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MARINEBIOLOGICAL INVESTIGATIONS AT THE ENIWETOK TEST SITE

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Abstract

The results of marine biological investigations conducted at the Eniwetok Test Site since 1952 are summarized. Radioisotopes introduced into the sea from the tests at various times since then include fission products and other radioisotopes (U^{237}, Np^{239}, Mn^{54}, $Fe^{55, 59}$, $Co^{57, 58, 60}$, Zn^{65} and W^{185}). The levels of radioisotopes in plankton samples taken 4 days to 6 weeks after contamination are reported and the distribution of the radioactivity between plankton and water is given. Grazing fishes contained Zn^{65}, Fe^{55}, $Co^{57, 58, 60}$ and Mn^{54}. Carnivorous fishes contained mostly Fe^{55} and Zn^{65}.

RECHERCHES DE BIOLOGIE MARINE AU CENTRE
D'ESSAIS D'ENIWETOK

Résumé

L'auteur fait le bilan des recherches de biologie marine effectuées depuis 1952 au Centre d'essais d'Eniwetok. Les radioisotopes qui se sont répandus dans la mer à la suite des essais auxquels on a procédé à plusieurs reprises depuis cette date comprennent des produits de fission et d'autres isotopes (^{237}U, ^{239}Np, ^{54}Mn, $^{55,59}Fe$, $^{57,58,60}Co$, ^{65}Zn et ^{185}W). L'auteur indique les quantités des radioisotopes présents dans des échantillons de plancton prélevés de quatre jours à six semaines après la contamination, et la répartition de la radioactivité entre le plancton et l'eau. Les poissons herbivores contenaient du zinc—65, du fer—55, du cobalt—57, 58, 60 et du manganèse—54. Les poissons carnassiers contenaient surtout du fer—55 et du zinc—65.

БИОЛОГИЧЕСКИЕ ИССЛЕДОВАНИЯ МОРСКОЙ ВОДЫ В
РАЙОНЕ ПРОВЕДЕНИЯ ИСПЫТАНИЙ ЯДЕРНОГО ОРУЖИЯ
НА АТОЛЛЕ ЭНИВЕТОК

Резюме

Суммируются результаты биологических исследований морской воды в районе испытательных взрывов ядерного оружия на атолле Эниветок, проведенных с 1952 года. Радиоизотопы, полученные морской водой в результате испытательных взрывов, проведенных в различное время с 1952 года, представляют собой продукты распада, а также другие радиоизотопы (U^{237}, Np^{239}, Mn^{54}, $Fe^{55, 59}$, $Co^{57, 58, 60}$, Zn^{65} и W^{185}). Сообщается об уровнях радиоактивности образцов планктона, взятых через 4 дня — 6 недель после радиоактивного загрязнения, а также о распределении радиоактивности между планктоном и водой. Питающиеся планктоном рыбы содержат цинк—65, железо—55, кобальт—57, 58, 60 и марганец —57, а плотоядные рыбы в большинстве случаев — железо—55 и цинк—65.

INVESTIGACIONES SOBRE BIOLOGIA MARINA EN LA ZONA DE ENSAYO DE ENIWETOK

Resumen

En la memoria se resumen los resultados de las investigaciones sobre biología marina que se vienen realizando desde 1952 en la zona de ensayo de Eniwetok. A consecuencia de los diversos ensayos nucleares efectuados desde esa fecha se han introducido en el mar una serie de productos de fisión y otros radio-isótopos (^{237}U, ^{239}Np, ^{54}Mn, $^{55, 59}$Fe, $^{57, 58, 60}$Co, ^{65}Zn y ^{185}W). El autor indica la concentración de los radioisótopos en las muestras de plancton tomadas entre cuatro días y seis semanas después de la contaminación, así como la distribución de la radiactividad entre el plancton y el agua. Los peces herbívoros contenían ^{65}Zn, ^{55}Fe, $^{57, 58, 60}$Co y ^{54}Mn. Los peces carnívoros contenían principalmente ^{55}Fe y ^{65}Zn.

MARINE BIOLOGICAL INVESTIGATIONS AT THE ENIWETOK TEST SITE

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The disposal of radioactive waste products in the sea is of primary interest to man insofar as it constitutes a potential hazard in his food materials derived from marine sources. At the present time, the principal source of artificial radioactive elements in the ocean is past weapons tests. In the future, however, the wastes from nuclear power development and the great volume of associated contaminated materials will far surpass the present burden of artificial radioactive elements in the sea.

Almost certainly, the radioactive wastes consigned to the sea in the future will be deposited in chemical forms different in varying degrees from those in fallout material. At first consideration comparison of the fates in the marine biotic mass of radioisotopes derived from the two sources might appear to be of limited use. However, investigations on the cycling of radioisotopes through the marine food webs and the levels of various isotopes in representative samples have been conducted for the past eleven years at the Eniwetok Test Site in areas contaminated by many different types of nuclear devices fired under a wide variety of conditions. The transmission of given radioelements from one organism to another follows similar paths whether the fallout is mixed with calcium compounds from pulverized coralline islands and reefs, or sodium chloride from evaporated sea-water, or is essentially free of either of those non-radioactive materials. Indeed, groups of radioelements with similar chemical characteristics tend to follow similar paths in marine organisms. The uptake of radioisotopes from areas contaminated by fallout is controlled primarily by two factors; the physical factors which determine the distribution of the radioactive material in time and space, and the chemical factors which control

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the accumulation of the radioelements by the organisms in the sea. The chemical factors include the interaction of the radioelements with sea water and its contained salts, and the biochemical factors.

Physical factors

The distribution and availability of radioactive contamination with time in the sea is controlled primarily by the times at which the individual radio-elements are condensed from a gas to a liquid within the fireball of the nuclear device, and by the physical half-lives of the constituent radioisotopes. ADAMS, FARLOW and SCHELL (1), in a study of fallout particles at the Eniwetok Test Site, found that different ratios of radioisotopes were associated with different types of fallout particles. Unmelted calcium oxide particles collected their radioactivity in cooler parts of the fireball and at later times than did spherical particles formed of melted calcium oxide. The latter particles lost the porous structure characteristic of the unmelted particles, so that hydration in the particles of melted origin proceeded at a much slower rate. The particles also differed in their chemical constitution. Since iron and fission-product vapours from the nuclear device and associated structure were concentrated near the centre of the fireball, they tended to become incorporated more in the particles of melted origin than in the unmelted particles.

Because the radioactive material condensed on to the unmelted particles in cooler parts of the fireball and at later times than on to the originally melted particles, a fractionating effect occurred in which the unmelted particles contained more of the volatile radioactive elements and, in the case of short-lived elements, their daughter products. The unmelted particles thus contained more radioactive barium (daughter of xenon) and strontium (daughter of krypton).

The effect of the physical half-life of the radioisotopes upon the distribution and availability of the contamination to the biomass in the sea is self-evident. Radioisotopes of short half-life will be available to the organisms for a limited time, after which the isotopes of longer half-lives will be of major importance.

The distribution of radioactive contamination in space within the sea is largely determined by oceanographic effects and gravity. Distribution is altered to a much lower extent by the movement of organisms in and out of the contaminated area.

The geographical distribution of the major masses of radioactive contamination in the sea is probably determined primarily by ocean currents, although in the long run meteorological effects upon global fallout may be more important in world-wide distribution of marine contamination.

Horizontal dispersion is dependent upon surface winds, currents, vertical and horizontal density gradients, and the size of the contaminated area. The rate of horizontal dispersion of radioactive material in sea-water was reported by REVELLE and SCHAEFER (2) to be about one million times the rate of molecular diffusion. Within the thermocline most of the motion of soluble and colloidal material occurs along surfaces of equal density, and thus dispersion in the lateral direction would be much greater than in the vertical. In observations made at the Pacific Proving Ground, DONALDSON et al. (3), SEYMOUR et al. (4), and PALUMBO and LOWMAN (unpublished) found the radioactivity mostly in the mixed layer and in some instances below the thermocline, but again the horizontal dispersion was much greater than the

vertical. In most of the surveys at the Eniwetok Test Site the westward drift of the radioactivity was approximately equal to that of the current speed, although in some instances it was lower. This may be attributed to the effect of daily vertical migration of the macroplankton into deeper water, where the current flow is slower than that at the surface, and to eddy effect, if this exists.

A measure of the transport by surface currents of radioactivity in the Pacific Ocean was reported by MIYAKE, SUGIURA and KAMEDA (6), by HARLEY (7), and by SEYMOUR et al. (4).

Miyake et al. reported that 4 months after the nuclear tests at Eniwetok and Bikini the radioactivity had moved west to a distance of 1,200 miles, which is equal to a drift of more than 9 miles per day. Harley reported the westward drift to extend approximately 4,300 miles west from Bikini, which gives approximately the same rate of drift.

In an oceanic survey made with the USS *Marsh* approximately six weeks after the 1956 test series at the Eniwetok Test Site (Seymour *et al.*), the highest observed centre of radioactivity was derived from a surface detonation at Bikini Atoll. Forty-three days before the *Marsh* survey, the centre of this activity was located almost due north of Bikini Atoll. During the intervening time the contaminated centre moved a total of 300 miles at an average rate of approximately 7 miles per day. The radioactive material in this area of high activity was almost entirely associated with the surface waters (above 25 m), and the direction of drift (WSW) of the contaminated water from the original site, at which it was first detected, was in the direction of the prevailing winds in that area. Thus the major part of the radioactivity was carried by the surface currents in the direction of the wind and remained in the surface water for at least a month and a half. The original contaminated deeper waters from this detonation appear to have moved almost due west, and according to the *Marsh* survey contained low levels of activity in comparison with the surface waters. However, the deeper waters may have initially contained appreciable amounts of radioactivity which by the time of the second survey had sunk to a depth beyond the depth of sampling (300 m).

Revelle and Schaeffer reported that the rate of vertical diffusion above the thermocline is about 1,000 times that of molecular diffusion and about 1,000 times less than that of horizontal dispersion. The extent of vertical stirring within the mixed layer depends upon the surface winds and the vertical density gradient.

The thermocline is a layer of rapid temperature change and separates the surface layer, which in the area of the Eniwetok Test Site is less than 100 m thick, from the deeper waters. Because the temperature is fairly consistent throughout the upper or mixed layer, mixing in this layer should occur easily and require only small amounts of energy. Because the thermocline is a layer of high stability, transfer of materials across this layer by turbulent diffusion would be expected to be much less rapid than in the upper layer. Therefore radioactive materials introduced above the thermocline should remain in the mixed layer for a long time and be subjected to great horizontal distribution and small amounts of vertical distribution. However, radioactive materials may cross the thermocline by at least two methods:

1. If the radioactive material is accompanied by large amounts of stable elements, such as calcium compounds, as where weapons are fired over coralline islands or reefs, the calcium may precipitate into particles of

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fairly large size and scavenge the accompanying radioisotopes out of the
mixed layer and through the thermocline into the deeper layers.
2. Plankton undergoes diurnal vertical migrations of distances that exceed
the depth of the thermocline, and thus may carry with it radioelements
adsorbed or ingested in the mixed layer.
Thus three layers of water in the ocean are normally present—the surface
layer or mixed layer, in which mixing should occur easily; the thermocline,
which is very stable and across which radioisotopes may exhibit appreciable
movement only under special conditions; and the body of water below the
thermocline, in which vertical mixing is limited.
REVELLE, FOLSOM, GOLDBERG and ISAACS (8) (1955) reported that
when fission products were introduced at the surface of an area where the
mixed layer was approximately 300 feet thick, the radioactivity moved down
at a rate of about 11 feet per hour, so that within 28 hours the radioactivity
was uniformly distributed to the thermocline.
Any study concerning the influence of gravity on radioactive materials in
the open sea is complicated by the effects of currents and turbulent diffusion.
If the leading edge of the contaminated body of water is sampled with passage
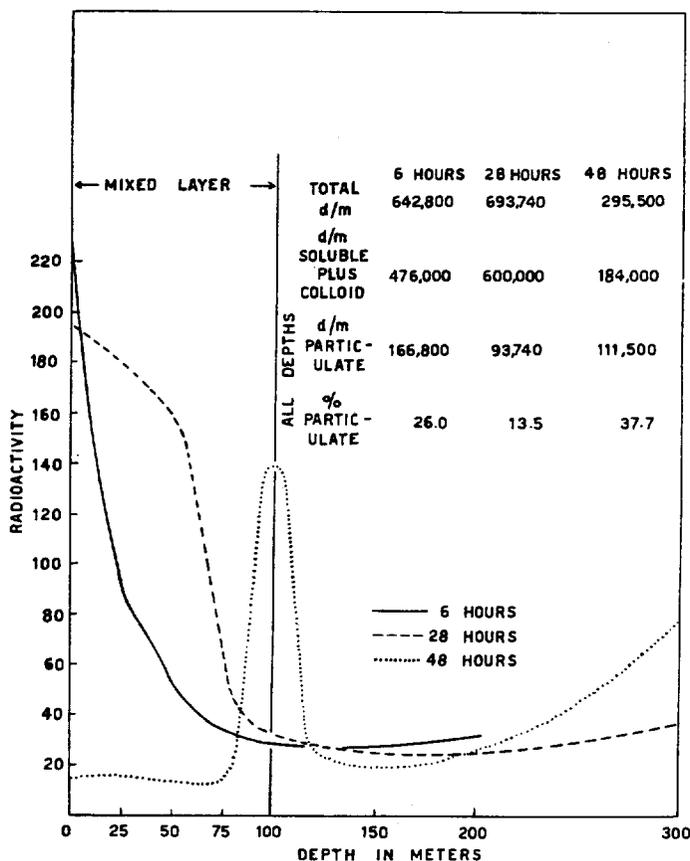

Fig. 1
Distribution of total radioactivity in sea-water with depth at 6, 28 and 48 hours

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of time, almost all the radioactive materials appear to remain in the surface waters. If the samples are taken at the original site of contamination over an extended period, the ocean currents, eddies and vertical turbulence tend to obscure the effects of gravity. However, the obscuring effects of these factors may be reduced in areas in the lee of land masses.

The effect of gravity upon radioactive materials introduced into the sea was studied during the 1958 test series at Eniwetok Atoll. Measurements were made within the contaminated area less than one hour after the detonation, and a series of three sets of water samples at depths to 300 m were taken over a period of 48 hours in the same area. The radioactive material in the water samples was divided by filtration into a particulate fraction greater than $0.45\,\mu$, and the colloidal-soluble fraction, less than $0.45\,\mu$. The distribution of total radioactivity with depth at the three times is shown in Fig. 1. During the first 6 hours the major part of the radioactivity was in the top 25 m. The radioactivity decreased with depth, so that through the upper edge of the thermocline the contamination was about one-eighth that at the surface. At
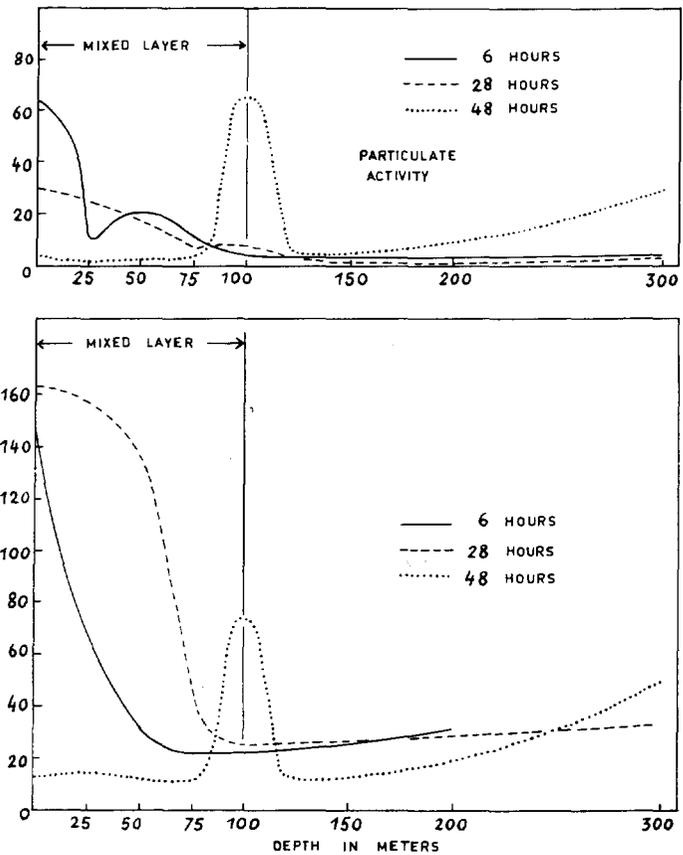
Fig. 2

Distribution of particulate activity and soluble-colloidal activity with depth at 6, 28 and 48 hours after an underwater detonation

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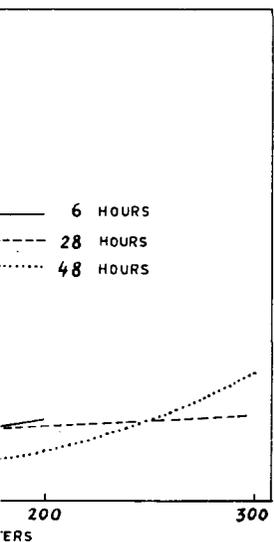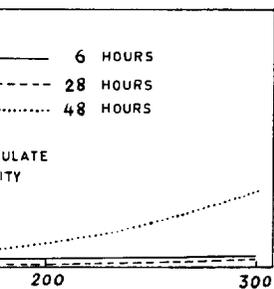
28 hours the radioactivity was distributed throughout the upper half of the mixed layer, and dropped in value rapidly between the depths of 50 m and the upper edge of the thermocline at 100 m. At 48 hours the major part of the total radioactivity was concentrated at the upper edge of the thermocline (100 m); the activity fell to a low value at approximately 150 m and then rose gradually to a level approximately one-half that at 100 m. Thus the radioactivity was distributed throughout the mixed layer between 28 and 48 hours. Probably at no time, however, was it evenly distributed throughout the mixed layer.

The distribution of activity with depth for the particulate fraction and the soluble-colloidal fraction at the three times following contamination is shown in Fig. 2. During the first 6 hours approximately one-fourth of the radio-activity was in the particulate fraction and was located in the top 25 m. The radioactivity associated with particles at this time was highest in the top 15 m of water, and a lower level of activity (approximately one-third of the radioactivity at the surface) was at approximately 50 m depth. At 28 hours only one-eighth of the total radioactivity was associated with the particulate fraction, and the major part of the total activity was located in the top 60 m of water. The two peaks of activity noted in the particulate fraction at 6 hours had migrated downward a distance of approximately 50 m each during the 22-hour period, and the deeper peak of activity coincided with the lower edge of the mixed layer. At 48 hours a major part of the radio-activity in both fractions was concentrated at a depth of 100 m (at the upper edge of the thermocline). At 300 m the radioactivity in both the particulate and the soluble-colloidal fractions rose to a value of about one-half that at 100 m. At 48 hours the particulate activity accounted for approximately two-fifths of the total.

During the period 6—28 hours following detonation, the activity associated with particulate matter dropped from one-fourth to one-eighth of that in the soluble-
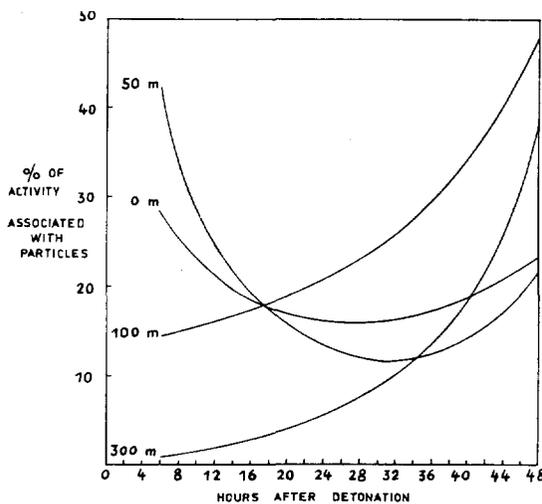
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-colloidal activity with depth at 6,
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Fig. 3
Percentage of radioactivity in sea-water in the particulate form at different depths with increasing time

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colloidal form. However, at 48 hours particulate radioactive material accounted for two-fifths of the total radioactivity. A major part of this activity was concentrated at the upper edge of the thermocline. Fig. 3 shows the percentage of radioactivity in the sea water in the particulate form at different depths with increasing time after detonation. In the two samples collected within the mixed layer (0 m and 50 m) the percentage of particulate radioactivity in the water decreased between 6 and 28 hours and rose slightly at 48 hours. However, in the upper edge of the thermocline (100 m) and inside the thermocline (300 m) the percentage of particulate activity rose at an increasing rate during the interval 6—48 hours post-detonation. The percentage of radioactivity in the particulate form at 300 m rose at an exponential rate from a value of approximately 1% at 6 hours to 37% at 48 hours.

During the interval 6—28 hours the leading edges of the particulate radioactivity moved downward approximately 50 m, and those of the soluble-colloidal activity approximately 60 m. During the time 28—48 hours the leading edges of both the particulate and the soluble activity in the mixed layer descended approximately 50 m. Thus the activity in the mixed layer during the entire period of sampling descended at a rate of approximately 2.5 m per hour.

Below the mixed layer, however, the radioactivity in the particulate form appeared to descend much faster than that in the mixed layer. During the period 28—40 hours the peak of radioactivity moved down at least 200 m,
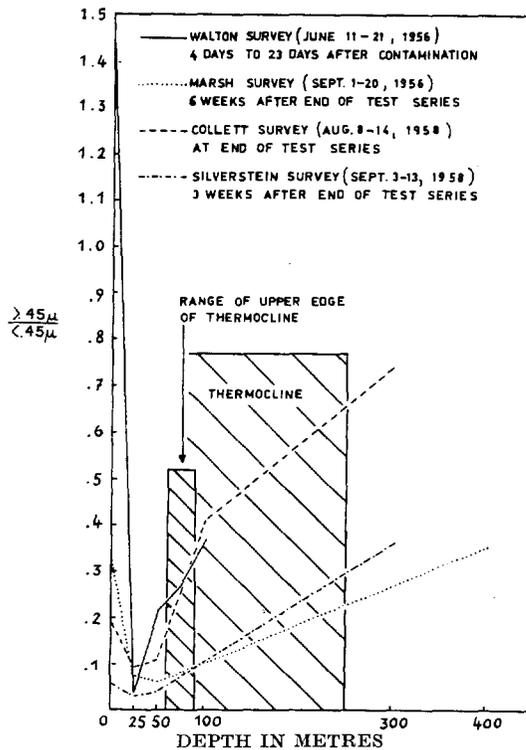

Fig. 4

Ratio of activity in particulate fraction to that in soluble plus colloidal fraction in all water samples from four oceanic surveys made during 1956 and 1958 between the Eniwetok Test Site and Guam

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or approximately 10 m per hour. This may reflect the separation of the particul-
ate matter into a heavier fraction, which was observed at 6 and 28 hours as
deeper concentrations of radioactive particles and which sank below the mixed
layer ahead of the remaining particulate activity. During the last 20 hours
the heavier particles descended rapidly through the thermocline.

The failure of the radioactive contamination to become evenly distributed
throughout the mixed layer might be attributed to the short duration of the
observations on the movement of the radioactive material in the water,
although this is probably not the case. In the area of the test site, at least, the
combination of the effect of gravity on the particulate activity plus the rapid
movement of the surface waters (above 25 m depth) tends to cause stratifica-
tion in the mixed layer of sufficient magnitude to overcome, in part, homo-
geneous mixing in this layer. The ability of phytoplankton to concentrate radio-
active materials and to remain near the surface probably adds to the tendency
towards stratification in the mixed layer.

Homogeneous dispersion of the radioactive contamination throughout the
mixed layer may occur after sufficient time has elapsed, as indicated in the
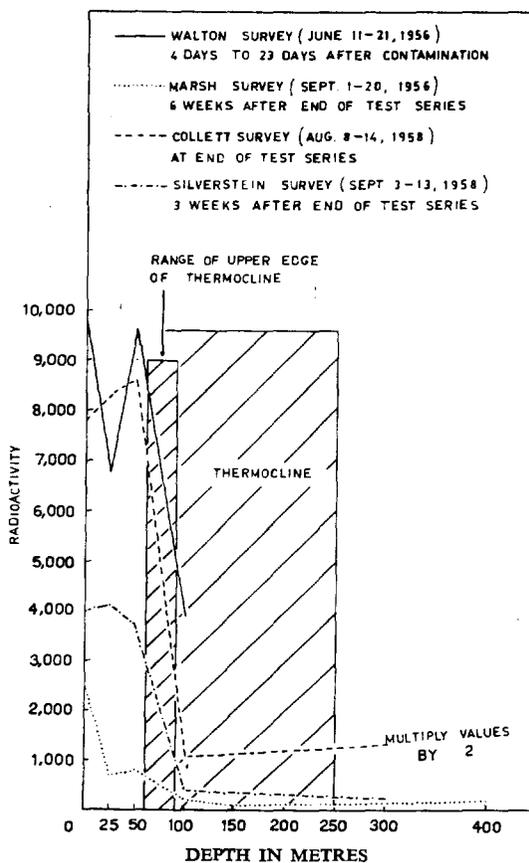

Fig. 5

Total radioactivity in water samples from four oceanic surveys made during 1956 and
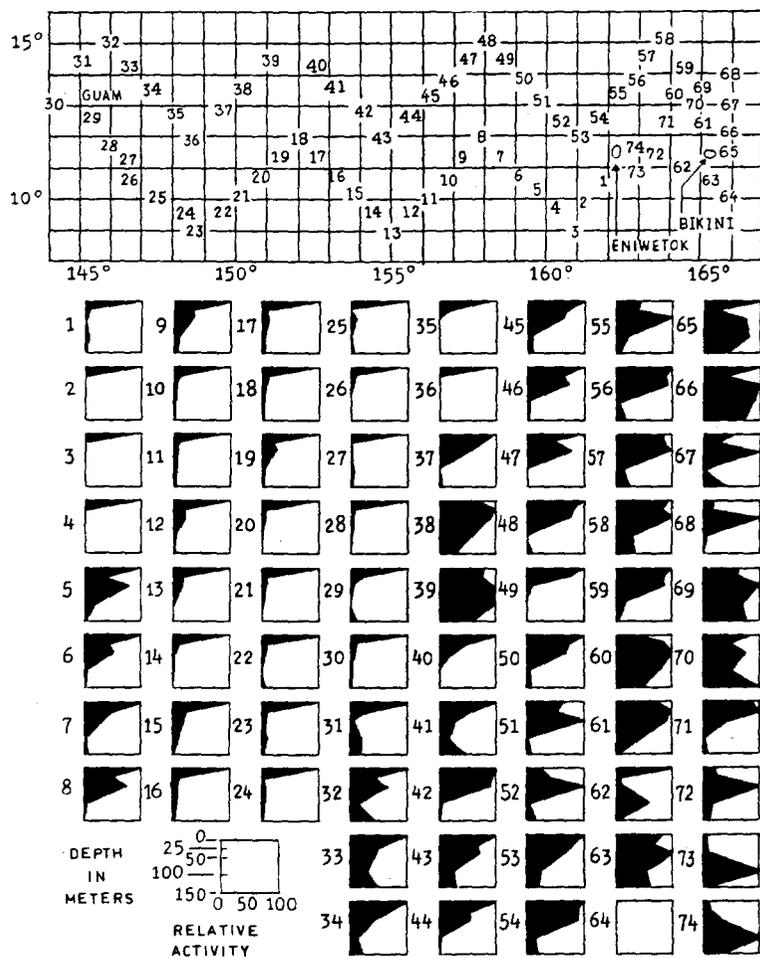1958 between the Eniwetok Test Site and Guam

soluble plus colloidal fraction in all
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Fig. 6

Collection stations of the *Marsh* survey (top) and the relative radioactivity of the wate (filter and filterable fraction less K^{40}) at each depth sampled. Maximum value at eac station taken as 100

Troll Report (7). However, in the area of the Marshall Islands and as far wes as Guam this dispersion does not occur within 6—8 weeks after contaminatio of the water mass. According to samples taken during four different surve in 1956 and 1958 the radioactive material was not uniformly distributed in th mixed layer. In samples taken a few days after contamination and 6 week later, the ratio of the radioelements in the particulate matter to those in th soluble-colloidal form varied throughout the upper mixed layer (Fig. 4). Al the total radioactivity in the water varied throughout the depths above th thermocline (Fig. 5). The values illustrated in these two figures are averages all stations sampled and show only overall trends. The lack of homogeneit within the mixed layer was even more marked at individual collection statio (Fig. 6).
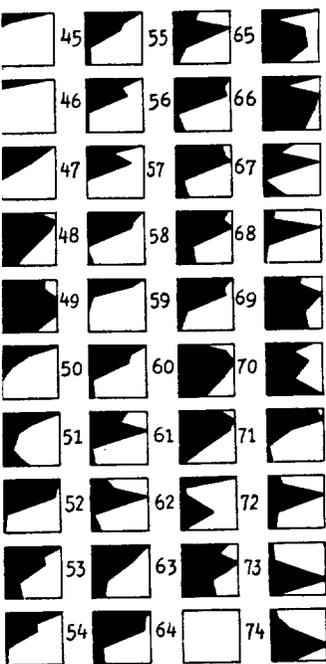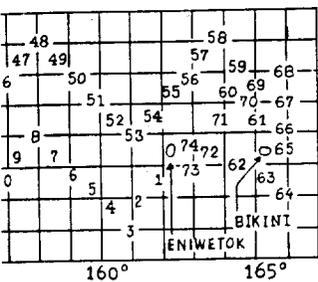
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In the four oceanic surveys made at the Eniwetok Test Site during 1956 and 1958, the ratios of particulate activity to soluble activity in water samples exhibited similar patterns (Fig. 4), with ratios relatively high at the surface, dropping to a minimum value at 25—50 m, and rising again near the thermocline. Only in the 1956 (*Walton*) surface samples, taken 4—23 days after contamination, did the particulate fraction contain a greater amount of radioactivity than the soluble fraction. The major part of the radioactivity observed during the *Walton* survey resulted from a surface land detonation which, at the time of sampling (23 days after detonation), included an area approximately 100 miles in diameter.

Another fallout area with much lower activity, observed during the *Walton* survey, resulted from two devices fired over water 4 and 6 days before sampling. The contaminated area was approximately 30 miles in diameter. The fallout material from the surface shot contained large amounts of calcium compounds, whereas the material from the over-water shots was mixed, for the most part, with evaporated salts from sea water and the material surrounding the nuclear device. The radioactivity was not distributed evenly throughout the mixed layer in either contaminated area, and in the areas of high activity from both the surface shot and the over-water detonation the activity in the water tended to be concentrated near the thermocline (Fig. 7a).
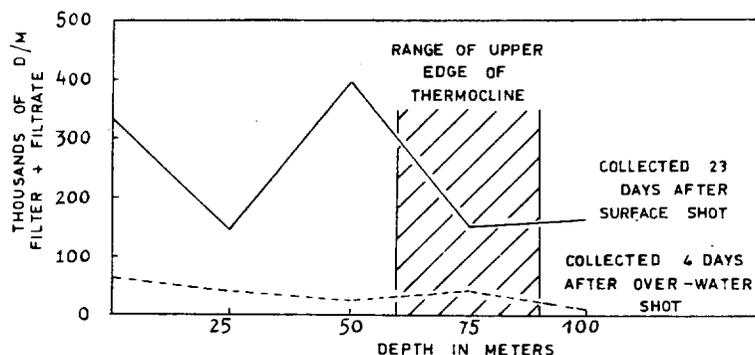

Fig. 7a

Total activity in water from two contaminated areas: one from a surface shot and one from an over-water detonation

The radioactivity in the water was about six times as high in the area contaminated by the surface detonation as in that contaminated by the over-water shot. The water samples from these two areas were combined into two samples to determine if any differences in distribution of activity with depth was apparent between the two types of fallout. In the area of fallout containing relatively large amounts of calcium compounds, the radioactivity in the water was high at the surface, low at 25 m, and again high just above the thermocline. Within the upper thermocline the radioactivity was about the same as that at 25 m. The ratio of radioactivity associated with particulate matter to that associated with matter in solution was high at the surface (ratio = 1.17), low at 25 m (ratio = 0.03), and rose with increasing depth into the thermocline to a value of 0.39 at 100 m (Fig. 7b).

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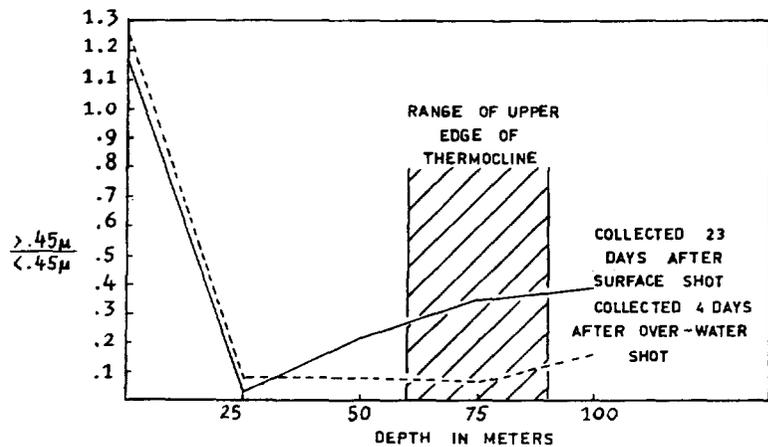ked at individual collection stations

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Fig. 7b

Ratio of activity in particulate fraction to that in solution plus colloid fraction from two contaminated areas: one from a surface shot, and one from an over-water detonation

In samples collected in the area from the over-water detonation, the total radioactivity in the water was highest at the surface, decreased gradually at 50 m to a value about one-third of the surface activity, and rose to a value about two-thirds of the surface activity in the upper edge of the thermocline. At 100 m depth the radioactivity was about one-tenth of that at the surface. More than half of the radioactivity at the surface was associated with particles. and at 25, 50 and 75 m less than one-tenth of the activity was in the particulate form. At 100 m about one-sixth of the activity was associated with particles.

In both contaminated areas the particulate matter at the surface is probably primarily comprised of microplankton. In the area contaminated with fallout containing calcium compounds, the rise of particulate activity at and in the upper edge of the thermocline is probably due, to a large degree, to very small inorganic particles hindered from settling through the thermocline because of the density gradient. Because few particles would be present in fallout from the over-water shot, a rise in ratio of particulate activity at the thermocline would not be expected, and the observed rise was only approximately one-fourth that observed from the ground detonation. The fallout area from the surface detonation was sampled 23 days after contamination; thus the larger particles present in the fallout would have already sunk to depths below those sampled by the time of the survey.

Chemical factors

When radioactive materials in the particulate form are introduced into sea-water, the particles may go into solution either because of their high solubility constant or from increased hydrostatic pressure as they sink through the water by gravity. If initially in the soluble form, they may be precipitated as particles of varying size by interaction with salts in the sea or with accompanying materials, or by adsorption to biological or inorganic particles. The particles. whether inorganic or organic, would tend to be removed from the mixed layer by gravity, although the planktonic organisms tend to offset this effect
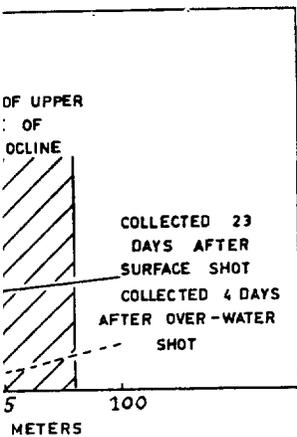
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by swimming upward. Because of the effect of gravity on particles, the soluble and insoluble fractions would probably act as two independent systems as far as concentration or dispersal processes are concerned.

In the waters of the open sea, the naturally-occurring trace elements zinc, cobalt, ruthenium, manganese, caesium, strontium, iodine, and possibly zirconium and cerium are present for the most part in solution. The small amount of natural iron occurs in the colloidal and particulate form and may have associated with it a limited amount of manganese and zirconium. All the above-named elements except caesium, strontium and iodine, however, when introduced to the sea in fallout, would most probably be present in the particulate form (36). The fallout elements strontium, caesium and iodine, which occur mostly in the soluble form, were found in the least amount in plankton. In contrast, the radioactive fission products with the least solubility, Zr^{95} and Ce^{144}, were present in plankton in the greatest amount (36, 7). LOWMAN (36) reported that the non-fission-product radioelements zinc, cobalt, and iron were present in plankton from one survey in approximately equal amounts of about 24% each and manganese at a level of less than one per cent. All of these elements introduced as fallout would exist initially in the insoluble form.

The physical state of any given element in sea-water will depend upon whether or not the solubility product of its least soluble compound has been exceeded (2, 10), and precipitation will occur when it is exceeded. In order to predict the form that a given radioelement will assume in sea-water, the ionic activity of the compounds likely to be present must be known. Very few data are available concerning the activities of the ionic forms of the fission-product elements in the sea. Limited information is available, however, for the neutron-induced radioelements found in fallout from thermonuclear devices.

Precipitation of a radioelement in the sea will occur only when precipitation of its stable element occurs normally or is induced by accompanying non-radioactive débris swept up into the fireball of the weapon at the time of detonation. This débris at the Eniwetok Test Site may be calcium compounds from the islands and reefs, materials from nearby structures associated with the nuclear devices or from the nuclear devices themselves, or salts from evaporated sea water. Several of the abundant fission products belong to the rare-earth series, and most of these elements are very insoluble in sea-water. In addition, sea-water is probably naturally saturated with rare-earth elements, so that any added material will be present in the particulate form (10).

Radioactive contamination may also occur in the sea in particulate form as a result of another process which is neither precipitation nor coprecipitation, but rather involves the adsorption of radioactive ions to organic detritus in the sea water. This process may be distinguished from the adsorption of radioactive ions or particles on to microorganisms. Both organic detritus and microorganisms have exposed polar groups with which transition elements such as manganese, iron, cobalt, nickel, copper and zinc, and anions such as zirconium, ruthenium, tungsten, neptunium, uranium, tellurium and molybdenum, may become complexed. A difference between organic detritus and microorganisms is an increased tendency of the former to sink to the depth of the sea, thus removing the radioelements from the biosphere. However, for the most part organic detritus probably sinks at a slower rate than particles of calcium compounds, and would thus tend to be removed from the surface waters at a slower rate than radioelements coprecipitated with calcium oxide or scavenged by calcite.

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Isotopes of a given element behave similarly in their uptake and metabolism by organisms, except the isotopes of hydrogen and possibly some of the other light elements. Thus one may assume that a radioactive isotope of a given element will follow the stable isotopes of the same element through the food web.

Because different isotopes of the same element generally behave identically, the degree at which a given radioisotope is taken up and retained by an organism depends to a large part on the amount of the stable counterpart of the same element present in the environment. The inhibition of uptake of a radioisotope by a stable isotope of the same element is known as isotope dilution, and is important in regard to health hazards from radioelements introduced into the sea. Moreover, elements closely related to each other in their ionic properties will tend to behave similarly and thus simulate isotope dilution. In this way the presence of stable calcium tends to reduce the uptake of radioactive strontium by organisms, and naturally-occurring potassium would be expected to have a similar effect on the uptake of radioactive caesium. Naturally-occurring potassium, which is similar chemically to caesium, is present in sea-water at the relatively high level of about 380 ppm. In comparison with potassium, Cs^{137} would be present in extremely small amounts by weight even in an area of heavy fallout. Because animals exhibit a limited ability to differentiate between the two elements, the uptake of caesium would be low in the presence of an abundance of potassium.

Natural calcium occurs in the sea at a level of about 440 ppm, and is very similar to strontium with regard to uptake by marine organisms. In the case of strontium and calcium, however, the uptake of Sr^{90} is not directly proportional to its occurrence in the water. Sr^{90} is discriminated against with reference to and in the presence of calcium by factors of about 3 in calcareous algae and foraminiferans, 2 in arthropods, 2—7 in molluscs, 2.5 in bryozoans (11), and 3—10 in marine fish (12). In addition to chemical competition, isotope dilution by stable strontium would result in reduced uptake of Sr^{90} by marine organisms. Stable strontium is present in the sea at a level of 6 to 1,300 times that of the naturally-occurring forms of the other major radioelements represented in fallout. As a consequence, the isotopic dilution of radiostrontium would be at least 6 to 1,300 times that to which the other radioelements would be subjected because of the presence of their stable counterparts.

The apparent discrimination by marine organisms against the uptake of radiostrontium may also be enhanced by a purely physical factor, including scavenging and coprecipitation. Granules of calcite resulting from pulverized islands effectively scavenge radioactive strontium as they sink through the sea into the deeper waters (36). SUITO, TAKIYAMA and UYEDA (13) reported that the ashes from the 1 March 1954 detonation which fell on the *No. 5 Fukuryu Maru* consisted of white granules of calcite approximately 100–400 μ in diameter. Identification of the chemical form was made by electron microscopy and X-ray diffraction techniques.

It is usually assumed that particles sink in the sea according to Stoke's Law, in which the assumption is made that the particles are smooth and rigid, are of given diameter and density, and do not interact with each other as they sink through the water. Calculations by Miyoshi indicate that particles of calcite 400 μ in diameter would sink from the surface to 260 m in 26 minutes, and particles 100 μ in diameter in 7 hours. Calcium from the pulverized islands

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and reefs on which the nuclear detonations occurred would probably be in the form of calcite and calcium oxide.

Thus the mechanism for rapid scavenging of Sr^{89-90} in fallout introduced into the sea is provided by calcite settling through the thermocline. Co-precipitation with calcium compounds would also scavenge radiostrontium from sea-water. When $CaCo_3$ is precipitated in sea-water, strontium is co-precipitated and carried down in the precipitate, a technique commonly used in chemical separations for sea-water. Because an appreciable amount of the calcium in fallout resulting from surface detonation is in the form of CaO and $Ca(OH)_2$, a considerable amount of radioactive strontium would be carried to deeper waters by this action. When strontium is coprecipitated with calcium in the particulate form, it would be available to filter-feeding organisms. However, an appreciable amount of the precipitate would probably settle out of reach of the biosphere into the deeper water. Some of the radiostrontium would undoubtedly be ingested by the filter-feeders. However, a rapid turn-over rate for this element has been observed in all marine invertebrates and fishes studied up to now (14, 15). Because of the rapid turnover rate, ingested radioactive strontium would be continually recycled to the sea in solution, where it would become more and more diluted with the naturally-occurring non-radioactive form.

Biological factors

Biological factors affecting the uptake of radioisotopes include biomass, surface area of the biomass, adsorption, absorption, feeding habits and ingestion, physiological selectivity, deposition, excretion rates, reproductive rates, average life-spans and growth rates, mobility, including horizontal and vertical migrations, and symbiotic relationships.

The ultimate source of all energy for living material is derived from the sun by photosynthetic processes in plants. This energy is utilized to convert basic inorganic raw materials into organic compounds, which are then passed throughout the food web to the various organisms of the biomass. Since the efficiency of conversion at each stage in the food web is low, usually 10 to 15%, the mass at each trophic level decreases with each step of conversion. Thus, with equal reproductive rates, the amount of total organic material and the average total of biochemically-combined minerals decreases as one passes through the food web, and the degree at which the mass decreases is dependent primarily upon the reproductive rate and average life-span of the populations making up a given level in the food web. The marine biosphere may be divided into the following levels: (1) the primary synthesizers, which are mainly comprised of photosynthetic plants; (2) the omnivores, which in the sea are usually filter-feeders, and (3) the carnivores, which include fishes such as tuna and sharks.

Metabolism in all forms of life at the cellular level is similar, although the organisms may differ markedly in body form and complexity. This similarity is present in organisms of both the plant and the animal kingdoms. Marine organisms tend to concentrate transition elements such as manganese, iron, copper, nickel, cobalt, zinc, and some of the anions. The transition elements, at least, are strongly bound to the organisms and are not easily removed by placing the organisms in sea-water containing lower amounts of these elements. They may be concentrated by factors up to 100,000.

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GOLDBERG (34) stated that two general processes operate in the uptake of trace materials from the marine environment: (1) a direct transfer of ionic species and dissolved substances from the hydrosphere to the organism, and (2) the uptake of particulate matter, including adsorbed surface ions.

LEHNINGER (16) discussed the physical bases of the specificity of metal ions in enzyme systems in the light of ion structure and properties. Many of these properties are also applicable to the study of the specificity of metal ions for proteins and other biological substrates, e. g. (1) mass, (2) ionic charge, (3) ionic radius, (4) oxidation-reduction potential, (5) the configuration and stability of the hydrates of the metallic ion in solution, and (6) the configuration and stability of coordination complexes of the metallic ion with substances other than water (i. e. organic detritus, bottom clays and muds).

Manganese, iron, cobalt and nickel are closely related, since they are members of the first transition series of the periodical table; thus they differ little in atomic weight, ionic radius, oxidation-reduction potential or mobility, and are equal in charge. All are capable of forming coordination complexes with many organic functional groups, and may form either ionic or covalent linkages.

It is probable that both Mn^{++} and Mg^{++} form aquocations of the type $Mg(H_2O)^{++}$ as do Fe^{++}, Ni^{++}, and Co^{++}, and all may form coordination complexes with organic compounds. Mn^{++} and the other transition metal ions would appear to be more versatile in this respect than Mg, since the unfilled 3-d electron orbitals allow formation of covalent as well as essentially ionic complexes.

The type of electrostatic bonding (ionic bonding) of general importance in biological adsorption is the ion-dipole bond, which results from electrostatic attraction between the electrically-charged metal ion and a dipolar molecule. Many such complexes are known, ranging from the simple aquocations to very complex forms. The ability of metal ions to form such complexes generally increases with ionic potential, except for the ions of the transition series, which show ability to form complexes out of proportion to their ionic potentials. Ions of low ionic potential such as Cs^+, Rb^+ and K^+ show the least tendency to hydrate or form complexes. Those of somewhat higher potential, such as Na^+, Ca^+, Sr^+ and Ba^+, show intermediate activity.

The transition elements are far more active in forming coordination complexes. They differ in that they may form essentially covalent linkages between the metal and the complexing molecule as well as ionic complexes. This difference is due to the fact that the metals of the first transition series have a tendency to borrow electrons from other molecules to fill out their 3-d orbitals, thereby establishing essentially covalent linkages in which a pair of electrons is shared between the metal and the group bound. The transition elements may also form ionic complexes. However, many coordination complexes have properties which suggest that the bond must be regarded as a hybrid between the two extremes of both types (16).

The relative interactions of the transition elements with any given biological substrate do not for the most part depend upon the chemical composition of the substrate. The stability usually occurs in the following order:

$$Mn^{++} < Fe^{++} < Co^{++} < Ni^{++} < Cu^{++} > Zn^{++}.$$

SALTMAN (17) observed that the accumulation of the trace metal ions, iron, copper and zinc, by different cells was unique when compared with other

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transport mechanisms. The transition elements were taken up by cells from a solution of very low concentration of the metals without apparent expenditure of energy by the cells, and was thus a non-metabolic process. Neither did the process obey the laws of diffusion normally observed for other elements, in that they did not respond to concentration gradients. Saltman concluded that for the transition elements no diffusion barrier was presented by the cell membrane, but that the rate of uptake was limited by the rate at which the ions found binding sites inside the cell. This process would exert only a minor effect on the external surfaces of planktonic organisms, but would be of major importance where cell surfaces were exposed.

KORRINGA (18) noted that oysters and other lamellibranchs concentrated considerable quantities of the metals Al, Mn, Fe, Cu, Zn and Pb, especially during periods of active feeding, although they occurred in very small concentrations in the environment. Korringa stated that the electrical properties of the food particles and the mucus feeding sheets in the oyster determine whether or not particles are rapidly caught. The positive polyvalent ions such as Al^{+++}, Cu^{++}, Fe^{++}, Zn^{++}, Hg^{++} and Mn^{++} were observed to be caught and accumulated by the oyster, but not positive monovalent ions such as Na^+ and K^+, though present in greater amounts.

The ability of plankton organisms to form complexes with heavy metal anions and the transition elements is illustrated by observations on the uptake of radioelements by these organisms in areas of radioactive fallout. The levels of different radioisotopes in plankton change with time after introduction of contamination. The change in ratio depends upon at least two main factors—the physical decay of the radioisotopes in question, and the velocity at which concentration of the individual isotopes occurs within the organisms. During the first 48 hours after detonation the principal isotope present in the plankton was Np^{239} (69 %, Table 1). Mo^{99}—Tc^{99m} and Te^{132}—I^{132} contributed approximately 10 % each of the total activity. I^{132} was the only iodine radioisotope found in measurable amount in marine plankton, and was present only as a consequence of being the daughter of Te^{132}. Radioactive iodine would be in solution in sea-water and, unless taken up selectively, would undergo dilution by stable chlorine, bromine and fluorine, as well as isotope dilution by the stable iodine. Other isotopes present in plankton within the first 48 hours after contamination at levels of 3 % or less included U^{237}, radioactive Ru-Rh, Ce-Pr, Ba-La, and Zr-Nb. Thus the radioisotopes associated with the plankton during the first 48 hours consisted principally of radioactive anions (96 %). Later than one week after contamination the ratio of the different isotopes in the plankton changed so that only 15 % of the total radioactivity was contributed by the anions—Zr^{95}—Nb^{95}, 6 %; radioactive Ru—Rh, 5 %; Np^{239}, 2 % and U^{237}, 2 %. At this time $Co^{57, 58, 60}$ contributed 43 % of the total radioactivity; Zn^{65} accounted for 3 % and Fe^{55+59} for 16 %. Ba^{140}—La^{140} contributed the remaining 23 %. In samples collected six weeks after detonation in 1956, the major isotopes were Zn^{65} (25 %), Fe^{55} (24 %), and $Co^{57, 58, 60}$ (24 %). Most of the remaining activity was contributed by Zr^{95}—Nb^{95} (20 %), Ce^{144}—Pr^{144} (5 %), and Ru^{106}–Rh^{106} (1.4 %); Mn^{54} was present only in trace amounts.

Therefore the initial uptake of radioisotopes by plankton included, for the most part, short-lived radioactive anions. However, within a short time the radioactive transition elements cobalt, zinc, iron and manganese were accumulated and retained by plankton, so that at an interval greater than

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TABLE 1

Average values of radioisotopes in plankton from two surveys made during 1958 and one in 1956 (*Marsh*). The six samples collected during the period less than 48 hours post-shot were from one detonation. The four samples collected at greater than one week had been contaminated by fallout from several nuclear devices. The five samples taken approximately six weeks after contamination had been subjected to radioactive fallout from several detonations.

	half-life	Rehoboth Survey < 48 hours	Collett Survey > 1 week	Marsh Survey 6 weeks
Mo^{99}—Tc^{99m}	66 h	12	0	0
Ce^{141}—Pr^{141}	33 d	2	0	0
Ce^{144}—Pr^{144}	285 d	< 1	0	5
Ru^{103}—Rh^{103}	40 d			
Ru^{105}—Rh^{105}	4.5 h, 36.3 h	3	5	1.4
Ru^{106}—Rh^{106}	1 y			
Ba^{140}—La^{140}	12.8 d	2	23	0
Te^{132}—I^{132}	77 h	8	0	0
Zr^{95}—Nb^{95}	65 d	1	6	20
Sr^{89}—Sr^{90}	50 d, 28 y	0	0	0
Cs^{137}	26.6 y	0	0	0
Np^{239}	2.3 d	69	2	0
U^{237}	6.8 d	3	2	0
Co^{57}	270 d	0	7	9
Co^{58}	71 d	0	33	14
Co^{60}	5.2 y	0	3	1
Zn^{65}	245 d	0	3	25
Fe^{55}	2.6 y	0	15	24
Fe^{59}	45 d	0	1	0
Mn^{54}	291 d	0	0	< 1

one week after contamination, these radioelements comprised 62 % of the total radioactivity, and at the end of six weeks accounted for 74 % of the contamination associated with the plankton.

The two anions accumulated by plankton (Zr^{95}—Nb^{95} and Ru^{106}—Rh^{106}) probably follow the metabolic path of some chemically similar anion or anions normally present in the marine environment. Evidence that radioactive anions are not retained *per se* is found in observations on the uptake by plankton of a radioactive anion with no known metabolic function.

In an oceanic survey made in 1958, LOWMAN et al (19) found a neutron-induced radioisotope of tungsten, W^{185}, for the first time. The isotope was found in high amount in water and plankton (~ 50 % of the total activity) in an area of high total radioactivity about 180 miles in diameter. About one-half of the W^{185} in the plankton was associated with silica, probably from the skeletons of marine diatoms.

Almost all the radioactivity in the water samples was contributed by Zr^{95}—Nb^{95} and W^{185}. A major part of the material containing the latter radioelement in the water samples passed through a filter with a pore size of 0.45μ.

A second survey was conducted three weeks later. During the time between the two surveys the centre of contamination moved 150 miles to the WSW, a net advance of 7.3 miles per day. In plankton samples collected during the second survey no W^{185} was found, although almost all the radioactivity

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...hoboth Survey ...8 hours	Collett Survey > 1 week	Marsh Survey 6 weeks
12	0	0
2	0	0
< 1	0	5
3	5	1.4
2	23	0
8	0	0
1	6	20
0	0	0
0	0	0
69	2	0
3	2	0
0	7	9
0	33	14
0	3	1
0	3	25
0	15	24
0	1	0
0	0	< 1

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...r samples was contributed by ...rial containing the latter radio- ...a filter with a pore size of 0.45μ. ...later. During the time between ...moved 150 miles to the WSW, ...kton samples collected during ...gh almost all the radioactivity

in the water samples was contributed by this radioisotope. $Zr^{95}—Nb^{95}$ were present in the plankton.

Although there was a high initial uptake of W^{185} by the plankton, probably by surface adsorption, the loss of the isotope was rapid. W^{185} also forms complexes with the surfaces of terrestrial plants and marine algae. In terrestrial plants it remains complexed to the surfaces of leaves exposed to almost daily rainfall, and the degree of binding appears to be directly related to the waxiness of the leaf surface. It was not present in the internal parts of the plants.

In marine algae the level of W^{185} was related to the exposed surface area. Radioactive tungsten is probably removed from organic surfaces in the sea by exchange with the ions in the water.

The second process of accumulation of radioisotopes by marine organisms is brought about by filter-feeding planktonic organisms which would accumulate both inorganic particles and microscopic organisms with their adsorbed ions. Some of the radioactivity contributed by the transition radioelements would probably be present in the area in combination with the hydrated oxides of iron. Also the radioactive anions accumulated by plankton would be largely associated with the same particles. GOLDBERG (34) showed that a marine diatom was unable to take up ionic iron in the complexed form but readily assimilated particles of hydrated iron oxide. Thus, in addition to the concentration of transition elements by the filter-feeding plankton by direct surface adsorption, a further concentration of these elements would occur through ingestion by these organisms of particles of hydrated iron with associated radioelements, particles of organic detritus with adsorbed radioelements, and contaminated microplankton.

Limited data from both control experiments (15) and field observations (4) suggest that an important route for uptake of fallout radioisotopes by marine animals is through the ingestion of particulate matter containing the radioactivity, whether the particles be inorganic or phytoplankton. CHIPMAN (15) noted that, in addition to the filter-feeding animals, two species of algae were also able to concentrate Ce^{144} several times over the levels in the water although the radioisotope was in the particulate form. The uptake of particulate material may either involve adsorption of the particle on to the organism or the ingestion of the particulate material, which would include both food particles and non-living particulate detritus.

The fractionation of introduced radioactive materials in the sea between the macroplankton, microplankton, nannoplankton, non-living particulate matter and colloidal and soluble fractions is important in the interpretation of the deposition of radioisotopes in marine organisms or in the bottom of the sea. In surveys at the Eniwetok Test Site the total activity in the samples was subdivided into three fractions—the macroplankton, the microplankton plus particulate matter greater than 0.45μ, and the soluble-colloidal material less than 0.45μ. The macroplankton and some microplankton were collected by means of half-metre nets of No. 6 mesh, and the nannoplankton plus inorganic particulate matter was collected by filtering 500 ml of sea-water, taken at various depths, through a millepore filter with a pore size of 0.45μ. A portion of the filtrate was then treated with concentrated $CaCo_3$ to precipitate practically all the radioelements from the water. The latter fraction included colloidal and soluble material.

Although some microplankton was undoubtedly retained by the filter

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paper, the ratio of radioactivity in the macro-microplankton to that in the nannoplankton plus particulate matter is insignificant (on the average 0.00025 of the particulate matter), and all the material on the filter paper may be considered to be from nannoplankton and inorganic particulate matter. Also, no macroplankton was retained on any of the filter papers.

The radioactivity of the macro-microplankton per unit volume of sea water was calculated by using data on plankton volumes reported by the Pacific Oceanic Fisheries Investigation from surveys made in 1956 and 1957 in the Marshall Island areas. In this study, the volumes of water strained by plankton nets were measured and compared with the volumes of plankton obtained. In the open sea approximately one ml of plankton was collected for every 100,000 l of water filtered.

In four different surveys made at the Eniwetok Test Site in 1956 and 1958, the radioactivity in the macro-microplankton was low in comparison to that in the particulate material and to that in the dissolved material (Table II).

TABLE II

RATIO OF ACTIVITY IN WATER TO ACTIVITY IN PLANKTON

	Filter paper plankton	CO_3 plankton	Filter paper + CO_3 plankton	
Walton survey 11—21 June 1956	2,900	7,600	10,500	4—23 days after detonation
Marsh survey 1—21 Sept. 1956	7,800	28,000	36,000	∼90 days after detonation
Collett survey, 8—14 Aug. 1958	1,500	9,400	10,900	at end of series
Silverstein survey, Sept. 1958	5,100	100,000	105,000	∼3 weeks after end of test
Walton—device detonated over water	3,400	10,500		series
			13,900	4 and 6 days post-shot
Walton—fired on ground	2,900	7,400		
			10,300	23 days post-shot

The *Walton* survey was made during the middle two weeks of June 1956 (during the test series) and included 53 sampling stations. Two centres of contamination were included in the survey area and were sampled 4–6 days and 23 days respectively after detonation of the devices producing the fallout. In an average of the *Walton* samples, the particulate matter greater than 0.45 μ contained 2,900 times and the dissolved material 7,600 times as much radioactivity as did the macro-microplankton per litre of water. The samples were also subdivided to include (1) the fallout area resulting from contamination produced by two closely-spaced over-water shots, and (2) a contaminated area with fallout containing large amounts of calcium compounds from a device fired on the ground. No appreciable difference existed between the ratios of particulate activity to macro-microplankton nor the dissolved activity to macro-microplankton in these two areas.

Another survey was conducted with the USS *Marsh* approximately $2^1/_2$ months after the *Walton* survey. The filter paper/plankton ratio was

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tok Test Site in 1956 and 1958,
·n was low in comparison to
he dissolved material (Table II).

TIVITY IN PLANKTON

CO_3 plankton	Filter paper + CO_3 plankton	
7,600	10,500	4—23 days after detonation
28,000	36,000	~90 days after detonation
9,400	10,900	at end of series
100,000	105,000	~3 weeks after end of test series
10,500	13,900	4 and 6 days post-shot
7,400	10,300	23 days post-shot

·iddle two weeks of June 1956
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7,800 and the carbonate/plankton ratio 28,000. The ratio of the total activity in the sea-water to that in the plankton increased by a factor of about $3\frac{1}{2}$ during the $2\frac{1}{2}$-month interval between the surveys. A greater increase in activity occurred during the time between the two surveys conducted in August and September 1958. The first survey was done aboard the USS *Collett* during the period 8–14 August 1958, and the ratio of activity associated with the particulate matter to that in the plankton was 1,500 and that for the soluble activity to that in the plankton 9,400. During the survey made three weeks later aboard the USS *Silverstein*, the ratios were 5,100 and 100,000 respectively. The total activity in the water compared to that in the plankton increased by a factor of about 9.5 during the three-week interval.

The mechanism involved in the apparent increase of activity in the water with respect to that in the plankton is not known. In order to determine the factors involved, techniques must be devised to divide the particulate matter in the water into its organic and inorganic fractions and to separate the colloidal and soluble fractions from each other. In addition, accurate measurements must be made of the total volume of water strained during each plankton haul. At the present time these problems have not been solved.

Radioactivity in fish samples from Bikini and Eniwetok Atolls cannot be related to any specific date of origin of contamination, since the weapons test programme at the atolls has continued over a period of years and residual

TABLE III

RESULTS OF RADIOCHEMICAL ANALYSES ON ORGANS AND TISSUES OF FISH TAKEN IN AND NEAR THE ENIWETOK TEST SITE

Sample	Isotope*							
	Mn^{54}	Fe^{55}	Fe^{59}	Co^{57}	Co^{58}	Co^{60}	Zn^{65}	Zr^{95}—Nb^{95}
Yellowfin tuna—Eniwetok Lagoon, June 1958 (during tests)								
Red muscle	trace	70	0	trace	trace	trace	21	8
White muscle	0	33	0	0	0	0	40	26
Liver	4	41	0	3	11	1	36	4
Kidney	1	72	0	4	17	2	1	3
Spleen	< 1	86	0	1	5	< 1	7	< 1
Fish liver—Eniwetok Lagoon, Sept. 1956 (six weeks post-shot)	4	35	trace	6	3	5	47	trace
Reef fish liver—Ailinginae Atoll, July 1957 (3 years post-shot**)	1	26	0	22	4	3	40	0
Tuna—Marshall Islands, open sea, July—Sept. 1958 (during tests)								
Red muscle	0.2	11.1		0.2	1.1	0.2	87.0	0
White muscle	0.3	6.9		0.2	1.2	0.3	90.5	0
Liver	0.1	18.1		0.1	0.6	< 0.1	80.9	0
Skin	0	6.1		0.3	1.7	0.4	91.5	0
Bone	2.7	5.4		< 0.1	0.5	0.1	90.5	0

* Sr^{90}—Y^{90}, Ru^{106}—Rh^{106} Cs^{137}—Ba^{137}, Ce^{144}—Pr^{144} not found in samples.
** Major contamination during March 1954. Recontaminated slightly during 1956.

contamination from previous tests is always present in the environment. However, comparisons of levels of radioelements in samples collected at different times do show the overall trend of uptake of radioactive materials by these organisms, and the observed ratios of the different radioisotopes in the organs of inidividual specimens indicate the sites of deposition in organs and tissues of the fish, and in some instances give evidence concerning turnover rates.

In Table III the results of radiochemical analyses on organs and tissues of fishes from the Eniwetok Test Site are shown.

In a yellowfin tuna sample from Eniwetok Lagoon, taken during the testing period in June 1958, Fe^{55} was the predominant isotope, with red muscle, kidney and spleen containing more than 70 % of the total radioactivity as this isotope. The highest level of $Co^{57, 58, 60}$ was in the kidney, suggesting a high turnover rate for radiocobalt in this fish.

The low value of Zn^{65} in the kidney suggests a low excretion rate for this element. The high level of radioactive iron in the kidney is probably not caused by a high excretion rate, but rather indicates a concentration of iron, which would be expected, since the kidney in fishes is a blood-forming organ. Similarly a high level of radioactive iron in the spleen is probably the result of active concentration of iron, since the spleen, too, has haematopoietic functions. In the red muscle Fe^{55} contributed 70 % of the total radioactivity, but in the white muscle only 33 %. Red muscle is known to contain greater concentrations of iron than white muscle, and therefore the deposition of radioiron in this organ would be expected. Mn^{54} apparently is not concentrated in the red muscle, white muscle, kidney or spleen, and was found in the liver at a level of only 4 % of the total radioactivity.

In fish-liver samples collected from Eniwetok Lagoon in September 1956, approximately six weeks after the test series, Zn^{65} contributed 47 % and Fe^{55} 35 % of the total radioactivity. Radioactive cobalt was present at a level of about 14 % of the total radioactivity, about the same percentage as that observed in the liver of the yellowfin tuna collected during the tests. Thus in fish-liver samples collected six weeks after the test series in 1956, the Zn^{65} was slightly higher and the Fe^{55} slightly lower than that found in yellowfin tuna liver collected during the 1958 test series. In the livers of reef fish collected at Ailinginae Atoll during July 1957, approximately three years after contamination, the levels of radioisotopes were similar to those in the two liver samples from Eniwetok collected during the test period and six weeks after.

During the test period in the summer of 1958, tuna samples taken in the open sea near the test site had different ratios of radioisotopes from those taken in the lagoons. In these samples Zn^{65} contributed 81—91 % of the total radioactivity in five different organs and the levels of Fe^{55-59} ranged from 5 to 18 %. Thus, in the fish samples collected in the open sea, the level of Zn^{65} in relation to the total activity is approximately twice that found in the lagoon fishes, and the levels of Fe^{55} and $Co^{57, 58, 60}$ are correspondingly lower. In addition, whereas small amounts of Zr^{95}—Nb^{95} were present in some of the lagoon samples, this radioisotope was not detected in the tuna samples from the open sea.

The plankton samples collected during the *Collett* survey (Table I) were taken at approximately the same time as the tuna samples from the open sea near the Marshall Islands (Table III). A comparison of the levels of the

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radioisotopes in the plankton and tuna samples gives some indication of the relative concentration factors for the different isotopes in the tuna fish. The two radioactive anions ruthenium and zirconium were present in the plankton at a level of approximately 11 % but were absent in the fish samples; 100 % of the radioactivity present in the fish samples was contributed by radioactive cations. $Co^{57, 58, 60}$ contributed 43 % of the total radioactivity in the plankton, and on the average only 1.4 % of that in the fish organs and tissues. Although Zn^{65} contributed only 3 % of the total radioactivity in the plankton, it was present at an average level of 88 % in the fish tissues. Fe^{55-59} contributed 16 % of the total radioactivity in the plankton and an average of 9.5 % in the tuna fish. Mn^{54} was not detected in the plankton samples and was present at an average level of 0.7 % in the fish samples.

The highest percentage of Zn^{65} in the tuna samples in comparison with that in the plankton suggests a low turnover rate for this radioelement in the fishes, and the very low percentage of radioactive cobalt in the tuna samples with respect to the percentage observed in the plankton suggests a high turnover rate for this element. The percentage of Fe^{55-59} in the tuna fish samples and in the plankton samples suggests a turnover rate for iron intermediate between that for radioactive cobalt and zinc.

Indirect evidence concerning the relative turnover rates of the three radio-elements iron, cobalt and zinc in fish was found in observations on the levels of these three radioelements in plankton samples collected at increasing time intervals following contamination of the sea during the 1956 test series (Fig. 8). The ratios of Co^{57}/Zn^{65} and Co^{57}/Fe^{55} were determined from plankton samples collected to the north and to the west of Eniwetok Test Site at distances of

Fig. 8
Ratio of Co^{57}/Zn^{65} (top) and of Co^{57}/Fe^{55} (bottom) in plankton taken approximately six weeks after the end of the test series in 1956

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150–480 miles. Since the major part of the radioactivity was deposited in the vicinity of Eniwetok Test Site, the samples collected at increased distances represented older contamination carried to the west by the water currents. With increased distance from the test site, the ratios of Co^{57}/Zn^{65} gradually changed from a value of 0.1 at 150 miles to a value of 5.5 at 480 miles, and the ratio of Co^{57}/Fe^{55} followed a parallel course. Thus, with increased time after contamination, the level of radioactive cobalt in the plankton increased by a factor of approximately 50 over that of Zn^{65} and Fe^{55}.

Grazing fishes feed upon the plankton and in turn are eaten by the pelagic carnivores such as tuna, bonitos and sharks. The rate of movement of these fishes is great in comparison with that of the plankton, and thus they would tend to disperse the radioactive material accumulated by feeding upon the contaminated plankton. If the pelagic fishes have a high turnover rate for cobalt and a low turnover rate for iron and zinc, they would tend to excrete the radioactive cobalt back into the water during their migrations through the contaminated area and would retain the radioactive zinc and iron. Thus the radioactive cobalt would again become available to the plankton, whereas the radiozinc and iron would be effectively tied up in a reservoir. The ratio of Co^{57}/Zn^{65} and Co^{57}/Fe^{55} in plankton would therefore increase with time after initial contamination.

In the marine plankton and fish exposed to fallout contamination, the principal radioelements concentrated after a few weeks' interval belonged to the transition series. In terrestrial organisms these radioelements are either absent or present in very low amounts. Sr^{90}—Y^{90} and Cs^{137}—Ba^{137m} contributed a major part of the activity in the land plants. Thus either a concentrating mechanism operates in the sea for the transition elements, or an exclusion mechanism operates on land. Conversely, in the case of Sr^{90} and Cs^{137} concentration occurs on land and exclusion occurs in the sea. Strom et al in 1958 reported the Co^{60}/Sr^{90} ratios in fallout samples from the 1956 test series. The samples were collected on large trays which had been exposed at various locations in the fallout pattern. The average ratio of abundance of Co^{60}/Sr^{90} at zero time was approximately 1/4, and the Co^{60}/Cs^{137} ratio would thus be approximately 1/6. Because Co^{60} has a half-life approximately one-fifth that of Sr^{90}, the ratio in favour of the presence of Sr^{90} would increase with time. If there were no exclusion against Sr^{90} in the sea, one would expect it to be present in the samples at a level four times or more that of Co^{60}. The same ratios should hold for land plants and land animals. However, although Sr^{90} and Cs^{137} are found in plants at Eniwetok at levels of approximately 7% and 85% of the total radioactivity, Co^{60} is absent, or present in only trace amounts. In the case of radioactive cobalt, zinc and manganese, the previously-discussed factors that limit uptake of Sr^{90} and Cs^{137} in marine organisms, except the scavenging action by CaO, $Ca(OH_2)$, and calcite, would tend to cause increased uptake of the non-fission-product elements. Thus the cobalt, manganese, and zinc that occur in fallout in the sea in the particulate form are not subject to chemical competition by similar elements nor to appreciable isotope dilution by their stable counterparts, neither should coprecipitation occur. However, cobalt, manganese and zinc would tend to precipitate with Fe^{55} and stable iron into a finely-divided form with only a limited tendency to sink through the thermocline, and thus these elements along with radioiron would remain available to the plankton in the mixed layer.

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Many of the radioisotopes concentrated by plankton organisms are biologically important to man. However, they do not necessarily have a long biological half-life in man, as has been stated elsewhere (2). Of the isotopes present in plankton in significant amounts for the first 48 hours after contamination, only $Ce^{141-144}$—$Pr^{141-144}$ (biological half-life 500 days) and Np^{239}—Pu^{239} (biological half-life 4.3×10^4 days)* have a biological half-life in man greater than one year. The other isotopes present include Mo^{99}—Tc^{99m} (150 days), Te^{132}—I^{132} (15 days), Ba^{140}—La^{140} (approximately 200 days) and $Ru^{103-105-106}$—$Rh^{103-105-106}$ (20 days). Of the isotopes present in plankton six weeks after contamination, only Ce^{144}—Pr^{144} has a long biological half-life. All of the other radioisotopes found in plankton at this time have biological half-lives of 65 days or less. These include Zr^{95}—Nb^{95} (50 days), Ru^{106}—Rh^{106} (20 days), Zn^{65} (23 days) $Co^{57, 58, 60}$ (9 days), Fe^{55} (65 days), and Mn^{54} (5 days) (Handbook-52).

Two isotopes which would be of long-term consequence as hazards to man because of their long biological half-life if taken up by plankton are Sr^{90}—Y^{90} (3.9×10^3 days) and Eu^{155} (1.4×10^3 days). However, these radioisotopes are not accumulated in any significant amount by marine plankton.

The biological half-lives of all of the isotopes concentrated by plankton are short in comparison with those of two radioisotopes commonly used in medicine, Ca^{45} (18×10^5 days) and P^{32} (3.2×10^3 days). However, the biological half-life is not the only criterion for determination of hazards of radioisotopes in humans. Another important consideration is that of radioactive half-life. The two isotopes Ca^{45} and P^{32} are relatively innocuous because of their short physical half-lives.

In fish tissues the predominant radioisotopes have short biological half-lives in man. They may therefore be tolerated in food or water for human consumption at a much higher level than Sr^{90}—Y^{90}. Fe^{55} may be present at a level of 8,000 times, Zn^{65} 1,000 times, Co^{57} 5,000 times, Co^{58} 1,000 times, and Co^{60} 500 times that allowed for Sr^{90} (Handbook 69).

The most important fission products in fallout on land, from the standpoint of hazard to human consumption, are Sr^{90}—Y^{90}. Cs^{137}—Ba^{137m} are potential hazards to a lower degree (21, 22, 23). The principal route for these radioelements is from the soil through the plants to man. The plants at Eniwetok and in the near vicinity tend to accumulate Sr^{90}—Y^{90} and Cs^{137}—Ba^{137m} from the soil with greater efficiency than they do the other radioisotopes (Table IV).

In soil from Eniwetok Atoll, Sr^{90}—Y^{90} accounted for 5.9% and Cs^{137}—Ba^{137m} for 3.8% of the total radioactivity. In three plants from the same island, Sr^{90}—Y^{90} accounted for an average of 7.3% and Cs^{137}—Ba^{137m} for 85% of the total radioactivity. On a direct comparison basis of disintegration rates of these isotopes in soil and in wet plant material, however, Sr^{90} was present in the plants at a level approximately one-sixth of that in the soil, and Cs^{137} at a level, on the average, of approximately $5\frac{1}{2}$ times that in the soil. In 12 samples of scaevola leaves collected at Rongelap Atoll in March 1958, Sr^{90}—Y^{90} accounted for approximately 26% and Cs^{137}—Ba^{137m} for 34% of the total radioactivity. In pandanus plant tissues from Rongelap collected in 1958, the leaves contained the lowest amounts of Sr^{90}—Y^{90} (0.37 to 3.1%) and Cs^{137}—Ba^{137m} (1.2 to 3.5%). In fruit, Sr^{90} contributed 5—7% of the total radioactivity and Cs^{137} 77—95%. Wood from the pandanus tree contained the

* Pu^{239} in the soluble form.

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TABLE IV

LEVELS OF Sr^{90}—Y^{90} AND Cs^{137} IN TERRESTRIAL SAMPLES IN AND NEAR THE ENIWETOK TEST SITE

		Sr^{90}—Y^{90}		Cs^{137}	
		d/m/g	%	d/m/g	%
*Eniwetok Atoll**					
Soil	dry weight	375	5.9	242	3.8
Messerschmidia	wet weight	96	11.9	667	83
Fimbristylis	wet weight	76	2.0	3,303	86
Sandburrs	wet weight	9.4	7.9	100	86.8
	Average		7.3		85
Rongelap Atoll					
Scaevola leaves (March, 1958)**					
(Average of 11 samples)			26.7		33.7
Pandanus fruit (August, 1958)**			4.6		95.4
Pandanus leaves			3.1		3.5
Pandanus trunk borings			71.4		28.6
*Kabelle Island (Rongelap Atoll)**					
Pandanus fruit (August, 1958)			6.5		92.6
Pandanus leaves (August, 1958)			0.37		1.2
Pandanus stems (August, 1958)			32.4		49.2
Pandanus bark (August, 1958)			5.4		4.6
*Eniaetok Island (Rongelap Atoll)**					
Pandanus fruit (August, 1958)			7.0		77.0
Pandanus leaves (August, 1958)			1.6		1.4
	Average		15.9		38.7

* D.J. South, 1957 (unpublished)
** D.J. South manuscript

highest percentages of Sr^{90}—Y^{90} (71%), and Cs^{137}—Ba^{137m} accounted for the remaining 29%. Thus on land the radioisotopes of strontium and caesium and their daughter products accounted for a significant fraction of the total radioactivity in the plants. This is not the case in the sea, although these isotopes have been considered to be also of primary importance in the contamination of marine organisms.

In a series of plankton samples collected at the Eniwetok Test Site in 1958 3 to 36 hours after contamination, the ratios of observed Sr^{90} to expected Sr^{90} based on the observed levels of Mo^{99}—Tc^{99m}, Te^{132}—I^{132}, Zr^{95}—Nb^{95}, Ce^{141}—Pr^{141}, and Ba^{140}—La^{140} were calculated. The results are shown in Table V.

In the plankton collected three hours after contamination, no Sr^{90} was detected in the large plankton. The level of Sr^{90} in the nannoplankton was, on the average, 0.43 of that expected on the basis of the levels of the above-named isotopes. In plankton collected later, no detectable difference in ratios of radioisotopes existed between the large and the small plankton. At 21 hours, Sr^{90} existed at a level 0.06 of that expected, and at 36 hours 0.03 of

^{90}—Y^{90} /m/g %	Cs^{137} d/m/g %	
5.9	242	3.8
11.9	667	83
2.0	3,303	86
7.9	100	86.8
7.3		85
26.7		33.7
4.6		95.4
3.1		3.5
71.4		28.6
6.5		92.6
0.37		1.2
32.4		49.2
5.4		4.6
7.0		77.0
1.6		1.4
15.9		38.7

^{7}—Ba^{137m} accounted for the
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TABLE V

RATIO OF OBSERVED Sr^{90} TO EXPECTED Sr^{90} IN PLANKTON BASED ON OBSERVED LEVELS OF Mo^{99}—Tc^{99m}, Te^{132}—I^{132}, Zr^{95}—Nb^{95}, Ce^{141}—Pr^{141} and Ba^{140}—La^{140}

Hours after contamination	Plankton	Mo^{99}	Te^{132}	Zr^{95}	Ce^{141}	Ba^{140}	Average
3	large	0	0	0	0	0	0
	small	0.16	0.23	0.42	0.18	1.18	0.43
21	large	0.14	0.06	0.06	0.06	0.08	0.06
36	large	0.07	0.05	0.02	0.01	0.02	0.03

that expected. This steady decrease in the expected levels of Sr^{90} with increased time is probably caused by a combination of two mechanisms: one, there was active concentration of the radioisotopes used for comparison, and two, there was little or no concentration of the Sr^{90}. Since Sr^{90} was not detected in any plankton samples collected six weeks after contamination, although in many of the samples the total amount of radioactivity was high, Sr^{90} was probably not concentrated in plankton organisms, and discrimination against its uptake may have occurred.

Strontium-90 is also found at low levels in marine invertebrates collected at the Eniwetok Test Site (Table VI) (South, unpublished). In spider-snail muscle and mantle, Sr^{90} accounted for 0.03% and 0.01% of the total activity. In sea-cucumber gonad, Sr^{90} contributed 0.06% of the total radioactivity. However, in the hermit crab, which lives on land and derives its food therefrom, Sr^{90} contributed 4.9% of the total radioactivity in the muscle and 40% of that in the carapace.

TABLE VI

PERCENTAGE OF TOTAL RADIOACTIVITY IN INVERTEBRATES FROM ENIWETOK ATOLL CONTRIBUTED BY Cs^{137}, Sr^{90} AND Ce^{144} (South, unpublished)

Eniwetok	Date	Cs^{137}	Sr^{90}	Ce^{144}
Spider-snail muscle	2/9/55	not done	0.03%	54%
Spider-snail mantle	2/9/55	not done	0.01%	57%
Hermit-crab muscle	2/9/55	84.7%	4.9 %	1%
Hermit-crab carapace	2/9/55	not done	40.0 %	1%
Sea-cucumber gonad	2/9/55	not done	0.06%	66%

Neither is Sr^{90}—Y^{90} found in high amounts in marine fishes collected in the contaminated area near the Eniwetok Test Site. NAGASAWA et al (25), in an analysis of muscle of yellowfin tuna collected at N 4° E 140° on 8 July 1954, found only 0.003 d/m of Sr^{90}/g of wet tissue. Sr^{90} accounted for 0.2% of the total activity. In the kidney of the same animal no Sr^{90} was detected. KAWABATA (35) reported in a study on contaminated fish tissues collected near the Eniwetok Test Site: "It is a noticeable fact that the activity detected in various samples of Group IV was slight or even negligible." This group includes strontium.

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HARDY and HARLEY (26) reported the strontium levels in eleven samples of albacore and tuna collected in the Western Pacific. The samples were canned tuna and contained Sr^{90} at an average level of 0.00226 d/m/g of wet tissue.

HIYAMA (27) reported Sr^{90} data on fish collected near the Eniwetok Test Site. Of 25 fish taken during the *Shunkotsu Maru* survey during June 1956, 19 contained no detectable Sr^{90} in the tissues, 5 contained approximately 0.14 d/m of Sr^{90}/g dry weight of bone (about 0.06 d/m/g wet), and one contained approximately 1.4 d/m of Sr^{90}/g of bone (about 0.6 d/m/g wet). Of nearly 100 fish collected during July 1954, the most highly contaminated contained 0.003 d/m of Sr^{90}/g wet weight of muscle.

SOUTH (unpublished) did strontium analyses on 9 samples of fish tissues collected in Eniwetok Lagoon between 6 July 1954 and 12 February 1955, and found that $Sr^{90}-Y^{90}$ contributed from 0.01% to 0.9% of the total radioactivity, an average of 0.13%. The highest level (0.9%) was found in shark cartilage, the lowest levels in muscle and liver (Table VII).

TABLE VII

PERCENTAGE OF $Sr^{90}-Y^{90}$, AND TOTAL DISINTEGRATION RATE PER GRAMME WET WEIGHT, OF FISH SAMPLES COLLECTED AT ENIWETOK DURING 1954—1955

Eniwetok*	Date	Percentage $Sr^{90}-Y^{90}$	Total d/m/g wet weight
Baracuda muscle	7/6/54	0.08	3
Bonito muscle	7/27/54	0.02	64
Shark muscle	10/27/54	0.1	1,500**
Shark skeleton	10/27/54	0.9	4,900**
Shark muscle	10/27/54	0.01	500**
Butterfly-fish liver	1/18/55	0.01	3,700
Mullet liver	2/10/55	0.02	160,000
Blue Bonito muscle	2/12/55	0.02	83
Blue Bonito liver	2/12/55	0.03	1,400

* South, unpublished
** d/m/g dry weight

LOWMAN, PALUMBO and SOUTH (9), in ion-exchange separations on bonito liver collected at Bikini Atoll, did not find Sr^{90}, although the total radioactivity in the sample was 50,000 d/m/g wet weight.

SUYEHIRO et al (29), in laboratory studies of the uptake of Sr^{90} in aquatic animals, reported that fresh-water fish took up Sr^{90} much more readily than did marine fish, which accumulated less than one per cent of the amount available. Suyehiro attributed the reduced uptake in sea water to salinity. However, other factors, including isotope dilution and dilution by stable calcium, were probably the primary causes for reduced uptake.

BOROUGHS et al (14) found that approximately one to two per cent of Sr^{89} which had been fed to 10 yellowfin tuna was retained after a period of 24 hours, and that another fish (*Tilapia*) contained on the average only about 7 1/2% of the ingested amount after 4 days. These animals were subjected to acute exposures of Sr^{90}, and the results would probably differ from those from a chronic exposure. Because marine organisms are able to concen-

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um levels in eleven samples
Pacific. The samples were
vel of 0.00226 d/m/g of wet

ted near the Eniwetok Test
u survey during June 1956,
.5 contained approximately
6 d/m/g wet), and one con-
: (about 0.6 d/m/g wet). Of
most highly contaminated
nuscle.
n 9 samples of fish tissues
954 and 12 February 1955,
, to 0.9% of the total radio-
(0.9%) was found in shark
(Table VII).

ATION RATE PER GRAMME
NIWETOK DURING 1954—1955

Percentage Sr^{90}—Y^{90}	Total d/m/g wet weight
0.08	3
0.02	64
0.1	1,500**
0.9	4,900**
0.01	500**
0.01	3,700
0.02	160,000
0.02	83
0.03	1,400

-exchange separations on
id Sr^{90}, although the total
et weight.
e uptake of Sr^{90} in aquatic
^{90} much more readily than
le per cent of the amount
e in sea water to salinity.
n and dilution by stable
duced uptake.
ely one to two per cent
vas retained after a period
ained on the average only
s. These animals were sub-
would probably differ from
anisms are able to concen-

trate many radioelements from relatively low levels of environmental con-
tamination, radioelements may be concentrated in the organisms to a greater
degree from a long-term than from an acute exposure.

Weapons tests are of use in the study of radioisotopes in the marine environ-
ment in that fairly large amounts of radiomaterials may be introduced into
a given area in a relatively short time and at levels so high that they may
be detected in the water and organisms for an appreciable time after cont-
amination. Because large areas are contaminated from the fallout, most
of the animals collected in the contaminated area may be considered to
have been exposed to a chronic rather than an acute exposure of radioactive
material.

However, the marine organisms analysed by LOWMAN, PALUMBO
and SOUTH (9), WELANDER (30), and SOUTH (24) had been subjected
to relatively long-term exposures of Sr^{90}, and the observed levels were not
significantly different from those reported by NAGASAWA et al (25) nor
HIYAMA (28) for fish collected in the open sea in the vicinity of the Marshall
Islands. Thus Sr^{90} is probably not concentrated to any extent by most marine
organisms, even under conditions of chronic exposure.

In laboratory experiments utilizing radioactive strontium under conditions
approaching chronic exposure, CHIPMAN (15) observed that Sr^{90} in Artemia
larvae reached a steady state at about 0.7 that of the sea-water,
and BOROUGHS, TOWNSLEY and HIATT (14) reported a value of about
0.3 in Tilapia. In fish bone and scales, however, the concentration factors
may be higher. In the croaker (Micropogon undulatus), concentration factors
for strontium in vertebrae and scales were 2.5 and 2.1 respectively those
of sea-water.

Little is known about the uptake and retention of radioisotopes by symbiotic
organisms. On the reefs at the Eniwetok Test Site the corals and their asso-
ciated algae are symbiotic and comprise the major part of the biotic mass.
The effect of the symbiotic relationship between coral and algae on the uptake
and retention of radioisotopes by these two organisms is not known.

Studies on uptake of radioisotopes by symbiotic organisms have been
made, however, on two related genera of tropical clams. Both Tridacna,
the giant clam, and Hippopus, the horse clam, contain symbiotic algae, zoo-
xanthellae, within their phagocytic blood cells. The algae are "farmed"
in great numbers in a mantle that is well developed in both species but more
highly in Tridacna. Both genera of clams are further modified for their speci-
alized mode of feeding by having the abnormally large kidneys required to get
rid of the waste material from the digested zooxanthellae. Because the digestive
tract contains a constriction which blocks passage of all except microscopic
particles and there is thus no faecal excretion, at least part of the waste
materials are stored in the kidneys, mostly in the form of spherical con-
cretions. Tridacna "is the supreme example of exploitation of associated
algae by animals" (30).

Tridacna and Hippopus are the only animals known to the author in which
radioactive cations are deposited in one organ and radioactive anions in
another. In Tridacna (Table VIII) 95% of the total radioactivity in the kidney
was from cations, 85% from Co$^{57, 58, 60}$, and 12% from Mn^{54}. A total of
2.5% of the activity was contributed by the anions Ru^{106}—Rh^{106} and
Zr^{95}—Nb^{95}. The radioisotopic content of the kidney of Hippopus was similar
to that of Tridacna except that small amounts of Zn^{65} were present.

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TABLE VIII

PERCENTAGE OF RADIOACTIVITY CONTRIBUTED BY DIFFERENT ISOTOPES IN TRIDACNA VISCERAL MASS AND KIDNEY, AND IN THE LIGHT AND HEAVY FRACTIONS OF THE KIDNEY

	Tridacna visceral mass % of activity	Tridacna kidney % of activity
$Ru^{106}-Rh^{106}$	72.0	2.2
$Zr^{95}-Nb^{95}$	22.0	0.3
$Ce^{144}-Pr^{144}$	0	0
$Sr^{90}-Y^{90}$	0	0
$Cs^{137}-Ba^{137m}$	0	0
Mn^{54}	0	11.6
Fe^{55}	0	0
$Co^{57, 58, 60}$	6.0	85.8
Zn^{65}	0	0

	Light fraction % of activity	Heavy fraction % of activity
$Ru^{106}-Rh^{106}$	3.6	0.3
$Zr^{95}-Nb^{95}$	0.5	0.04
Mn^{54}	8.7	16.4
$Co^{57, 58, 60}$	87.0	83.0

In the visceral mass only 6% of the total radioactivity was from $Co^{57, 58, 60}$. $Ru^{106}-Rh^{106}$ (72%) and $Zr^{95}-Nb^{95}$ (22%) accounted for the major part of the activity.

The deposition of radioisotopes in the concretions of the kidney was studied by separating the kidney into a light fraction and a heavy fraction through homogenization and centrifugation. The heavy fraction contained more concretions than the light fraction. In the heavy fraction, Mn^{54} contributed approximately twice as much and the radioactive anions $Ru^{106}-Rh^{106}$, $Zr^{95}-Nb^{95}$ one-tenth as much of the total radioactivity as they did in the light fraction.

The deposition of radioactive cations in one organ and radioactive anions in another appears to be related to the metabolism and retention of stable sodium, potassium, calcium, magnesium, phosphate, bromide, and chloride in these organs. Metabolism of the heavy metals probably also exerts an effect on the deposition pattern of radioelements.

The levels of stable elements in the organs tend to parallel those of the radioelements. Zn^{65} was found in *Hippopus* kidney but not in that of *Tridacna*. Stable zinc is present in *Hippopus* kidney at a level 17 times that in *Tridacna* kidney. Mn^{54} contributed twice the percentage of the total radioactivity in the heavy fraction of *Tridacna* kidney as it did in the light fraction, and stable manganese was present in the heavy fraction at a level six times that in the light fraction. The amount of phosphate in the kidney fractions paralleled the levels of manganese, which is probably stored in the concretions as a manganese-phosphate-protein complex.

The tridacnid clams differ from those in which symbiosis does not occur in that almost none of the accumulated ions are lost to the environment

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D BY DIFFERENT ISOTOPES IN
IN THE LIGHT AND HEAVY
DNEY

sceral mass ctivity	Tridacna kidney % of activity
.0	2.2
.0	0.3
)	0
)	0
)	0
)	11.6
)	0
.0	85.8
)	0

raction etivity	Heavy fraction % of activity
.6	0.3
.5	0.04
.7	16.4
.0	83.0

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ch symbiosis does not occur
are lost to the environment

in the symbiotic forms (31). Conversely the tridacnids, especially *Tridacna*, are able to reject almost completely at least one ion, zinc, which is taken up in large amounts by other marine organisms.

Summary

Two physical factors which control the distribution and the availability in time of radioactive contamination in the sea are the distribution on fallout particles of the individual radioelements, and the half-lives of the radionuclides.

Two principal factors control the geographical distribution of radioactive contamination in the sea: oceanographic effects and gravity. The dominant oceanographic influence is that of ocean currents, and the degree to which these affect the body of contamination depends upon surface winds, magnitude of subsurface currents, vertical and horizontal density gradients, and the size of the contaminated area.

The horizontal dispersion of the radioactive contamination is much greater than the vertical dispersion. The rate of drift of radioactivity is about equal to that of the ocean currents, although the vertical migration of plankton may reduce the rate of movement with respect to that of the surface currents.

Near the Eniwetok Test Site most of the contamination was carried by the surface currents at depths less than 25 m in the direction of the wind. Contamination in the deeper water moved in the direction of the north equatorial current.

In an area of contamination near Eniwetok Atoll the radioactive material associated with the soluble-colloidal and the particulate fractions moved down through the mixed layer at a rate of 2.5 m per hour, and at 48 hours most of the radioactivity was concentrated at the upper edge of the thermocline. At depths of 100 and 300 m the percentage of total activity associated with particulate material increased at an almost exponential rate during a period of 48 hours following contamination.

In the area of the Marshall Islands and west to Guam, homogeneous vertical dispersion of the radioactive contamination throughout the mixed layer did not occur within a period of six to eight weeks following the introduction of contamination into the sea.

The uptake of radioisotopes by plankton appears to depend upon the physical form of the individual radioelements in sea water. Radioactive iodine, strontium and caesium are deposited in the soluble form and are not concentrated by planktonic organisms. Radioelements in the particulate form are concentrated by plankton.

Factors that determine whether or not a radioelement is present in the particulate form include solubility products, coprecipitation, and adsorption to inorganic and organic detritus or to micro-organisms.

The uptake of a given radioisotope by plankton depends upon the amount of isotope dilution and competition by chemically similar elements. Also, organisms may actively discriminate against certain radioelements. In the sea, discrimination against the uptake of $Sr^{90}-Y^{90}$ occurs in most organisms.

Several biological factors affect the uptake and retention of radioisotopes by marine plants and animals. The biomass at each trophic level determines the total amount of radioactive material contained within that level. Because the efficiency of conversion is low between ascending trophic levels, the

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higher the level the lower the total amount of radioelements contained therein. Reproductive rates, average life spans, and average size of the organisms comprising each trophic level modify the total accumulation of radioactive material in that level.

The initial stage in the uptake of radioactive elements by nannoplankton probably occurs by adsorption of the material on to the surface of the organism by means of ionic or covalent linkages.

In plankton collected within 48 hours after contamination, the principal isotopes were Np^{239}, Mo^{99}—Tc^{99m}, Te^{132}—I^{132}, U^{237}, $Ru^{103, 105, 106}$—$Rh^{103, 105, 106}$, $Ce^{141, 144}$—$Pr^{141, 144}$, Ba^{140}—La^{140}, and Zr^{95}—Nb^{95}. At one week after contamination the principal isotopes in descending order of abundance were Co^{58}, Ba^{140}—La^{140}, $Fe^{55, 59}$, Co^{57}, Zr^{95}—Nb^{95}, $Ru^{103, 105, 106}$—$Rh^{103, 105, 106}$, Co^{60}, Zn^{65}, Np^{239}, and U^{237}. At six weeks Zn^{65}, Fe^{55}, Zr^{95}—Nb^{95}, Co^{58}, Co^{57}, Ce^{144}—Pr^{144}, Ru^{106}—Rh^{106}, and Co^{60} were present in the plankton in the above order of occurrence.

The initial uptake of radioelements by plankton included, almost entirely, radioactive anions. Within a week, however, radioactive transition elements of the first series (cations) accounted for the major part of the radionuclides in the plankton.

W^{185}, which has no known metabolic function, was not retained by plankton, although the initial contamination of these organisms by radiotungsten was high.

The second stage of accumulation of radioactive contamination by marine plankton occurs through the filter-feeders, which probably ingest both inorganic and organic particles.

In the contaminated area near the Eniwetok Test Site, only a small fraction of the total radioactivity in the sea was associated with the plankton. In four surveys taken up to 8 weeks following contamination, the average total activity in the water was 40,000 times that in the contained micro-macroplankton. However, the radioactivity per unit volume of plankton was much higher than that for an equal volume of water.

In the fish samples collected in the lagoons at Eniwetok and Ailinginae Atolls, Fe^{55}, Zn^{65}, $Co^{57, 58, 60}$, Mn^{54} and Zr^{95}—Nb^{95} accounted for the major part of the radioactivity. In tuna fish samples taken in the open sea near the test site, cations contributed 100 % of the activity. Zn^{65} was highest and Fe^{55} was second in abundance. $Co^{57, 58, 60}$ and Mn^{54} were present in low amount, and the low level of the cobalt isotopes suggests a rapid turnover rate for this element in marine fishes. The amount of radioactive cobalt in plankton increases, with respect to that of Zn^{65} and Fe^{55}, with time after contamination. This may be related to a rapid turnover rate for cobalt and a low rate for zinc and iron in pelagic fishes which feed indirectly upon the plankton.

The uptake of radioisotopes by symbiotic organisms was studied in two related genera of tropical clams, both of which contain symbiotic algae. One clam is more closely associated with the algae than the other, but both obtain a major part of their food through their symbionts. Waste products from digested algae are stored in the kidneys of the clams, mostly as concretions. Of the radioisotopes deposited in these organs, 95 % are cations. However, in the visceral mass 94 % of the contained radioisotopes are anions.

The amount of Zn^{65} deposited in the kidneys of the clams is inversely related to the closeness of the symbiotic association between the clam and the algae.

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of abundance were Co^{58},
—$\text{Rh}^{103, 105, 106}$, Co^{60}, Zn^{65},
^{5}, Co^{58}, Co^{57}, Ce^{144}—Pr^{144},
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