of stages is not meant to preclude the existence of great deviations caused by developmental processes within this general trend. Henshaw (1940, p. 919) showed for the sea urchin Arbacia that the resistance fluctuated by factors or two to three in a cyclic manner from fertilization, through the second cleavage. Neifakh and Rott (1958, Figure 3, curve 1) indicated that the synchronous early cleavages are accompanied by 11 cycles of oscillation involving about a five-fold fluctuation in the radioresistance to 500 roentgens of the egg of Misgurnus fossilis during the first six hours after fertilization. In the early cleavages of the silver salmon we have tried to discern any possible relationship between low values of lethal dose-50 and known or suspected periods of susceptibility, for example, the anaphase of mitosis. Although an association of radiosensitivity with phase of mitosis or other developmental stages undoubtedly exists, it is not shown by our data. More study of the early cleavage period is needed.

SUMMARY

Experimental data and analysis of published results indicate that the radioresistance of the embryos of fishes and amphibia increases with advancing development in a way that may be expressed mathematically. Eggs of silver salmon were x-irradiated in 23 stages of development randomly selected from one-cell to late-eyed stage, at each stage using seven dosages incremented by a factor of about two. Lethal dose-50's at hatching and at 150 days post-fertilization were both calculated and estimated from graphs. The regression of log lethal dose-50 in roentgens upon log percentage of incubation attained at time of irradiation was computed at both hatching ($Y = 240\ X^{0.42}$) and at 150 days ($Y = 125\ X^{0.33}$). These regression coefficients (the exponents of X) had confidence limits determined by $t_{.05}S_b = 0.194$ and 0.188, with 18 and 19 degrees of freedom respectively. The regressions represent general trends rather than the most sensitive stage. Estimates from the literature give similar relationships for rainbow trout, Fundulus and Misgurnus, although the regression for Misgurnus was non-significant. Amphibian radioresistance increased very slowly from fertilization to neurulation, and then sharply from neurulatiom almost to metamorphosis.

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FACTORS CONTROLLING THE DISTRIBUTION OF THE RARE EARTHS IN THE ENVIRONMENT AND IN LIVING ORGANISMS

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INTRODUCTION

The rare earths include the elements in the periodic table from lanthanum to lutetium (atomic numbers 57 through 71) and, because of their similar chemical behavior, yttrium (atomic number 39) and scandium (atomic number 21) usually are included in a discussion of the rare earths. This closely knit group of elements, often referred to as the lanthanides or lanthanons, received little attention until radionuclides of the group became available for study. Under normal conditions the rare earths exist in aqueous solutions only as the trivalent ions. This constancy of oxidation state arises from the fact that beyond lanthanum added electrons enter an inner shell (4f) where space for 14 electrons exists. The filling of the 4f shell leads to the rare earth series and causes the unusual properties of the members of this series (Moeller, 1956). The availability of radiolanthanides and recent improvements in ion exchange and solvent extraction techniques have made feasible the previously difficult separation of these elements from one another. A comprehensive review of the radiochemistry of the rare earths has been prepared recently by Stevenson and Nervik (1961). Because of their special characteristics and their abundance in radioactive fallout, these nuclides deserve detailed consideration from the standpoint of their distribution in the environment and their uptake by living organisms.

There is little information regarding the presence of the rare earth elements in nature. In Vinogradov's (1953) compilation of the chemical composition of marine organisms only a few data are given for the rare earths. Trace amounts of praseodymium, neodymium, samarium, cerium, lanthanum, and yttrium have been found in the coralline alga, Lithothamnion. Yttrium has been found in radiolarians, globigerina silts and gorgonaceans and samarium has been found in two species of corals. In the land environment small amounts of rare earths have been found in soils and in crops and native plants (Robinson, 1943).

The rare earths occur usually in insoluble form and thus are not readily available for uptake by living systems. The factors which control the rare earth nuclides in the environment and in the biota can be divided for the sake of convenience into two groups, the physico-chemical and the biological. The biological factors relate to the ability or inability of the organisms to absorb nuclides in the particulate state, and these factors depend in turn on the types of organisms and the methods by which nutrients enter them.

The physico-chemical factors govern the distribution and availability of the nuclides and include the radioactive half-life, the physical state and the chemical behavior of the element and certain other forces in nature which will be described.

Although many radioactive lanthanides are formed in the production of nuclear energy, only a few of these have been studied in relation to their distribution in the environment and their fate in living organisms. This discussion will be concerned primarily with Ce^{144}-Pr^{144}, lanthanum-140, promethium-147, and yttrium-90, 91.

PHYSICO-CHEMICAL FACTORS

The physical half-life in combination with fission yield affects the abundance of the rare earth nuclides produced during nuclear detonations. Thus as shown in Table 1 cerium-141 and lanthanum-140, the rare earth daughter of barium-140, are present for a relatively short time after fission, whereas Ce^{144}-Pr^{144} and promethium-147 persist for some time. The results of radiochemical analyses of plankton samples illustrate the fact that the composition of rare earth nuclides changes with time. Plankton samples collected one week after a series of nuclear tests in 1958 in the Pacific contained 23 per cent Ba^{140}-La^{140} and no detectable Ce^{144}-Pr^{144}. Samples collected at six weeks, however, contained no Ba^{140}-La^{140} while Ce^{144}-Pr^{144} contributed five per cent of the total activity (Lowman, 1960). About a year after the 1954 test series Harley (1956) found that 80 to 90 per cent of the radioactivity in the plankton was due to Ce^{144}-Pr^{144}; no other rare earth nuclides were reported.

Table 1. Percentage abundance and physical half-life of some rare earth fission products.

Nuclide	Percentage abundance at:[1] 10 days	30 days	1 year	10 years	Physical[2] half-life
Lanthanum-140	12.0	12.5			40.2 hours
Cerium-141	6.3	11.2			33.1 days
Cerium-144		2.0	26.5		285 days
Praseodymium-144		2.0	26.5		17.3 minutes
Promethium-147			5.7	15.8	2.64 years
Yttrium-90			1.8	21.8	64.2 hours
Yttrium-91	3.4	7.6	3.9		57.5 days
Neodymium-147	4.8	4.1			11.1 days
Praseodymium-143	10.0	11.2			13.8 days
Samarium-151				2.5	approx. 93 years

[1] From Hunter, H.F., and N.E. Ballou (1951).

[2] From Strominger, D., J.M. Hollander, and G.T. Seaborg, (1958).

The physical state of the rare earth nuclides is a most important factor in determining their uptake and distribution in the environment. When the rare earth nuclides are formed in the high temperatures associated with nuclear detonations, they exist probably as oxides; and upon contact with other matter and with the sea or with fresh water, they retain their particulate nature, primarily as the hydroxides of the type $R(OH)_3$. Greendale and Ballou (1954) determined the physical states of various fission products following their vaporization in sea water and distilled water and found that the major portion of the cerium and yttrium was in the particulate state (Table 2).

Table 2. Physical state of cerium and yttrium following an underwater vaporization.[1]

Element	Solution	Physical state (per cent) ionic	colloidal	particulate
Cerium	sea water	1	4	95
		2.7	4	94
	distilled water	0	0.5	99.5
		0	2	98
Yttrium	sea water	0.4	2.2	98
		0.2	6.6	93
	distilled water	1.6	13	86
		1.3	19	80

[1] From: Greendale, A.E., and N.E. Ballou (1954).

other rare earth nuclides, praseodymium-144 and promethium-147 probably occur in the particulate state when added to sea water (Carritt and Harley, 1957), and all of the elements of this group, having similar chemical characteristics, probably exist in this form. Rice and Willis (1959) found that the precipitation of cerium-144 proceeded at a faster rate in distilled water than it did in water of higher salinities, an indication also that cerium exists in the particulate form in fresh waters. The extremely low percentage of stable cerium, yttrium, and lanthanum in solution in the oceans compared with the total amount that has been supplied, a relationship that has been designated the "transfer percentage" by Goldschmidt, reflects the low solubility of these elements. The solubilities in water of some rare earth elements as determined by Moeller and Kremers (1945) and the "transfer percentages" for these elements are given in Table 3.

Table 3. Solubility product[1] and transfer percentage[2] of some rare earth elements.

Element	Hydroxide solubility product (at 25° centigrade)	Total supplied (ppm)	Amount present in ocean (ppm)	Transfer percentage
Yttrium	5.2×10^{-22}	16.9	.0003	.002
Lanthanum	1×10^{-19}	11	.0003	.003
Cerium^{+3}	$0.8\text{-}1.5 \times 10^{-19}$	27.7	.0004	.001

[1] From: Moeller, T., and H.E. Kremers (1945).

[2] From: Goldschmidt, V.M., quoted in Rankama, K., and T.G. Sahama (1950).

As stated by Carritt and Harley (1957) "...it would appear...that marine waters are saturated with respect to these elements and that a major portion of the rare earth elements are dispersed in the sea as solids." From the above data it can be concluded that any radioactive rare earth nuclide added to the sea or to fresh water will exist mostly in the particulate state.

In the hydrosphere the spatial distribution of the particulate rare earth nuclides is governed by physical factors such as gravity, turbulence, and adsorption to other matter. Rare earths adsorbed to large particulate matter will tend to sink to the bottom or to the appropriate density layer, and except for bottom-feeding organisms will be removed, essentially, from further distribution. Martin (1958), for example, found relatively high amounts of cerium-144 (189 micromicrocuries per square centimeter) in a bottom core sample taken in September, 1956, in the open sea near the Eniwetok Proving Ground. Also, Bowen and Sugihara (1958) showed that the cerium-144 and promethium-147 content of deeper waters of the Atlantic Ocean in some cases was higher than that of the shallow waters. They interpreted these data as indicating that the silt particles in the water are "sweeping down rare earths" at a faster rate than the microplankton are retaining them in the surface waters. In contrast to removal by sedimentation, part of the rare earths become adsorbed to small organic and inorganic debris suspended in the water, and, because of the turbulence produced by wind and ocean currents, this particulate matter may remain in the mixed or shallow layers for some time and eventually may become associated with living organisms.

In the land environment also the physico-chemical form of the lanthanides govern their spatial distribution. For example, in the soil solution these nuclides are insoluble and consequently they do not move readily through the soil. Investigations conducted by Nishita and Larson (1957) showed that less than 12 per cent of the cerium-144 and yttrium-91 added to seven different types of soil was removed with distilled water. More than 90 per cent of these radionuclides remained fixed to four of the soils after leaching with neutral ammonium acetate. If the pH of the leaching solution was raised above 5.5 the extractability of cerium-144 and yttrium-91 decreased sharply. However, when bentonite, a clay mineral, was pretreated with acid, adsorbed cerium-144 was released more readily. The low availability of these nuclides under normal soil conditions is also demonstrated by the small amount in the tissues of land plants. Ce^{144}-Pr^{144}, yttrium-91 and promethium-147 are taken up in about the same order of magnitude and usually in much smaller amounts than radiostrontium or radiocesium and usually in lesser amounts than Ru^{106}-Rh^{106} (Nishita and Larson, 1957; Jacobson and Overstreet, 1948; Rediske et al., 1955). Also, Selders et al. (1953) found that although the rare earths comprised most of the radioactivity in Nevada Test Site soil, they were not preferentially absorbed by plants growing in this soil.

The presence of rare earth radionuclides in leaf samples collected at Eniwetok Atoll during and immediately following the nuclear test series in 1956 (Operation REDWING) was reported by Thomas et al. (1958). The high levels of rare earths found in these samples can be explained only on the basis of surface adsorption, a mechanism believed to be of major importance in the accumulation of these nuclides by land plants.

The presence of stable isotopes of elements in the environment is also known to affect the uptake of various nuclides by organisms. Rediske et al. (1955) found that the addition of stable yttrium to the substrate caused a significant increase in radioyttrium in the plant tissues. It is probable that addition of the stable isotope of yttrium displaced the radioisotope from the soil and increased its availability to the plant.

BIOLOGICAL FACTORS

In discussing the factors involved in the ability of an organism to absorb rare earth nuclides, it is essential to consider the type of organism as well as the method by which the organism obtains nutrients. The particulate nature of the lanthanides in the aquatic environment makes difficult their absorption by autotrophic organisms. Rice (1956), Rice and Willis (1959), and Chipman (1958) have shown that many species of planktonic algae adsorb particulate lanthanum-140, cerium-144, and yttrium-91 rapidly. However, Rice and Willis (1959) showed that a diatom (Nitzschia closterium) absorbed ionic cerium-144, although at a slower rate than particulate cerium-144, and Spooner (1949) suggested that yttrium-90 uptake by sessile algae might be an ion-exchange process.

Although the filter-feeding zooplankton remove radioactive particles from water by surface adsorption, the primary source of these rare earth nuclides is ingested organic and inorganic particulate matter. Chipman (1958) showed that marine copepods and other filter-feeding zooplankton exposed to sea water containing phytoplankton cells and other cerium-144-bearing particles ingested this material rapidly. Similar studies by Chipman (1958) and Boroughs et al. (1957) with filter-feeding invertebrates, such as oysters, scallops, and clams, showed that particulate radionuclides, including cerium-144, were concentrated markedly but were not accumulated in the body tissues. Apparently the insolubility of these particles at the physiological pH of the digestive tract makes their absorption difficult.

The principal source of rare earth nuclides for fish and terrestrial animals, also, is the food they ingest. The physical state of the nuclide in the food governs its fate in the organism. In fish there appears to be some absorption of cerium-144 after ingestion, indicating that at least a portion of this nuclide is in the soluble form. Chipman (1958), for example, found that cerium-144 was accumulated in small amounts by the bone and liver of crabs. In addition, Rudakov (1958) reported that high amounts of Ce^{144}-Pr^{144} were absorbed by carp fingerlings after a one-hour immersion in a radioactive solution, and that with the passage of time there was a redistribution of the nuclides in the tissues. At one day they were found mainly in the liver. At one month the radioactivity in the bone was twice as high as that in the liver. Other results, including those obtained from samples collected at the Eniwetok Proving Ground show that Ce^{144}-Pr^{144} is incorporated in small amounts in muscle, liver, kidney, lung, gills, and skeletal tissues of fish (Thomas et al., 1958; Broughs et al., 1956; Seymour et al., 1957; University of Washington, 1955; Rinehart et al., 1955).

In terrestrial animals there appears to be very little absorption of the rare earth nuclides from the digestive tract. The results of Durbin et al. (1956) showed that cerium-144, europium-152 and europium-154, terbium-160, and thulium-170 administered orally to rats were not absorbed significantly. However, parenteral injection of several radiolanthanides, complexed with an organic acid to increase their solubility, showed that absorption from the site of injection was almost complete within four days. The primary sites of deposition initially were the liver and skeleton. After ten months, however, the site of greatest concentration was the bone; other tissues contained only very small amounts of the rare earth nuclides (see also Kyker and Anderson, 1956). Similarly, in samples of rats collected at Eniwetok Atoll shortly after a nuclear detonation, the rare earth activity was highest in the skeleton and liver (Thomas et al., 1958). Samples of rat tissues collected approximately two and one-half years after the last test series in 1958 (Operation HARDTACK I) contained no detectable rare earth radioactivity although there was some in the soil and plants (Lowman and Palumbo, unpublished data). These data indicate that very little of the rare earth nuclides ingested under natural conditions is retained in the tissues of terrestrial animals.

The inhalation of particulate matter is also a factor to be considered. This process could be important, especially during periods of heavy fallout from nuclear tests and accidental discharge of radioactive materials from reactor stacks. Experiments have shown that rats subjected to aerosols containing lanthanum-140 (Cohn et al., 1957) and cerium-144 (Hennacy, 1961) had a relatively high initial concentration of these nuclides in their gastrointestinal tracts and low concentrations in other tissues. Cerium-144 was eliminated rapidly from the gastrointestinal tract but increased in the liver. The relatively rapid uptake by the liver suggested that some of the cerium was in the soluble form.

As in other animals, the rare earth elements enter man during the ingestion of food and the inhalation of air and dust. Based on the data of J. G. Hamilton and co-workers, Morgan (1960) estimated that less than 0.01 per cent of the rare earth elements pass from the gastrointestinal tract to the blood. It was found also that only a small fraction of the longer-lived rare earth nuclides taken in by inhalation reaches the critical organs for these nuclides.

These and similar data have been considered in establishing for man the maximum permissible concentrations in air, water, and food. The values for some of the rare earth nuclides are presented in Table 4.

The last biological factor to be considered here is the selectivity and concentration of the rare earths by different species. Organisms in the lower trophic levels concentrate them to a greater degree than do organisms in the higher trophic levels. For example, Krumholz and Foster (1957) estimated that the concentration factor was 1,000 for phytoplankton, 500 for filamentous algae, and 100 for fish. Also, it has been shown by Spooner (1949), Rice (1956), and Rice and Willis (1959)

Table 4. Maximum permissible concentrations of some rare earth radionuclides in air and in water for occupational exposure (From: K.Z. Morgan, 1960).

Nuclide	Half-life (days) Physical	Bio-logical	Effec-tive	Critical organ[1]	Maximum burden in total body (microcuries)	Maximum permissible concentration for 40-hour week (microcuries per cubic centimeter) Water[2]	Air	Fraction reaching organ By ingestion	By inhalation
Lanthanum-140	1.68			GI -(s)		7×10^{-4}	2×10^{-7}		
				GI-(i)		7×10^{-4}	10^{-7}		
Cerium-144	290	1500	243	Bone-(s)	5	0.2	10^{-8}	3×10^{-5}	0.075
				GI-(s)		3×10^{-4}	8×10^{-8}		
	290	293	146	Liver-(s)	6	0.3	10^{-8}		
				Lung-(i)			6×10^{-9}		
Promethium-147				GI-(i)		3×10^{-4}	6×10^{-8}		
				GI-(s)		6×10^{-3}	10^{-6}		
	920	1500	570	Bone-(s)	60	1.0	6×10^{-8}	3×10^{-5}	0.09
				Lung-(i)			10^{-7}		
Samarium-151				GI-(i)		6×10^{-3}	10^{-6}		
				GI-(s)	100	0.01	2×10^{-6}		
	3.7×10^{4}	1500	1442	Bone-(s)		2.0	6×10^{-8}	3.5×10^{-5}	0.09
				Lung-(i)			10^{-7}		
Europium-152				GI-(i)		0.01	2×10^{-6}		
				GI-(s)		2×10^{-3}	5×10^{-7}		
	4.7×10^{3}	1480	1125	Kidney-(s)	20	0.3	10^{-8}	3×10^{-6}	7.5×10^{-3}
				Lung-(i)			2×10^{-8}		
				GI-(i)		2×10^{-3}	4×10^{-7}		

[1] GI - gastrointestinal tract, lower large intestine; s = Soluble; i = insoluble.

[2] Air ... includes the water in food.

t different species of algae concentrate trium-90 and cerium-144 to markedly different degrees. In fact, Spooner (1949) found that one variety of Fucus vesiculosus, a sessile brown alga, accumulated nearly all of the yttrium-90 in the medium, whereas another variety accumulated very little. Also, Bowen and Rubinson (1951) found that a yeast selected lanthanum-140 from the medium, they suggested that lanthanum uptake might be a common property of yeast growth. Later investigations, however, showed that this apparent selectivity was in reality a surface adsorption phenomenon (Bowen, personal communication). Since a biological requirement for the rare earth elements has not been demonstrated positively, the selectivity described in the examples above is probably due to differences in the external surfaces of the organisms. Chipman (1958) and Davis et al. (1958) suggested that the extent of accumulation of the rare earth nuclides by aquatic organisms is at least partly related to the surface area of the organism.

SUMMARY

Many radioactive lanthanides are made available in the production of nuclear energy; however, studies of their distribution in the environment and their uptake by living organisms have been confined primarily to Ce^{144}-Pr^{144}, lanthanum-140, promethium-147, and yttrium-90, 91.

Ecological factors governing the distribution of the rare earths are physico-chemical and biological. The first are concerned with the properties of the nuclide and the nature of the environment, and the second with the selectivity and retention of the nuclide by the organism.

Because the rare earth nuclides usually exist in the particulate state, they are not readily available to and are poorly absorbed by terrestrial or aquatic organisms. They are accumulated principally by surface adsorption, and the degree of accumulation can be related to surface area and to species differences.

Laboratory studies indicate that animals, including man, are unable to absorb the rare earths to any extent from the gastrointestinal tract and that elimination of them is rapid. The principal sites of deposition are the bone and the liver.

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IRON AND COBALT IN ECOLOGY

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The interrelationships between organisms and their environments are usually assumed to be reciprocal the environment controls the conditions under which the organisms exist and the organisms, in turn, may influence the state of the environment.

The effects of the organisms upon the environment are usually more subtle than the reciprocal relationship, primarily as a result of the relatively small total mass of the organisms. However, one of the salient characteristics of the biosphere is its chemical reactivity and, although the generation of organisms may produce an almost undetectable alteration, significant changes in the environment may be brought about by the organisms over a period of many generations.

On the other hand, many effects of the environment upon the organisms are easily observed because they are usually effective during the lifetime of any one organism and may be detected during a reasonable period of observation.

The dominant environmental factors often produce immediate effects upon the organism that are sometimes considered to be of the greatest importance. However, within the environment, some physical factors may cause minor changes which are cumulative over a long period of time, resulting in greatly altered environmental conditions from those which would have existed had these processes not been in operation.

The environment is the result of the interplay between matter and energy. In the material phase of the environment, low availability of a given chemical element may exert a limiting effect upon local organisms. Thus, a low level of phosphate in marine waters may exert a limiting effect upon phytoplankton production, and deficiencies of cobalt or copper in pasture lands may produce anemia (wasting disease) in grazing stock (Underwood, 1956).

In addition to the immediate effects, the characteristics and availability of a chemical element throughout the history of the biosphere may have resulted in detectable patterns of utilization or non-utilization within organisms now living. These patterns may not be easily discerned unless a large variety of organisms from different environments are compared.

A given chemical element may also exert secondary effects. A few elements have produced significant alterations in the availability of other chemical elements to the organisms during the history of the biosphere. If some of these alterations had not been effected, several ecological relationships would now be different from those that are observed.

Iron and, to a lesser degree, cobalt are elements which have exerted primary and secondary effects upon the environments, and thus upon the organisms. Some of these mechanisms have extended back to times earlier than the origin of the biosphere.

The roles of iron and cobalt in ecosystems are primarily dependent upon their availabilities to the organisms. The availability of these elements is determined by their distribution and chemical forms in the lithosphere and hydrosphere. the present elemental composition of these environmental spheres has been determined by: (a) the amounts of individual elements produced by the reactions which formed the original material of the earth, (b) fractionation which occurred early in the formation of the earth, and (c) fractionation which has occurred and is occurring as the result of geochemical processes.

In Table 1 are shown the abundances of several elements in the solar atmosphere, the total

Table 1. Abundances of elements in the solar atmosphere, total earth, crust of the earth, and in sea water. The amounts of the elements supplied to the sea and the percentages of the amounts supplied now in solution. Plus (+) signifies trace amounts.

Element	Solar Atmosphere[1] (grams per 100 grams)	Total earth (grams per 100 grams)	Crust of earth[1] (grams per 100 grams)	Sea water[2] (grams per 100 grams)	Total supplied to sea[3] (grams per 100 per grams)	Percentage of Total Element Supplied to Sea Water now in Solution[3]
Hydrogen	53.0	+	0.14	10.8		
Helium	42.0	+	0.00000003	0.0000000005		
Boron	+	+	0.0003	0.0005	0.0002	250.0
Carbon	.012	+	0.03	0.003	0.019	16.0
Nitrogen	.031	+	0.005	0.00005	0.0028	1.8
Oxygen	4.7	28.0	46.6	87.5		
Sodium	.0024	0.14	2.8	1.05	1.70	62.0
Magnesium	.043	17.0	2.1	0.13	1.25	10.0
Aluminum	.0031	0.[illegible]	8.1	0.000001	4.88	0.0002
Silicon	.029	13.6	27.7	0.0003	16.63	0.002
Phosphorus	+	0.03	0.12	0.000007	0.071	0.01
Sulfur	0.014	2.7	0.05	0.09	0.03	300.0
Chlorine	+	+	0.02	1.90	0.019	10,000.0
Potassium	0.00033	0.07	2.6	0.04	1.55	2.6
Calcium	.0036	0.61	3.6	0.04	2.18	1.8
Vanadium	.000031	+	0.02	0.0000002	0.009	0.002
Manganese	.00086	0.09	0.10	0.0000002	0.06	0.0003
Iron	.167	35.0	5.0	0.000001	3.0	0.00003
Cobalt	.00034	0.20	0.002	0.00000005	0.0014	0.004
Nickel	.0029	2.7	0.01	0.0000002	0.005	0.004
Copper	.000058	+	0.01	0.0000003	0.004	0.007
Zinc	.00021	+	0.01	0.000001	0.008	0.01
Molybdenum	+	+	0.0015	0.000001	0.0009	0.11
Iodine	+	+	0.00003	0.000006	0.000018	33.0

[1] Adapted from Mason, 1958

[2] Isaacs, J.D. personal communication, after Goldberg, 1961

[3] Adapted from Goldschmidt, in Rankama and Sahama, 1950.

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earth, the crust of the earth, in sea water, the total amounts of elements supplied to sea water and the percentage of those supplied which are now in solution.

The composition of the material from which the earth was formed may be estimated from the abundance of the elements in the solar atmosphere. In this material hydrogen, helium, and oxygen account for 99.7 per cent of the total mass. Of the remaining elements only iron is present in relatively high (0.167 per cent) amount and contributes 97.4 per cent of the total mass of the transition elements vanadium, manganese, iron, cobalt, nickel, and copper. Had iron not been produced in relatively large quantities, due to its high nuclear binding energy, the subsequent role of iron in the formation of the earth and its availability for use by organisms probably would have been significantly reduced.

Fractionation of elements occurred in the early molten stages of the earth and, the predominant elements at this time were iron, oxygen, magnesium, and silicon (Table 1). The amount of oxygen was not sufficient to convert the cationic elements into oxides so that a major part of the iron and nickel in elemental form sank to the center forming the core. Most of the cobalt, molybdenum, and other heavy metals were carried along and a major part of the iron and other heavy metals was thereby effectively removed from the environment which was later to be inhabited by the biosphere (Mason, 1958).

A mantle and crust solidified over the core and the latter was enriched in sodium, aluminum, potassium, chlorine, calcium, phosphorus, silicon, and oxygen. Iron contributed only about one-seventh and cobalt about one-one hundredth as much of the crust as they did of the total earth, and in addition, nickel, sulfur, and magnesium were reduced in amount. Although the percentage of iron was reduced in the crust of the earth in relation to that in the core, it was present in appreciable amounts with only oxygen, silica, and aluminum being more abundant.

With the formation of the hydrosphere many of the elements of the crust were redistributed. The mobility and chemical forms of iron and cobalt were and are influenced by the environmental conditions on land and in the water and were affected significantly by physical and chemical action of the atmosphere. The paths of iron and cobalt were probably from weathered rock, to fresh water, to the seas where a major part of the iron and a lesser part of the cobalt was precipitated and deposited at the bottom. In many instances the bottoms of the seas were raised to altitudes higher than the surface of sea and the cycle was repeated.

Thus, leaching of the earth's crust was an important factor in the distribution and deposition of iron and cobalt in both the lithosphere and hydrosphere.

In natural water, iron is present as Fe^{3+}, Fe^{2+}, and $Fe(OH)^{2+}$. Iron, in the ferrous form, remains in solution with precipitation occuring after its oxidation to the ferric state. The transport of ferric hydroxide is dependent upon its solubility which, in turn, is dependent upon the pH of the medium. At pH six, ferric hydroxide is about 100 times as soluble as at pH seven and is 100,000 times as soluble as at pH 8.5 (Cooper, 1937). Iron in weakly acid streams flowing into the seas therefore is precipitated in the slightly alkaline sea water. This fact is illustrated by the observations that the average iron content of fresh waters is about 100 times the level found in marine waters (Mason, 1958).

The precipitation of iron in sea water from acidic iron wastes was described by Ketchum and Ford (1952). After the introduction of the wastes, the sea water first became green from the formation of ferrous hydroxide. Later the oxidation of the iron to ferric hydroxide resulted in a flocculent red precipitate.

In addition to direct observations, an estimate of the behavior of chemical elements in the oceans may be made from the geochemical balance of the elements in sea water. If the comparison is made of the total amount of the elements supplied to the seas from the lithosphere with the amounts now found in sea water, the transfer percentage for each element may be determined. The larger the transfer percentage for a given element, the more of that element has remained in solution. In Table 1 are given the amounts of several elements in sea water, the amounts supplied to the sea, and the percentage of each element which is in solution. Iron has the lowest transfer percentage of any of the elements with only one part in three million of that originally supplied remaining in solution. Manganese exhibits a transfer percentage ten times and the other transition elements cobalt, nickel and copper 100 to 200 times that of iron. In absolute amounts, however, iron is present in sea water at a level about three times that of copper, five times that of vanadium, manganese and nickel, and twenty times that of cobalt. However, all of the transition elements, including iron, are present in very low amounts.

The role of iron in the respiratory pigments of animals may be related to the distribution of this metal in the different environments and to the levels of available oxygen. The two principal metals contained in respiratory pigments are iron and copper in that order of occurrence. The copper containing pigment (hemocyanin), probably originated mostly among plankton and free swimming forms of ancient invertebrates which inhabited marine waters containing low amounts of iron and relatively high oxygen content. The iron containing pigment (hemoglobin) probably originated and developed at about the same time in organisms that lived at the ocean bottom, especially in those organisms which burrowed in the bottom sediments, where iron was and is available in relatively high amounts and oxygen was at a reduced level (Vinogradov, 1953). In most of those animals which made the transition from salt to brackish or fresh water or onto the land mass, the respiratory pigments contain iron. The high levels of iron available to terrestrial, fresh water, and bottom dwelling marine organisms, in contrast to the reduced amounts available to free living marine organisms, probably influenced the incidence of utilization of iron by the inhabitants of the different environments.

Iron, when precipitated in sea water, forms a positively charged colloid and the precipitate carries scavenged ions with it as it settles to the bottom. Many ions, including arsenic, molybdenum, nickel, vanadium, phosphorus, antimony, copper, selenium, and lead, are removed from solution by this mechanism. In addition, the manganous ion may be catalytically oxidized by a gel of ferric hydroxide hydrate and this reaction probably accounts in part, at least, for the low transfer percentage of manganese in comparison to vanadium, cobalt, nickel, and copper. The co-precipitate of manganese and iron is capable of removing additional ions from the sea water, including cobalt, zinc, thorium, tin, and silver (Arrhenius, 1959).

The removal of heavy ions by ferric hydroxide is of great importance in the survival of past

and present organisms in the sea. Sufficient amounts of arsenic, copper, selenium, molybdenum, and lead have been supplied to marine water during geological history to have resulted in a poisoned marine environment had they not been scavenged from the water (Mason, 1958).

The chemical characteristics of iron and cobalt which influence their distribution in the lithosphere and hydrosphere are also important in their utilization by organisms. Iron and cobalt are transition elements and exhibit the characteristics of variable valence, easy oxidation and reduction, and a strong tendency to form complexes with organic material.

Thus, they are potentially useful in physiological processes in both animals and plants, in which these chemical characteristics are required.

Although the amount of iron required by plants is low, it is nevertheless essential for growth. It is a constituent of several enzymes and carriers which operate in the cell respiratory mechanisms of which catalase, peroxidase, cytochromes, and cytochrome oxidase are examples (Miller, 1957). It is physiologically active in the ferrous state and when taken in as the ferric ion, is rapidly reduced in the cells. It is one of the most immobile of all elements in plants (Biddulph, 1951; Meyer et al., 1960), and often cannot be absorbed from the soil although it may be plentiful. With low pH and low internal phosphate, iron is readily absorbed by roots or leaves and rapidly becomes distributed in the plant. However, at neutrality and high levels of phosphate, iron, applied to leaves, is precipitated in the veins of the leaves (Rediske and Biddulph, 1953). Iron is toxic to plants only in the case of low soil pH, lack of aeration, or combinations of these conditions (Meyer et al, 1960).

Cobalt has not been proven to be essential for higher plants although it is required by lower plants such as algae and fungi (Miller,1957). Cobalt does aid in the accumulation of chlorophyll in the leaves of some higher plants. It decreases the decomposition of chlorophyll in the dark (Solovera and Makorova, 1961), and activates some plant enzymes including carboxylases and peptidases (Meyer et al., 1960).

In animals, iron can be ingested only in the ferrous state. It is an active component of hemoglobin and myoglobin and is associated with the activity of cytochromes, cytochrome oxidases, and catalase. Thus, it is important in animals in oxidative processes, transport of oxygen, storage of oxygen in muscle, and in intermediate cell metabolism. It is usually strongly bound in the animal body and, therefore, has a low turnover rate (Underwood, 1956).

Cobalt in animals is active in vitamin B_{12}. Neither higher plants nor animals can synthesize the vitamin which is formed primarily by bacteria and actinomycetes (Sherman, 1957). Cobalt is poorly retained by most animal tissues and is rapidly eliminated by many animal species (Underwood, 1956).

Thus the functions of iron and cobalt in plants and animals are at least partly known and the geochemical characteristics of these elements have been determined. However, these types of observations do not necessarily provide the information required to determine the mechanisms which control the uptake of these elements by organisms from their environments. Even if the amounts of iron and cobalt were measured in representative samples of the organisms and their environments, only the static condition at the time of sampling would be determined.

The biological factors which control the levels of iron and cobalt in various trophic levels are incompletely known and experimental data are needed on the movements of these elements through given ecosystems. Some of these factors include: (a) population sizes, (b) population biomasses, (c) ratios of population surface areas to biomasses, (d) chemical characteristics of surface areas of different species, (e) average movements of populations, (f) movements of individuals, (g) feeding habits, (h) physiological selectivity, (i) turnover rates, (j) reproductive rates, (k) growth rates, and (l) average life spans.

The characteristics of the biota in a given environment are usually complex and subject to continued changes in balance. The interactions of the great number of variables upon each other result in an over-all problem of such complexity that the solution probably cannot be achieved by the determination of the variables and applying them to mathematical models.

A simplified approach to the complex problem of iron and cobalt metabolism in a given ecosystem is that of producing a perturbation in the system and studying the resulting fluctuation through the system. The introduction of one or more radioisotopes of the element into the system at one or more trophic levels will simultaneously produce the perturbation and the marker by which the fluctuation may be followed. An approximation of mass transfer within the system may also be achieved if the amount of isotope dilution in samples from all trophic levels is determined with increased time after the introduction of the radionuclide.

The interrelationships of the environment and the various trophic levels may be more accurately determined, however, if tracer experiments in an ecosystem are correlated with selected investigations in the laboratory to determine accurately some of the above mentioned characteristics of the organisms.

The use of radionuclides as tracers depends on the premises that: (a) the chemical properties of all isotopes of a given element are identical and that the introduced radioelement is in the same chemical and physical form as the naturally occurring element, (b) the added radionuclide does not result in sufficient radiation damage to alter the viability of the organisms, and, (c) the added radionuclide does not significantly increase the total amount of the element under consideration.

Most of the observations on the cycling of radionuclides have been made in areas of fallout from nuclear weapons tests, at sites contaminated by reactor incidents, in areas receiving cooling water from large reactors, or downstream and downwind from installations in which radioactive materials are processed.

In the cooling waters from reactors several neutron-induced radionuclides have been identified, including iron-59, Cobalt-58, and cobalt-60 (Heath, 1956; Moeller, 1957; Conley, 1954; Foster and Rostenbach, 1954; Rebeck et al., 1954). In fallout from nuclear tests the nuclides iron-55, iron-59, cobalt-57, cobalt 58 and cobalt-60, have been reported (Kawabata, 1954; South and Lowman, 1955; Rinehart et al., 1955; Yamada et al., 1955; Saiki et al., 1955; Mori and Saiki, 1956; Yoshii, 1956; Nagasawa et al., 1956; Seymour et al., 1957; Lowman, et al., 1957; Lowman, 1958; Palumbo and Lowman, 1958; Welander, 1958; Lowman, 1960).

Only a limited number of observations have

been made concerning the levels of radioactive iron and cobalt in areas contaminated by reactor effluents, primarily because relatively small amounts of these radionuclides are produced.

Krumholz and Foster (1957) reported the concentration factors for iron-59 in several organisms collected in the Columbia River. The concentration factors were as follows:

phytoplankton	200,000,
filamentous algae	100,000,
insect larvae	100,000, and
fish	10,000.

The primary producers, the phytoplankton and filamentous algae, concentrated radioiron to a high degree. The concentration factor for zooplankton was not reported but probably was about the same as that of the primary producers since the concentration factor for insect larvae was of the same order of magnitude. In the fish, however, which accumulate radionuclides principally by ingestion, the concentration factor was about one-tenth that of the other organisms.

Several observations have been made in the tropical oceanic and terrestrial areas of the Central Pacific Ocean regarding the amounts of radioactive iron and cobalt within organisms and their environments and have been reviewed by Lowman (1960). In these studies the levels of radioactivity in solution, in colloidal form, and associated with small particles in sea water have been compared with the radioactivity in plankton, sessile invertebrates, filter feeding and carnivorous fishes, and birds whose source of food is marine organisms. On land the amount of radioactivity have been measured in soils, plants, and herbivorous animals. In addition, analyses of soils under the nesting areas of birds have been made in which the radionuclide content has been altered by the excrement of the birds which feed upon marine organisms.

Iron and cobalt are microconstituents of living material and, although present in small amounts, are important in the normal metabolism of most organisms (Table 2). All of the microconstituents except boron and silicon have high specific gravities and atomic numbers in comparison with the primary and secondary constituents.

In sea water, iron and cobalt are also microconstituents, contributing only 0.000001 and 0.00000005 percent respectively of the total mass. The radioactive forms of these elements introduced in fallout are therefore subject to limited isotope dilution by the corresponding stable element in the water. A comparison of the

Table 2. The invariable primary constituents of living matter according to percentage of body weight (Webb and Fearon, 1937; Mason, 1958).

Primary	Secondary	Microconstituents
(1 to 60 per cent)	(0.05 to 1 per cent)	(less than 0.05 per cent)
Hydrogen	Sodium	Boran
Carbon	Magnesium	Iron
Nitrogen	Sulfur	Silicon
Oxygen	Chlorine	Manganese
Phosphorus	Calcium	Copper
		Iodine
		Cobalt
		Molybdenum
		Zinc

amount of isotope dilution in sea water for radioisotopes of biologically important elements is as follows (cobalt isotope dilution evaluated to one):

Primary		Secondary		Micro-constituents	
Hydrogen	2.2×10^8	Sodium	2.1×10^7	Manganese	4.0
Carbon	5.6×10^4	Sulfur	1.8×10^6	Iron	20.0
Phosphorus	1.4×10^2	Chlorine	3.8×10^7	Cobalt	1.0
		Calcium	8.0×10^5	Copper	6.0
				Zinc	20.0
				Molybdenum	20.0

Radioisotopes of all of the microconstituents of biological material are subject to less isotope dilution in sea water than are the primary and secondary constituents. Thus, for equal amounts of radionuclides added to sea water an organism would have to concentrate 2.2×10^8 times as much stable hydrogen and 8.0×10^5 times as much stable calcium as it would stable cobalt to contain an equal amount of each of the three radionuclides.

Marine organisms do exhibit the ability to concentrate many elements from sea water (Donaldson et al., 1956; Boroughs et al., 1957; Krumholz et al., 1957; Lowman et al., 1957, 1959; Seymour et al., 1957; Lowman 1958; Palumbo et al., 1959), and it is usually assumed that the organisms rapidly acquire a major fraction of the radioisotopes of these elements introduced in fallout. At the Eniwetok Proving Ground this does not occur, probably as a result of the plankton contributing only one part per million of the total volume of sea water plus organisms. Although plankton exhibit high concentration factors for some elements the relatively small amount of these organisms in the tropical waters resulted in their containing only 1/10,000 of the total radioactivity at one week and 1/30,000 at six weeks after the water in

Table 3. Percent of total radioactivity in sea water and plankton contributed by fission products and neutron-induced radionuclides at approximate times of 48 hours, one week and six weeks (Knapp, 1960; Lowman, 1960). The percents of total are underlined.

	Less than 48 hours		Greater than One week		Six weeks	
	Water	Plankton	Water	Plankton	Water	Plankton
Molybdenum - 99, Technetium - 99m	5.9	12.0	5.0	0	0.02	0
Cerium - 141,144, Praesodymium - 141,144	0.83	3.0	2.1	0	20.0	5.0
Ruthenium - 103,105,106, Rhodium - 103,105,106	2.1	3.0	2.2	5.0	16.0	1.4
Barium - 140, Lanthanum - 140	2.6	2.0	6.8	23.0	18.0	0
Tellurium - 132, Iodine - 132	6.2	8.0	5.6	0	0.05	0
Zirconium - 95, Niobium - 95	0.38	1.0	1.0	6.0	18.0	20.0
Strontium - 89,90	0.30	0	0.76	0	8.2	0
Cesium - 137	<0.01	0	0.05	0	0.08	0
Neptunium - 239	54.0	69.0	33.0	2.0	0.02	0
Uranium - 237	24.0	3.0	43.0	2.0	18.0	0
Cobalt - 57,58,60	0.02	0	0.05	43.0	0.67	24.0
Zinc - 65	0.02	0	0.05	3.0	0.84	25.0
Iron - 55,59	0.02	0	0.05	16.0	0.93	24.0
Manganese - 54	trace	0	0.01	0	0.12	0.6

which they were taken had been contaminated.

The plankton contained only a small fraction of the total radioactivity, however, the amounts of radioactivity per unit volume, including radioactive iron and cobalt, were much greater in plankton than water. In Table 3 the average percentage of total radioactivity contributed by the principal radionuclides in sea water and plankton at times less than 48 hours, one week, and six weeks after contamination are shown. At 48 hours the percentage compositions of water and plankton were similar, indicating little or no selectivity. However, at one week cobalt-57,58,60, and iron-55,59 contributed 43 percent and 16 percent, respectively, of the total radioactivity in plankton and 0.05 percent each of the total radioactivity in the water. Therefore, at one week the radioisotopes of iron and cobalt accounted for 59 percent of the radioactivity in the plankton but only 0.1 percent in the water.

At six weeks, radioactive iron and cobalt were present in plankton at a level which accounted for 48 percent of the total radioactivity, each element contributing 24 percent of the total radioactivity. In the water, iron and cobalt accounted for 0.93 and 0.67 percent respectively of the total.

The concentration factors exhibited by the plankton at six weeks after contamination were approximately 90,000 for iron and 100,000 for cobalt. The iron value is approximately one half of that reported by Krumholz and Foster (1957), for freshwater phytoplankton which have a much larger surface to volume ratio than do the zooplankton.

The plankton considered above comprise primarily two trophic levels in the food chain, the phytoplankton or primary producers and the zooplankton. The zooplankton are eaten by omnivorous fishes (flying fishes) which are in turn eaten by the carnivorous fishes (tunas). The radionuclide composition of the two types of fishes have been discussed in another paper of this series (Lowman, 1962), and will not be considered in detail, however, the distribution pattern of cobalt-57,58,60, and iron-55,59, in the three trophic levels are as follows:

	Time after contamination (weeks)	Per cent of total Radioactivity Cobalt-57,58,60	Iron-55,59	Total radioactivity (disintregrations per minute per gram of dry weight)
Plankton	1	43	16	2.3×10^6
Omnivorous fish				
Muscle	1	10	31	2.2×10^4
Liver	1	9	81	1.1×10^6
Carnivorous fish				
Muscle	6	0.9 to 2.5	6 to 8	3.3×10^3 to 1.2×10^4
Liver	6	0.0 to 2.3	15 to 25	3.6×10^4 to 2.0×10^5

On a dry weight basis the total radioactivity in the plankton was approximately two and 100 times that in the liver and muscle respectively of the omnivorous fish. The levels of radioactivity in the plankton and omnivorous fish cannot be compared directly with those in the carnivorous fish (tuna) because the latter fishes are capable of migrating great distance in short periods of time and the length of time they had been in the area of contamination before they were taken cannot be determined.

If the per cent of total radioactivity contributed by cobalt-57,58,60, and iron-55,59 are compared in the plankton, the omnivorous fish, and the carnivorous fish, however, a pattern of accumulation may be seen. Whereas in the plankton radiocobalt contributed 43 per cent of the total radioactivity, in tissues of the omnivores it contributed only nine to ten per cent and in the carnivores zero to 2.5 per cent. Radioiron accounted for 16 per cent of the total radioactivity in the plankton, 31 to 81 per cent in the omnivores, and six to 25 per cent in the carnivores.

The reduction in the percentage of total radioactivity contributed by radioiron in the carnivorous fish was caused by a high concentration factor for zinc-65 in the tissues of these fishes.

Thus, the percentage of total radioactivity contributed by radiocobalt decreased through the two higher trophic levels. Radioactive iron, however, increased in percentage of total radioactivity in the muscle and liver of the omnivorous fish over that in plankton and was concentrated in the liver at an absolute level double that found in the plankton.

In addition to the food chain represented by the plankton, omnivorous fishes, and carnivorous fishes, another food chain is comprised primarily of the plankton, omnivorous fishes, and sea birds. Among the birds at Eniwetok are the terns or sea swallows (fairytern, Gygis alba; common noddy tern, Anous stolidus) which feed primarily upon small fish and to a lesser degree upon squid. Analyses for iron-55,59 have not been made on the tissues of these animals, and in gamma spectra of the liver, gut, kidney, muscle, and lung tissues of the terns, only the gamma peak of zinc-65 was detected (Lowman, et al., 1957). Iron-55 is probably present in the tissues of the terns in large amounts, however. Indirect evidence has been found that this is the case in analyses of soil samples from areas in which these birds nest (Palumbo and Lowman, 1958). A comparison is shown below of soil samples taken in bird nesting areas and from an island where nests were not present.

	Per cent of Total Radioactivity in Soil Non-nesting area (Lowman, et al., 1957)	Nesting area (Palumbo and Lowman, 1958)
Ce^{144}-Pr^{144}	43.6	41.7
Iron - 55,59	0	40.6
Cobalt - 57,58,60	1 to 2	0.4
Manganese - 54	0	0.8
Zinc - 65	0	0

In the non-nesting area the soil did not contain the radioisotopes of iron, manganese, or zinc, but did contain one to two per cent of radiocobalt. In the nesting area, zinc-65 was not found in the soil, which contained excretory products from birds although it was present in high amount in the tissues of the birds. However, approximately 41 per cent of the total radioactivity in the soil was contributed by iron-55,59 with only trace amounts of radiocobalt and manganese. On the basis of the soils data and the gamma spectrum data on the birds, these animals excreted high amounts of iron-55,59, almost all of the cobalt-57,58,60, and manganese-54, and retained the zinc-65. The fact that the gamma peaks of cobalt-57,58,60, and manganese-54 were not found in the bird tissues and that these radionuclides were present in low amounts in the soil, indicates that the birds ingested relatively low amounts of radioactive cobalt and manganese and large amounts of iron-55,59 and zinc-65 and retained the radiozinc to a

greater degree than the radioiron, cobalt, or manganese.

SUMMARY

The roles of iron and cobalt in ecology are dependent upon their physical and chemical characteristics in the environment and upon the biological demand for these elements. Iron and, to a lesser degree, cobalt are important elements in the physiology of plants and animals.

The introduction of radioisotopes of iron and cobalt may be used to determine the roles of these elements in the metabolic patterns and mass transfers within a given ecosystem. Most investigations of this type have been done in marine environments near nuclear weapons test sites.

Plankton, collected from an area of sea water which had been contaminated with radioactivity for a period of six weeks, exhibited average concentration factors for radioactive iron and cobalt of 90,000 and 100,000, respectively, and contained the stable counterparts of these elements in about the same ratio as they were present in the water.

The roles of iron and cobalt in two food chains have been investigated in the marine environment in the Central Pacific. These include:

Water ⟶ plankton ⟶ omnivorous fish ⟶ carnivorous fish

and

Water ⟶ plankton ⟶ omnivorous fish ⟶ birds

In the first food chain, discrimination against cobalt-57,58,60,was progressive throughout the trophic levels. Iron-55,59 was actively concentrated in the omnivorous fish, and it accounted for a reduced percentage of the total radioactivity in the carnivorous fish due to the presence of large amounts of zinc-65.

In the second food chain the birds, which feed primarily upon omnivorous fish and secondarily upon squid, did not retain significant amounts of the radioisotopes of cobalt or iron but retained a major part of the ingested zinc-65.

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SOME ASPECTS OF THE BIOLOGY OF ZIRCONIUM-95

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Zirconium-95 with a sixty-five day half life has become of biological interest primarily because of its relatively high yield in nuclear fission, 15 per cent of the total radioactivity 90 days after fission and 7.3 per cent one year after fission (Hunter and Ballou, 1951).[1] Stable zirconium has no known biological function although it has been found in trace amounts in plants (Rankama and Sahama, 1950). Vinogradov (1959), states that there have been very few quantitative determinations of zirconium in soils; on the basis of about 500 determinations the average content is at most 0.04 per cent. Two thirds of the zirconium is found as zircon (zirconium oxide), a stable mineral (Rankama and Sahama, 1950). It does not seem likely, therefore, that isotopic dilution need be of any great concern when considering the uptake of zirconium-95 by organisms under ordinary conditions.

The only published review of the radiochemistry of zirconium is that by Steinberg, (1960). Confusion exists as to the behavior of zirconium in solutions because of the formation of colloids and extensive hydrolysis and polymerization of zirconium ions. Zirconium tracer is strongly coprecipitated with most precipitates in the absence of complex-forming ions. In evaluating biological uptake, particularly in aqueous media, an important point made by Steinberg should be kept in mind, i.e., zirconium is easily carried on foreign matter and adheres to glassware. Thus, in studies of the uptake of zirconium-95 by organisms it is more than usually important to know the chemical state of the zirconium both at the time of administration and at the conclusion of the experiment. It should also be especially pertinent to establish a "balance sheet," for the total system, including such portions as the sides of containers used. Unfortunately, such information is not always available in the literature.

Gofman (1949) has described the preparation of colloids of zirconium isotopes for use in the selective localization of radioisotopes in tissues. Dobson et al., (1949), using these preparations in the mouse and rabbit by intravenous injection, found that colloids of zirconium of relatively large particle size ("Sedimentible in large part with ordinary centrifuges...") rapidly disappeared from the blood (half time was 30 seconds to one minute) and were deposited mainly in the liver and spleen. Colloids of smaller particle size disappeared from the blood more slowly (half time was 30 to 80 minutes) and were deposited mainly in the bone marrow and spleen and secondarily in the liver. Once deposited, both types of colloids retained their distribution patterns for the duration of the experiments, two to four weeks. The objective of these experiments was to establish methods for radiotherapy. Similar preparations could also be useful in studies of the uptake of colloids by various organisms and in studies of the filtering capacity of appropriate aquatic organisms.

Following intramuscular administration of zirconium-95 (state not given) to eels kept in both fresh and sea water, Tomiyama and Kobayashi (1957) found a different distribution of the nuclide than has just been described for the mouse and rabbit. Twenty-five hours after administration the nuclide was widely distributed among the tissues and organs but found in the largest amount in the blood, kidney, and spleen. It is stated that the excretion was very slow. When zirconium-95 was added to the water about 70 per cent of that in the eel was found in the gills and most of the remainder in external portions such as the surface mucus. The authors interpret the high level in the gills as indicating uptake through the gills.

Various organic acids influence the distribution of zirconium-95 in the rat regardless of the route of entry (Hamilton, 1949). Zirconium citrate was first used to replace plutonium and yttrium in skeleton by Schubert (1949). This and subsequent work of this kind has been reviewed by Rosenthal (1960). These metals are concentrated in the osteoid matrix rather than being deposited in the mineral structure as are the alkaline earths.

Langham (1960) has reviewed the significance of the portals of entry of fission products. Gastrointestinal absorption of zirconium-95 is very low, 0.01 per cent. Pulmonary deposition of the oxide is similar to that of other oxides and nitrates. Material remaining in the lungs remains in the pulmonary lymph nodes. Under most circumstances ingestion is probably the primary portal of entry.

In plants zirconium-95 is found mainly in the roots if supplied in the soil or in aqueous solution (Klechkovskii and Gulyakin, 1958; Nishita et al., 1960). If foliar application is made the zirconium remains near the site of application.

In experiments with the alga Porphyra sp., Foreman and Templeton (1958) have reported concentration factors between 200 and 470 and comparatively rapid loss of the zirconium-95, 50 per cent in six days, 96 per cent in 65 days. Only the abstract was available and the authors' conclusions were not given but it seems most likely that a surface adsorption phenomenon is involved.

Timofeeva-Resovskaya and Timofeeva-Resovskii (1958) have reported a concentration factor for zirconium-95 of 315 by the snail, Limnea stagnalis. Again only the abstract was available and experimental details were not given.

Zirconium-95 in environmental contamination has been found in a wide variety of organisms[2] but always appears to be associated with adsorption, surface contamination, or the ingestion of particulate matter. Although still detectable in soil several years after contamination by local fallout from nuclear tests, zirconium-95 does not appear in the food web in significant amounts after less than a year.

Summing up, the biological half-life of zirconium-95 is much longer than its 65-day physical half-life. Uptake from environmental contamination is mainly by the oral route in animals and

[1] Since the reading of this paper Collins et al. have reported that zirconium-95 in accumulated fallout during 1958 and 1959 in New York City produced gamma doses comparable to doses from cesium-137. (Collins, W.R., Jr., G.A. Welford, and R.S. Morse, 1961. Fallout from 1957 and 1958 nuclear test series. Science 134 (3484): 980-984.)

[2] A large number of reports have appeared and will not be cited individually. Those available at this writing are included in the reference cited and can be identified by their titles.

probably by foliar contamination in plants, and the fraction absorbed is very small. Surface contamination of plants is probably the main source to mammals. In aquatic systems adsorption and the ingestion of particulate matter are of major importance. Due to its availability, half-life, and ability to form complexes and colloids, zirconium-95 should be particularly useful as a tracer in biological studies of aquatic systems.

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