THE EFFECTS OF ATOMIC RADIATION ON OCEANOGRAPHY AND FISHERIES

National Academy of Sciences—
National Research Council

Publication 892

Given in Loving Memory of

Raymond Braislin Montgomery
Scientist, R/V Atlantis maiden voyage
2 July - 26 August, 1931
****
Woods Hole Oceanographic Institution
Physical Oceanographer
1940-1949
Non-Resident Staff
1950-1960
Visiting Committee
1962-1963
Corporation Member
1970-1980
******
Faculty, New York University
1940-1944
Faculty, Brown University
1949-1954
Faculty, Johns Hopkins University
1954-1961
Professor of Oceanography,
Johns Hopkins University
1961-1975

R B Mayer

THE BIOLOGICAL EFFECTS OF ATOMIC RADIATION

THE EFFECTS OF ATOMIC RADIATION ON OCEANOGRAPHY AND FISHERIES

Report of the
Committee on Effects of Atomic Radiation
on Oceanography and Fisheries
of the
National Academy of Sciences
Study of the Biological Effects
of Atomic Radiation

Publication No. 551
NATIONAL ACADEMY OF SCIENCES—NATIONAL RESEARCH COUNCIL
WASHINGTON, D. C.

LIBRARY OF CONGRESS CATALOG CARD NO.: 57-60049

COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION ON OCEANOGRAPHY AND FISHERIES

FOREWORD

The studies of the biological effects of atomic radiation, of which the report published in this volume is a part, were undertaken by the National Academy of Sciences in 1955. The first formal reports issuing from the study were published by the National Academy of Sciences—National Research Council in June 1956 as "The Biological Effects of Atomic Radiation—Summary Reports." These noted briefly the findings of six committees established to review broadly the status of knowledge in this field of vital importance to the welfare of man at the threshold of the atomic age. They considered the problem from the points of view of genetics, pathology, agriculture and food supplies, oceanography and fisheries, meteorology, and the disposal and dispersal of radioactive wastes.

The Academy study is a continuing one. Each of the Committees in a manner appropriate to its area of concern is pursuing its work.

The Committee on the Effects of Atomic Radiation on Oceanography and Fisheries held two meetings prior to the publication of its "Summary Report": the first on March 3-5, 1956 and the second on April 13-16, 1956. Rough drafts of most of the materials published in this volume were prepared at the second meeting. These reports, which give the detailed technical background of the committee's findings and recommendations, have been completed during the past year. Although the different chapters are signed by individual authors, all members of the committee participated in planning and outlining the materials covered. Valuable editorial assistance was given by Dr. George A. Rounsefell and Mr. Charles I. Campbell.

A similar report was prepared by the Committee on Pathologic Effects of Atomic Radiation and published in the Fall of 1956 by the NAS-NRC as Publication Number 452. The Committee on the Disposal and Dispersal of Radioactive Wastes has nearly completed a similar detailed report of its considerations.

After the publication of its Summary Report in June 1956, the Committee on the Effects of Atomic Radiation on Oceanography and Fisheries met informally with scientists from the United Kingdom on September 27 and 28, 1956. The discussions centered around the recommendations that could be made on the basis of existing knowledge and the nature of the research needed in planning disposal of radioactive waste at sea.

Members of this Committee have also participated in the preparation of a report by Unesco to the UN Scientific Committee on the Effects of Atomic Radiation, concerning the oceanic disposal of radioactive wastes.

As the use of atomic energy becomes more and more a part of our daily life it is essential that thoughtful attention in broad perspective be paid to the often subtle and perhaps profound effects of this new technology on man and his environment. The Academy study will continue to provide this review and to report its findings to the public when appropriate.

The facts upon which the study's conclusions are based result from more than two decades of research which has been sponsored by the Academy and other private organizations as well as by various government agencies. The current study has been financed by a grant from the Rockefeller Foundation. It has been greatly assisted by the generous and whole-hearted co-operation of the U. S. Atomic Energy Commission and other government agencies.

DETLEV W. BRONK,
President, National Academy of Sciences.

TABLE OF CONTENTS

GENERAL CONSIDERATIONS CONCERNING THE OCEAN AS A RECEPTACLE FOR ARTIFICIALLY RADIOACTIVE MATERIALS[1]

ROGER REVELLE and MILNER B. SCHAEFER, *Scripps Institution of Oceanography*
and
Inter-American Tropical Tuna Commission, La Jolla, California

I. INTRODUCTION

IN THIS report, we have attempted to summarize both the present knowledge and the areas of ignorance concerning the oceans that must be taken into account in considering the biological effects of radiation.

The oceans of the world furnish essential sources of food and other raw materials, vital routes of transportation, recreation, and a convenient place in which to dispose of waste materials from our industrial civilization. These different ways in which men use the sea, however, are not always compatible. The use of the sea for waste disposal, in particular, can jeopardize the other resources, and hence should be done cautiously, with due regard to the possible effects. Waste products from nuclear reactions require special care: they constitute hazards in extremely low concentrations and their deleterious properties cannot be eliminated by any chemical transformations; they can be dispersed or isolated, but they cannot be destroyed. Once they are created, we must live with them until they become inactive by natural decay, which for some isotopes requires a very long time.

Waste products from nuclear reactions arise in two ways: (1) from the slow controlled reactions involved in laboratory experimentation, in the production of materials for nuclear weapons, the production of reactor fuels, and the "burning" of fuels in power reactors; (2) from the rapid, uncontrolled reactions involved in testing of weapons or in warfare. Up to the present time, the largest quantities of fission products introduced into the aquatic environment have been from weapons tests; most of the products from controlled reactions have been isolated on the land, and only relatively small quantities have been introduced into the sea or fresh water. In the future, however, industrial nuclear wastes will present difficult disposal problems and the sea is a possible disposal site, particularly for small, densely populated nations with long sea coasts. We have, therefore, given particular attention to the long-range problems that may arise from the large-scale disposal of both high-level and low-level industrial wastes, as well as to the effects of weapons tests.

Among the variety of questions generated by the introduction of radioactive materials into the sea, there are few to which we can give precise answers. We can, however, provide conservative answers to many of them, which can serve as a basis of action pending the results of detailed experimental studies. The large areas of uncertainty respecting the physical, chemical, and biological processes in the sea lead to restrictions on what can now be regarded as safe practices. These will probably prove too severe when we have obtained greater knowledge. It is urgent that the research required to formulate more precise answers should be vigorously pursued. Fortunately, the use of radioactive isotopes is one excellent means of acquiring the needed information, and the quantities of these isotopes required for pertinent experiments are well within limits of safety. Moderate quantities of the very waste products we are concerned with can, therefore, provide one means of attacking the unsolved scientific problems.

II. THE NATURE OF THE OCEAN AND ITS CONTAINED ORGANISMS

The ocean basins cover 361×10^6 square kilometers and have an average depth of 3,800 meters, giving a total volume of 1.37×10^9 cubic kilometers. They are characteristically bordered by a continental shelf, which slopes gently out to a depth of about 200 meters. Inside it is a steeper slope extending down to the

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 901.

deep sea floor with depths of 4,000 meters or more. The average width of the continental shelf is about 30 miles, varying from almost nothing off mountainous coasts, such as the West Coast of South America, to several hundred miles in the China Sea. The shelf is not everywhere smooth, but is often intersected by submarine valleys and canyons. In the deep ocean basins there are high mountains and long, deep trenches, features larger than any on land. Some of the deeper parts are isolated by submarine ridges which restrict the exchange of water between adjacent areas.

The waters of the oceans are stratified. Within a relatively thin layer at the surface, varying in thickness in different places but averaging about 75 meters, vertical mixing caused by winds is fairly rapid and complete. In consequence, the temperature, salinity and density are nearly uniform from top to bottom. Relatively fast wind-driven currents exist in this upper mixed layer; these are the "surface" currents of the oceans depicted on many charts. Here also the horizontal mixing is relatively rapid. The mixed layer is the region of the sea in which most of man's activity takes place.

Below the mixed layer is a zone within which the temperature decreases and the density increases rapidly with depth. This thermocline, or pycnocline, separates the surface mixed layer from the layers of intermediate and deep water, the latter extending to the bottom, within which there are gentle gradients of decreasing temperature and increasing salinity and density with depth. Vertical movement in the intermediate and deep layers is much slower than in the mixed layer, and horizontal currents are more sluggish. The strong density gradient across the pycnocline tends to inhibit physical transport across it, because work is required to move water vertically in either direction, and thus the pycnocline acts as a partial barrier between the mixed layer and the lower layers. There is, however, some interchange of both living and non-living elements; indeed the continued existence of some marine resources depends on such interchange.

MARINE RESOURCES

Living resources

The most important extractive industry based on the resources of the sea is the harvesting of its living resources.

On land the cycle of life is relatively simple; we may describe it in four figurative stages. First is the grass, which by a subtle and complex chemistry captures the energy of sunlight and builds organic matter. Sheep and cows live on the grass; tigers and men eat them. The cycle is closed by bacteria, which decompose the dead bodies and the excreta of all living creatures, making their constituent substances again available as building materials for the plants. In the sea, the cycle is longer. Instead of grass there are the tiny floating plants called phytoplankton; in place of cows, the zooplankton animals that eat the plants are small crustaceans, no bigger than the head of a pin. Many kinds of tigers eat the cows, but they are mostly also zooplankton, only a fraction of an inch in length. Other intermediate flesh-eaters exist between them and the fishes of our ocean harvest. Because every link in this long food chain is inefficient, we reap from the sea only a small fraction of its organic production.

Other characteristics of the ocean also tend to limit the harvest as compared to that from the land. One is its giant size; more than 70 per cent of all the sunlight that penetrates the atmosphere falls on the sea; moreover, this sunlight can act throughout the top 20 to 100 meters, thus the living space for plants and animals is far greater than on land. This great areal extent and volume, combined with the fluidity of the oceans, results in a low concentration of organisms per unit volume and therefore inefficiency in harvesting.

On land, the standing crop of plants and animals is of the same order of magnitude as the amount of organic production per year, while in the ocean the crop is very small, compared to the production, because of rapid turnover. The average rate of organic production per unit area is probably about the same on land and in the sea, but the efficiency of harvesting depends more on the size of the crop than on the total amount of organic matter produced.

The plants of the sea, on which all other living things depend, grow only in the waters near the surface where bright sunlight penetrates. These waters differ widely in fertility. Like the land, the ocean has its green pastures where life flourishes in abundance, and its deserts where a few poor plants and animals barely survive.

The fertility of the land depends on four things: water, temperature, intensity of sunlight, and available plant nutrients—substances

that usually occur in very small amounts but are essential for plant growth. In the sea, water is, of course, always abundant; the plants are well adapted to the narrow range of temperature; the intensity of sunlight determines the length of the growing season and the depth of growth, but usually not the differences in fertility. These depend only on the plant nutrients in the waters near the surface. As in any well-worked soil on land, the nutrients in the waters must be replenished each year. They are continually depleted by the slow sinking of plant and animal remains from the brightly lighted near-surface layers into the dark waters of the depths.

Men plow the soil to restore its fertility; the fertility of the sea is restored when nutrient-rich deeper waters are brought up near the surface. The "plowing" of the sea is accomplished in three ways. In some regions winds drive the surface waters away from the coast or away from an internal boundary, and nutrient-rich waters well up from mid-depths. In other areas, the surface waters are cooled near to freezing in the winter, become heavy and sink, and mix with the deep waters. Elsewhere, violent mixing occurs along the boundaries between ocean currents, and deeper waters are thereby brought into the brightly lighted zone.

The influx of nutrients to the upper layer, and the corresponding loss from this layer by sinking of plant and animal remains, do not directly involve the deep waters. Upwelling and vertical mixing take place only in the upper few hundred meters. The exchange between these mid-depths and the abyssal deep is a very much slower process, of the scale of hundreds of years.

Most of the commercially important marine organisms are harvested in coastal waters or in offshore waters not very far from land. Several factors are involved: (1) Profitable fisheries can be conducted more easily near ports and harbors; (2) the coastal waters are of high fertility, because of greater upwelling and turbulent mixing and the ease of replenishment of plant nutrients from the shallow sea floor, and perhaps also because of the supply of nutrients and organic detritus from land; (3) the standing crop of plants and animals attached to or living on the bottom in coastal areas is large, relative to the total organic production.

None of the animals of the great depths are the objects of a commercial fishery. Even the truly pelagic, high seas fisheries, such as the great offshore fisheries for tuna, herring, redfish and whales, harvest animals that live primarily in the surface layer. Some of these animals, however, do much of their feeding in the deeper layers. The sperm whales, for example, feed on deep-sea cephalopods at great depths. Moreover, much of the food for commercially harvested organisms consists of small animals, including crustaceans, squids, and fishes, that perform vertical diurnal migrations from several hundred meters depth to the surface.

The sea fisheries produce about 25 million metric tons per year of fishes and marine invertebrates, in addition to about 4 million tons of whales. The great bulk of the harvest is taken, at present, from the waters of the northern hemisphere, despite the fact that the southern oceans constitute 57 per cent of the world's sea area. The following table indicates the production in 1954 by latitude zones:

TABLE 1 HARVEST OF FISHES AND MARINE INVERTEBRATES IN 1954, BY LATITUDE ZONES (FROM FAO, 1957)

Zone	Millions of metric tons	%
Arctic region	1.2	5
Northern hemisphere-temperate zone	17.5	72
Tropical zone	4.1	17
Southern hemisphere-temperate zone	1.4	6
Antarctic regions	0*	0*

* About 4 million tons of whales were taken in the Antarctic, but few fish or marine invertebrates.

The disproportionately large yield in the northern hemisphere is related to three factors: (1) Human populations are heavily concentrated there; (2) the major fishing nations are the industrialized maritime nations, which are mostly located in the north; (3) except for some of the fisheries for tuna, salmon, herring, and whales, the important fisheries are located in the relatively shallow areas along the continents, and the extent of these areas is much greater in the northern than in the southern hemisphere.

The sessile algae of shallow coasts are also the object of important industries in Japan, the United States, the United Kingdom, Norway, and some other countries. Some of these plants are used directly for human consumption, while

others are employed indirectly in pharmaceutical and food products.

Petroleum and natural gas

It is estimated that about 30 million cubic meters of possible oil-bearing sediments underlie the 11.8 million square miles of the submerged continental shelves. These sediments contain some 400 billion barrels of recoverable crude oil.

Exploitation of these deposits of petroleum and the associated natural gas has commenced in the waters of the Gulf of Mexico; intensive geophysical prospecting has been conducted offshore from California and in the Persian Gulf. It may be expected that this source of fossil fuels will be extensively utilized in the near future. The resource is confined to the subsoil of the marginal seas, since only there do we find oil-bearing sediments.

Minerals

Extraction of sea salt for sodium chloride is an ancient industry, and is now highly developed also for production of sodium sulfate, potassium chloride, and magnesium chloride. Bromine is extracted directly from sea water for the manufacture of ethylene dibromide. Magnesium metal has been produced commercially from seawater by chemical and electrolytical procedures for nearly two decades.

All of these industries use sea water taken from near the surface at the shore but the quantity of water utilized is insignificant. For example, a single cubic kilometer of sea water contains over a million tons of magnesium, about five times the peak world annual production of this metal.

The floor of the deep sea is known to contain low-grade deposits of cobalt, nickel and copper (0.1 to 0.7 per cent by weight of the metals) associated with deposits of iron and manganese. The problems of mining these materials, in the face of the great depths and pressure, have not been solved, and they certainly will not soon be economically useable.

Ocean transportation

Long-distance transportation of large cargos by sea is the indispensable basis of international commerce. The economy of the United States and of other industrial nations is in large part dependent on the sea-borne commerce that flows through the seaports.

Contamination of the sea by nuclear wastes will certainly not present a hazard to shipping, since acceptable levels of such materials in the surface layer of the sea will be limited by other considerations (such as the effects on the fisheries) to much lower levels than would constitute a hazard to ships' personnel. On the other hand, it is almost certain that nuclear power plants will be extensively used in merchant vessels; they are already in use in naval craft.

Serious hazards may arise in confined waters from collisions in which the reactor is damaged and the fuel elements with their contained fission products are lost in the water. Suppose for example that a 50,000 kilowatt reactor (probably fairly typical for a large fast freighter) has been in service without refueling for one year on a ship that has spent half its time under way. Approximately 10 kilograms of fissionable material will have been used up and the total amount of fission products will be approximately 10^7 curies. If, owing to a collision, the reactor is lost in a harbor, say 8 miles long by 3 miles wide by 50 feet deep, and the fission products become uniformly distributed, the water in the harbor would contain 10^{-2} curies per cubic meter giving an almost constant radiation dose of about 0.5 r per day on the surface. Dock pilings, ship bottoms and other structures covered with fouling organisms would accumulate a much higher level of radioactivity, and local concentration in the water might be extremely high.

Recreation

For coastal populations in the temperate, subtropical, and tropical regions, the sea and its contents provide healthful sports and satisfaction of men's curiosity and their desire for beauty. Boating, swimming, sport fishing, and other recreations are engaged in by millions of people, and are the basis of tourist and service industries of very considerable monetary value.

Waste disposal

Disposal of domestic sewage and industrial wastes is often conveniently accomplished near coastal population centers by running them into the sea. The large volume and rapid mixing of the ocean waters dilute the wastes, and the bac-

teria in the sea break down the organic constituents. Unless care is exercised, however, this discharge into inshore sea areas may be deleterious to other resources. Dumping of excess volumes of sewage and industrial wastes, without proper regard to the local characteristics of the sea bottom, currents, and other factors, has already resulted in ruining some harbors and beaches for recreation, damage to the living resources of adjacent areas, and even serious problems of corrosion to ships.

The use of the sea for the disposal of atomic wastes has, fortunately, been so far approached with great caution and with due regard to the possible hazards. The problems, because of the dangerous character of small amounts of atomic wastes, are of a different order of magnitude than those of the disposal of other kinds of wastes.

III. Potential Hazards From Radioactive Materials

Direct hazards

A direct hazard to human beings from radiation may exist if the levels of radiation in the environment are sufficiently high.

The natural radioactivity of the sea is very much lower than that of the land. According to Folsom and Harley (Chapter 2 of this report), a man in a boat or ship receives only about half a millirad per year from the radio isotopes in the sea, compared with about 23 millirads per year from sedimentary rock or 90 millirads per year from granite. Thus, it would be necessary to increase the radioactivity of the sea many fold to equal the radiation that man normally receives from the land on which he lives. Due to the rather rapid mixing in the upper layers of the sea, and to its very large volume, even large quantities of activity introduced at the surface in the open sea become sufficiently dispersed to constitute no direct hazard after a relatively short time, as has been shown by the dispersion of the activity resulting from weapons tests in the Pacific. If the direct hazard were the only consideration, sea disposal of radioactive wastes would give rise to difficulties only in small areas near the disposal sites.

Some radioactive wastes have been disposed of in the sea by placing them in containers designed to sink to the sea bottom. In this way, the wastes are isolated and not dispersed by the ocean currents. Direct hazards could arise if the containers in some manner were to come into contact with humans, such as through accidental recovery during fishing or salvage operations or if, through improper design, the containers were to float to the surface and come ashore.

Indirect hazards

The most serious potential hazards to human beings from the introduction of radioactive products into the marine environment are those that may arise through the uptake of radio isotopes by organisms used for human food. There are several reasons why these indirect hazards are more critical than the direct hazards: (1) The radiation received from a given quantity of an isotope ingested as food is much greater than the dose from the same quantity in the external environment; (2) many elements, including some of those having radioactive isotopes resulting from nuclear reactions, are concentrated by factors up to several thousand by the organisms in the sea; (3) the vertical and horizontal migrations of organisms can result in transport of radioactive elements and thereby cause distributions different from those that would exist under the influence of physical factors alone; for example, certain elements may be carried from the depths of the sea into the upper mixed layer in greater amounts than could be transported by the physical circulation.

It is quite certain that the indirect hazard to man through danger of contamination of food from the sea will require limiting the permissible concentration of radioactive elements in the oceans to levels below those at which there is any direct hazard. Any part of the sea in which the contamination does not cause dangerous concentrations of radioactive elements in man's food organisms will be safe for man to live over or in.

A reduction of the harvestable living resources of the sea could conceivably occur through the effects of atomic radiations on the organisms that are the objects of fisheries, or on their food. This might result from mortality induced by somatic effects, or from genetic changes. There is no conclusive evidence that any of the living marine resources have yet suffered from either of these, and they are not likely to be undesirably influenced at radiation levels safe from other standpoints. The knowl-

edge of radiation effects on marine organisms is, however, inadequate for firm conclusions.

Pollution in general

The introduction of atomic wastes into the aquatic environment is but one aspect of the general problem of pollution.

Man's record with respect to pollution of lakes, streams, and parts of the sea by sewage and industrial wastes has not been good. In many places, the waters have been ruined for recreation and useful living resources have been destroyed or made unfit for human consumption. This unhappy record results from two factors: (1) the insidious nature of pollution of the aquatic environment, and (2) the fact that the waters and most of their resources are not private property, but are the common property of a large community (in the case of the high seas, the whole world); what is everyone's business often becomes no one's business.

The ruin of an aquatic resource by pollution seldom has been rapid. Quantities of waste products, at first very small, increase year by year until finally the concentrations become so large as to have obvious deleterious effects. For example, in the depletion of oxygen by organic wastes, sharp critical levels of tolerance of low oxygen content exist for some of the living resources, so that there is little adverse effect until a critical concentration of pollutant is reached, whereupon catastrophic mortality occurs. In other cases, the effects are more or less proportional to the concentrations. The destruction of a resource may then proceed gradually and it may not even be clear whether the pollutant has, indeed, been the cause rather than some other environmental change. For these reasons, it is necessary that the introduction of waste materials of any kind into the aquatic environment be carefully monitored, so that the effects may be detected before they become serious. Unfortunately, such monitoring is seldom the concern of those who produce the pollutants.

The record of the control and monitoring of the disposal of atomic pollutants has, so far, been excellent. We are, however, at the threshold of a tremendous growth of the atomic energy industry, and it behooves mankind to make sure that as much caution is exercised in the future as in the past.

Ordinary pollutants in sewage and industrial wastes are rapidly neutralized by the chemical and biological processes in the sea, and when effects of pollution are detected they can be rather quickly reversed by the cessation of introduction of the waste. A number of the radio isotopes, on the other hand, are very long-lived. Having reached harmful concentrations in the sea, they will diminish only by very slow decay, so that the effect of any serious pollution is not reversible. For this reason, the *prevention* of atomic pollution is of paramount importance.

URGENCY OF THE PROBLEM

Estimates of the rate of economic development of nuclear power vary widely. This source of power is already competitive with conventional sources in some places, and research on reactor development with consequent reductions in cost is proceeding rapidly. Thus, we can expect that very large quantities of nuclear power will be generated in the quite near future, even though the relative urgency of nuclear power requirements differs greatly in different countries. In countries with high costs from conventional (fossil) fuels there is encouragement to proceed immediately with the commercial construction of reactors of proved design. In such countries as the United States, where conventional power costs are low, major efforts are being devoted to experimental construction of new types of reactors that hold promise of economical operation in the future.

One megawatt-year of heat produced by a nuclear reactor results in 365 grams of fission products. The Committee on Disposal and Dispersal of Radioactive Wastes, also a part of the National Academy of Sciences' study of the biological effects of atomic radiation (1956), estimates that by 1965 the United States will be generating about 11,000 megawatts of reactor heat, some 20 per cent of which will be for naval vessels. This will result in the production of about 4 tons per year of fission products. According to recent statements of government officials, reported in *Nucleonics* (1957), the United Kingdom has a 1965 target of 6,000 megawatts of electricity from Calder Hall-type reactors; "Euratom" has a goal of 15,000 megawatts by 1967, and Japan will produce 1,000 megawatts by 1965 and 10,000 megawatts by 1975. If the reactors are of 25 per cent efficiency in conversion of heat to electricity (the Calder Hall reactor has a net thermal efficiency of 21.5 per cent, *Nucleonics* 1956), for each

1,000 megawatts of electrical power there will be produced 1.46 tons per year of fission products. Thus, the fission products from the foregoing programs will amount to: United Kingdom 8.8 tons, "Euratom" 21.9 tons, Japan 1.5 to 14.6 tons.

If we further assume that all other areas of the world will in the next ten years develop nuclear power equal to the sum of that generated in the United States, Japan, the United Kingdom, and "Euratom," there will be a total of some 80 tons per year of fission products. This represents, after 100 days' cooling, according to the values given by Renn (see Chapter 1, Tables 2 and 3), 3.9×10^4 megacuries of beta radiation and 2.5×10^4 megacuries of gamma radiation, or over $\frac{1}{10}$ of the total natural radioactivity of all the oceans (Revelle, Folsom, Goldberg and Isaacs 1955). The annual production of the isotope of greatest long-range hazard, strontium 90, will be 200 megacuries. Craig (Chapter 3) has shown that a thousand tons of fission products per year would result from a 2.7-fold increase in the present world energy consumption of about five million megawatts, if 10 per cent of this energy were derived from the heat of nuclear fission at 50 per cent efficiency. World energy consumption is now doubling once every thirty years and a 2.7-fold increase would be expected by about the year 2000. An annual production of a thousand tons of fission products corresponds to an equilibrium quantity of 7.7×10^5 megacuries of radiation or about 1.6 times the total natural radioactivity of the oceans. The equilibrium amount of strontium 90, plus its daughter yttrium 90, would be 2.2×10^5 megacuries. Carritt and Harley (Chapter 6) have made calculations based on an annual production of 4,000 tons of fission products, corresponding to two million megawatts per year of nuclear heat production from fission. If no new sources of power, such as thermonuclear reactions, become available, this production would be expected in the very early part of the twenty-first century because of the limited world fossil fuel reserves.

Our knowledge of just what share of these fission products can be safely introduced into the oceans is woefully incomplete because we simply do not know enough about the physical, chemical, and biological processes. If the sea is to be seriously considered as a dumping ground for any large fraction of the fission products that will be produced even within the next ten years, it is urgently necessary to learn enough about these processes to provide a basis for engineering estimates.

As shown in the several chapters of this report, the necessary information can be obtained only by extensive fundamental research. In the next decade we should attempt to learn far more about the ocean and its contents than has been learned since modern oceanography began 80 years ago.

Some of the required investigations of physical, chemical, and biological processes involve the employment of naturally occurring or experimentally introduced radioactive tracers. Pollution of the seas by the dumping of atomic wastes, even at levels that are "safe" from the standpoint of human health hazards, will make such experiments progressively more difficult because the presence of introduced pollutants will add an unknown background variability. The sooner the work can be commenced and the cleaner our oceanic laboratory, the more precise will be the experimental results. At the very least, it is urgent that the details of any interim introductions of radio isotopes be carefully documented, so that researchers can take account of them in their investigations.

INTERNATIONAL IMPLICATIONS

The oceans and their resources cannot be separated into isolated compartments; what happens in one area of the sea ultimately affects all of it. Moreover, the greater part of the oceans and their contained resources are the common property of all nations. Even the relatively narrow territorial seas are amenable only to juridical and not physical control; no nation can effectively modify the natural interchange of the biological and physical contents of its territorial sea with those of the high seas or of the territorial seas of other nations. The continuity of the oceans, and their status as international common property require that the oceanic disposal of radioactive wastes be treated as a world problem.

It is, first of all, urgent that the nations of the earth formulate agreements for the safe oceanic disposal of atomic wastes, based on existing scientific knowledge. Second, because of the vastness, complexity, and immediacy of the underlying scientific problems, it is important that pertinent oceanographic research be intensified on a world-wide basis. Third, from the

standpoint both of research and of proper control of this new kind of pollution, careful records should be maintained of the kinds, quantities, and physical and chemical states of all radio isotopes introduced into the seas, together with detailed data on locations, depths and modes of introduction. This can probably best be done by national agencies reporting to an international records center.

Although we are urgently concerned with preventing possible deleterious effects of atomic wastes, atomic radiations can also be of benefit. Large-scale experiments employing radioactive isotopes might contribute importantly to our knowledge of the flux of materials through the food chains from the phytoplankton to the harvestable fishes, invertebrates, and whales (Schaefer, Chapter 13 of this report). Such knowledge will not only make possible assessment of the ocean's potential for providing food to mankind, but is a basic prerequisite for the effective conservation of marine populations, to permit maximum harvests to be taken year after year. Other experiments using radioactive tracers could lead to improved knowledge of the processes of circulation and mixing in the sea (Folsom and Vine, Chapter 12; Craig, Chapter 11). In both types of experiments, inter-

TABLE 2 Elements in Solution in Sea Water (Except Dissolved Gases)[1, 2]

Element	mg/kg Cl = 19.00‰	Total in oceans (tons)	Natural activities		
			Nuclide	Total (tons)	Curies
Chlorine	18,980	2.66×10^{16}			
Sodium	10,561	1.48×10^{16}			
Magnesium	1,272	1.78×10^{15}			
Sulfur	884	1.23×10^{15}			
Calcium	400	5.6×10^{14}			
Potassium	380	5.3×10^{14}	K^{40}	6.3×10^{10}	4.6×10^{11}
Bromine	65	9.1×10^{13}			
Carbon	28	3.9×10^{13}	C^{14}	56	2.7×10^{8}
Strontium	13	1.8×10^{13}			
Boron	4.6	6.4×10^{12}			
Silicon	0.02 –4.0	$0.028–5.6 \times 10^{12}$			
Fluorine	1.4	2×10^{12}			
Nitrogen (comp.)	0.01 –0.7	$0.14 –9.8 \times 10^{11}$			
Aluminum	0.5	7×10^{11}			
Rubidium	0.2	2.8×10^{11}	Rb^{87}	1.18×10^{11}	8.4×10^{9}
Lithium	0.1	1.4×10^{11}			
Phosphorus	0.001–0.1	$0.014–1.4 \times 10^{11}$			
Barium	0.05	7×10^{10}			
Iodine	0.05	7×10^{10}			
Arsenic	0.01 –0.02	$1.4 –2.8 \times 10^{10}$			
Iron	0.002–0.02	$0.28 –2.8 \times 10^{10}$			
Manganese	0.001–0.01	$0.14 –1.4 \times 10^{10}$			
Copper	0.001–0.01	$0.14 –1.4 \times 10^{10}$			
Zinc	0.005	7×10^{9}			
Lead	0.004	5.6×10^{9}			
Selenium	0.004	5.6×10^{9}			
Cesium	0.002	2.8×10^{9}			
Uranium	0.0015	2.1×10^{9}	U^{238}	2.8×10^{9}	3.8×10^{9}
Molybdenum	0.0005	7×10^{8}	U^{235}	2.1×10^{7}	1.1×10^{8}
Thorium	<0.0005	$<7 \times 10^{8}$	Th^{232}	1.4×10^{7}	8×10^{6}
Cerium	0.0004	5.6×10^{8}			
Silver	0.0003	4.2×10^{8}			
Vanadium	0.0003	4.2×10^{8}			
Lanthanum	0.0003	4.2×10^{8}			
Yttrium	0.0003	4.2×10^{8}			
Nickel	0.0001	1.4×10^{7}			
Scandium	0.00004	5.6×10^{7}			
Mercury	0.00003	4.2×10^{7}			
Gold	0.000006	8.4×10^{6}			
Radium	$0.2–3 \times 10^{-10}$	28 –420	Ra^{226}	4.2×10^{2}	1.1×10^{9}

[1] Sverdrup, H. U., M. W. Johnson, and R. H. Fleming, OCEANS (1942).
[2] Revelle, R., T. R. Folsom, E. D. Goldberg, and J. D. Isaacs (1955).

national scientific cooperation will often be essential for optimum results.

IV. Chemical Processes and Radioactive Materials

Elements in sea water

Sea water is a solution of a large number of dissolved chemicals containing small amounts of suspended matter of organic and inorganic origin. The ratios of the more abundant elements are very nearly constant, despite variations in absolute concentrations in different parts of the sea. Lower than average absolute amounts are encountered in coastal areas and near river mouths, while higher amounts are encountered in areas of high evaporation, such as the Red Sea. Vertical variations are usually small; in general, in the open ocean in mid-latitudes, the quantity of dissolved materials, measured by the *salinity,* first decreases slightly with depth, then increases slowly in the deep water.

Table 2 (from Carritt and Harley, Chapter 6) shows the concentrations of some of the elements in solution in sea water at a chlorinity of 19.00‰, which is near average for the sea, and the total amounts in the ocean as a whole. Also shown are the total amounts and total radioactivity of the principal naturally occurring radio isotopes. In addition to the listed elements, there are variable amounts of dissolved gases, including nitrogen, oxygen, and the noble gases. A range of values is given for some elements present in small quantities, such as nitrogen, phosphorus, silicon, iron, and copper. These are substances necessary for living organisms, and the inorganic phases may be reduced to nearly zero in surface waters where they have been almost completely removed by organic uptake.

Behavior of introduced materials

A number of things can happen to materials introduced into the sea either in solution or as particles. The particles may go into solution. The dissolved substances may be precipitated as particles of colloidal or larger size either by coprecipitation with other elements, by sorption on organic or inorganic particles already present in the sea, or by interaction with other sea water constituents. Both dissolved materials and particles may be ingested by organisms and enter into the biochemical cycles.

Particles in the sea are continually removed by settling out on the bottom. The rates of settling depend on the size and density of the particles, as modified by various physical and biological factors.

Normal removal of elements from sea water

The results of geochemical studies provide very approximate estimates of the fractions of some elements supplied to the ocean that are eventually removed from solution. In Table 3

TABLE 3 Geochemical Balance of Some Elements in Sea Water (From Goldschmidt, Quoted in Rankama and Sahama, 1950, Table 16.19)

Element	Total supplied (ppm)	Amount present in ocean (ppm)	Transfer percentage
Na	16,980	10,560	62
K	15,540	380	2.4
Rb	186	0.2	0.1
Ca	21,780	400	1.8
Sr	180	13	7.2
Ba	150	0.05	0.03
Fe	30,000	0.02	0.00007
Y	16.9	0.0003	0.002
La	11	0.0003	0.003
Ce	27.7	0.0004	0.001

are listed a number of elements, including some of the elements having long-lived fission-product isotopes, with their concentrations in the supply to the ocean and in the ocean itself. Assuming steady-state equilibrium, the ratio of the concentration in the ocean to the concentration in the supply, the *transfer percentage,* indicates what share of the supply stays in solution. Large values of the transfer percentage indicate that a relatively large fraction remains dissolved; small values indicate that relatively much is removed.

These data give no information on the removal processes or on the time rate of removal. The latter can be obtained from estimates of rates of natural sedimentation together with chemical analysis of sediments or from study of rates of sedimentation of radio isotopes following weapons tests or waste disposal operations (Carritt and Harley, Chapter 6).

Goldberg and Arrhenius (in press), from a study of natural sediments, have estimated residence times in the ocean for several elements. They conclude that one half the amount of

strontium present at a given time is deposited in the sediments in about ten million years. For other elements the residence times relative to strontium are roughly proportional to the transfer percentages. Thus they estimate that the residence time for iron is of the order of a hundred years.

Introduction of radioactive materials

Radioactive materials in large quantities can be introduced into the sea from reactor wastes, from weapons tests, or in warfare.

gradients of specific activity decreasing from the sites of introduction, and depending on the mixing characteristics of the ocean.

Nuclear explosions have been the principal source of fission products introduced into the sea to date. The total quantity of fission power from such explosions so far may be estimated at 40 to 60 megatons of TNT equivalent, from the data summarized by Lapp (1956). This corresponds, with 20 kilotons equal to 1 kilogram of fission products (Libby, 1956a), to two to three metric tons of fission products.

TABLE 4 Fission Product Activity After 100 Days Cooling From 10^{11} Megawatt Hours of Nuclear Power Production [1]

Isotope	Half-life	Tons (metric)	Curies at 100 days	Specific activity curies per ton [2]
Kr^{85}	94 y	7.3	3.3×10^{9}	—
Sr^{89}	55 d	86	2.3×10^{12}	0.128
Sr^{90}	25 y	463	7.5×10^{10}	0.0042
Y^{90}	62 h	—	7.48×10^{10}	178
Y^{91}	57 d	111	2.8×10^{12}	6,660
Zr^{95}	65 d	152	3.2×10^{12}	—
Nb^{95}	35 d	161	6.3×10^{12}	—
Ru^{103}	45 d	46	1.3×10^{12}	—
Rh^{103}	57 m	—	1.3×10^{12}	—
Ru^{106}	290 d	35	1.5×10^{11}	—
Rh^{106}	30 sec	—	5.15×10^{10}	—
I^{131}	8.0 d	—	5.2×10^{9}	0.0743
Cs^{137}	33 y	705	5.63×10^{10}	20.1
Ba^{137}	2.6 m	—	5.1×10^{10}	0.728
Ba^{140}	12.5 d	2	1.5×10^{11}	2.14
La^{140}	1.7 d	—	2.5×10^{11}	595
Ce^{141}	28 d	45	1.5×10^{12}	268
Pr^{143}	13.8 d	2	1.4×10^{11}	—
Ce^{144}	275 d	490	1.6×10^{12}	386
Pr^{144}	17 m	—	2.4×10^{12}	—
Pm^{147}	94 y	7.3	3.3×10^{9}	—
Sm^{151}	73 y	0.7	2.0×10^{7}	—

[1] Adapted from data of Culler (1954) and Revelle et al. (1955).
[2] Based on tonnage shown in Table 2.

In Table 4 is a listing of the important fission products, their half-lives, and the quantities resulting from 10^{11} megawatt hours of nuclear power production (Carritt and Harley, Chapter 6). The column "specific activity" shows the ratio of the quantity of radioactivity of a particular isotope to the total amount of isotopes of that element in the sea for this amount of energy. The specific activity will, of course, be lower for smaller amounts of fission. It is also obvious that a uniform specific activity in all parts of the sea would be obtained only if the fission products were evenly distributed. Since, under any practical method of introduction, this will not occur, there are bound to be

The amount of fission products reaching the sea from nuclear explosions depends on a number of factors such as the location of the burst, the distance above (or below) the surface, and the size of the weapon or device. For the smaller devices with a TNT equivalent of several kilotons, most of the fallout is immediate and local, although an appreciable fraction remains in the troposphere for a few weeks (Libby, 1956a, b). Subsurface explosions will result in local deposition of a larger fraction of the fission products; a deep underwater burst will deposit practically all of the activity locally, with nearly $\frac{1}{3}$ being in the surface layer and about $\frac{2}{3}$ below (Revelle, 1957). In the case of

large, megaton devices, half or more of the total fission products are injected into the stratosphere from which there is a slow leakage into the troposphere (of the order of 10 per cent per year) and subsequent fallout fairly evenly over the entire northern hemisphere, with lesser amounts in the southern hemisphere (Libby, 1956a, b). Of this long-term fallout, up to 71 per cent falls on the oceans, since this is the proportion of the earth's surface covered by them. (The proportion of land to sea is higher in the northern hemisphere than in the southern, and since most of the long-term fallout occurs in the northern hemisphere, the amount entering the ocean will be less than 71 per cent.) On the other hand, some of the fallout on the land will eventually reach the sea through land drainage or river runoff.

Except in the case of deep underwater bursts, all of the fission products reaching the sea from weapons tests are deposited in the upper layer of the ocean. Removal into the deeper water is relatively slow. Despite the rapid mixing within the upper layer by vertical and horizontal wind stirring, the products from a large weapon remain in measurable concentrations over many months. A survey made 13 months after the 1954 weapons tests in the Pacific showed low-level activity over a vast area (Harley, 1956).

Radio isotopes in fallout on the land remain largely in the upper few inches of the soil. Fallout on the sea, on the contrary, is rapidly diffused through the upper mixed layer, some 75 meters deep on the average. Consequently, for conditions of equal fallout, the concentrations of radio isotopes in the part of the sea from which they are taken up by man's food organisms are less than in the soil. Thus, even though the calcium concentration of sea water is lower than in most soils, the ratio of strontium 90 to calcium in the marine environment is now much less than in agricultural lands of the mid-western United States. In 1955 (Libby 1956b) these soils contained about .025 microcuries of strontium 90 per kilogram of calcium available to growing plants. Revelle (1957) has calculated that for an equal amount of widely distributed fallout (from approximately 25 megatons TNT equivalent of fission) about .00015 microcuries of strontium per kilogram of calcium would be present in the upper mixed layer of the sea, half of one percent of the amount in soils.

In addition to fission products, neutron irradiation of elements in the environment immediately after the detonation produces other radioactive isotopes. With ordinary land or marine materials, the amounts of this neutron-induced radioactivity are small (Libby, 1956a). However, soon after the 1954 tests in the Pacific, quantities of zinc 65 were discovered in marine fishes, and subsequently cobalt 60 was recovered from clams in the Marshall Islands. These isotopes probably originated from neutron irradiation of metals, other than the fissionable materials, in the test device.

Comparison of table 2 and table 4 demonstrates that the mass of radioactive isotopes introduced into the sea from weapons tests, or which might be introduced from disposal of waste products, will be very small compared with the amounts of their normal isotopes already present. The introduction of the radioactive material does not, therefore, appreciably modify the chemical and physical properties of normal seawater, so that the chemistry of the introduced radioactive substances is the same as for the corresponding non-radioactive isotopes in the sea.

Introduced radioactive isotopes will partition into a soluble and an insoluble fraction. The physical states of a given element under equilibrium conditions depend upon whether or not the solubility product of the least soluble compound has been exceeded. Since the ionic activities of the elements in the complex chemical mixture that is sea water are not accurately known, it is difficult to attack this problem from theory. Greendale and Ballou (1954) have determined the distribution among soluble, colloidal, and particulate states of important fission product elements by simulating the conditions of an underwater detonation; their results are given in Table 5. Elements of Groups I, II, V, VI and VII usually occur as ionic forms, while other elements, including the rare earths, occur as solid phases. Some of these results have been confirmed by field observations following weapons tests (see Chapter 6 of this report by Carritt and Harley, and Chapter 7 by Krumholz, Goldberg and Boroughs). Those elements in Table 5 that have sufficiently long half-lives to contribute a significant share of the total activity after one year of decay are marked with an asterisk. Cesium 137 and strontium 89 and 90

remain in solution, while ruthenium 106, cerium 144, zirconium 95, yttrium 90 and 91, and niobium 95 are largely in the solid phase.

The solid fractions, whether they be chemical precipitates or solids produced by accumulation in the bodies of organisms, will tend to settle out. As they settle, they may encounter environmental conditions which will prevent or hinder deposition. There will be, however, some net transport toward the deeper water and the bottom from the settling process. Because of biological uptake, the removal of the particles from the upper layers of the sea may be quite slow. For example, cerium 144, a rare earth which has a half-life of 275 days, and which is present in the sea primarily in particulate form, and its daughter Pr 144 were found to account for 80 to 90 per cent of the activity in plankton samples from the upper layer taken in the Pacific by the TROLL survey 13 months after weapons tests (Harley, 1956).

TABLE 5 PHYSICAL STATES OF ELEMENTS IN SEA WATER [1] (FROM GREENDALE AND BALLOU, 1954)

Element	Percentage in given physical state		
	Ionic	Colloidal	Particulate
Cesium *	70	7	23
Iodine	90	8	2
Strontium *	87	3	10
Antimony	73	15	12
Tellurium	45	43	12
Molybdenum	30	10	60
Ruthenium *	0	5	95
Cerium *	2	4	94
Zirconium *	1	3	96
Yttrium *	0	4	96
Niobium *	0	0	100

[1] Elements introduced by simulated underwater detonation of atomic bomb, Greendale and Ballou, 1954.

* Indicates element has important fission product isotope.

A very rough idea of the reduction in activity that would eventually be obtained by removal from the ocean can be gained from the transfer percentages of Table 3. The fraction of an introduced fission product remaining in the sea will, at equilibrium, be equal to or greater than the transfer percentages for the corresponding element. (The transfer percentage reflects, in part, retention on land as well as sedimentation from the sea.) An important factor is the time required for equilibrium to be reached; if it is very long in relation to the half-life of the element in question, reduction of activity may be negligible. The long-lived and dangerous isotope, strontium 90, has a relatively high transfer percentage and a long equilibrium or "residence" time; the same would be expected for cesium 137, which is an alkali and should behave somewhat like potassium or rubidium.

Disposal of atomic wastes by deep sea burial in various sorts of packages has been proposed. Dispersion of the activity would then be by slow diffusion from concreted wastes, or would be delayed until rupture of an impermeable container occurred. Because the deep ocean sediments have appreciable exchange capacities, much of the wastes would be retained in this highly absorptive environment. The upper layers of the sediments would, presumably, tend to become saturated, and the further removal of radioactive elements by exchange or absorption would be controlled by the rate of diffusion into the deeper sediments.

There are wide gaps in our knowledge of many of the processes mentioned above. These, and suggestions for research needed to fill them, are discussed by Carritt and Harley (Chapter 6). Much of the required information can be obtained by the use of radioactive tracers, introduced in weapons tests and experimental waste disposal operations, as well as in purposive experiments.

V. PHYSICAL PROCESSES AND RADIOACTIVE MATERIALS

Physical structure of the sea

The physical properties of sea water of importance to the present study are functions of temperature, salinity, and pressure. The temperature ranges from about 30° C to about −2° C, which is the initial freezing point. The highest temperatures occur at the surface or in the mixed near-surface layer; below this the temperature decreases to about 5° C at 1,000 meters and to 1° to 2° at greater depths. In the deepest parts of the ocean there is a slight increase of temperature due to adiabatic heating. Hydrostatic pressure increases about one atmosphere for each 10 meters of depth. In the open ocean in mid-latitudes the salinity generally decreases slightly with depth in the upper few hundred meters, then increases slowly. In high latitudes the salinity normally increases with depth throughout the water column.

The density of sea water increases with decreasing temperature and with increasing salinity and pressure. Except in quite dilute sea water, the temperature of maximum density is lower than the freezing point. The range of density in the open sea is between about 1.02 and 1.06. It may, of course, be lower in inshore waters in the vicinity of river mouths. At constant pressure the major changes in density in the sea are associated with temperature, so that to a first approximation the change of density (computed for constant pressure) with depth is inversely proportional to the change of temperature.

Many processes in the sea depend on the density distribution. The ocean basins are largely filled with water of relatively high density formed in high latitudes; overlying this dense water in middle and low latitudes, and separated from it by the pycnocline, is the subsurface mixed layer, varying from a few meters to several hundred meters in thickness but averaging about 75 meters, of water of high temperature and low density. The relative rate of change of density with depth may be taken as a measurement of the vertical *stability* of the water (Sverdrup, Johnson and Fleming, 1942, p. 417). Stability in the region of the pycnocline is much higher than above or below it, so that exchange of water across it tends to be small.

All parts of the ocean and its bordering seas are in communication with each other, and are in continuous motion. The rates of movement, however, differ greatly in different areas. Thus, although there is eventual complete interchange of water between all oceans and seas, some parts are partially isolated from others, the exchange between these parts being much slower than within them.

Near-surface currents and mixing within the upper layer

Currents in the upper, mixed layer of the sea are primarily generated by winds, and, consequently, the major horizontal surface currents of the ocean correspond to the field of wind stress (Munk, 1950). The average locations and velocities of the important surface currents are well known from numerous observations of merchant ships and research vessels, and appear on many charts.

The velocities and volume transports of the major near-surface currents are large. For example, the mean speed of the Florida Current is about 193 cm/sec. and of the Kuroshio about 89 cm/sec. The volume of water flowing through the Florida Straits in 15 years is equal to the volume of the upper 500 meters of the whole North Atlantic, and the transport of water by the Kuroshio between the Northern Ryukus and Kyushu in 50 years is equal to the upper 500 meters of the whole North Pacific.

Because of the large surface currents, introduced materials tend to be carried away from the sites of introduction to other parts of the upper mixed layer of the sea. Thus, no area of surface water in the ocean is isolated for long periods from the remaining areas.

The currents are not steady streams, but have a complicated fine structure, with many eddies, jets, and filaments. In consequence of this turbulence, on both large and small scales, dissolved materials in seawater are rapidly dispersed horizontally. The rate of dispersion is about a million times the rate of molecular diffusion, and depends on wind speed, current shear, vertical and horizontal density gradients, direction of dispersion, and the dimensions of the area considered. Because of this large number of variables and the lack of knowledge of turbulent processes, it is not possible to predict accurately the horizontal dispersion in particular areas. If even moderately precise values are required, experiments must be conducted in the area of interest. Some of the results of such studies are reported by Wooster and Ketchum (Chapter 4).

The rate of vertical diffusion in the upper, mixed layer, although much less than that for horizontal dispersion, is nevertheless about a thousand times greater than molecular diffusion. The extent of vertical stirring in the upper layer depends on the magnitude and uniformity of the wind stress and on the vertical density gradient. Convective processes, and, in coastal areas, strong tidal currents, also contribute to vertical mixing. The mixing rate in the upper layer has been measured by changes in the vertical distribution of radio isotopes following weapons tests. Revelle, Folsom, Goldberg, and Isaacs (1955) report that in one such test the lower boundary of the radioactive water moved downward at about 10^{-1} cm/sec. until it reached the thermocline, where it abruptly stopped.

Circulation and mixing within the intermediate and deep layer

Within the pycnocline and for some distance below it, it is believed that most of the motion takes place along surfaces of constant potential density, so that transport and diffusion in the lateral direction are very much greater than in the vertical. This belief has been confirmed by experiments with radioactive tracers, reported by Revelle, Folsom, Goldberg, and Isaacs (1955), in which it was shown that the radioactive water spread out over an area of about 100 square kilometers while maintaining a thickness of the order of a few meters.

Much of our knowledge of deep and intermediate currents has been inferred from the distributions of properties. These indicate that the average velocities of the deep currents are only a few cm/sec. or less. However, Wüst (1957) has recently made calculations on data from the Atlantic which indicate velocities of meridional currents of 3 to 17 cm/sec. in the deep sea, along the western margin of the western trough, in depths between 3,000 and 5,000 meters. The calculated currents on the eastern side of the deep South Atlantic, especially in the region of the Angola Basin were, on the contrary, very weak. Dietrich (1957) has likewise computed mean current velocities of about 10 cm/sec. for the deep Antarctic Circumpolar Current, and for the Subarctic Bottom Current in the northern North Atlantic, the latter increasing to as much as 40 cm/sec. when flowing across the Greenland-Scotland ridge. He states, however, that in the largest part of the ocean the bottom currents are below 2 cm/sec.

Direct measurements of deep currents are technically difficult. The few successful measurements summarized by Bowden (1954) show mean velocities from less than a cm/sec. to 13 cm/sec. Recently Swallow (1955 and unpublished data) has measured subsurface currents by tracking a neutrally buoyant float at a fixed depth. His measurements in the North Atlantic give mean resultant velocities of 1.7 to 9.1 cm/sec. Tidal currents of about 10 cm/sec. have been obtained by Swallow and others in deep water. It appears that the mean current in many parts of the deep sea may be less than the periodic variable currents.

The turbulence of these variable tidal currents, especially near the bottom, contributes to vertical and horizontal mixing in deep water. Mixing should also occur along the boundaries of the rapid deep resultant currents indicated by Wüst and Dietrich, where there must be considerable shear.

Dietrich (1957) also suggests that horizontal spreading of near-bottom water may occur in regions of turbidity currents, which occur especially on the continental slopes.

Exchange between the open sea and coastal areas

In coastal areas and estuaries where precipitation and land runoff exceed evaporation, there is a net seaward drift of dilute surface water and an inshore drift of sub-surface water from the open sea. This is superimposed on the flow of wind-driven currents through the coastal areas.

Some idea of the average time involved in interchange of coastal waters can be obtained from the volume in and transport through various areas along the American Atlantic seacoast. Calculations give a mean age of $2\frac{1}{2}$ years for the waters over the Continental Shelf from Cape Hatteras to Cape Cod, about 3 months for the Bay of Fundy, and 3 to 4 months for Delaware Bay (Wooster and Ketchum, Chapter 4).

Exchange between the deep and intermediate layers and the mixed sub-surface layer

Evidence of local cross pycnocline interchange was obtained from measurements of the vertical distribution of radioactivity following the 1954 Pacific weapons tests (Japanese Fishery Agency, 1955 and Harley, 1956); it is not, however, clear whether the observed phenomena were entirely the result of physical exchange of the water and its contents or were in part due to settling of particles and to biological transport.

The major exchange between the near-surface and deeper waters takes place in the following regions:

In areas where the pycnocline is maintained, by the distribution of mass related to the general circulation, at a sufficiently shallow depth to be eroded away by wind stirring. Such areas exist near the equator, along the north edge of the Equatorial Counter Current, and at the centers of strong cyclonic eddies.

In regions of upwelling, where vertical currents carry water toward the surface and stir the surface and intermediate layers. Water from as deep as about 500 meters may be

brought to the surface by this process. Upwelling occurs along the western coasts of continents in intermediate and low latitudes, wherever the wind-driven circulation removes surface water from the coast. This water is replaced by deeper water moving upward. Such coastal upwelling has been found to be of the order of 1 to 3 meters per day. Upwelling also occurs in mid-ocean where there are surface current divergences, most notably along the equator in the eastern and central Pacific.

In regions of surface convergence, where sinking waters may extend to the oceanic depths, or may spread out at intermediate levels, according to their density. In tropical and temperate latitudes such sinking is confined to the upper few hundred meters, but at high latitudes the waters may reach great depths. Indeed, it is in the convergence regions of high latitudes that much of the intermediate and deep water of the oceans are formed.

In regions where increase of surface density by evaporation, freezing out of ice, or cooling, causes the surface waters to sink and be replaced by the formerly deeper water. Deep thermal convection occurs in high latitudes and extends in some areas to the bottom; for example, Antarctic bottom water is formed in the Weddell Sea by the cooling and sinking of the surface waters, and the Atlantic deep water is formed in a similar manner east of Greenland. Haline convection takes place in regions where evaporation exceeds precipitation or where freezing prevails over melting. The latter in high latitudes increases the intensity of the thermal convection. Haline convection in winter is responsible for the characteristics of the deep water of the Mediterranean Sea. This water flows out into the North Atlantic at depths of 1,000 to 1,500 meters, and can easily be identified even on the western side of the ocean.

The exchange between the surface layer of the ocean and the deeper layers may be either continuous or discontinuous. Some idea of the rate of exchange can be obtained from various estimates of the "age" or average residence time of the water in the deeper layers. These estimates, which differ widely depending on the data and assumptions used, have been summarized by Wooster and Ketchum (Chapter 4 of this report) and by Craig (Chapter 3).

Three estimates for the water in the intermediate layer of the Atlantic Ocean give residence times between 7 and 140 years. Estimates for the water below 2,000 meters vary from 50 to 1,000 years. An estimated upper limit based on the measured heat flow through the sea floor under the Pacific Ocean indicates that the Pacific deep water is replenished in less than 1,000 years. The deep water in the Pacific may be older than in the Atlantic because of the larger volume of the Pacific.

EXCHANGE FROM CONFINED BASINS

The few data available for estimating the age of water in confined basins have been considered by Wooster and Ketchum (Chapter 4). These indicate that the mean residence time of water in the Mediterranean Sea is about 75 years. In the Caribbean Sea the mean age cannot be less than 6 years and, in the deeper part, may be as much as 140 years. The deep waters of the Black Sea apparently remain isolated for very long periods. Transport considerations lead to an estimated age of at least 2,500 years, while, from consideration of phosphorus accumulation, the age has been estimated at 5,600 years.

VI. Biological Processes and Radioactive Materials

Uptake and accumulation of elements in organisms

Organisms take up from their environment and their food and incorporate into their bodies those elements required for their maintenance, growth, and reproduction. The proportion of various elements required by the organisms are different than the proportions in the environment, and this results in concentrations of some elements in the biosphere.

The energy that drives the whole life cycle is the energy of sunlight. This energy is bound chemically in organic compounds by the photosynthesis of plants, and is passed along, through the food chain, in the food of all the organisms beyond the plants. The flux of energy, and hence the flux of carbon, through the various trophic levels measures the *productivity* of the organisms at each level. Since the efficiency at each stage of the chain is low (of the order of 10 per cent to 30 per cent) the flux decreases at each step. The standing crop, or biomass, of organisms at the different levels, or, in other words, the amount of carbon present in the organisms at each level, may be greater or less

than the amount at the next lower level, depending on the rates of turnover of the populations involved.

In addition to the abundant elements carbon, oxygen and hydrogen, the bodies of organisms contain a number of elements in smaller amounts, such as nitrogen, phosphorus, calcium, strontium, copper, zinc, and iron, which are essential to the life processes. These may be obtained by organisms above the plants in the food chain either from their ingested food, or by direct uptake from the sea water. Since the requirements for different elements are different in different kinds of organisms, the fluxes of the of the populations of a particular part of the sea, and any quantities added will be soaked up by the biosphere very rapidly.

Both dissolved and particulate materials can be taken up from the environment. Iron, for example, occurs in the sea almost entirely in particulate form and is used in that form by diatoms. Fishes can take up ionic calcium and strontium directly from the sea water. Observations in conjunction with weapons tests, reported in Chapter 7 of this report, have shown that particulate feeders among the zooplankton ingest particles of inorganic compounds and retain them.

TABLE 6 Approximate Concentration Factors of Different Elements in Members of the Marine Biosphere. The Concentration Factors Are Based on a Live Weight Basis (From Krumholz, Goldberg and Boroughs, Ch. 7 of This Report)

Element	Form in sea water	Concentration in sea water (micrograms/l)	Concentration factors: Algae (non-calcareous)	Concentration factors: Invertebrates: Soft	Concentration factors: Invertebrates: Skeletal	Concentration factors: Vertebrates: Soft	Concentration factors: Vertebrates: Skeletal
Na	Ionic	10^7	1	0.5	0	0.07	1
K	Ionic	380,000	25	10	0	5	20
Cs	Ionic	0.5	1	10		10	
Ca	Ionic	400,000	10	10	1,000	1	200
Sr	Ionic	7,000	20	10	1,000	1	50
Zn	Ionic	10	100	5,000	1,000	1,000	30,000
Cu	Ionic	3	100	5,000	5,000	1,000	1,000
Fe	Particulate	10	20,000	10,000	100,000	1,000	5,000
Ni *	Ionic	2	500	200	200	100	0
Mo	Ionic-Particulate	10	10	100		20	
V	?	2	1,000	100		20	
Ti	?	1	1,000	1,000		40	
Cr	?	0.05	300				
P	Ionic	70	10,000	10,000	10,000	40,000	2,000,000
S	Ionic	900,000	10	5	1	2	
I		50	10,000	100	50	10	

* Values from Laevastu and Thompson (1956).

various elements are variable from one to another, and at different trophic levels.

The concentration factors of some of the important elements in different kinds of organisms are tabulated in Table 6, taken from Krumholz, Goldberg and Boroughs (Chapter 7 of this report). Certain elements, for example, sodium, occur in some organisms at lower concentrations than in the water; they are selected against. On the contrary, those elements, such as phosphorus, that are essential to the organisms but occur in low concentration in the sea water, are concentrated by several orders of magnitude. In some parts of the sea, the phosphorus may be nearly completely removed from the water by the organisms. Such elements are often limiting constituents for further increase

The uptakes of various elements by organisms are not entirely independent of one another. Elements of similar chemical properties tend to be taken up together very roughly in the same proportions as they exist in the environment. This is true, for example, of calcium and strontium. Sometimes one element has an inhibiting effect on another. There can also be synergistic effects, such as the enhancement of phosphorus uptake of diatoms by increased concentration of nitrogen.

Certain elements are deposited, in large part, in particular organs. Perhaps the best known examples are the deposition of iodine in the thyroid glands of vertebrates, or the deposition of calcium and strontium in the bones of verte-

brates and in the shells and other hard parts of invertebrates.

The length of time an organism retains the average atom of a given element varies greatly from one element to another. This is sometimes measured as the biological half-life, although the relative rate of loss is not a simple linear function of time as is the case with radioactive decay. Much is known about the retention times of different elements in man (see, for example, Handbook 52 of the National Bureau of Standards, 1953), but there are few data for most marine organisms. The rate of excretion of an element and the amount ultimately retained, will be quite different if the element is taken up quickly from a single dose or is taken up slowly over a long time.

The processes of uptake, accumulation, and loss of elements by marine and other aquatic organisms, are discussed in more detail by Boroughs, Chipman and Rice (Chapter 8 of this report), Krumholz, Goldberg, and Boroughs (Chapter 7), and by Krumholz and Foster (Chapter 9).

Effects of organisms on spatial distributions of elements in the sea

Those elements of which a large proportion is cycled through organisms are modified profoundly in their spatial distributions by the effects of the biosphere, so that they are quite differently distributed in the sea than elements in which the distribution is determined only by physical and inorganic chemical processes. We have already mentioned phosphorus as a notable example. Ketchum (Chapter 5 of this report) has written a detailed discussion of the general effects of the ecological system on the distribution of elements in the sea.

The marine biosphere acts as a reservoir for those elements that are removed selectively from sea water by organisms. This reservoir is not stationary in space, however, because many of the living organisms make both vertical and horizontal migrations of large extent, while their dead bodies and fecal materials continually fall toward the bottom under the influence of gravitation. The effects of the living reservoir in the distribution of elements vary not only from one part of the sea to another, but also seasonally in the same area.

Because organisms in the sea are more abundant in the upper layers than deeper down, those elements in scarce supply that are essential to life tend to be retained by the biosphere in the upper layers and to be returned to solution in the deeper layers. Stationary populations, such as attached benthic organisms, act as a fixed reservoir.

Where there are currents at different levels in opposite directions, the accumulation of elements by pelagic organisms, together with gravity effects on their dead bodies and fecal materials, can result in local concentrations of elements at intermediate depths greater than the concentrations in either the overlying or the deeper waters. This pattern, as noted by Ketchum, is common in estuaries, continental shelves, and in the vicinity of coastal upwelling.

Migration of organisms may result in a net transport of elements from areas of high concentration to areas of lower concentration. Thus, for example, the vertical migrations of the organisms of the deep scattering layer can result in a transport from the deeper layers into the upper mixed layer. Salmon which spawn and die in fresh waters after accumulating elements in the sea can transport significant quantities of some elements from the sea to fresh waters.

Finally, the remains of organisms, falling out as particulate matter, are an important component of the sedimentation process in the deep sea, and are thus important in the geochemical cycle, as noted by Carritt and Harley (Chapter 6) and others.

Although we have some understanding of the various processes involved, data for making useful quantitative assessments are almost entirely lacking.

Effects of introduction of radioactive elements

Since the isotopes of most chemical elements are similar in chemical behavior, it can be assumed that organisms do not appreciably distinguish between the radioactive and non-radioactive isotopes, and that, to a good degree of approximation, the path of a radioactive element through the biological system is the same as that of its non-radioactive isotopes.

The accumulation of radio isotopes in organisms will, therefore, depend on the same factors as the accumulation of normal isotopes (their concentration in the water where the organisms are located, the concentrations of other elements by which uptake is influenced, the size of the population of organisms concerned, the concen-

tration factors of the organisms for each element, and the rates of excretion, and *in addition* will depend on the decay rates of the radioactive isotopes).

The most important radio isotopes from the standpoint of accumulation in organisms are, therefore, those which are concentrated in large degree by organisms, are retained by them for relatively long periods of time, and have slow decay rates. An additional consideration from the standpoint of human hazards is the uptake and biological half-life of the elements in humans who may consume the marine organisms as food.

The most important fission product from all these considerations is strontium 90 and its daughter yttrium 90. This isotope has a large fission yield and a long physical half-life, is concentrated by organisms, and can be tolerated in human food only in very low amounts.

Ce 144 is another isotope with a large fission yield, which is concentrated by organisms (Harley, 1956), and has a moderately slow decay rate. Due to its small uptake and low retention by humans, it can, however, be tolerated in human food in much greater concentrations than Sr 90.

Zn 65 and Co 60, although not fission products, are sometimes produced in relatively large quantities in weapons tests. They are concentrated by very large factors in fish and mollusks used for human food, but fortunately they possess a relatively high tolerance level in humans.

Because of its biological role both in marine organisms and in humans, strontium 90 dominates consideration of depositing mixed fission products in the sea. For other radioactive wastes, and for mixed fission products from which Sr 90 has been removed, other elements will be the critical determinants, but in most cases, prior removal of Sr 90 will permit the safe disposal in the sea of larger quantities than would otherwise be possible.

The safe quantity of fission products depends on the concentrations that reach man's food organisms. The quantity will be greater if sites of introduction are chosen to give either long periods of isolation of the wastes or high dispersion (and thus low concentration) of the fractions that come into the environment (both physical and biological) of human food organisms.

Somatic and genetic effects on marine organisms

It is sometimes suggested that sufficient quantities of radioactive elements may be accumulated by marine organisms to endanger their populations, either by direct somatic effects or through genetic changes. Some aspects of this problem are discussed by Donaldson and Foster in Chapter 10 of this report.

So far as somatic effects are concerned, experimental data indicate that primitive forms are more resistant to ionizing radiation than the more complex vertebrates. It has not been possible to demonstrate any large-scale radiation damage to marine populations in the vicinity of large weapons tests. Levels of radiation safe from the standpoint of human hazards are also probably safe for the populations of marine organisms that are used as human food.

By analogy with results from genetic studies on laboratory animals, it may be inferred that significant genetic population effects will occur in marine organisms at much lower levels of radiation than will produce somatic effects. These genetic effects might be related to the increase in amount of total body radiation above the natural background. As shown by Folsom and Harley (Chapter 2), the normal radiation background of organisms in the deep sea is very low, so that appreciable quantities of radioactive wastes would significantly increase the radiation received by them. Craig (Chapter 3) has shown that the deposition of 1,000 tons per year of fission products in the deep sea would, at secular equilibrium, almost triple the average radiation level in the deep water. This could, conceivably, result in genetic effects in the marine populations in these waters, which might seriously upset the ecological system of the oceans. At the present state of knowledge, however, this is pure speculation. The matter does require, nevertheless, serious investigation.

VII. Predicted Effects of Introduced Radioactive Materials

Prediction of the effects of the introduction of radioactive materials into the different domains of the oceans must take into account the various physical, chemical, and biological processes discussed above. While our knowledge of these processes is very imperfect, we can make rough evaluations of the effects of disposal of fission products in different parts of the sea. Because of the limitation of knowledge, these

evaluations must, of necessity, be conservative. Under some circumstances this necessity could involve considerable cost to society. Those sites and methods of disposal, both on the land and in the sea, that provide the least hazard may also involve the greatest disposal costs, so that, to the extent we must include a safety factor because of ignorance, there can be economic loss.

In disposing of radioactive materials in the sea, we aim at two things: (1) isolation of the materials, so that their entry into the part of the sea and its contents used by man is limited, (2) dispersal of the materials that do enter the domain important to man, to keep the concentrations of radioactive elements at tolerable levels. Depending on the quantity of materials to be dealt with, we may need to consider either or both of these possibilities.

Introduction in the upper mixed layer

Radioactive materials introduced into the upper mixed layer will, because of the rapid transport and large horizontal and vertical mixing within this layer, be carried away from the site of introduction and rapidly dispersed. Dispersion may be more rapid in coastal areas than in the open sea, but in some situations there may be a net transport inshore, particularly in or near estuaries, if the materials are introduced below the surface.

Direct evidence of near-surface transport and dispersion of fission products in the open sea has been obtained by the surveys of the "Shunkotsu Maru" (Miyake, Sugiura and Kameda, 1955) and the "Taney" (Harley, 1956), respectively four and thirteen months after the Pacific weapons tests of March 1954. The indicated transport of these products was in good agreement with current velocities measured by conventional means. These data from the open sea and earlier measurements on the partially confined waters of Bikini Lagoon (Munk, Ewing and Revelle, 1949) demonstrate the rapid dispersal of fission products in the surface layer.

Dispersion in an inshore situation (the Irish Sea) was measured with fluorescein by Seligman (1955) as a preparatory study for the discharge of low-level wastes from a power reactor installation. Subsequent experience with liberation of the radioactive wastes (Anon., 1956) confirmed that they were rapidly dispersed.

Radioactive materials introduced into coastal waters enter directly into that part of the ocean most utilized by man, from which he removes the greater share of his harvest of marine food organisms. The sessile algae, bottom living invertebrates, and fishes of these waters heavily concentrate certain of the elements, such as strontium, cesium, zinc, and cobalt that has radioactive isotopes most hazardous to man. While dispersion due to physical transport and dispersion in these waters is high, they are usually shallow, so that the volume is limited and there can also be considerable accumulation in shallow bottom sediments from which the isotopes can be again taken up by man's food organisms.

In some coastal areas the combination of physical and biological processes can result in local concentrations of radioactivity in the waters themselves (Ketchum, Chapter 5).

Because of the above considerations, the quantity of radioactive materials that can be introduced safely into coastal waters near shore is very limited, of the order of a few hundred curies per day. The particular physical, chemical, and biological factors vary so widely from one coastal area to another, that careful study is required to determine the safe amount in any particular locality, and continuous monitoring should be conducted to guard against effects of unforeseen variability in environmental factors.

The rather low level of discharge of radioactive products that can be tolerated in coastal waters imposes the necessity of providing adequate safeguards against discharge of high-level atomic wastes from accidents to power reactors, either at locations on the shore or shipborne reactors.

The quantity of radioactive material that can be safely deposited in the mixed layer in the open sea depends on such local characteristics as the direction and rate of transport, the rate of horizontal dispersion, the rate of uptake by organisms, and the contiguity of fishing areas. However, in general, the quantities will be much greater than those permissible for coastal waters. An idea of the order of magnitude of mixed fission products that can be safely introduced in a fairly typical situation is given by the results of weapons tests in the Pacific where a quantity of mixed fission products of the order of half a ton was introduced into the mixed layer in a short time period. That this was near the limit of safety is evidenced by the capture in adjacent areas of specimens of tunas and other fishes with sufficient radioactivity to be doubt-

ful for human consumption (Kawabata, 1956, and Hiyama and Ichikawa, 1956).

Deep water introduction

The only place in the ocean in which we can be confident at this time that radioactive wastes of the order of some tons a year can be safely deposited is in the depths of the sea. Knowledge is, however, insufficient to determine whether radioactive materials of the order of the expected production from power reactors in the next few decades could be disposed of in this way.

Radioactive materials introduced into the deeper layers will be partially isolated from the upper layer for time periods related to the residence time of the water in the deeper layer. During this time there will be a decrease of radioactivity due to decay, and dilution due to dispersion. Since, as we have noted above, the residence times are variable in different depths and different locations, a much greater time of isolation will be obtained in some places than others.

The longest average time of isolation will be obtained in deep nearly enclosed basins such as the Black Sea. It has been suggested by Wüst (1957) that there may also be a long isolation period in the abyssal trenches of the central equatorial regions, such as the Romansch Deep or the Tonga Trench, but no data on currents in these deeps are now available.

Craig (Chapter 3 of this report), assuming an estimated average residence time in the deep sea of 300 years, the introduction into the deep sea of 1,000 tons per year of fission products after 100 days cooling, and complete uniform mixing within the deep water, has calculated the activity in the deep and surface layers at secular equilibrium. This calculation indicates that the total fission product activity in the mxed layer would be about equal to that at present from natural sources (primarily K^{40}). The concentration of Sr 90 would, however, be about 6.5×10^{-5} microcuries per liter, or 0.16 microcuries per kilogram of calcium in solution in sea water.

Studies of the uptake of strontium by marine fishes indicate a discrimination against strontium with respect to calcium approximately by a factor between 3 and 10. Thus for human populations such as the Japanese (Hiyama, 1956), in which much of the dietary calcium is obtained from marine fishes (including the bones and skin of some species), the amount of strontium 90 ingested per unit weight of calcium would be of the order of .04 microcuries per kilogram of calcium. A human population that obtained all its calcium from marine fishes after equilibrium was established with about 1,000 tons of fission products per year (1.1×10^5 megacuries of strontium 90) in the deep sea would have a burden, primarily in the bones, of approximately .005 microcuries of strontium 90 per kilogram of calcium. This is 5 per cent of the maximum permissible concentration for the population at large, estimated by the National Bureau of Standards (1955).

Weapons tests resulted in an average amount of .025 microcuries of strontium 90 per kilogram of calcium available to growing plants in the United States in 1955. By 1970, the amount will be .08 microcuries per kilogram of calcium even in the absence of further weapons tests (Kulp, Eckelmann, and Schulert, 1957). Because of discrimination against strontium with respect to calcium in food grains and grasses, and the additional discrimination in cows' milk and in human beings, it is expected that by 1970 an average of about .002 microcuries of strontium 90 per kilogram of calcium will exist in the United States population, 2 per cent of the maximum permissible concentration.

From the above considerations it is uncertain whether reactor-fuel wastes of the order of 1,000 tons a year could be deposited safely in the deep sea. Craig's calculation is most useful in orienting our thinking, but is, of course, very much oversimplified. No account is taken of the removal of activity from the sea by sedimentation. On the other hand, it does not take into account any biological transfer of material across the pycnocline, nor can we assume that effective concentration of Sr 90 per unit weight of calcium for some commercially important organisms will not be greater than the values we have taken.

Moreover, such a calculation assumes even distribution of the radioactive materials throughout the deep layer. This could only occur if they were evenly distributed when introduced, or if there were uniform and complete mixing in all parts of the deep layer.

A priori we should expect that neither the physical circulation and mixing in the deep sea nor the transfer between the deep layer and the mixed layer would be uniform. There is

some evidence, however, from carbon 14 measurements made by the Lamont Geological Observatory that in fact fairly complete mixing occurs within the deep sea during the average residence time of a water particle.

Another calculation, based on very conservative assumptions concerning the mixing processes, was made in the report of a meeting of scientists from the U.S. and U.K. (Anon., 1956). It was assumed that fission products deposited on the ocean floor in mid-latitudes would drift and disperse for at least 10 years before surfacing, at which time the contaminated area would be a disc about 2 km. thick and 70 km. in diameter, which would be subsequently dispersed throughout the surface layer. Repeated deposits of 1 megacurie of Sr 90 (0.4 tons of mixed fission products) made at the rate of ten per year would result in an average concentration of Sr 90 of not over 10^{-5} microcuries per liter in the mixed layer, or .025 microcuries per kilogram of calcium.

Although we cannot say at this time with any precision what quantities of reactor-waste products can be safely deposited in the deep sea, it appears certainly safe to employ quantities up to a few tons a year in careful experimental studies. It is not impossible that 1,000 tons a year can be safely disposed of in deep, isolated basins where the residence time is much greater than the 300-year average estimated for the deep sea generally. For quantities of the order of 100 tons a year or more, effects on the animal populations of the deep sea, and resulting effects on the whole ecology of the sea could become important; as to this no information is at present available.

VIII. What We Need to Know

Our knowledge of most of the processes in the oceans is altogether too fragmentary to permit precise predictions of the results of the introduction of a given quantity of radioactive materials at any particular place. In order to obtain the necessary knowledge, an adequate, long-range program of research on the physics, chemistry, and geology of the sea, and on the biology and ecology of its contained organisms is required. Such research must be directed toward the understanding of general principles, not simply to the *ad hoc* solution of a particular local problem for immediate application. The latter sort of study is, of course, desirable in order to provide engineering solutions to particular waste-disposal problems as they arise. Such engineering solutions must necessarily be of limited application and, moreover, they must always be conservative, at least until sufficient broad understanding is obtained.

Major Unsolved Problems

Some of the major basic problems that should be included in the research program can be briefly outlined:

1. Dispersion in the upper mixed layer

Fairly extensive information is available on the mean velocities and transport of the major surface currents. The transient currents and eddies that result in dispersion in both the horizontal and vertical directions are, on the contrary, not understood. Some empirical parameters approximately describing the relationships of diffusivity to time and to size of area have been developed, but understanding of the detailed physical principles is lacking. In consequence, it is not possible to predict on the basis of more elementary properties the dispersion of materials introduced into the upper layer at a given point. Direct measurements must be made, and these are costly and not necessarily reliable. Basic research on the turbulent motion of water in the upper layer is needed.

2. Circulation in the intermediate and deep layers

For the region of the sea below the surface layer, we not only do not understand the nature of the turbulent motion, we do not even have a description of the mean currents. The charting of the deep currents, and investigations toward elucidating the physical principles involved should be vigorously pursued.

3. Exchange between the surface layer and deeper layers

It is important to determine the average rate of exchange of water between the surface and the deep layers, as a basis of estimating average "hold up" times of dissolved materials deposited in the deep layer. It is probably even more important to measure the heterogeneity in the exchange system, that is to measure the rates of exchange in different areas and depths. We

know that vertical exchange is much more rapid in some parts of the oceans than others, but describing it in quantitative terms can be done only in a very sketchy manner. Quantitative data on this subject are required as one basis of arriving at estimates of the amount of atomic wastes that can be deposited safely in specified parts of the deep sea.

4. Sedimentation processes

Sedimentation processes constitute an important mechanism for removing atomic wastes from the waters of the oceans. In order to evaluate their role, however, we need to measure the average times that different elements remain in the sea before being deposited in the sediments, the rates of sedimentation in different parts of the deep sea, and the ability of the sediments to capture and retain various fission products.

5. Effects of the biosphere on the distribution and circulation of elements

As we have noted, marine organisms have profound effects in modifying the distribution and circulation of elements in the sea. It is vitally necessary that the biological processes be studied in sufficient detail to enable their effects to be quantitatively evaluated. Such investigations need to include: The flux of various elements through the different trophic levels, and the variations in different ecological realms such as inshore coastal waters, offshore surface waters and the deep sea; the effects of vertical and horizontal migrations of organisms on redistribution of elements; the effects of the uptake, modification of the physical state, and elimination of elements by members of the marine biosphere on their subsequent distribution in the sea.

6. Uptake and retention of elements by organisms used as food for man

Related to the foregoing, but of separate importance, is the study of the quantities of radioactive elements deposited in different situations in the sea that can be expected to be taken up by organisms harvested for food, the length of time such elements are retained in the food organisms, and, consequently, the levels of concentration. Some parts of some organisms are not eaten by man, but are discarded or used for other purposes. The sites of accumulation of different radioactive elements in the organisms must therefore be determined.

7. Effects of atomic radiation on populations of marine organisms

In order to determine what quantities of atomic wastes can be safely deposited in the sea without upsetting the ecology of the sea through destruction of important populations of organisms, research is needed on the somatic and genetic effects of atomic radiation on marine populations. This is especially important for organisms of the deep sea which may come in contact with very high concentrations of radioactive elements, if deep sea disposal of large quantities proves feasible in other respects.

RESEARCH METHODS

Much of this required research can be accomplished by the intensive application of classical techniques of physics, chemistry, geology, and biology. In addition, however, the availability of radioactive isotopes provides us with a powerful new tool, which is especially valuable for studying *processes*. The use of radioactive elements as tracers permits the paths of various elements, both in the physical environment and within the biosphere, to be determined, and the fluxes of the elements through various parts of the system to be measured.

Radioactive tracers are useful both in laboratory experiments and in field studies of various kinds. The use of tracers in the laboratory and in small scale field experiments is already familiar. Information from the tracers introduced into the sea by weapons tests has provided valuable information. What has not yet been done, and what we believe will be a fruitful approach, is the employment of fairly large quantities of radio isotopes to study the various processes in the open ocean in a planned fashion. In Chapters of this report by Folsom and Vine and by Schaefer, suggestions are made for some experiments that should be useful and are currently feasible.

Naturally occurring radioactive isotopes can also provide a fruitful means of attack. Craig, in Chapter 11, discusses some of these avenues of research in detail.

FACILITIES REQUIRED

The Committee has not attempted to draw up detailed estimates of men, ships, and facili-

ties which will be required for an adequate attack on this problem. These requirements will, however, be large. The problems outlined above are among the most difficult in the marine sciences. Adequate solutions will demand the collection of much more knowledge about the sea and its contents than the total obtained in the past hundred years.

Because of the urgency of these problems, and because of the large costs involved, it is essential that research be coordinated on both the national and international levels. Coordination among scientists engaged in these studies should be easier in the future than it has been in the past.

OTHER BENEFITS OF THE RESEARCH TO MANKIND

The potential requirement for disposal of atomic wastes in the sea is sufficient reason for pursuit of these investigations. However, mankind will derive additional, and perhaps even greater, benefits in other ways. For example, the flux of materials through the various trophic levels of the biosphere is the fundamental process underlying the harvest of the sea fisheries. This process must be studied to provide part of the basis for atomic waste disposal, but its elucidation will also provide much of the scientific base for the optimum exploitation and conservation of the seas' living resources by man.

IX. Conclusions and Recommendations

We repeat here the conclusions and recommendations that were agreed upon by the members of the Committee at the time they prepared the Summary Report published by the Academy in 1956:

1. Tests of atomic weapons can be carried out over or in the sea in selected localities without serious loss to fisheries if the planning and execution of the tests are based on adequate knowledge of the biological regime. The same thing is true of experimental introduction of fission products into the sea for scientific and engineering purposes.

2. Within the foreseeable future the problem of disposal of atomic wastes from nuclear fission power plants will greatly overshadow the present problems posed by the dispersal of radioactive materials from weapons tests. It may be convenient and perhaps necessary to dispose of some of these industrial wastes in the oceans. Sufficient knowledge is not now available to predict the effects of such disposal on man's use of other resources of the sea.

3. We are confident that the necessary knowledge can be obtained through an adequate and long-range program of research on the physics, chemistry, and geology of the sea and on the biology of marine organisms. Such a program would involve both field and laboratory experiments with radioactive material as well as the use of other techniques for oceanographic research. Although some research is already under way, the level of effort is too low. Far more important, much of the present research is too short-range in character, directed towards *ad hoc* solutions of immediate engineering problems, and as a result produces limited knowledge rather than the broad understanding upon which lasting solutions can be based.

4. We recommend that in future weapons tests there should be a serious effort to obtain the maximum of purely scientific information about the ocean, the atmosphere, and marine organisms. This requires, in our opinion, the following steps: (1) In the planning stage committees of disinterested scientists should be consulted and their recommendations followed; (2) funds should be made available for scientific studies unrelated to the character of the weapons themselves; (3) the recommended scientific program should be supported and carried out independently of the military program rather than on a "not to interfere" basis.

5. Ignorance and emotionalism characterize much of the discussion of the effects of large amounts of radioactivity on the oceans and the fisheries. Our present knowledge should be sufficient to dispel much of the overconfidence on the one hand and the fear on the other that have characterized discussion both within the Government and among the general public. In our opinion, benefits would result from a considerable relaxation of secrecy in a serious attempt to spread knowledge and understanding throughout the population.

6. Sea disposal of radioactive waste materials, if carried out in a limited, experimental, controlled fashion, can provide some of the information required to evaluate the possibilities of, and limitations on, this method of disposal. Very careful regulation and evaluation of such operations will, however, be required. We, therefore, recommend that a national agency,

with adequate authority, financial support, and technical staff, regulate and maintain records of such disposal, and that continuing scientific and engineering studies be made of the resulting effects in the sea.

7. We recommend that a National Academy of Sciences—National Research Council committee on atomic radiation in relation to oceanography and fisheries be established on a continuing basis to collect and evaluate information and to plan and coordinate scientific research.*

8. Studies of the ocean and the atmosphere are more costly in time than in money, and time is already late to begin certain important studies. The problems involved cannot be attacked quickly or even, in many cases, directly. The pollution problems of the past and present, though serious, are not irremediable. The atomic waste problem, if allowed to get out of hand, might result in a profound, irrecoverable loss. We, therefore, plead with all urgency for immediate intensification and redirection of scientific effort on a world-wide basis towards building the structure of understanding that will be necessary in the future. This structure cannot be completed in a few years; decades of effort will be necessary and mankind will be fortunate if the required knowledge is available at the time when the practical engineering problems have to be faced.

9. The world-girdling oceans cannot be separated into isolated parts. What happens at any one point in the sea ultimately affects the waters everywhere. Moreover, the oceans are international. No man and no nation can claim the exclusive ownership of the resources of the sea. The problem of the disposal of radioactive wastes, with its potential hazard to human use of marine resources, is thus an international one. In certain countries with small land areas and large populations, marine disposal of fission products may be essential to the economic development of atomic energy. We, therefore, recommend: (1) that cognizant international agencies formulate as soon as possible conventions for the safe disposal of atomic wastes at sea, based on existing scientific knowledge; (2) that the nations be urged to collaborate in studies of the oceans and their contained organisms, with the objective of developing comparatively safe means of oceanic disposal of the very large quantities of radioactive wastes that may be expected in the future.**

10. Because of the increasing radioactive contamination of the sea and the atmosphere, many of the necessary experiments will not be possible after another ten or twenty years. The recommended international scientific effort should be developed on an urgent basis.

11. The broader problems concerned with full utilization of the food and other resources of the sea for the benefit of mankind also require intensive international collaboration in the scientific use of radioactive material.

REFERENCES

ANON. 1956. Report of a meeting of United Kingdom and United States scientists on biological effects of radiation in oceanography and fisheries. Nat. Acad. Sci.—Nat. Research Council, Oct. 31, 1956, 8 pp. (mimeographed).

BOWDEN, K. F. 1954. The direct measurement of subsurface currents in the oceans. *Deep Sea Research,* Vol. 2, pp. 33-47.

CULLER, F. L. 1954. Notes on fission product wastes from proposed power reactors. ORNL Central File No. 55-4-25.

DIETRICH, G. 1957. Selection of suitable ocean disposal areas for radioactive waste. (A preliminary report with 6 charts.) M.S., 10 pp.

FOOD AND AGRICULTURE ORGANIZATION OF UNESCO. 1957. Yearbook of fishery statistics. FAO, Rome, Vol. 5 (1954-55).

GOLDBERG, E. and ARRHENIUS, G. O. S. 1957. Chemistry of Pacific pelagic sediments. In press.

GREENDALE, A. E., and N. E. BALLOU. 1954. Physical state of fission product elements following their vaporization in distilled water and sea water. USNRDL Document 436, pp. 1-28.

HARLEY, JOHN E. (Editor). 1956. Operation Troll. U.S., A.E.C., N.Y. Operations office 1956. 37 pp.

* The President of the Academy, Dr. Detlev W. Bronk, has requested that the present committee undertake to develop and carry forward this continuing program.

** As a first step in this direction an informal discussion was held by members of this committee with scientists from the United Kingdom at North Falmouth, Massachusetts, on September 27 and 28, 1956. A brief summary of the meeting was published by the National Academy of Sciences (Anon., 1956).

HIYAMA, Y. 1956. Maximum permissible concentration of Sr 90 in food and its environment. Records of Oceanographic Work in Japan, Vol. 3, No. 1, March 1957, pp. 70-77.

HIYAMA, Y., and R. ICHIKAWA. 1956. Movement of fishing grounds where contaminated tuna were caught. Japan Society for the Promotion of Science; Research in the Effects and Influences of the Nuclear Bomb Test Explosions, pp. 1079.

JAPANESE FISHERY AGENCY. 1955. Report on the investigations of the effects of radiation in the Bikini region. Res. Dept., Jap. Fish. Agency, Tokyo, 191 pp.

KAWABATA, T. 1956. Movement of fishing grounds where contaminated tuna were caught. Japan Society for the Promotion of Science; Research in the Effects and Influences of the Nuclear Bomb Test Explosions, pp. 1085.

KRAUSKOPF, K. B. 1956. Factors controlling the concentration of thirteen rare metals in sea water. *Geochim. et Cosmochim.* Acta 9, pp. 1-32.

KULP, J. L., ECKELMANN, W. R., and A. R. SCHULERT. 1957. Strontium 90 in man. *Science,* Vol. 125, No. 3241, pp. 219-225.

LAEVASTU, T., and T. G. THOMPSON. 1956. The determination and occurrence of nickel in sea water, marine organisms, and sediments. *Jour. du Cons.,* Vol. 21, pp. 125-143.

LAPP, RALPH E. 1956. Strontium limits in peace and war. *Bull. Atomic Scientists,* Vol. 12, No. 8, pp. 287-289, 320.

LIBBY, W. F. 1956a. Radioactive fallout and radioactive strontium. *Science,* Vol. 123, pp. 657-660.

1956b. Radioactive strontium fallout. *Proc. Nat. Acad. Sci.,* Vol. 42, No. 6, pp. 365-390.

MIYAKE, J., SUGIURA, Y., and K. KAMEDA. 1955. On the distribution of radioactivity in the sea around Bikini Atoll in June 1954. *Pap. Meteorol. Geophys.,* Tokyo, Vol. 5, No. 3-4, pp. 253-262.

MUNK, W. H. 1950. On the wind-driven ocean circulation. *Jour. Meteorol.,* Vol. 7, No. 2, pp. 79-93.

MUNK, W. H., EWING, G. C., and R. R. REVELLE. 1949. Diffusion in Bikini lagoon. *Trans. Am. Geophys. Union,* Vol. 30, No. 1, pp. 59-66.

NATIONAL BUREAU OF STANDARDS. 1953. Maximum permissible amounts of radio isotopes in the human body and maximum permissible concentrations in air and water. U.S. Dept. of Commerce, Nat. Bureau Standards. Handbook 52, 45 pp.

1954. Radioactive waste disposal in the ocean. Nat. Bureau of Standards. Handbook 58, 31 pp.

NUCLEONICS. 1956. Calder Hall, over-all description. *Nucleonics,* Vol. 14, No. 12, pp. S10-S11.

1957. Roundup of key developments in atomic energy. *Nucleonics,* Vol. 15, No. 6, pp. 17-28.

RANKAMA, K., and T. C. SAHAMA. 1950. Geochemistry. Univ. of Chicago Press, 1950.

REVELLE, R. R. 1957. Statement by Professor Roger Revelle before the joint Committee on atomic energy, 28 May 1957. The Nature of Radioactive Fallout and its Effects on Man; *Hearings before the Special Subcommittee on Radiation of the Joint Committee on Atomic Energy.* Congress of the United States, 1957.

REVELLE, R. R., FOLSOM, T. R., GOLDBERG, E. D., and J. D. ISAACS. 1955. Nuclear Science and Oceanography. Int. Conf. on the Peaceful Uses of Atomic Energy. A/Conf. 8/P/277, 22 pp. (mimeographed).

SELIGMAN, N. 1955. The discharge of radioactive waste products into the Irish Sea. Part I: First experiment for the study of movement and dilution of released dye in the sea. Proc. Int. Conf. on Peaceful Uses of Atomic Energy, United Kingdom paper number 418, 25 pp.

SVERDRUP, H. U., JOHNSON, M. W., and R. H. FLEMING. 1942. The Oceans. Prentice Hall, New York, 1942, 1060 pp.

SWALLOW, J. C. 1955. A neutral-buoyancy float for measuring deep currents. *Deep Sea Research,* Vol. 3, pp. 74-81.

VINOGRADOV, A. P. 1953. The elementary composition of marine organisms. Sears Foundation for Marine Research, Memoir No. 2, 647 pp.

WÜST, G. 1957. Report on the current velocities, volume transports and mixing effects in the Atlantic deep sea as physical processes important to the transport and dispersal of radioactive wastes. M. S. (mimeographed), 19 pp.

CHAPTER 1

PHYSICAL AND CHEMICAL PROPERTIES OF WASTES PRODUCED BY ATOMIC POWER INDUSTRY

CHARLES E. RENN, *The Johns Hopkins University*
Department of Sanitary Engineering and Water Resources

THE ULTIMATE forms and radioactivities of wastes delivered for sea disposal will be determined by conditions that have not yet been fully evaluated. Present and projected wastes will undoubtedly be modified by requirements for storage, transport, and economical handling, and the ultimate form of wastes with which we may be concerned will be further conditioned by what we learn in early disposal practice. The following represents the characteristics of high-level reactor wastes that now exist, and which are likely to appear soon.

The primary radioactive wastes result from the chemical extraction of inhibitory fission products from metallic reactor elements. A strong nitric acid solution of aluminum heavily contaminated with a variety of fission products is obtained after the useful reactor fuel is recovered. To conserve tank space and shielding, the solutions are concentrated by evaporation. Where storage is to be made in steel containers, the solution may be neutralized and made slightly alkaline with commercial caustic. A neutral or alkaline salt solution or slurry is developed — the concentration of salts may approach or exceed saturation values at storage temperature. The neutral salt concentration of the waste determines its density. Some types of reactor elements are not directly soluble in nitric acid and require solution in combinations of other mineral acids and catalysts; most ultimately require conversion to nitrates before complete extraction, however.

The cladding and alloying metals of the reactor elements are also discarded in the wastes. Aluminum is the most common and abundant of the metals used; it appears in concentrations as high as 80,000 ppm. in final wastes. Zirconium will also be present.

Of the various non-radioactive components in the wastes, the properties of the high-density-producing salts, of the high nitrate concentrations, and of aluminum are of greatest interest. The presence of these at present limit the practical production of selectively adsorbed fission waste products. If the wastes are concentrated for economical storage and transportation and neutralized to limit corrosion, the densities of the waste liquids will exceed that of sea water.

The temperatures for precipitation of supersaturated salts in the various wastes are not known, but it may be assumed that further sludges will be formed on cooling to deep sea temperatures — some corrosion-product sludges already exist.

The solubilities of both normal and radioactive components of the waste will be conditioned by the presence of nitrates in concentrations exceeding equivalence. Aluminum nitrate precipitates as a light floc in sea water at concentrations as low as 1 ppm. Al. At present there are no data on its solubility in a sea water waste mixture. Neither do we know what the adsorption characteristics of the aluminum floc in sea water may be.

The range of physical and radiochemical characteristics that may be anticipated in concentrated fuel re-processing wastes and approximate quantities of wastes produced are indicated in the three tables following.

TABLE 1 GROSS PHYSICAL AND CHEMICAL CHARACTERISTICS OF STRONG AQUEOUS WASTES FROM REACTOR FUEL RECOVERY PROCESSES [1]

(Concentrations of non-radioactive components before evaporation, neutralization, and treatment for fission removal.)

Component	Range of Molar Concentrations
H	0.07 – 7.0
Al	0.04 – 1.6
Fe	0.05
Zr	0.03 – 0.5
NH_4^+	0.05 – 2.0
Cr	0.01 – 1.0
Ni	0.03
Sn	0.02
Mn	0.001
Hg	0.001– 0.01
F	0.05 – 3.0
NO_3^-	0.14 – 7.0
$SO_4^=$	0.2 – 0.5
Specific Gravity (unconcentrated)	1.07 – 1.25
Curies/gal. (100 days cooling)	80 –5200
BTU/hr./gal. (10 days cooling—50% gamma, 50% beta)	1.37 – 29.4

[1] From Tables 4 and 5, Status Report on the Disposal of Radioactive Wastes, ORNL-CF-57-3-114, F. L. Culler.

TABLE 2 SHORT-LIVED FISSION PRODUCTS PER 1000 GM U^{235} REACTOR CHARGE AT 100 DAYS COOLING WITH 30 PER CENT BURNUP [1]

Fission products	Half life [2]	Grams	Beta curies	Gamma curies
Y-90	62 h	4.63	748	—
Rh-106	30 s	0	1,514	515
Ce-144	275 d	4.90	16,332	4,900
Zr-95	65 d	1.52	32,647	62,356
Nb-95	35 d	1.61	63,657	65,657
Y-91	57 d	1.11	28,239	—
Sr-89	55 d	0.86	23,253	—
Ru-103	45 d	0.46	13,236	6,618
Ce-141	28 d	0.45	10,004	20,008
Ba-137	2.6 m	—	—	508
Ru-106	290.0 d	0.35	1,514	—
Pr-143	13.8 d	0.02	1,465	—
Ba-140	12.5 d	0.02	1,222	305
La-140	1.7 d	—	1,222	1,331
I-131	8.0 d	—	23	29
Total		15.93	195,076	162,227

[1] From presentation by F. L. Culler, Oak Ridge National Laboratory, before Meeting on Ocean Disposal of Reactor Wastes, Woods Hole Oceanographic Institution, August 5–6, 1954.

[2] Abbreviations are s for seconds, m for minutes, h for hours, and d for days.

TABLE 3 LONG-LIVED FISSION PRODUCTS PER 1000 GM U^{235} REACTOR CHARGE AT 100 DAYS COOLING WITH 30 PER CENT BURNUP [1]

Fission products	Half life	Grams	Beta curies	Gamma curies
Cs-137	33 y	7.05	563	—
Sr-90	25 y	4.63	748	—
Pr-144	17 m	4.90	16,333	17,966
Te-129	72 m	0.03	1,217	2,435
Total long-lived		16.61	18,861	20,401
Inactive fission products		230.00		
[2] Short T $\frac{1}{2}$		15.93	198,564	152,325
Grand total		262.54	217,425	172,726

[1] From presentation by F. L. Culler, Oak Ridge National Laboratory, before Meeting on Ocean Disposal of Reactor Wastes, Woods Hole Oceanographic Institution, August 5–6, 1954.

[2] Short-term fission products from table 2.

CHAPTER 2

COMPARISON OF SOME NATURAL RADIATIONS RECEIVED BY SELECTED ORGANISMS [1]

THEODORE R. FOLSOM, *Scripps Institution of Oceanography, La Jolla, California*

and

JOHN H. HARLEY, *Health and Safety Laboratory, U. S. Atomic Energy Commission*

IN ATTEMPTING to consider in numerical terms possible consequences to populations from mutations caused by very low levels of artificial radioactivity, it is instructive to collect for quick comparison some estimates of the natural doses to which certain organisms have been exposed for geological periods. These data emphasize that doses from natural sources vary widely and depend not only upon the habitat but also upon the physical size of the organism; this natural radiation background varies particularly widely amongst aquatic organisms.

A very useful summary of natural and artificial radiation to which human beings are now exposed has been published by Libby (1955); it has already been quoted and some of his comparisons will be repeated here. Nevertheless, additional radiological factors must be included whenever the natural exposures of marine organisms are to be evaluated.

Only sources contributing substantially to the average dose to the organisms as a whole will be listed here. The major contributors are (a) cosmic rays, (b) radioactivity in local surroundings, and (c) radioactivity spread through the tissue inside the organism itself.

Cosmic rays

Cosmic ray intensity decreases far more rapidly from sea level downward than it increases with increasing elevation above the earth. Figure 1 and Table 1 show the trend of the ionizing component of these rays with elevation above sea level, and with depth in water. The absolute dose which is used in Table 3 and Figure 2 is the average of the two values Libby (1955) uses for the geomagnetic equator and for 55° geomagnetic north latitude. (See Figure 1 and Table 1.)

External activity

Most organisms live close to either (a) igneous or metamorphic rock, (b) sedimentary rock, or (c) water. Sea water has a characteristic natural radioactivity — much lower than that of terrestrial rocks but quite appreciable when

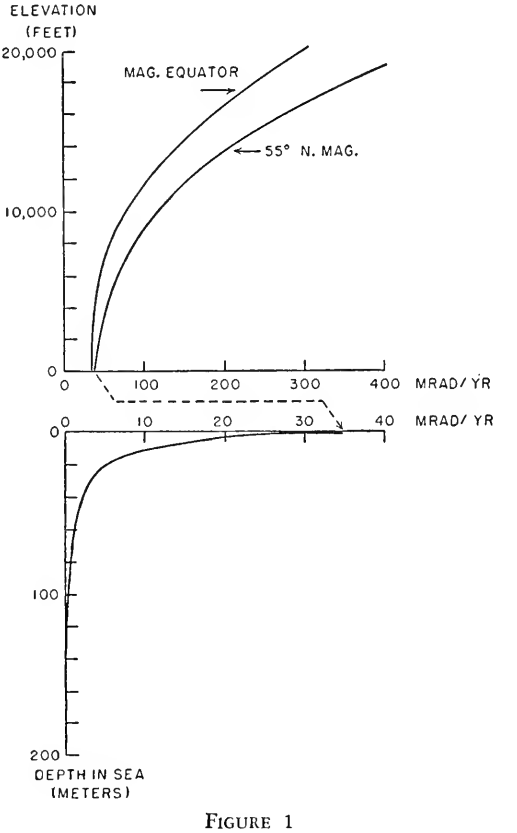


FIGURE 1

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 904.

TABLE 1 TREND OF COSMIC RAYS WITH DISTANCE ABOVE AND BELOW SEA LEVEL

Variation with elevation above sea level, values of intensity of ionizing component (in mrads/year) taken from Libby (1955).

Elevation in feet	Mrad/year	
	Equator	Latitude 55° N (mag)
0	33	37
5,000	40	60
10,000	80	120
15,000	160	240
20,000	300	450

Variation with depth in water, values computed from average attenuation compiled by George (1952) using Libby's average absolute intensity for mean sea level.

Depth in meters	Mrad/year	Percent of surface value
0	35	100
10	10.1	28.8
20	4.86	13.9
50	1.40	4.0
100	0.47	1.35
200	0.15	0.42
300	0.074	0.21
500	0.030	0.087
1,000	0.009	0.025
4,000	0.007	0.002

compared to that of most natural fresh waters. The major activity in sea water comes from radiopotassium (Revelle, Folsom, Goldberg and Isaacs 1955), and only this constituent will be considered here. Of the metamorphic and igneous rocks, granite has the highest activity; for our comparisons the same average radioactivities used by Libby (1955) are used here for granite and sedimentary rocks.

Internal sources of activity

The bodies of large animals contain a much higher concentration of potassium than is found in sea water. A value of 0.2 per cent is used herein for human tissue (Burch and Spiers, 1954) and 0.3 per cent is used for the potassium concentration of large fish (Vinogradov, 1953). Since radio-potassium contributes the major portion, aside from cosmic rays, of the radiation contributing to the average dose to the total body of any marine organisms, the character and distribution of this important natural activity has been compiled in Table 2.

Geometrical factors influencing dose

A man standing above a granite plane surface receives from the granite roughly one half the radiation which might strike him if he were completely surrounded by granite; likewise a man in a row boat receives from the sea only one half the dose which the sea gives to any submerged organism.

Potassium yields both beta and gamma activity; roughly three fourths of the total energy comes from the beta rays. Nevertheless, because of its short range, the beta particle from the potassium in the surrounding sea contributes

TABLE 2 POTASSIUM RADIATION DATA

Distribution and Intensities

Material	Potassium content		Beta rays		Gamma rays		
			d/m/g	mrad/yr	d/m/g	mrad/yr	
Sea water (35‰ salinity)	0.038%	(1)	0.66	2.7	0.068	0.9	
Man	0.2%	(2)	3.5	15	0.36	2.3	(4)
Fish (large)	0.3%	(3)	5.8	24	0.3	3.7	(4)

Physical Nature of Potassum Activity
Beta activity = 29 d/s/gram of total potassium
Beta ray energy (average) = 0.5 mev
Gamma activity = 3 d/s/gram of total potassium
Gamma ray energy = 1.5 mev

Sample Calculations for Potassium Activity

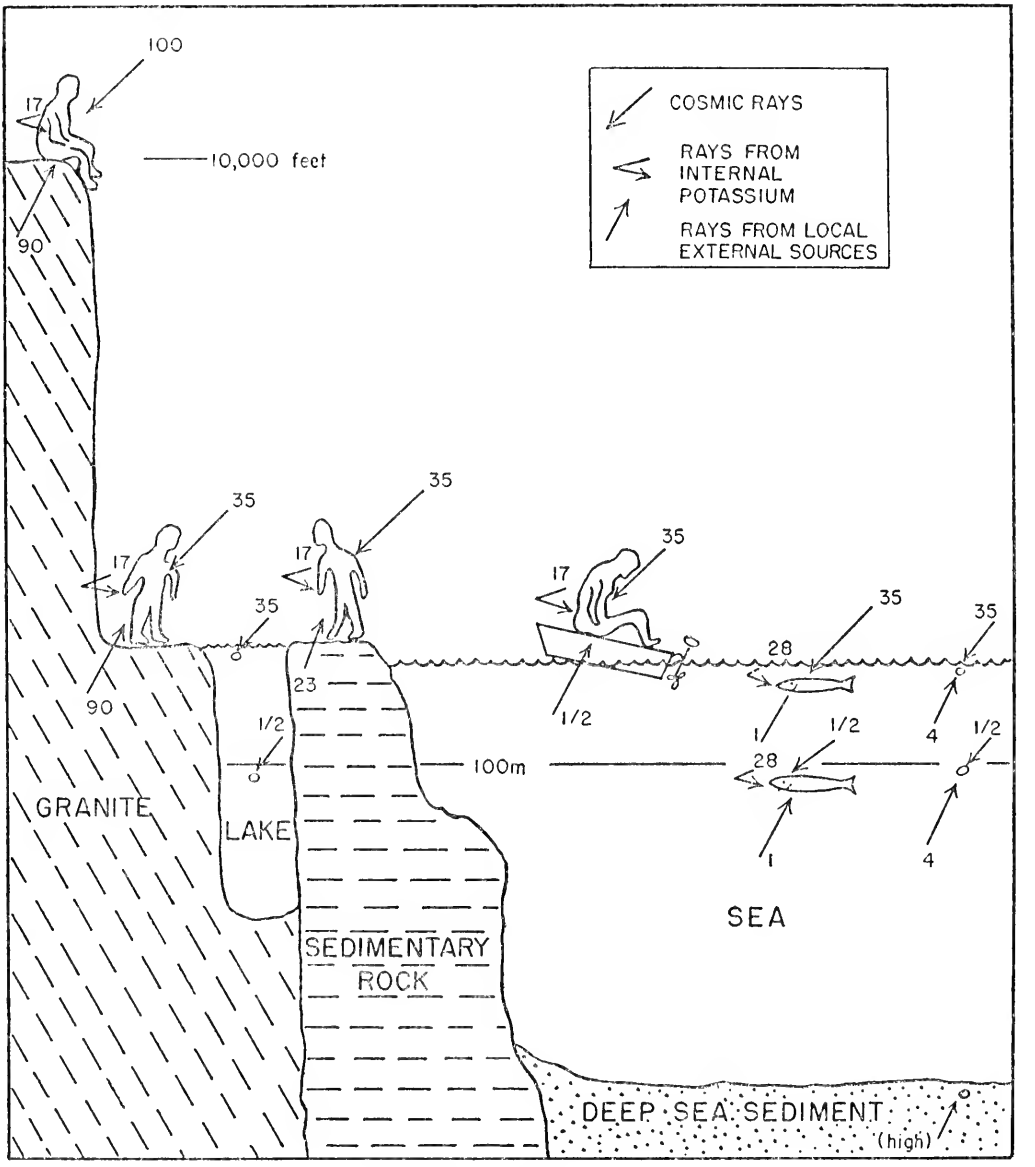
$$\frac{\text{Beta d/m/g} \times 1440 \times 365 \text{ m/yr} \times 0.5 \text{ mev/d} \times 1.6 \times 10^{-6} \text{ erg/mev } 1000}{100 \text{ erg/rad}} = \text{mrad/yr beta which reduces}$$

to, Beta d/m/g $\times$ 4.2 = mrad/yr beta; and correspondingly, Gamma d/m/g/ $\times$ 12.6 = mrad/yr gamma.

(1) Sverdrup, Johnson and Fleming (1942).
(2) Sherman (1941).
(3) Vinogradov (1953).
(4) Assume half of the gamma rays from internal activity are absorbed inside the body.

very little to the total dose of a large animal. On the other hand the beta rays from the surroundings can appreciably affect very small organisms and can in fact become the predominant contributor to dose whenever the organism has dimensions much smaller than the range of the beta particles in water and tissue.

The effect of beta rays starting from internal sources also depends upon the size of the organism. If the organism is very small the beta bombardment from the outside sources may contribute much more than does internal activity even though the source of activity is more concentrated in the tissue than it is in the surrounding water. It would appear from the character of beta penetration (Friedlander and Kennedy,



TOTAL NATURAL DOSES (mrad/year)

Man over granite		Man over sedimentary rock	Man over sea	Large fish in sea		Micro-organism in sea	
10.000	m.s.l.			at surf.	100 m.	at surf.	100 m.
207	142	75	52	64	30	39	5

FIGURE 2

1949) that any potassium beta particle which originates inside a small organism will deposit most of its energy outside the organism; apparently less than 10 per cent of the total ionization can take place inside a sphere having a mean radius of 0.1 mm, and perhaps from the activity concentrated inside a phytoplankter having a mean radius of 0.01 mm only 1 per cent of the energy would be felt by the organism itself. Thus we see that the constitution of the surrounding medium dominates the life of the marine microorganism in a radiological sense as well as in those other manners more familiar to biologists.

Units used

For quantitative statements concerning such feeble radiations as these it is logical to use a very small unit and preferably one which is defined in terms of energy absorbed; the millirad per year (mrad/yr) is such a unit and is used here. The rad unit is only slightly larger than the more familiar roentgen unit, since 1.0 rad by definition causes 100 ergs to be absorbed per gram of matter, and this is approximately the energy deposited by 1.1 roentgen of gamma rays. For converting beta activity to equivalent rad dosage the average beta energy of potassium has been taken as being 0.5 mev.

Comparison of natural doses in several domains

Figure 2 attempts to bring into a single picture the magnitudes of the main components making up the radiation in each of several domains of interest. The approximate total dose to the organism is listed below the figure so that numerical comparisons can be made. In the sea and in deep lakes the dose to small organisms must be evaluated separately from that experienced by large organisms. Circumstances in each domain are given in more detail in Table 3. (See Figure 2 and Table 3.)

Discussion

Small organisms must be considered separately from large ones. Only a small fraction of the energy coming from activity inside a very small organism can be absorbed by the organism, whereas a large organism cannot escape so well from its own radioactivity.

Near the sea surface a large fish receives about half its total natural exposure from the rays originating in the radio-potassium in its own tissues. On the other hand near the sea surface cosmic rays appear to outweigh all other radiations received by a microorganism.

At depths of the order of 100 meters the attenuated cosmic rays no longer contribute significantly to marine organisms either large or small. However, the beta and gamma rays from potassium in sea water can give small organisms doses amounting to about ten per cent of the total dose they receive at the sea surface; the small marine organism cannot escape this exposure to radioactivity in the surrounding water.

It is the deep fresh water which makes possible the most extreme variation in natural exposure. In the deeper waters living things can hide from external bombardment; fresh water generally contains such small amounts of radioactivity that this source can be neglected even in comparison with the feeble effect of cosmic rays remaining at depths of several hundred meters or more.

In pure fresh water the total dose from strongly ionizing rays depends largely upon the size of the organism and upon its living habits. If the organism is small in the sense already discussed, if it lives in deeper waters, if it stays away from the bottom sediments, if it avoids the neighborhood of large masses of living tissue or of detritus, and if it avoids as far as possible accumulating excessive amounts of those elements which can be radio-active — then it can remain remarkably free from the ionizing bombardment received by all other living things.

It would be interesting to find out how the phytoplankton that seek the deeper portion of the euphotic zone of clear lakes respond to their extremely low external dose. If morphological or other differences are discovered between surface specimens and deep-water specimens, then one of the origins of these differences might possibly be the extremely different amounts of strongly ionizing rays in the two biospheres.

Geneticists should not overlook another aspect of the minute cell in feeble radiation; an individual cell has an extremely small probability of being struck at all during one generation. In a deep lake the radiation intensity can be so low that only one phytoplankter in about five hundred would experience an ionizing ray before it divided; at least this is the probability of a cosmic ray hitting an area 0.1 mm square

at 100 meters depth. Furthermore, should an individual plankter accidentally concentrate an excessive amount of radioactive material in its tissue there is little probability that this individual would ever pass along any effect of it; there would be very little chance of a disintegration occuring before division. Purely physical reasoning therefore indicates that mutations leading to a capability for accumulating relatively large amounts of activity might be carried to offspring for ten or more generations before any nuclear energy would be released in any cell whatever.

Because of the "patchiness" of the radiation, the use of a unit like the millirad per year for feeble doses of strongly ionizing radiation unfortunately cannot convey the complete picture of the interesting bombardments which must be experienced by the very small organism.

TABLE 3 Radiations in Eleven Radiological Domains

	Total mrads/year
Man over granite	
1. At 10,000′ elevation	
Cosmic rays 100 + granite 90 + internal 17	= 207
2. At sea surface	
Cosmic rays 35 + granite 90 + internal 17	= 142
Man over sedimentary rock	
3. At sea level	
Cosmic rays 35 + rock 23 + internal 17	= 75
Man over sea	
4. Cosmic rays 35 + sea 0.5 [1] + internal 17	= 52
Large fish in sea	
5. Near surface	
Cosmic rays 35 + sea 0.9 [1] +internal 28	= 64
6. 100 meters deep	
Cosmic rays ½ + sea 0.9 [1] + internal 28	= 30
Micro-organism (mean radius 0.01 mm or less) in water	
7. Near sea surface	
Cosmic rays 35 + sea 3.6 [1] + internal [3]	= 39
8. 100 meters deep in sea or more	
Cosmic rays 0.5 + sea 3.6 [1] + internal [3]	= < 5
9. Buried in deep sea sediments	
Cosmic rays 0.000 + clay 40–620 + internal [4]	= 40–620
10. Near fresh water surface	
Cosmic rays 35 + water activity [2] + internal [2]	= 35
11. 100 meters deep in a fresh lake	
Cosmic rays < 0.5 + water activity [2] + internal [2]	= < 0.5

[1] For every radiopotassium disintegration there are 10 betas having average energy 0.5 mev and also one gamma ray having 1.5 mev. The man receives half the gammas from activity in the sea; the large fish, substantially all the gammas; while the micro-organism receives gammas and betas together.

[2] In fresh water natural activity is extremely low and little of this energy stays in the cell. For example (Robeck et al., 1954) in the Columbia River the beta background of the water is at or below 1×10^{-8} microcurie per ml (2×10^{-4} d/m/g) while the activity of aquatic organisms is at or below 1×10^{-6} microcuries per gram (2×10^{-2} d/m/g). For comparison, the beta activity in normal sea water is 0.66 d/m/g.

[3] The marine microplankton probably carries more internal activity than does the lake plankton, nevertheless effect can be neglected unless activity is concentrated more than 100 fold.

[4] All deep-water organisms have not escaped radiations. Micro-organisms buried in true deep-sea sediments have exceptionally high exposure to radium (Love, 1951); they receive 40-620 mrads/year depending upon the type of sediment.

CONCLUSIONS

1. Some humans actually live under exposure levels surprisingly near the magnitude, 10 roentgen during 40 years, which has been suggested as a genetic tolerance level, i.e., see Figure 2 and Table 2 (domain 1, high elevation over granite).

2. A man may experience 207 mrad/year on high mountains, or 142 on a sandy shore; he may reduce this further by half, say, by staying aboard a ship.

3. A large fish experiences a 50 per cent reduction in dose when going to a depth of 100

meters; it carries along its own source of internal radiation, however.

4. A marine microorganism, having a mean radius of 0.01 mm, receives only about 10 per cent of the surface dose at a depth of 100 meters in the sea; most of the dose comes from sea water activity unless exceptionally high internal activities are accumulated.

5. In a deep fresh water lake those microorganisms living in deep water (but not right at the bottom) receive from their surroundings what is probably the lowest natural ionizing dose within the biospheres of the earth. It would appear that geneticists should consider seeking evidence of abnormal mutation rates amongst microorganisms which live in deep waters of clear lakes, particularly amongst those which have low affinity for radioactive elements.

REFERENCES

BURCH, P. R. J., and F. W. SPIERS. 1954. Radioactivity of the human being. *Science* **120**:719–720.

FRIEDLANDER, G., and J. W. KENNEDY. 1949. Introduction to radiochemistry. J. Wiley and Sons, New York: xiii+412.

GEORGE, E. P. 1952. Progress in cosmic rays. J. C. Wilson, ed. '52 Interscience, North-Holland Publ. Co.: xviii+557.

LIBBY, W. F. 1955. Dosages from natural radioactivity and cosmic rays. *Science* **112** (3158): 57–58.

LOVE, S. K. 1951. Natural radioactivity of water. *Ind. Eng. Chem.* **43**:1541.

REVELLE, R. R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear science and oceanography. International conference on the peaceful uses of atomic energy, Geneva. Paper no. **277**:22.

ROBECK, G. G., C. HENDERSON, and R. C. PALANGE. 1954. Water quality studies on the Columbia River. U. S. Dept. of Health, Education, and Welfare. Robert A. Taft Sanitary Engineering Center; Cincinnati, Ohio: viii+294.

SHERMAN, H. C. 1941. Chemistry of food and nutrition. 6th Ed., McMillan, New York: x+611.

SVERDRUP, H. U., M. W. JOHNSON, and R. H. FLEMING. 1942. The oceans, their physics, chemistry and general biology. Prentice-Hall, Inc.: x+1087.

VINOGRADOV, A. P. 1953. The elementary chemical composition of marine organisms. Trans. Julia Efron and Jane K. Setlow, Sears Foundation for marine research, Yale Univ., New Haven: xiv+647.

Chapter 3

DISPOSAL OF RADIOACTIVE WASTES IN THE OCEAN: THE FISSION PRODUCT SPECTRUM IN THE SEA AS A FUNCTION OF TIME AND MIXING CHARACTERISTICS [1]

Harmon Craig, *Scripps Institution of Oceanography, University of California,* La Jolla, California

I. *Introduction:* Estimated output of nuclear heat and fission products at "steady state" nuclear power production

In two other papers in this report, Wooster and Ketchum discuss mixing rates in the oceans on the basis of oceanographic data, and the present writer reviews the natural isotopic studies which bear on the problem. In this paper we attempt to construct a detailed quantitative picture of the fission product spectrum in the ocean, in steady state with a given fission rate. Such an attempt may well be termed premature, in view of our sketchy knowledge of the internal mixing rate in the sea. Nevertheless, we know a good deal more today than was known five years ago, enough at least to make some simple model calculations which may well yield correct results to an order of magnitude. Moreover, the construction of a model and the calculation of its characteristics are often highly informative, and, at the very least, provide a basis for the orientation of future studies.

The following figures, available in various sources, are pertinent to the estimation of future consumption rates of nuclear power.

Present U. S. electrical energy:

$$6\times10^{8}\ \text{mwh/yr.}$$

Present world electrical energy:

$$10^{9}\ \text{mwh/yr.}$$

Present world energy consumption (all sources) is about 4.5×10^{10} mwh/yr, doubling every 30 years.

For the present calculations, we shall assume a stationary world fission rate of U^{235} equal to 1000 metric tons/yr, supplying all the fission products to be disposed of in the sea. We shall then attempt to construct as reasonable a picture as possible of the fission product activity in the sea, when this activity reaches steady state with the rate of fission, i.e., when the decay rate of each fission product in the sea is equal to the rate at which it is being dumped into the sea, so that its concentration remains constant. We shall also make some calculations for a linear build up to such a fission rate in 50 years.

Since 1 gram of U^{235} is equivalent to 24 mwh, our assumed fission rate of 1000 tons of U^{235} per year is equivalent to 2.4×10^{10} mwh/yr of nuclear heat. At 50 per cent efficiency, this is equivalent to a world nuclear power consumption of 1.2×10^{10} mwh/yr. If this latter figure represents 10 per cent of the total world energy utilization, we are then assuming a world consumption of 1.2×10^{11} mwh/yr, which seems not unreasonable as an estimate for the year 2000 A. D.

Thus a fission rate of 1000 tons of U^{235}/yr represents a 2.7 fold increase in the present world energy consumption, 10 per cent being derived from nuclear heat with 50 per cent efficiency, which should be reached in about the year 2000 based on the present trend in energy consumption (see above). Our calculations will all be linear with the fission rate, so that data for other fission rates are easily derived from the present calculations.

The build up of fission products in a reaactor is given by:

$$\frac{dN}{dt}=fR-\lambda N$$

where f = fission yield (per cent of fissions yielding an individual fission product, the sum equalling 200 per cent), R is the rate of fission (atoms U^{235}/yr) here assumed constant and equivalent to 1000 tons of U^{235}/yr, and N = the

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 902a.

number of atoms of an individual fission product present in the reactor at any time.[2]

Integration with appropriate limits gives the number of atoms of a given fission product in the reactor as a function of time:

$$N=\frac{fR}{\lambda}(1-e^{-\lambda t}) \qquad (1)$$

where the build up factor $(1-e^{-\lambda t})$ varies from 0 to 1 as t varies from 0 to infinity, and gives the fraction of the equilibrium amount attained at any time. At secular equilibrium in the reactor, $dN/dt=0$, and $\lambda N=fR$; we then have:

$$N_{eqlb}=\frac{fR}{\lambda} \qquad (2)$$

from which one sees that at any time in the reactor, $N=N_{eqlb}(1-e^{-\lambda t})$.

The assumed fission rate of 1000 tons U^{235}/yr is equivalent to 2.2×10^6 megacuries of fission (1 curie$=3.7\times10^{10}$ disintegrations/sec), and since the sum of the fission yields is 200 per cent, at equilibrium the total activity of all fission products present in the world, in megacuries, could be roughly estimated by multiplying 4.4×10^6 by the average number of radioactive members per fission chain. The amount of an individual fission product would be fR/λ, using the appropriate decay constant, and its activity would simply be fR, using the appropriate fission yield.

The lengths of the fission chains are difficult to estimate because of the extremely short half-lives of the first members. However, Dr. E. C. Anderson (personal communication) has studied the experimental data on the activity of fission product mixtures directly after fission, and concludes that for times beyond one day after cessation of fission, on the average only $\frac{1}{6}$ of the chains are still active (i.e. from this time on there are left only about 0.3 radioactive members per pair of fission chains initiated). Thus he points out that assuming a fission rate of 1000 tons U^{235}/yr as used above, and taking one day as an assumed minimum delay between accumulation and disposal, the steady activity in the sea for continuous stripping and disposal after one day would be roughly 7×10^5 megacuries. This is about the same total activity as that found below for an average irradiation time of one year with a 100-day cooling period before disposal, namely 7.7×10^5 megacuries (see calculations in Section IV and Table 1). The rough agreement of these numbers merely emphasizes the great predominance of the few long-lived isotopes of high fission yield in the fission product activity after very short times.

II. *Rate of introduction of fission products into the sea*

A more realistic picture is obtained by considering the irradiation time, or reactor holding time for uranium slugs, which is limited by structural weakening from irradiation, poisoning by fission products, etc., and the cooling period necessary for safe handling and for the growing in of plutonium in breeder piles. We assume the fission products of the world are distributed between (1) reactors, (2) cooling pits, and (3) the oceans (or any gross disposal site for that matter). The distribution among these reservoirs and the fission product spectrum in each depends on the irradiation and cooling times.

We shall assume an irradiation time of t_r years, equivalent to any of the following physical interpretations:

1. The reactors of the world are operated, on the average, t_r years, then stripped down and rebuilt.
2. The reactor slugs are continuously pushed through the reactors, each spending, on the average, t_r years in the reactor.
3. Continuous stripping into a holding tank which is opened every t_r years for removal of fission products.

[2] The above equation actually applies only to the first member of a fission chain; for the build up of the second member (y) of a chain with initial member (x), the correct expression is:

$$\frac{dN_y}{dt}=[f_x(1-e^{-\lambda_x t})+f_y]\,R-\lambda_y N_y$$

where f_x and f_y are the individual direct fission yields, and so forth for the succeeding members of each mass number chain. However the decay constants are very large for the first members of a chain, and thus one can neglect the exponential terms and assume a fission yield which is the total yield of the isotope under consideration plus all preceding members of the chain, for all irradiation times with which we shall be concerned. The experimental fission yield figures generally refer to the total chain yield, but because of the very low production of the later members of a chain by direct fission, there is no error involved in applying them to the first significantly long-lived chain member.

From these sources, the fission products are assumed to enter the cooling pits, from which they are dumped into the sea.

At the end of the irradiation time t_r, the amount of a fission product is given by (1) as:

$$N_{t_r}=\frac{fR}{\lambda}(1-e^{-\lambda t_r}) \qquad (3)$$

Assuming for the moment no cooling time, the fission products are stripped out every t_r years and dumped into the sea. Thus the introduction rate into the sea of a given fission product is equal to its activity A_s in the sea at steady state, and is given by N_{t_r}/t_r or:

$$A_{s(t_c=0)}=\frac{fR}{\lambda t_r}(1-e^{-\lambda t_r}) \qquad (4)$$

where t_c denotes the cooling time, here assumed to be 0.

The activity of the fission products in the world reactors at any time, A_r, may be evaluated in the following way. The fission products are stripped out every t_r years, and N_r, the amount in the reactors, varies from 0 to N_{t_r} in cycles, as t varies from 0 to t_r. For many reactors operating independently (the sum of the fission rates being R) with random distribution on the t_r cycle, we take the average of N_r consistent with R by integrating equation (1) from 0 to t_r and dividing by t_r; i.e., the steady state value of N_r is:

$$\bar{N}_r=\frac{1}{t_r}\int_0^{t_r}\frac{fR}{\lambda}(1-e^{-\lambda t})\,dt$$

Performing the integration, and setting $A_r=\lambda N_r$, we have for the steady state activity of a fission product in the reactors of the world:

$$A_r=\frac{fR}{\lambda t_r}[\lambda t_r-(1-e^{-\lambda t_r})] \qquad (5)$$

and from equations (4) and (5) we see that $N_s+N_r=fR/\lambda=N_{eqlb}$, the total amount of the fission product in the world, as of course it must.

Still neglecting cooling time, the fraction of the world total of a fission product which is in the sea is given by $N_s/N_{eqlb}=A_s/A_{eqlb}=F_s$, and:

$$F_{s(t_c=0)}=\frac{(1-e^{-\lambda t_r})}{\lambda t_r} \qquad (6)$$

Neglecting cooling time, the effect of irradiation time may be demonstrated by considering the long and short-lived radioisotopes of strontium, calculating the fraction of the world totals, for the assumed fission rate, which is in the sea, as a function of t_r, as given below.

	F_s (%)	
t_r (years)	Sr^{90} (28y)	Sr^{89} (54d)
0.1	99.9	79.8
0.5	99.4	38.6
1	98.8	21.2
2	97.5	10.7
10	88.5	2.1

Equation (6) shows the following characteristics:

For long half-lives (λt_r small): $F_s=1-\frac{\lambda t_r}{2}$ . . . (approaching 1).

For short half-lives ($\lambda t_r>5$): $F_s=\frac{1}{\lambda t_r}$

For $t_r=1$ year, and for any isotope with a half-life of less than 60 days:

$F_s=0.4t_{1/2}$ (where $t_{1/2}$ is here in days, $t_c=0$).

Thus, as shown above, increasing the irradiation time from 0.1 to 1 year cuts the fraction in the sea of a 60 day isotope by $\frac{1}{4}$, neglecting cooling time effects, but does not affect the long-lived isotopes.

We next interpose the cooling time between the reactor stripping and the disposal in the sea. The amount of an isotope left after the cooling period is:

$$N_c=Nt_re^{-\lambda t_c}$$

and from (4), the steady state activity of a given fission product in the sea, equal to its introduction rate, now becomes:

$$A_s=\frac{fR}{\lambda t_r}(1-e^{-\lambda t_r})\,(e^{-\lambda t_c}) \qquad (7)$$

and F_s is reduced to:

$$F_s=\frac{(1-e^{-\lambda t_r})\,(e^{-\lambda t_c})}{\lambda t_r} \qquad (8)$$

III. *Fission product concentration in the sea as a function of linearly increasing fission rate*

We can get some idea of the transient characteristics of the fission product spectrum in the sea by examining the build-up of fission products with an increasing rate of fission. We

shall take R, the world rate of fission, as 0 at the present time ($t=0$) and increasing linearly from the present time until it reaches the 1000 ton rate in 50 years. We shall further assume *continuous stripping* of fission products into the sea, and examine the transient characteristics of long-lived and a short-lived fission product.

The rate of increase of a fission product in the sea is given by:

$$\frac{dN}{dt}=f\left(\frac{R}{t}\right)t-\lambda N$$

where (R/t) is a constant by virtue of the assumed linear increase from R = 0. N is now the amount of a fission product in the sea at any time t. We thus have:

$$dN+[\lambda N-(fR/t)t]dt=0$$

Multiplication by $e^{\lambda t}$ makes the equation exact, and the solution is:

$$N_t=ce^{-\lambda t}+\left(\frac{fR}{t}\right)\frac{(\lambda t-1)}{\lambda^2}$$

Evaluating the constant from $N=0$ at $t=0$, we have the general solution:

$$N_t=\frac{fR}{\lambda^2 t}[\lambda t-(1-e^{-\lambda t})] \qquad (9)$$

where N_t is the amount in the sea at the time t. Multiplication by λ to give the activity is seen to give an equation of the same form as (5) for the steady state amount in reactors, except that in (9) both R and t are variables, with R/t being constant.

We take $R=0$ at the present time, increasing linearly to 1000 tons U^{235}/year in 50 years. As noted previously, this rate is equivalent to 2.2×10^6 megacuries of fission, and thus $R/t=4.4\times10^4$ megacuries/year. Thus the activity of a fission product in the sea at any time t is given by:

$$A_t=4.4\times10^4\frac{f}{\lambda}[\lambda t-(1-e^{-\lambda t})] \qquad (10)$$

where A_t is in megacuries, $\lambda=\text{yrs}^{-1}$, t is in years, and f is the fission yield. We tabulate below the increasing activity in the sea for a long-lived and a short-lived isotope with continuous stripping into the sea.

Activity (megacuries) in the sea

t (years)	Sr^{90} $t_{1/2}=28y$ $f=0.05$	I^{131} $t_{1/2}=8d$ $f=0.028$
1	26.4	1200
10	2640	12,300
50	4.8×10^4	6.2×10^4
100	1.4×10^5	1.2×10^5
200	3.5×10^5	2.4×10^5
1000	2.1×10^6	1.2×10^6

At 50 years, when the fission rate of 1000 tons/year is reached, the Sr^{90} activity is half the amount which would be in steady state with this fission rate with an irradiation time of 1 year (see below and Table 1). If R continues to increase at the same rate, the steady state Sr^{90} activity for constant R is reached in about 100 years, and thereafter the activity increases linearly at a rate given by: $A_t=2200(t-40)$, the mean life of Sr^{90} being 40 years. The factor $(1-e^{-\lambda t})$ grows in to 95 per cent at 3 mean lives or 4 half-lives.

With a constant fission rate of 1000 tons U^{235}/year, irradiation time one year, and no cooling time, the I^{131} steady state activity in the sea would be 2000 megacuries (calculated as in Table 1, but with no cooling time). With the linear increase of fission rate and continuous stripping as shown above, this level is surpassed in two years. These data illustrate rather strikingly how rapidly the short half-life isotopes build up to secular equilibrium with an increasing fission rate. Sr^{90} does not equal the I^{131} activity until after 100 years of dumping into the sea, under the above conditions. For all species which have grown into secular equilibrium with the increasing fission rate, the activity ratios in the sea are simply given by the fission yield ratios.

IV. *Steady state fission product spectrum in a homogeneous, rapidly mixed sea*

The first three columns of Table 1 list all the fission products of any significance, together with their half-lives and fission yields. Columns 4 and 5 show the total amounts of each isotope in the sea, in metric tons and megacuries of activity respectively, in secular equilibrium with a fission rate of 1000 tons U^{235}

per year (2.2×10^6 megacuries of fission), assuming an irradiation time (t_r) of one year, and a cooling time (t_c) of 100 days (0.274 years). With such conditions, the expression for the activity of each fission product in the sea, as given by equation (7), becomes:

$$A_s=\frac{1}{\lambda}(1-e^{-\lambda})\,(e^{-0.274\lambda})\times 2.2\times10^6 \text{ megacuries} \quad (11)$$

where λ is in years^{-1}.

For half-lives greater than 1 year there is essentially no reduction in the oceanic activity by the cooling time. For all isotopes with half-lives greater than 5 years, more than 90 per cent of the isotope will be in the sea at steady state.

Of the 30 isotopes shown, 22 are independent and 8 are short-lived daughters which come quickly into secular equilibrium with their parents, decaying thereafter with the activity of the parent. Cs^{137} has a branching decay with 8 per cent going directly to the ground state of Ba^{137}; thus the secular activity of Ba^{137m} is only 92 per cent of the parent activity. The activities listed are beta activities only, for all isotopes except Ba^{137m}, Te^{129m}, and Cd^{115m}, which decay from their excited states by gamma emission. The Sm and Eu activities depend on the actual rate of burn-up in the reactors, and may vary considerably with different reactor conditions.

In the calculations, the first long-lived member of each fission chain was taken, and the fission yield for the entire chain was used for this isotope. The direct fission yield for the 11-day Nd which lies above the 2.5-year Pm in the 147 fission chain is not known, and thus this isotope has been neglected; the Nd comes quickly into secular equilibrium in the reactor, so that the total chain fission yield can be used for the Pm calculation.

The fission products are listed in order of decreasing total activity in the sea, with radioactive daughters paired with their parents. The total amount of all fission products in the sea is found to be about 3200 metric tons, corresponding to almost one million megacuries of activity. This represents almost twice the present activity in the sea, which is mainly due to the radioactivity of potassium 40. The figures for K^{40} and Rb^{87} are shown for comparison, the activity of the other radioactive elements in the sea being negligible relative to these isotopes.

We shall now discuss the effects of the mixing barrier at the thermocline in the sea on the distribution of the fission products between the deep sea and the upper mixed layer of the sea.

V. *Distribution of fission products between the deep sea and the mixed layer*

We shall assume a simple model, convenient for calculation, in which we divide the ocean into two geophysical reservoirs: a mixed layer above the thermocline, and the bulk of the ocean, termed the "deep sea," below the thermocline. The exchange of fission products between these two reservoirs is assumed to be a first order process, the rate of removal of a fission product from a reservoir being simply proportional to the amount of the isotope in the reservoir. The thermocline is assumed to represent the boundary across which the hold-up in mixing takes place.

Thus, for example, the rate of transfer of water from the mixed layer to the deep sea is assumed to be k_mN_m, where N_m is the mass of water in the mixed layer and k_m is the exchange rate constant for transfer of material from the mixed layer to the deep sea. In general, we write k_i as the fraction of material in reservoir i removed per year.

The residence time of a molecule in a reservoir, τ, is defined as the average number of years a molecule spends in the reservoir before being removed by the physical mixing process. The meaning of τ may be shown by the following derivation which gives a rigorous definition.

Assume a reservoir with a steady-state fixed content of N molecules of a substance, and a continuous flux into and out of the reservoir of ϕ molecules/year. At a particular time, $t=0$, we have N_0 particular molecules in the reservoir, and at some later time t, we have N' of these original N_0 molecules still present. Then we define the average life of a molecule in the reservoir in the usual way, as

$$\tau=\frac{\Sigma_i t_i n_i}{\Sigma_i n_i}\approx-\frac{1}{N_0}\int_{t=0,\ N'=N_0}^{t=\infty,\ N'=0} t\,dN'$$

where n_i is the number of molecules of the original N_0 which remain in the reservoir for each time t_i, and dN' is the number of mole-

cules removed in the interval t and $t+dt$, i.e., the number of molecules with a reservoir lifetime equal to t.

The number of molecules of the original particular set of N_0 which are removed in any interval dt is simply given by the concentration of such molecules in the reservoir, multiplied by the total flux from the reservoir, i.e.:

$$dN' = -\frac{N'}{N}\phi dt$$

which yields on integration $N = N_0 \exp(-\phi t/N)$.

Substituting for dN' and then for N' in the integral expression for τ, and integrating between $t=0$ and infinity, we obtain:

$$\tau = \frac{N}{\phi}$$

and from the expression for N' one sees that τ, the average life, is also the time required for the original number of N_0 particular molecules to be reduced to $1/e$ times the initial number. τ is thus formally equivalent to a radioactive mean life.

In our particular model we are assuming the rate of removal to be dependent only on the total amount of substance, N, in the reservoir, so that the outgoing flux is given by $\phi = kN$. In such cases we see that $\tau = 1/k$, just as the radioactive mean life is equal to $1/\lambda$. The total removal rate of a radioactive isotope from a reservoir is of course the sum of the physical removal rate and the radioactive decay; τ as defined above refers only to the residence time relative to physical removal.

The symbols used in the following discussion are listed below, where i refers to the subscripts m and d for the mixed layer and deep sea.

N_i, V_i: mass and volume, respectively, of water in reservoir i.

N_i^* = amount of any fission product in reservoir i.

A_i = activity of any fission product in reservoir i ($=\lambda N_i^*$), in megacuries.

a_i = activity of any fission product per unit volume of sea water in reservoir i.

k_i = exchange rate constant, = fraction of material in reservoir i removed per year.

τ_i = residence time in reservoir i relative to physical removal, $= 1/k_i$.

m = average depth of mixed layer of the sea (taken as 100 meters).

D = average depth of the ocean (taken as 3800 meters).

We assume that the fission products are introduced into the deep sea after the 100 day cooling period, the disposal rate or flux of a given fission product isotope, termed ϕ, being equal to the steady-state total activity in the sea A_s as given by equation (11). ϕ is thus in "megacuries of flux," = atoms/sec divided by 3.7×10^{10}. We wish to ask what steady-state activity per unit volume of water will be in the mixed layer, as a function of the rate of cross-thermocline exchange of sea water and fission products.

The water balance between the reservoirs is given by:

$$k_m N_m = k_d N_d$$

or, neglecting density differences which are not important for these calculations,

$$\frac{N_d}{N_m} = \frac{\tau_d}{\tau_m} \approx \frac{D-m}{m} \qquad (12)$$

The fission products are introduced into the deep sea with a rate of introduction for any give isotope ϕ. The radioactive balance in the two reservoirs is then given by:

Deep sea: $\phi + k_m N_m^* = k_d N_d^* + \lambda N_d^*$ (13)

Mixed layer: $k_d N_d^* = k_m N_m^* + \lambda N_m^*$ (14)

Total: $\phi = \lambda(N_m^* + N_d^*) = A_s = A_m + A_d$ (15)

From (12) and (14)

$$\frac{N_d^*}{N_m^*} = \frac{\tau_d}{\tau_m} + \lambda\tau_d$$

or:

$$\frac{A_d}{A_m} = \frac{D-m}{m} + \lambda\tau_d$$

Thus for a stable element ($\lambda = 0$) the partitioning is simply statistical. From (15):

$$A_m = \frac{\phi}{D/m + \lambda\tau_d} \qquad (16)$$

which gives the total activity of any fission product in the mixed layer as a function of decay constant, relative sizes of the mixed layer and deep sea, and exchange rate between the reservoirs as given by τ_d.

Various estimates of the value to be assigned to τ_d may be obtained from the separate papers by Wooster and Ketchum, and by Craig, in this report, and are discussed in relation to this

particular model in the paper by Craig. From these discussions, we choose for the present calculations a value $\tau_d = 300$ years as perhaps the best guess. As discussed by the writer in a separate chapter of this report, radio carbon data indicate a residence time for water of about 1000 years, as a world-wide average. Mixing in the Atlantic is probably a good deal faster than in the Pacific, and 300 years is probably a safe lower limit estimate for the Atlantic, considering the material to be deposited on the bottom. Thus the mixed-layer activities we calculate should be upper limits, which would be approached more closely in the Atlantic than in the Pacific.

The average world-wide depth of the mixed layer, m, is taken as 100 meters, and the average depth of the sea is taken as 3800 meters. The volume of the sea is 1.4×10^{21} liters; thus the volume of the mixed layer is taken as 1/38 of this or 3.7×10^{19} liters. Putting these numerical values into (16), and noting that $\phi = A_s$, we have for the activity of any fission product per unit volume of sea water in the mixed layer:

$$a_m = \frac{10^{-3} A_s}{300\lambda + 38} \text{ dps/liter} \qquad (17)$$

in disintegrations per second per liter, where A_s is in megacuries, as tabulated in column 5 of Table 1, and λ is in years^{-1}. From this equation the values tabulated in column 8 of Table 1 were calculated, and were converted to microcuries per liter for column 9.

From the relation $a_d/a_m = (A_d V_m / A_m V_d) = (A_d/A_m)\,(m/D\text{-}m)$ we obtain:

$$\frac{a_d}{a_m} = \lambda\tau_m + 1$$

where τ_m, the residence time of a water molecule in the mixed layer, is given by (12) as 1/37 of $\tau_d = 8.1$ years. We thus write:

$$\frac{a_d}{a_m} = 8.1\lambda + 1 = \alpha \qquad (18)$$

from which, given the values of a_m computed above, the values of a_d tabulated in column 7 of Table 1 were computed. We call α the "oceanographic partition factor." It is a function of the mixing rate of the sea and the decay constant of the individual isotope, and is a measure of the effectiveness of the cross-thermocline exchange rate in buffering the mixed layer from the fission products introduced into the deep sea. Values of α are tabulated in column 6 of the table, and range from about 1 for the longest lived isotopes to about 250 for an isotope with a half-life of 8 days. For stable isotopes λ is 0, α is 1, and (18) reduces to simple statistical partitioning.

From (17) we see that as λ, the decay constant of an isotope, increases, the activity in the mixed layer decreases; i.e., if more of the isotope can be removed from the deep sea by decay, less needs to be transferred to the mixed layer to preserve the steady state. If the half-life were so long that the radioactivity did not affect the distribution between the mixed layer and the deep sea, we would have simply a statistical partitioning of the isotope between these reservoirs, such that the activity per unit volume in each reservoir would be the same. From the above equations we can derive the ratio of the activity in the mixed layer for an isotope to the activity per unit volume which would be observed if the partitioning were statistical:

$$\frac{a_m}{a_m(stat)} = \frac{\tau_d + \tau_m}{\alpha\tau_d + \tau_m} \approx \frac{1}{\alpha} \qquad (19)$$

and we see that α^{-1} is approximately the fraction of the statistical activity per unit volume attained by a fission product in the mixed layer. Equation (19) can be written exactly as:

$$\frac{a_m}{a_m(stat)} = \frac{t_{1/2}}{t_{1/2} + 5.5} \qquad (20)$$

where $t_{1/2}$ is the half-life of the isotope in years. The ratio $a_m/a_m(stat)$ is plotted in Figure 1 as a function of the half-life, and one reads, for example, that an isotope with a 5 year half-life attains about 48 per cent of the activity per unit volume in the mixed layer which it would have if its half-life were so long, relative to the mixing rate in the sea, that its radioactivity had no effect on its distribution.

The values of a_m, a_d, and α are tabulated in Table 1, in which the isotopes are arranged in order of their activity in the deep sea. For comparison, the activities of potassium 40 and rubidium 87, which provide essentially all the radioactivity in the sea, are also listed. In the deep sea, the predicted fission product activity is 19.3 disintegrations per second per liter, as compared with the natural activity of 12.2 dps/liter; thus the fission products in steady state with the 1000 ton fission rate would almost triple the deep-sea activity.

TABLE 1 FISSION PRODUCT SPECTRUM IN THE OCEAN AT STEADY STATE DISPOSAL INTO DEEP SEA. CALCULATED FOR FISSION RATE OF 1000 TONS U^{235}/YR (2.4×10^{10} MWH/YR OF NUCLEAR HEAT), IRRADIATION TIME OF 1 YR AND COOLING TIME OF 100 DAYS. AVERAGE LIFE OF A WATER MOLECULE IN THE DEEP SEA TAKEN AS 300 YEARS; AVERAGE DEPTH OF THE MIXED LAYER TAKEN AS 100 METERS.

			Total amount in ocean			Activity (dps/liter)		
Isotope	Half-life	Fission yield %	Metric tons	Activity megacuries	$\alpha = a_d/a_m$	a_d Deep sea	a_m Mixed layer	a_m Microcuries per liter
$_{55}Cs^{137}$	33 y	6.3	1750	1.4×10^5	1.17	3.64	3.12	8.4×10^{-5}
$_{56}Ba^{137m}$	2.6 m	—	—	1.3×10^5	—	3.35	2.87	7.7×10^{-5}
$_{38}Sr^{90}$	28 y	5.0	780	1.1×10^5	1.20	2.90	2.42	6.5×10^{-5}
$_{39}Y^{90}$	64 h	—	0.20	1.1×10^5	—	2.90	2.42	6.5×10^{-5}
$_{58}Ce^{144}$	280 d	5.3	19	6.0×10^4	8.32	1.62	0.19	5.2×10^{-6}
$_{59}Pr^{144}$	17.5 m	—	—	6.0×10^4	—	1.62	0.19	5.2×10^{-6}
$_{61}Pm^{147}$	2.5 y	2.6	48	4.6×10^4	3.24	1.23	0.38	1.0×10^{-5}
$_{62}Sm^{151}$	100 y	0.7	630	1.5×10^4	1.06	0.40	0.38	1.0×10^{-5}
$_{40}Zr^{95}$	65 d	6.4	0.58	1.2×10^4	32.5	0.33	1.0×10^{-2}	2.7×10^{-7}
$_{41}Nb^{95}$	36 d	—	0.32	1.2×10^4	—	0.33	1.0×10^{-2}	2.7×10^{-7}
$_{39}Y^{91}$	60 d	5.9	0.39	9.4×10^3	35.1	0.25	7.2×10^{-3}	1.9×10^{-7}
$_{44}Ru^{106}$	1 y	0.5	2.0	6.6×10^3	6.61	0.18	2.7×10^{-2}	7.2×10^{-7}
$_{45}Rh^{106}$	35 s	—	—	6.6×10^3	—	0.18	2.7×10^{-2}	7.2×10^{-7}
$_{38}Sr^{89}$	54 d	4.6	0.22	6.0×10^3	38.9	0.16	4.1×10^{-3}	1.1×10^{-7}
$_{44}Ru^{103}$	40 d	3.7	7.2×10^{-2}	2.3×10^3	52.0	6.1×10^{-2}	1.2×10^{-3}	3.2×10^{-8}
$_{45}Rh^{103}$	55 m	—	—	2.3×10^3	—	6.1×10^{-2}	1.2×10^{-3}	3.2×10^{-8}
$_{58}Ce^{141}$	32 d	5.7	6.3×10^{-2}	1.8×10^3	65.0	4.9×10^{-2}	7.6×10^{-4}	2.0×10^{-8}
$_{63}Eu^{155}$	2 y	0.03	0.44	5.1×10^2	3.8	1.4×10^{-2}	3.6×10^{-3}	9.7×10^{-8}
$_{52}Te^{129m}$	33 d	0.3	3.4×10^{-3}	1.0×10^2	63.4	2.8×10^{-3}	4.4×10^{-5}	1.2×10^{-9}
$_{52}Te^{129}$	70 m	—	—	1.0×10^2	—	2.8×10^{-3}	4.4×10^{-5}	1.2×10^{-9}
$_{59}Pr^{143}$	13.7 d	5.4	—	40	151	1.1×10^{-3}	7.2×10^{-6}	2.0×10^{-10}
$_{56}Ba^{140}$	12.8 d	6.1	—	30	161	8.0×10^{-4}	5.0×10^{-6}	1.3×10^{-10}
$_{57}La^{140}$	1.7 d	—	—	30	—	8.0×10^{-4}	5.0×10^{-6}	1.3×10^{-10}
$_{50}Sn^{123}$	130 d	1.2×10^{-3}	—	6.8	16.8	1.8×10^{-4}	1.1×10^{-5}	3.0×10^{-10}
$_{48}Cd^{115m}$	44 d	8×10^{-4}	—	0.64	47.4	1.7×10^{-5}	3.6×10^{-7}	9.8×10^{-10}
$_{53}I^{131}$	8 d	2.8	—	0.35	256	9.5×10^{-6}	3.7×10^{-8}	1.0×10^{-12}
$_{63}Eu^{156}$	15.4 d	0.01	—	0.15	134	4.0×10^{-6}	3.0×10^{-8}	8.0×10^{-13}
$_{55}Cs^{136}$	13.7 d	0.01	—	7.5×10^{-2}	151	2.0×10^{-6}	1.3×10^{-8}	3.6×10^{-13}
$_{50}Sn^{125}$	10 d	0.02	—	1.7×10^{-2}	206	4.7×10^{-7}	2.3×10^{-9}	6.2×10^{-14}
$_{47}Ag^{111}$	7.6 d	0.018	—	1.4×10^{-3}	268	3.9×10^{-8}	1.4×10^{-10}	3.9×10^{-15}
Totals:			3230	7.7×10^5		19.3	12.1	3.2×10^{-4}
Natural potassium and rubidium in the sea:								
K^{40}			6.3×10^{10}	4.6×10^5	1	12	12	3.2×10^{-4}
Rb^{87}			1.2×10^{11}	8.4×10^3	1	0.22	0.22	5.9×10^{-6}

All activity values are Beta activities only, except where isomeric transitions are indicated.

Conversion: 1 disintegration per second $= 2.7 \times 10^{-5}$ microcuries.
1 curie $= 3.7 \times 10^{10}$ disintegrations per second.

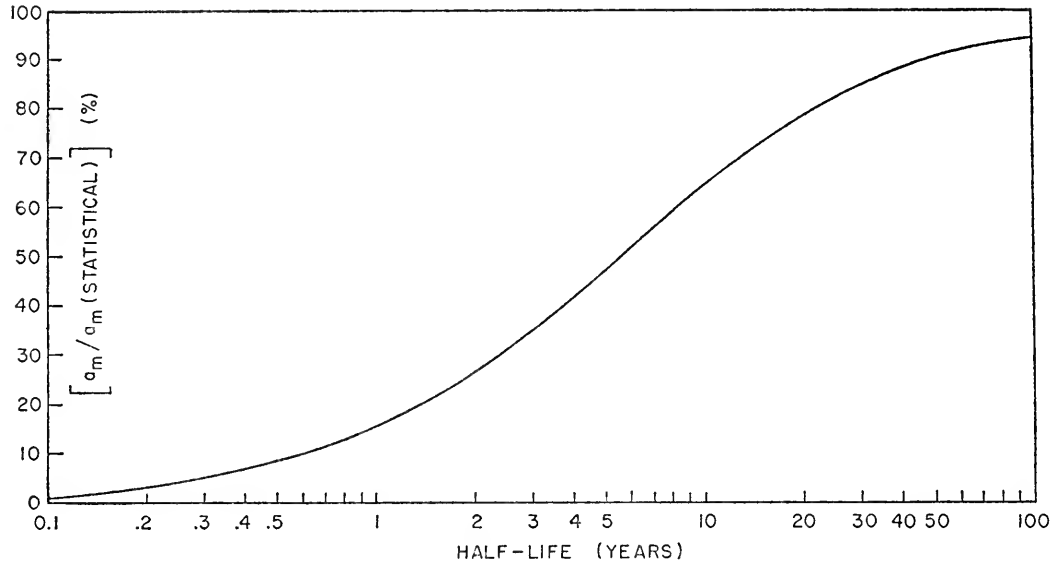


FIGURE 1

However, the effect of the internal mixing rate of the sea in the model adopted, is to cut the activity in the mixed layer down to 12.1 dps/liter which is, by coincidence, just equal to the natural activity and which would thus just double the activity in the mixed layer.

It should be noted that the figures given in the table for the predicted activities in the mixed layer refer only to cross-thermocline mixing by physical processes, exclusive of biological transfer through the thermocline. However, the figures listed provide a basis for speculation on the hazardous effects of the mixed layer activity, in that comparison may be made with biological concentration factors, discussed elsewhere in this report, to predict the activity levels in marine organisms. In this way, rough predictions may be made of the hazard to man, not only by direct exposure to the waters of the mixed layer of the sea, but by the activity concentrated in marine organisms used for food.

CHAPTER 4

TRANSPORT AND DISPERSAL OF RADIOACTIVE ELEMENTS IN THE SEA [1]

WARREN S. WOOSTER, *Scripps Institution of Oceanography,* La Jolla, California

and

BOSTWICK H. KETCHUM, *Woods Hole Oceanographic Institution,* Woods Hole, Massachusetts

THE FATE of radioactive elements in the sea differs from that of non-radioactive elements since they are subject to radioactive decay. Otherwise, concentrations of radioactive elements are changed by the same physical and biological processes as are those of other isotopes in the same physical state. Thus the fate of radioactive material introduced into the sea depends on:

1. What is introduced — the nuclide, its radioactive properties (half-life, nuclear reaction, kind and energy of radiation), its physical state in sea water (whether particulate, colloidal or ionic) and its chemical properties (including its role in biological processes).
2. Where it is introduced — position and depth with respect to the density and velocity structure of the sea.

This paper describes the physical processes whereby radioactive elements in true solution are diluted by mixing and are carried from one part of the ocean to another. Although all parts of the open ocean appear to be in continuous motion and in communication with each other, the rates of this motion and exchange cover such a wide range that it is convenient to consider separately the questions of near-surface vertical and horizontal exchange, intermediate and deep circulation, and the exchange between the deep sea, coastal areas and enclosed basins.

Near-surface circulation

In middle and low latitudes the surface layer of the ocean, from 10 to 200 meters thick, is separated from the colder deep waters by a layer of rapid density change and great stability, the pycnocline or thermocline. This intermediate layer varies in depth and stability from time to time and from place to place. At times there are two such layers, the seasonal thermocline and a deeper main thermocline. The surface layer is often called the "stirred" or "mixed" layer [2] because of its relative uniformity in temperature and in concentrations of dissolved substances.

It is believed that radioactive material introduced into this surface layer will be rapidly distributed vertically throughout the layer. The general uniformity of concentrations within this layer suggests that forces are present which tend to bring it about. Because density increases only slightly with depth through the layer, little energy is required for vertical stirring.

Some evidence of the rapidity of vertical mixing in the upper layer is given by Folsom (Revelle, Folsom, Goldberg and Isaacs, 1955), who observed that when fission products were introduced at the surface in an area where the surface layer was about 100 meters thick, the lower boundary of the radioactive water reached the bottom of this layer in about 28 hours. Within this period of time radioactivity had become uniformly distributed vertically throughout the layer.

Rapid vertical mixing in the upper layer is brought about primarily by the following two processes:

1. Convection: When the density of surface water is sufficiently increased, owing to either

[1] Contribution from the Scripps Institution of Oceanography, New Series, no. 903. Contribution no. 870 from the Woods Hole Oceanographic Institution. This paper, in part, represents results of research carried out by the University of California under contract with the Office of Naval Research. Reproduction in whole or in part is permitted for any purpose of the United States Government.

[2] A distinction is made here between stirring and mixing. In stirring, one causes relative motion of different parts of the liquid, and the average value of the gradient is increased. Mixing then takes place, the gradients disappearing and the liquid becoming homogeneous (Eckart, 1948).

a decrease in temperature or an increase in salinity, the surface water sinks and mixes with deeper water. Convection is maintained by (a) surface cooling due to long wave radiation and heat conduction to the atmosphere, (b) the loss of latent heat and water vapor in evaporation, or (c) the increase of surface salinity from freezing of surface water.

2. Wind stirring: Vertical turbulence in the upper layer results from wind action on the sea surface. The extent of wind stirring depends both on the magnitude and uniformity of wind stress and on the vertical density gradient. Stirring is effective only to a depth where there is sufficient energy to overcome the effect of stability. Both the homogeneity and the depth of the upper layer are affected by wind. Single gales have been observed to deepen the surface layer on the average by about 20–30 feet, (Francis and Stommel, 1953).

Rapid vertical mixing may be brought about by other processes. Thus in shallow coastal areas stirring by strong tidal currents is important. Stirring may also be accomplished by the vertical component of currents, particularly in regions of upwelling and sinking.

It should be noted that even above a well-developed pycnocline there is not complete homogeneity within the so called "mixed" layer. Concentrations of those elements affected by biological activity (such as oxygen and phosphorus) may show significant variation within the euphotic zone. Even so-called "conservative" concentrations (temperature and salinity) may not be uniform within the surface layer. Such heterogeneity may be attributed to incomplete vertical mixing or to vertical shear in the surface layer.

When radioactive materials are introduced into the near-surface layer, they are transported away from the area of introduction by surface currents. These currents extend, in general, through the entire depth of the upper layer and seem to be driven, directly or indirectly, by the wind.

The average locations and velocities of the important surface currents of the world ocean have been studied for many years and are well known (see, for example, Deutsche Seewarte, 1942; U. S. Navy Hydrographic Office, 1947 a and b, 1950; Sverdrup, Johnson, and Fleming, 1942, ch. 15). This knowledge comes primarily from averages of countless ship-drift observations, and from computations based on the observed subsurface distribution of density.

These calculations give mean speeds as high as 193 cm/sec (90 miles per day) in the Florida Current (Montgomery, 1938a) and 89 cm/sec (41 miles per day) in the Kuroshio (Koenuma, 1939). The volume of water flowing through the Florida Straits in 15 years is about equal to that of the upper 500 meters of the whole North Atlantic. Similarly, between the northern Ryukyus and Kyushu, the Kuroshio transports a volume equivalent to that of the upper 500 meters of the North Pacific in about 50 years. It seems likely that there is no area of surface water in the ocean that can be considered as isolated from the remaining surface waters.

Recent intensive studies of the Gulf Stream and other surface currents, using such modern instruments as the bathythermograph, electronic navigational aids, and geomagnetic electro-kinetograph (GEK), have revealed complicated fine structures, with filamentous jets and countercurrents not apparent in the average picture (Fuglister, 1951). Characteristic maximum surface velocities measured by GEK and Loran dead reckoning in the Gulf Stream were found to fluctuate between 150 and 300 cm/sec or 70 to 140 miles per day (von Arx, Bumpus and Richardson, 1955). Thus, in estimating the time at which radioactive materials will be found at various distances from the area of introduction, one must be cautious in the use of average surface current speeds.

Direct evidence of the transport of radioactive materials by surface currents in the western Pacific is given by "Shunkotsu-Maru" survey (Miyake, Sugiura and Kameda, 1955) and the "Taney" survey (U. S. Atomic Energy Commission, 1956) four months and thirteen months respectively after nuclear weapons tests in the Marshall Islands in March, 1954. The earlier survey found significant levels of radioactivity at a distance of 2000 kilometers from Bikini, suggesting a westward drift of more than 9 miles per day (about 20 cm/sec). The later survey found significant levels of radioactivity at least 7000 kilometers downstream from Bikini; this gives about the same minimum westward drift.

In addition to being drifted away from the area of introduction, radio-active materials are

dispersed by diffusion. Diffusion in the ocean is caused by turbulence or eddies, and the coefficient of eddy diffusivity is usually more than a million times the corresponding molecular coefficient. The rate of eddy diffusion depends on wind speed, current shear, density gradient, gradient of the diffusing concentration, direction of diffusion, and the dimensions of the phenomenon. The calculated rates depend upon the magnitudes of eddy diffusivity coefficients used, and they have been estimated by a number of methods (Sverdrup et al., 1942, p. 484–485; Munk, Ewing and Revelle, 1949).

Because of both the large number of variables concerned and the present unsatisfactory state of our quantitative knowledge of turbulence in the ocean, it is difficult to predict the diffusion of radioactive materials under any given circumstances. The most satisfactory approach at present is to conduct diffusion studies and experiments at the place and under the conditions of contemplated release. The results are only applicable to the particular areas.

During the 1946 preliminary survey in Bikini Lagoon, the state of turbulence was determined by a variety of measurements, and the subsequent observed distribution of radioactivity was in close agreement with the predicted values (Munk, Ewing and Revelle, 1949). A mean value for the radius of the contaminated area was 3 km., which approximately doubled between the first and second days after the burst. The initial distribution of radioactivity as deposited by the atomic bomb was patchy, and the turbulent eddies, which spread the contamination over a larger area, did not appreciably reduce this patchiness during the first three days.

Another pertinent study was made by Ketchum and Ford (1952) who examined the rate of dispersion of acid-iron wastes in the wake of a barge at sea. Computed mixing coefficients showed a tendency to increase with increasing time, and thus with the dimensions of the mixing field, and the radius of the contaminated area was observed to double in time periods ranging from 0.5 minutes to 35 minutes. It should be noted that the scale of this phenomenon was about 10^{-2} that of Munk, Ewing and Revelle (1949); they show that the ratio of lateral eddy diffusivity coefficient to the radius of the area considered is relatively constant over a range of radius between 10^3 and 10^8 cm.

A large scale tracer experiment was carried out in the Irish Sea prior to the discharge of radioactive effluent (Seligman, 1955). During each experiment, 10 tons of 6.7 percent fluorescein solution were introduced near the surface during a 20-minute period, and the sensitivity of subsequent detection was believed to be of the order of 1 part in 10^9. Maximum concentrations detected directly after release were 10^{-4} of the original concentration; 12 hours after release, they were down to 5×10^{-7} of the original concentration. The trial area was probably part of an eddy and was subject to tidal mixing, so the results may not be generally applicable.

Exchange between near-surface and intermediate waters

Since the surface layer is separated from deeper waters by a layer of rapid density increase, and hence of great stability, vertical transfer of materials across this layer by eddy diffusion must be much less rapid than is vertical diffusion in the upper layer. Thus radioactivity introduced at the surface by fallout may remain in the upper layer for a long time and be diluted by only a small part of the total volume of the sea. Conversely, radioactive materials introduced below the pycnocline should only slowly contaminate the upper layer where they are most likely to endanger human activities. However, organisms and particles of sufficient density may readily cross the pycnocline, due both to gravity and to vertical migrations.

There are few observations which show directly the existence of cross-pycnocline exchange on a local scale. In the western Pacific, both the "Shunkotsu-Maru" survey (Japanese Fishery Agency, 1955) and the "Taney" survey (U. S. Atomic Energy Commission, 1956) reported patches with significant concentrations of radioactivity below the thermocline four months and thirteen months, respectively, after mixed fission products were introduced at the surface in the Marshall Island area. It is not known, however, whether this exchange was effected by mixing processes, or by particulate or ecological processes.

Exchange of properties between the near-

surface and deeper waters is most likely to take place under the following conditions:

1. In regions where the pycnocline is sufficiently shallow to be eroded at the top by wind stirring. In coastal waters the pycnocline is usually shoaler than in midocean, and shallow pycnoclines may also be found in high latitudes, at the equator, along the north edge of the Equatorial Countercurrent, and at the center of strong cyclonic eddies. This process is not effective to great depths, but could serve to bring radioactive materials into the surface waters from the pycnocline layer.

2. In regions of upwelling, where the pycnocline is relatively weak and where vertical currents not only carry water toward the surface but also stir surface and deeper waters. It is unlikely that water from depths of more than 500 meters is ever brought to the surface by this process. Upwelling is common along western coasts of continents in the trade wind belt, such as the coasts of Peru and Northern Africa. In a simple sense, the persistent trade winds blowing parallel to or offshore develop an offshore component of transport in the surface waters, and deeper waters upwell to maintain the volume continuity. Upwelling may also occur along other coasts when the winds are suitable. The process has been extensively studied along the coast of California where it is not continuous because of the variability of the winds (Sverdrup et al., 1942, p. 725). The speed of coastal upwelling has been variously estimated as 0.6 m/day (McEwen, 1934), 2.25 m/day (Saito, 1951) and 2.7 m/day (Hidaka, 1954). However, since these estimates are theoretical mean values, they may differ significantly from actual instantaneous upwelling rates.

Midocean upwelling, associated with divergence of the surface currents, occurs in a band along the equator in the eastern and central Pacific Ocean (Cromwell, 1953). Observations indicate that the effects of this upwelling extend to 50 meters in the eastern Pacific and to 100–150 meters in the central Pacific (Wooster and Jennings, 1955). Similar but less pronounced upwelling has been observed in the equatorial Atlantic (Bohnecke, 1936).

3. In regions of surface convergence, where sinking waters may fill the depths of the ocean, or may spread at intermediate depths according to their density. In tropical and temperate latitudes such sinking is confined to the surface layer. In such regions mixing in the upper layer may be facilitated but exchange across the pycnocline probably is not, since the sinking water tends to increase the density gradient in the pycnocline.

In high latitudes, on the other hand, sinking waters may reach great depths, and it is in such regions that most of the intermediate and deeper water masses of the ocean are formed. The most extensive and pronounced of these convergences is the Antarctic Convergence which occurs at 50 to 60°S in a band around the entire Antarctic Continent. The cold, low-salinity water which sinks there forms an identifiable water mass, the Antarctic Intermediate Water, which spreads at depths between 800 and 1200 meters in all southern oceans. This water can be identified everywhere in the South Atlantic and extends across the equator as far as 22°N in the North Atlantic (Deacon, 1933; Iselin, 1936).

In the Irminger Sea, between Iceland and Greenland, and in the Labrador Sea, warm high salinity water of the Gulf Stream is partly mixed with cold low-salinity water flowing out of the Arctic Ocean. The resulting mixture may spread in small quantities as Arctic Intermediate Water, or when sufficiently dense may form the deep and bottom water of the North Atlantic (the possibility that the formation of this deep water is not a continuous process is discussed later). Intermediate waters of the North Pacific are probably formed in winter at the convergence between the Kuroshio Extension and the Oyashio (Sverdrup et al, ch. 15). There is apparently no deep or bottom water formed by this process in the Pacific.

4. In regions where the density of surface waters is so increased by evaporation, cooling or freezing, that they sink to intermediate or greater depths. Active formation of Antarctic Bottom Water takes place in the Weddell Sea due to the freezing of high salinity surface waters. In the Mediterranean and Red Seas, bottom water is formed by winter cooling of waters whose salinity has been greatly increased by evaporation. Mediterranean water flows out into the North Atlantic at depths of 1000 to 1500 meters and can readily be identified near Bermuda, 2500 miles from its source.

In summary, exchange between near-surface and deeper waters takes place most commonly (1) in high latitudes, (2) along the equator,

and (3) in coastal regions, particularly along the western coasts of continents. Conversely, such exchange is least likely in temperate and tropical latitudes in the vast central regions of the northern and southern oceans.

Exchange between the open sea and coastal areas

In coastal areas or enclosed basins where precipitation exceeds evaporation, there is a seaward surface drift of diluted water and a landward subsurface drift of water derived from the open sea. If radioactive materials were released in such a coastal area, the material which remained in the surface layer would be carried seaward, but the part of the material which mixed or settled to the deeper water would move toward shore and the estuaries of rivers. Conversely, if radioisotopes were liberated in the open sea, some would eventually be carried inshore as a result of the coastal and estuarine circulation.

It is clear that the ultimate distribution in coastal areas of radioactive materials added to the sea would depend on the location of the release, the vertical distribution of radioactivity and density in the area of release, the length of time required for the transport to the coastal area or estuary, and the location of the source sea water which provides for the counter drift. The number of variables involved makes it difficult to discuss the effects in general terms, but it is worthwhile to note that the circulation in coastal areas is rapid, and water bathing the North Atlantic beaches is not uncommonly 90 per cent sea water even off large rivers such as the Hudson and Delaware.

An idea of the lengths of time involved in the coastal circulation can be obtained from the mean age of waters in various parts of the Atlantic seacoast. Such mean ages are computed from the volume of water contained in the region and the estimated transport of water through the region. The waters of the continental shelf from Cape Hatteras to Cape Cod have a mean age of about $2\frac{1}{2}$ years, those of the Bay of Fundy about 3 months, and those of Delaware Bay from the ocean to the height of tide about 3–4 months (Ketchum and Keen, 1953, 1955). The source sea water for all of these circulations is the "slope water" which is formed between the Gulf Stream and the edge of the continental shelf.

A few data are available for confined basins and seas from which estimates of the mean age of the water can be derived. In most cases, however, the sources of water entering into the circulation are uncertain, and it should be emphasized that in all cases some of the waters within the basin will be older or younger than the mean age.

The source waters of the Florida Current are funnelled through the Caribbean Sea. The mass transport is 26 million cubic meters a second (Sverdrup et al., 1942, p. 638), so that this current carries annually a volume of water equivalent to one-sixth of the total volume of the Caribbean. However, there is evidence that the renewal of the deep water of the Caribbean proceeds at a much slower rate than the six year mean age that this ratio implies. Worthington (1955) has calculated, on the basis of loss of oxygen from this deep water during the last 30 years, that the age of the deep water in the various parts of the Caribbean may range from 93–142 years. The mean age of the waters above 2000 meters would be reduced to about 5 years if the deepest $\frac{1}{5}$ of the volume of the basin is isolated from the present circulation.

The same current passes through the Yucatan channel into the Gulf of Mexico, before emerging as the Florida Current. No estimate of the mean age of the waters of the Gulf of Mexico is possible, however, since the current data in the Gulf indicate an anticyclonic eddy in the western portion, and suggest that the waters of the Gulf of Mexico are drawn into the Florida Current to only a slight extent (Dietrich, 1939, Sverdrup et al., 1942, p. 642).

The Black Sea probably contains the most isolated and the oldest deep water to be found anywhere in the oceans. Precipitation and runoff exceed evaporation, and the surface waters are dilute (salinities less than 18 per cent) and isolated from the deep water by an intense density gradient. The deep waters are anaerobic; hydrogen sulfide reaches large concentrations below about 200 meters. The sill at the Bosporus is only 90 meters below the surface so that this deeper water is isolated from the more rapid surface circulation. The inflow of sea water is so small that it would take about 2500 years to replace the deep water in the basin (Sverdrup et al., 1942, p. 651). The mean replacement time for the surface layers

to a depth of 200 meters is equivalent to about 200 years. Gololobov (1949) has computed the mean age of the deep water on the basis of the annual contribution of phosphorus in the river inflow and the quantity accumulated in the depths. This computation indicates an accumulation time of 5600 years.

The Arctic Basin receives its major inflow north of Scotland and a much smaller inflow through the Bering Strait. Additional sources are from the river runoff and excess of precipitation over evaporation. The outflow is primarily through the Denmark Strait (Sverdrup et al., 1942, p. 655). These flows would provide a volume equal to that of the Arctic Ocean in about 160 years. The Arctic is also stratified because of the addition of fresh water from rivers and melting ice, and it is not known how isolated some of the waters in the deeper basins may be. However, recent analyses have shown that the deeper water in the Arctic Ocean is far from anaerobic, so that it seems unlikely that this water can be considered as isolated from the circulation.

The Mediterranean is a basin in which evaporation exceeds precipitation and runoff. Through the Strait of Gibralter there is an inflow of oceanic surface water and a subsurface outflow of high salinity Mediterranean water. The exchange is sufficiently rapid to replace the entire Mediterranean in about 75 years (Sverdrup et al., 1942, p. 647). The Mediterranean is divided into eastern and western basins by a 500-meter sill between Sicily and Tunisia, and it is not know to what extent the deep waters of these basins are involved in the over-all exchange.

Deep circulation

Most of our present knowledge of the intermediate and deep circulation (see Sverdrup et al., 1942, ch. 15) has been obtained indirectly from the observed distribution of properties. The general uniformity of temperature and dissolved substances in deep water suggests that deep currents are very slow, perhaps at most a few centimeters per second. But deep currents cannot be computed by the geostrophic method because only relative velocities can be thus obtained. Furthermore, small errors in the measurement of salinity or temperature produce uncertainties in velocity of the same magnitude as the currents being computed. The direction of movement in the deep and bottom water has been deduced from the observed distribution of properties such as salinity and potential temperature, but little can be learned about current speeds from such observations.

Existing direct measurements of subsurface currents have been summarized by Bowden (1954). Such measurements have been made since the time of the CHALLENGER Expedition (1873–76), but because of practical difficulties (such as the problem in the open sea of referring observations to a fixed frame of reference) they have taught us little about the deep oceanic circulation. The few successful measurements at depths greater than 1000 meters reported by Bowden showed mean speeds ranging from "negligible" to about 13 cm/sec. At nearly all stations and depths at which current measurements have been made, semidiurnal tidal currents of the order of 10 cm/sec. have been recorded.

Recently measurements of subsurface currents have been made in the North Atlantic by tracking for three days a neutral-buoyant float stabilized at a given depth (Swallow, 1955 and unpublished). These measurements show small resultant speeds (1.7 to 9.1 cm/sec or 0.8 to 4.2 miles/day at depths from 600 to 1900 meters), tidal components of about 10 cm/sec, and in two successive three-day measurements at 1900 meters, a change in direction of 124°. Thus it seems likely that motion below the pycnocline is characterized by more variation, periodic or otherwise, than previously supposed and indeed that the mean drift may represent only a small part of the total motion.

Little is known about the nature and extent of lateral and vertical mixing in the deep sea. It is generally believed, however, that flow and mixing take place along surfaces of constant potential density (isentropic surfaces) and that below the upper layer vertical mixing is very slow except near coastlines and areas where upwelling may occur (Montgomery, 1938). An observation supporting this belief was reported by Revelle, et al. (1955). Introduction of mixed fission products below the pycnocline led to the formation of a lamina of high radioactivity about one meter thick and 100 or more square kilometers in area. The radioactive water apparently spread out along an isentropic surface and resisted destruction by vertical mixing for at least three days.

Age of intermediate and deep waters

It is generally accepted that intermediate and deep waters in most parts of the oceans acquired their characteristics while at or near the surface. Thus the low temperature and relatively high oxygen content of deep water can only be explained by assuming an exchange between deep and surface waters. The problem of the disposal of radioactive wastes in the deep sea has stimulated the oceanographer's natural curiosity as to the rate of this exchange.

The North Atlantic receives surface waters from the South Atlantic and loses deep water to the South Atlantic. Assuming a surface flow from the South to the North Atlantic of 6 million cubic meters per second (Sverdrup et al., 1942, p. 685), and considering only the upper kilometer of the North Atlantic to be affected, the mean replacement time is about 140 years. The gyral in the North Atlantic, which includes the Gulf Stream, carries about ten times the volume of water exchanged between the South and North Atlantic, so that the mean circulation time is only about one-tenth the replacement time.

This surface exchange between the North and South Atlantic is balanced by a deep current from North to South. The mean displacement time for the deep water of the North Atlantic (2000–4000 meters) is calculated as about 250 years. This time is in reasonable agreement with more recent estimates of the age of the deep water discussed below.

Between these surface and deep layers are the intermediate waters which appear to circulate even more rapidly. Deacon (1933) calculated rapid rates of northward flow of the Antarctic intermediate water in the South Atlantic, based upon alternate maxima and minima in the concentrations of oxygen in the oxygen minimum layer. These were interpreted as representing annual cycles when the waters were formed at the surface. He estimated a transit time of about $4\frac{1}{2}$ years between the Antarctic convergence and the equator. Seiwell (1934) has similarly computed rapid flows and a mean transport time of 7–8 years for the drift of the oxygen minimum layer of the North Atlantic Ocean. Deacon's and Seiwell's interpretations have been questioned (see Riley, 1951, p. 77) on various grounds. However, their rates of flow agree with direct current measurements at comparable depths (see earlier) which also indicate rapid rates of circulation.

The deep outflow from the Mediterranean sinks from sill depth to 1000–1500 meters in the North Atlantic Ocean. This water, although much diluted by Atlantic water, is characterized by relatively high salinity and temperature, and spreads out in a sheet which may be identified in most of the temperate North Atlantic, and some spreads into the South Atlantic. It can be readily identified near Bermuda, 2500 miles from its source. Iselin (1936) computed that sufficient excess salt would be produced by the Mediterranean outflow to produce the observed anomaly in 12–15 years. He pointed out that the actual replacement would be more rapid because he neglected admixture of Atlantic water in the immediate vicinity of the Straits of Gibraltar. Defant (1955) has evaluated the mixing processes involved in dissipating the Mediterranean water within the Atlantic Ocean, and has concluded that the total accumulation in the Atlantic Ocean represents the contribution resulting from six years of flow through the Straits of Gibraltar. The rapid dissipation of this large water mass at mid depths suggests a more rapid circulation than had been generally accepted for intermediate waters.

During recent years other lines of investigation have led to the belief that the overturn of water in the ocean basin takes place in less than a thousand years and probably in 200 years or less. Evidence supporting this belief follows. (Carbon-14 and carbon dioxide exchange estimations are discussed in greater detail by Craig elsewhere in this report.)

1. *Heat flow measurements:* Measurements reported by Revelle and Maxwell (1952) have shown a heat flow through the floor of the Pacific Ocean of 1.2×10^{-6} calories per square centimeter per second, or 38 calories per square centimeter per year. If not dissipated by circulation and mixing, this heat flow would lead to warming of the deep and bottom water during its passage from the Antarctic to the equator. From considerations of meridional circulation, observed temperature gradients and mixing in the deep sea, Revelle and Maxwell estimate that the deep water is replenished in less than 1000 years.

2. *Secular change of oxygen:* Worthington (1954) has shown that the North Atlantic Deep Water has suffered a loss of dissolved

oxygen of about 0.3 ml/L over the last twenty years. Assuming a steady rate of attrition he computes that the date at which this water was saturated, presumably while at the surface, was about 1810. A further study (Worthington, 1955) suggests that the Caribbean Deep Water was formed at the same time. Thus it seems possible that formation of the North Atlantic Deep Water, which composes about half of the contents of the Atlantic, is not continuous but sporadic.

3. *Carbon-14 dating:* In recent years the techniques of carbon-14 age determination have been applied to deep sea water samples. The most reliable measurements (Rubin, unpublished), of samples from east of the Lesser Antilles, show the carbon at 1750 meters to be about 200 years older than the surface carbon.

Present estimates of the age of deep waters are based primarily on measurements in the North Atlantic and on geochemical calculations for the entire world ocean. That the deep circulation of the Pacific is significantly slower than that of the Atlantic is suggested by the apparent absence of regions of deep and bottom water formation in the Pacific and the relatively high nutrient salt content and low dissolved oxygen content of deep Pacific waters. In order to determine whether the deep waters of the Pacific would provide a longer period of isolation for radioactive wastes than elsewhere, deep Pacific oceanographic data must be carefully scrutinized.

REFERENCES

BOHNECKE, G. 1936. Atlas: Temperatur, Salzgehalt und Dichte an der Oberflache des Atlantischen Ozeans. Deutsche Atlantische Exped. *Meteor,* 1925–27, Wiss. Erg. **5**: vii+76 pp.

BOWDEN, K. F. 1954. The direct measurement of subsurface currents in the oceans. *Deep-Sea Res.* **2**:33–47.

CROMWELL, T. 1953. Circulation in a meridional plane in the central equatorial Pacific. *J. Marine Res.* **12**:196–213.

DEACON, G. E. R. 1933. A general account of the hydrology of the South Atlantic Ocean. *Discovery Rep.* **7**:171–238.

DEFANT, A. 1955. Die Ausbreitung des Mittelmeerwassers im Nordatlantischen Ozean. Pap. Mar. Biol. and Oceanogr., *Deep-Sea Res.,* suppl. to **3**:465–470.

DEUTSCHEN SEEWARTE. 1942. Weltkarte zur Ubersicht der Meeresstromungen. Deutschen Seewarte No. 2802.

DIETRICH, G. 1939. Das Amerikansiche Mittelmeer. Gesellsch. Erdkunde zu Berlin, Zeitschr., 108–130.

ECKART, C. 1948. An analysis of the stirring and mixing processes in incompressible fluids. *J. Marine Res.* **7**:265–275.

FRANCIS, J. R. D., and H. STOMMEL. 1953. How much does a gale mix the surface layers of the ocean. *Quart. J. Roy. Meteorol. Soc.* **79**:534–536.

FUGLISTER, F. C. 1951. Multiple currents in the Gulf Stream System. *Tellus* **3**(4): 230–233.

GOLOLOBOV, Y. K. 1949. Contribution to the problem of determining the age of the present stage of the Black Sea (in Russian). *Dokl. Akad. Nauk SSSR* **66**:451–454.

HIDAKA, K. 1954. A contribution to the theory of upwelling and coastal currents. *Trans. Am. Geophys. Union* **35**(3):431–444.

ISELIN, C. O'D. 1936. A study of the circulation of the western North Atlantic. *Pap. Phys. Oceanogr. Meteorol.* **4**(4):1–101.

JAPANESE FISHERY AGENCY. 1955. Report on the investigations of the effects of radiation in the Bikini region. Res. Dept., Japanese Fishery Agency, Tokyo, 191 p.

KETCHUM, B. H., and W. L. FORD. 1952. Rate of dispersion in the wake of a barge at sea. *Trans. Am. Geophys. Union* **33** (5):680–684.

KETCHUM, B. H., and D. J. KEEN. 1953. The exchanges of fresh and salt waters in the Bay of Fundy and in Passamaquoddy Bay. *J. Fish. Res. Bd. Can.* **10**:97–124.

1955. The accumulation of river water over the continental shelf between Cape Cod and Chesapeake Bay. Pap. Mar. Biol. and Oceanogr., *Deep-Sea Res.,* suppl. to vol. **3**: 346–357.

KOENUMA, K. 1939. On the hydrography of south-western part of the North Pacific and the Kuroshio. *Kobe Imper. Marine Observ., Memoirs* **7**:41–114.

MCEWEN, G. F. 1934. Rate of upwelling in the region of San Diego computed from

serial temperatures. *Fifth Pac. Sci. Congr.*, Toronto **3**:1763.

MIYAKE, Y., Y. SUGIURA, and K. KAMEDA. 1955. On the distribution of radioactivity in the sea around Bikini Atoll in June, 1954. *Pap. Meteorol. Geophys.*, Tokyo **5**(3, 4):253–362.

MONTGOMERY, R. B. 1938a. Fluctuations in monthly sea level on eastern U. S. coast as related to dynamics of western North Atlantic Ocean. *J. Marine Res.* **1**:165–185.

1938b. Circulation in upper layers of southern North Atlantic deduced with use of isentropic analysis. *Pap. Phys. Oceanogr. Meteorol.* **6**(2):1–55.

MUNK, W. H., G. C. EWING, and R. R. REVELLE. 1949. Diffusion in Bikini Lagoon. *Trans. Am. Geophys. Union* **30**(1):59–66.

REVELLE, R., and A. E. MAXWELL. 1952. Heat flow through the floor on the eastern North Pacific Ocean. *Nature* **170**:199.

REVELLE, R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear science and oceanography. United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, Paper no. 277: 22 pp.

RILEY, G. A. 1951. Oxygen, phosphate and nitrate in the Atlantic Ocean. *Bull. Bingham Oceanogr. Coll.* **13**, Art. 1:1–126.

SAITO, Y. 1951. On the velocity of the vertical flow in the ocean. *J. Inst. Polytech., Osaka City Univ.* **2**(1) Ser. B.:1–4.

SEIWELL, H. R. 1934. The distribution of oxygen in the western basin of the North Atlantic. *Pap. Phys. Oceanogr. Meteorol.* **3**(1):1–86.

SELIGMAN, H. 1955. The discharge of radioactive waste products into the Irish Sea. Part 1. First experiments for the study of movement and dilution of released dye in the sea. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva. Paper 9:701–711.

SVERDRUP, H. U., M. W. JOHNSON, and R. H. FLEMING. 1942. The oceans, their physics, chemistry and general biology. Prentice-Hall, N. Y., x+1087 pp.

SWALLOW, J. C. 1955. A neutral-buoyancy float for measuring deep currents. *Deep-Sea Res.* **3**:74–81.

U. S. ATOMIC ENERGY COMMISSION. 1956. Operation Troll. U. S. Atomic Energy Commission, New York Operations Office, NYO 4656, Ed. by J. H. Harley, 37 pp.

U. S. NAVY HYDROGRAPHIC OFFICE. 1947a. Atlas of surface currents, north Atlantic Ocean. H. O. Pub. 571, First Ed., reprinted 1947.

1947b. Atlas of surface currents, northeastern Pacific Ocean. H. O. Pub. 570, First Ed.

1950. Atlas of surface currents, northwestern Pacific Ocean. H. O. Pub. 569, First Ed., reprinted 1950.

VON ARX, W. S., D. F. BUMPUS, and W. S. RICHARDSON. 1955. On the fine structure of the Gulf Stream front. *Deep-Sea Res.* **3**:46–65.

WOOSTER, W. S., and F. JENNINGS. 1955. Exploratory oceanographic observations in the eastern tropical Pacific, January to March, 1953. *Calif. Fish and Game* **41**: 79–90.

WORTHINGTON, L. V. 1954. A preliminary note on the time scale in North Atlantic circulation. *Deep-Sea Res.* **1**:244–251.

1955. A new theory of Caribbean bottom-water formation. *Deep-Sea Res.* **3**:82–87.

Chapter 5

THE EFFECTS OF THE ECOLOGICAL SYSTEM ON THE TRANSPORT OF ELEMENTS IN THE SEA [1]

Bostwick H. Ketchum, *Woods Hole Oceanographic Institution,* Woods Hole, Massachusetts

Some elements may be profoundly influenced by the biological cycle and their resulting distribution in the sea may be quite different from the distribution of elements that are affected only by the circulation of the water. Numerous examples of the modification of distribution by biological activities could be given but it may suffice to review briefly the vertical distribution of phosphorus in the ocean.

The photosynthetic fixation of carbon is limited to the surface hundred meters or less of the sea by the penetration of light, and the plant nutrients, including phosphorus, are assimilated there. The surface concentration of these elements may be reduced to virtually zero. Below the photosynthetic zone, the concentrations of these nutrients increase, reaching maximum values at depths of 200 to 1000 meters, the actual depth depending upon location and the oceanic circulation. These maximum concentrations are produced by two processes. The water at intermediate depths is formed by cooling at high latitudes in the ocean, where it sinks and spreads out. At the time of sinking, it contains some inorganic phosphorus and organic matter which is decomposed, liberating the plant nutrients and decreasing the oxygen content. Additions to the organic matter from the surface waters occur everywhere, increasing the nutrient maximum concentration and decreasing the oxygen minimum. Below the nutrient maximum-oxygen minimum layer the concentration of phosphorus decreases again reaching values which are generally constant and uniform from a depth of about 1500 meters to the bottom (Redfield, 1942).

The general patterns of distribution of the elements important in plankton growth on an ocean-wide scale are thus quite different from the pattern of distribution of the major elements. The processes which must be considered in order to evaluate biological effects on the ultimate distribution of radioisotope wastes or contaminants in the sea include (1) the assimilation or adsorption of the elements by the biological populations, (2) the effects of gravity, (3) vertical migrations, (4) horizontal migrations, and (5) the effects of stationary populations in flowing systems.

It has been shown in another section of this report that biological populations may concentrate by several orders of magnitude various elements and their radioisotopes. To evaluate the possible significance of this in the oceans, it is necessary to determine the quantity of living material present (the biomass) and the rate of production of the populations of the ecological system. The biomass, when combined with the known concentration factor, will indicate how much of an element in the water may be combined in the living organisms. The most active concentration of elements may occur during the rapid growth of populations; consequently it is also essential to know the rate of production of the various populations involved. A few data on both the biomass and the rate of production of various populations in the sea are given in Tables 1 and 2.

The biomass figures indicate that concentration factors of 12,500 or more would be required, *under static conditions,* to incorporate half of an element in a cubic meter of water within the ecological system even in the high concentrations of living material found in red-tide blooms. However, the biological populations are not static; those movements which are independent of the motion of the water can, by repetition, transport larger proportions of elements than is indicated by static equilibrium conditions. The productivity values in Table 2 indicate that several times the standing crop of phytoplankton is produced annually. Both

[1] Contribution No. 871 from the Woods Hole Oceanographic Institution.

TABLE 1 Estimates of Biomass of Marine Populations. All Values Have Been Converted to Volume (Wet Weight) Per Cubic Meter (Parts Per Million)

Population	Location and character	Source	cc/m³
Phytoplankton	Maximum Atlantic	a	10
	Maximum Pacific	a	25
	Red Tide Blooms	b	41
	Long Island Sound	c	18.2
	Coastal Water	c	1.2
	Sargasso Sea	c	0.3
Zooplankton	Gulf of Maine	d	0.08 — 1.0
	Coastal Water	e	0.08 — 0.8
	Sargasso Sea	f,e	0.006 — 0.09
	N. African Upwelling	f	1.0
	Eastern North Pacific	g	0.042
	Eastern Tropical Pacific	g	0.055
	Peru Current	g	0.124

a. Complete utilization of maximum phosphorus concentrations; conversion P = 0.5 per cent of wet weight.
b. Ketchum and Keen, 1948, 17–21, Table 1. Conversion as in a.
c. Riley, Stommel and Bumpus, 1949, Table VI; conversion C = 10 per cent wet weight.
d. Redfield, 1941, drained volumes, vertical tows, assumed mean depth 100 meters.
e. Riley, et al., 1949, Table V, displacement weight.
f. Unpublished data, W. H. O. I., surface tows at night, drained volumes.
g. Unpublished data, S. I. O., oblique tows 200–300 meters to surface, wet plankton volumes.

the depth of the photosysthetic zone and the production rate at various depths, are variable, thus the values for production cannot be reduced without excess over-simplification to a volume basis which would permit direct comparison with Table 1. However, Riley's (1941) maximum value for the standing crop of phytoplankton in Long Island Sound, 1.82 gC/m³, showed a production of 0.187 gC/m³/day and the annual production was twenty times as great as the maximum standing crop observed at any one time. Estimates of the growth of zooplankton populations have given values ranging up to 5 per cent of the standing crop per day.

TABLE 2 Estimates of the Productivity of Marine Phytoplankton Populations

Location and character	Source	gC/m²/year	cc/m²/year [1]
Sargasso Sea (Atlantic)	a	18	180
Coastal Areas (Atlantic)	a	1100	11000
Open Ocean (Pacific)	a	50	500
Equatorial Divergence (Pacific)	a	140	1400
Coastal Areas (Pacific)	a	200	2000
Oceanic Mean	a	55	550
Long Island Sound min.	b	95	950
max.		1000	10000
N. Atlantic 3°-13°N.	b	278	2780
Oceanic Mean	c	340 ± 220	1200–5600

a. Steemann Nielsen (1954). Carbon-14 method. This is given as gross production, but Ryther (1954) suggests that it may be net (gross minus respiration) production in nutrient poor areas.
b. Riley (1941). Gross production, oxygen method.
c. Riley (1944).

[1] Conversion assuming one gram of carbon = 10 cc of wet plankton.

It is a truism in ecology that the total quantity of living material which can be produced decreases as the trophic level of the organisms considered increases. In some ecological systems the biomass reflects this progression, i.e., at any one time there will be a larger standing crop of plants than of herbivores and the standing crop becomes progressively smaller as one goes through the various higher steps of the food web. In the oceans, however, this is not necessarily true. It is common to find rather high concentrations of the herbivorous zooplankton when phytoplankton are scarce. Large populations of herbivores will quickly decimate the plants on which they feed. A balance may be maintained as a result of the different lengths of the life cycle of the various parts of the food web. A population of phytoplankton can double in a period of time ranging from hours to days, whereas the life cycles of zooplankton are more commonly measured in terms of weeks or months and the life cycles of the higher elements of the food web, such as fish, are

measured in terms of seasons or years. A comparatively small population of phytoplankton doubling rapidly can provide the energy and nutrients of an equivalent or even larger animal population which is increasing more slowly.

The size of various populations and their rate of production in the English Channel has been evaluated by Harvey (1950) and his results are given in Table 3. These illustrate the above conclusions, since the average biomass of animals exceed that of the plants, but the rate

TABLE 3 AVERAGE QUANTITY, THROUGHOUT THE YEAR, OF PLANTS AND ANIMALS BELOW UNIT AREA OF SEA SURFACE IN THE ENGLISH CHANNEL [1]

Organism	Dry wt of organic matter: Standing crop g./m²	Dry wt of organic matter: Production g./m²/day
Phytoplankton	4.00	0.4–0.5
Zooplankton	1.50	0.1500
Pelagic Fish	1.80	0.0016 [2]
Bacteria	0.04	—
Demersal Fish	1.25	0.0010
Bottom Fauna	17.00	0.0300 [3]
Bottom Bacteria	0.10	—

[1] From Harvey (1950), depth equals 70 meters.
[2] Based on estimated mortality of 30 per cent per annum.
[3] Based on estimated mortality of 60 per cent per annum.

of production of the plants exceeds that of the animal populations.

The plankton organisms in the open sea provide by far the largest quantity of living material and by even more the largest organic absorptive surface. Those radioisotopes which are adsorbed will become bound to the organisms, and they are as subject to the effects of gravitation and migration as if they had been assimilated and utilized.

Gravity affects the organisms in a population and can thus modify the distribution of elements which become incorporated in the biological cycle. Ultimately only two fates await most of the plankton which grows in the surface layers. It may be eaten by the herbivores or it may sink out of the illuminated zone and decompose at greater depths. If the plankton is eaten by a herbivore, a proportion of the organic matter is incorporated into the herbivore body but an even larger proportion is returned to the water as excretion or faecal pellets. The excretions may be present in the water inhabited by the plankton and reused *in situ*. The faecal pellets settle into the deeper water where they decompose. Gravity thus imposes on elements which become incorporated in the biological system a modification of the distribution which would be produced by movements of the water alone, since they tend to accumulate at some intermediate depth in the water column, or on the bottom.

One of the unsolved problems of marine biology is the definition of the proportion of organic matter which is decomposed by the time the particulate material sinks to various depths. This problem must be solved before an evaluation of the biological effects on the distribution of radioisotope contamination of the seas can be made. It may be worthwhile to summarize some of the present thinking on this problem.

In the first place, everywhere that samples have been taken in the deep sea, living organisms have been found. Since we know of no mechanism other than photosynthesis at the surface which can provide the organic material necessary to support these populations, it is clear that some of the surface produced material must reach all depths of the ocean. It may be argued by some that the bacterial chemosynthetic processes are a source of fixed carbon which has not been considered, but the conditions in the deep sea are not suitable for the formation of organic matter by any of these processes.

The presence of the nutrient maximum-oxygen minimum layer at intermediate depths in the sea has led to the conclusion that most of the organic matter formed at the surface must be oxidized by the time it has sunk to a depth of 1000 meters (Redfield, 1942). Analyses of organic phosphorus in the equatorial Atlantic Ocean showed considerable amounts in the waters above 1000 meters, but none at greater depths (Ketchum, Corwin, and Keen, 1955). There is no present evaluation of the quantity of organic carbon which can sink to greater depths, nor is it possible to evaluate whether this quantity would be sufficient to support the known populations of archibenthic organisms. These two extremes thus define the dilemma. Namely that some organic matter must reach the great depths, but, at the same time, most of the decomposition appears to occur above a depth of 1000 meters.

The secular change of oxygen in the deep

sea which has been found by Worthington (1954) in the North Atlantic, provides one means of computing the total quantity of organic matter required. Worthington observed a decrease of 0.3 milliliters of oxygen per liter in thirty years at depths between 2500 meters and the bottom. In the Atlantic Ocean this corresponds to an average thickness of 1500 meters and the total quantity of organic matter required to produce this change in oxygen is equivalent to the decomposition of 8 grams of organic carbon per square meter per year in this layer. This quantity of organic matter is nearly 15 per cent of the annual mean production according to Steemann Nielsen (1954) and from 1.4 to 7 per cent of the mean suggested by Riley (1944). Part of the secular change in oxygen may have been produced by eddy diffusion into the oxygen minimum layer, which would reduce the quantity of organic carbon reaching greater depths.

The effects of gravity may be accentuated when the surface currents are opposed to the currents in the deeper layers. This type of circulation pattern is very common in estuaries, on continental shelves, and in those areas where offshore winds produce upwelling of the deeper waters. In all of these cases the nutrient rich deep water is carried inshore in a sub-surface drift, and brought to the surface by upwelling or vertical mixing. The nutrients are assimilated by the plankton in the surface layers and are carried offshore in the surface current. When the organisms sink, they again reach the onshore sub-surface current where they decompose liberating more nutrients into water which is already relatively rich. Thus the elements involved in biological processes follow a different cycle from the circulation of the water and this cycle leads to an accumulation of elements greater than can be found in either of the source waters (Ström, 1936; Hulburt, In press).

Nutrient elements are commonly concentrated by this type of mechanism in fjords. Where the deepest water is relatively stagnant and isolated from the intermediate and surface layers, considerable concentrations of organic derivatives can be developed. In the Norwegian fjords with a relatively shallow sill, for example, anaerobic conditions may be produced in the bottom water and the nutrients are five to ten times as concentrated as in either of the source waters (Ström, 1936). In the Black Sea the deep water is isolated from the surface by a strong density gradient and its average age has been estimated at 2500–5000 years (Sverdrup, Johnson and Fleming, 1942, p. 651). Very large accumulations of organic derivatives are found in this deep water. (Gololobov, 1949.)

Opposed currents can, however, work in the opposite way and lead to a decrease in the concentration of elements involved in the biological cycle. The classic example of this type of circulation is the Mediterranean, where the nutrients available for plant growth are less than half of the concentration available in the adjacent parts of the Atlantic. In the Mediterranean the supply comes from the surface waters of the North Atlantic which are already impoverished by plant growth. Since evaporation exceeds precipitation in the Mediterranean the water becomes more saline, sinks and is lost as a deep outflow over the sill at Gibraltar (Thomsen, 1931). The accumulation of elements in sinking organisms transfers these elements from the inflowing surface water to the outflowing deep water. They are eventually lost from the Mediterranean. A similar process apparently applies to the entire North Atlantic. There is a large inflow of South Atlantic surface water which contains low concentrations of elements involved in the ecological cycle. The outflow from the North Atlantic required to balance the water budget occurs at depths and this water contains considerable quantities of the elements which had been returned to the water (Sverdrup et al., 1942).

In summary the various peculiarities of distribution which can be attributed to gravitational effects on the ecological cycle are therefore (1) the accumulation of elements at intermediate depths as a result of sinking and decomposition, (2) the concentration of elements in areas of opposed flow where the deep water is brought to the surface by upwelling or vertical mixing and (3) the impoverishment of areas where the supply of water is from the surface and the loss from greater depths.

In addition to the passive gravitational effects on organisms, animal plankton forms exhibit vertical migrations. A considerable literature has developed in this field over the last ten years, but the effects of these vertical migrations on the distribution of elements has not been studied directly and must be inferred from our knowledge of the ecological system.

Historically, a few studies of the vertical migration of zooplankton had been made prior to the war. Great impetus was given these studies when a false bottom was repeatedly observed on echo sounding recorders (Dietz, 1948; Hersey and Moore, 1948). This has been called the scattering layer. Although there is still controversy as to which organisms are the principal scatterers in the sea, it has been established that one or more layers are commonly found which migrate vertically over a depth of as much as 800 meters, being at or near the surface at night and at great depths at mid-day.

No observations of the changes of elements involved in the biological cycle which may be associated with vertical migrations have been made. Most of our analytical techniques are too insensitive to detect the day to day changes which might be expected in biologically active elements if our present evaluation of the density of the populations and their respiration and excretion rates is correct. It is known, however, that direct assimilation of some elements is possible by invertebrate forms and vertical transport of radioisotopes might be expected to result. Indeed, the transport of radioisotopes might prove an excellent tool for the study of vertical migrations if a source were provided at one depth within the range of the migration.

Ecologically the following effects might be expected as a result of vertical migration. The zooplankton are certainly in the area of the most dense concentration of their food, the phytoplankton, when they are at the surface at night. During the hours of darkness they may therefore be expected to consume the living material in the water, and some of this, at least, would be excreted or passed as faecal pellets at depth in the day time. This process would thus augment the effects of gravity on those elements incorporated in the biological system. There is also evidence that the zooplankton can assimilate dissolved elements from sea water. If elements were assimilated at depth they might be excreted or exchanged near the surface and thus directly modify the vertical distribution in the sea.

It should not be neglected that larger organisms can certainly migrate vertically over greater distances than we have discussed above. Certainly whales, tuna and sharks, and presumably the smaller forms upon which they feed are known to go to considerable depths in the ocean. Quantitatively, of course, these members high on the food chain are proportionally small compared to the plankton organisms. However, their effects on vertical distribution of materials may not be negligible over periods of several decades.

Horizontal migrations of organisms may also result in the transport of material involved in the biological cycle and are also independent of the currents of the ocean. Here again man does not know enough to assess these quantitatively, but their possible effects should not be ignored.

The migrations of pelagic fishes may be of considerable interest in this regard. The salmon for example reach maturity in the open sea, then migrate in enormous numbers to coastal areas to breed. Such a horizontal migration could transport radioisotopes, since the salmon could accumulate materials from large volumes of the sea and, by their migration, concentrate them many thousand-fold in the rivers and estuaries.

Many other fish also exhibit extensive migrations. Even though some of these do not enter the rivers to breed, they may enter the areas where they are available for commercial capture, thus becoming some of the food supply of the nation. Unfortunately, in many of these species we do not know the complete life history and most of our information concerning their occurrences and migrations is obtained only during the period of year when they are caught. The Atlantic tuna, for example, are caught in the early spring in the Caribbean and off the Bahama Banks. As spring and summer progresses they migrate northward along the coast, and maximum catches occur in New England in late summer and early fall. The winter habitat and breeding area of these large and important food fish is largely unknown, though preliminary data suggest that they practically circumnavigate the North Atlantic Ocean (Mather and Day, 1954). Similarly the mackerel catches are first concentrated in the southern part of the Atlantic coastline in the late spring and early summer. The large catches off New England occur in August and September. This species breeds on the Atlantic continental shelf during its summer northward migration (Sette, 1943, 1950).

Additional examples of mass migrations into

the coastal regions are found in the Pacific sardine and the North Atlantic herring. In all of these cases materials assimilated at sea may be concentrated in inshore waters as a result of these migrations, which may cover thousands of miles. Such migrations certainly make it difficult to select any area in the oceans as being sufficiently remote and isolated from human interest to insure that the discharge of radioisotope wastes might not be transported into those areas man is most interested in protecting. It should, however, be pointed out that this is a quantitative problem, and our knowledge is not sufficiently detailed to permit evaluating the quantity of radioisotopes which could be transported in mass migrations of fish.

In addition to the movements of organisms which are independent of the circulation of the water resulting from gravity and vertical and horizontal migrations, many populations remain stationary in a flowing stream of water. The organism is thus able to concentrate remarkably the constituents of the water masses which pass by. Harvey (1950) estimated, for example, that the bottom population was nearly 70 per cent of the total population at a station in the English Channel (see Table 3).

The most apparent of these stationary populations are those which live on or in the bottom. Much of our knowledge concerning such populations is confined to those which occupy shallow waters such as the clams, the oysters, and other economically important species. Stationary populations may be exposed to and feed on populations in many cubic miles of sea water during the course of an active growing season.

Although most of our knowledge is confined to shallow water forms, it is known that such stationary populations are a main source of food for many bottom-feeding commercial fishes. The haddock and cod fisheries of New England and the halibut fishery of the Pacific Coast, for example, are ground fisheries. These important species of fish feed on sedentary or stationary populations. Even in the great depths of the ocean such sedentary populations have been found wherever man has had the opportunity to search for them. Although little is known of their location in the food web and dynamics of the ocean, it seems certain that they play a part.

The importance of such stationary populations is that they can concentrate enormously the density of organic matter in those locations suitable for their survival. In unique situations they may concentrate by several orders of magnitude the available organic matter in the ocean.

Less obvious stationary populations are planktonic and unattached, and one would expect them to be transported away from a given area by the currents. It has been found in some cases, however, that in spite of horizontal currents of considerable velocity, the centers of some planktonic populations can remain relatively stationary. Presumably there is a constant drain from these populations as a result of the currents which carry away some of the organisms, but the rate of production of the population is sufficient to maintain the population in spite of this drain. Examples of such populations are to be found in almost all estuaries which tend to maintain endemic species different from those commonly found in the adjacent sea (Ketchum, 1954; Bousfield, 1955). Even in the open ocean similar stationary populations have been found (Redfield, 1939, 1940, 1941; Johnson and co-workers, unpublished observations). It is necessary to have a rate of reproduction of the population as a whole sufficient to balance the circulatory drain. This rapid rate of reproduction will, of course, lead to the concentration of materials from the water mass moving past.

A special case of biological concentration of materials which probably involves several of the above phenomena is found in the "red tide." It has been shown that the concentration of total phosphorus in the colored water of these dinoflagellate blooms is commonly ten to twenty times as great as the concentration which can be found in any of the adjacent waters (Ketchum and Keen, 1948). Most of this phosphorus is combined in the living cells, and very little is present in the inorganic form. One of the explanations for these high concentrations involves the accumulation of the organisms at the surface because of their buoyancy, and the subsequent further concentration of the surface film by convergence of water masses (Ryther, 1955). In the red tides which have occurred in recent years off the west coast of Florida, the organism involved, *Gymnodinium brevis,* produces a toxin which is lethal to the fish and other organisms in the water, and vast numbers of fish have been killed as a result of these dinoflagellate blooms (Gunter, et al.,

1948). Recent evidence indicates that the organisms are almost always present in the water (Collier, A., unpublished), but in such low concentrations that there is no marked fish mortality. It is only after the concentration produced by the biological and hydrographic system that mortalities result.

In evaluating the discharge of radioisotope wastes at sea, the factor of safety must be sufficient so that safe levels of radioactivity can be maintained, even after the various mechanisms of biological accumulation.

REFERENCES

BOUSFIELD, E. L. 1955. Ecological control of the occurrence of barnacles in the Miramichi Estuary. *Nat. Mus. Canada Bull.* No. 137, Biol. Ser. No. 46, pp. 1–69.

DIETZ, R. S. 1948. Deep scattering layer in the Pacific and Antarctic oceans. *J. Mar. Res.* **7**:430–442.

GOLOLOBOV, Y. K. 1949. (Contribution to the problem of determining the age of the present stage of the Black Sea) in Russian. *Dokl. Akad. Nauk SSSR.* **66**:451–454.

GUNTER, G., R. H. WILLIAMS, C. C. DAVIS, and F. G. WALTON SMITH. 1948. Catastrophic mass mortality of marine animals and coincident phytoplankton bloom on the west coast of Florida, November, 1946 to August, 1947. *Ecol. Monogr.* **18**:309–324.

HARVEY, H. W. 1950. On the production of living organic matter in the sea off Plymouth. *J. Mar. Biol. Assoc. U. K.* **29**: 97–137.

HERSEY, J. B., and H. B. MOORE. 1948. Progress report on scattering layer observations in the Atlantic Ocean. *Trans. Amer. Geophys. Union.* **29**:341–354.

HULBURT, E. M. In press. The distribution of phosphorus in Great Pond, Massachusetts. (Submitted to *J. Mar. Res.*)

KETCHUM, B. H. 1954. Relation between circulation and planktonic populations in estuaries. *Ecol.* **35**:191–200.

KETCHUM, B. H., N. CORWIN, and D. J. KEEN. 1955. The significance of organic phosphorus determinations in ocean waters. *Deep-Sea Res.* **2**:172–181.

KETCHUM, B. H., and D. J. KEEN. 1948. Unusual phosphorus concentrations in the Florida "red tide" sea water. *J. Mar. Res.* **7**:17–21.

MATHER, F. J., III, and C. G. DAY. 1954. Observations of pelagic fishes of the tropical Atlantic. *Copeia,* 1954, no. 3:179–188.

REDFIELD, A. C. 1939. The history of a population of *Limacina retroversa* during its drift across the Gulf of Maine. *Biol. Bull.* **76**:26–47.

1941. The effect of the circulation of water on the distribution of the calanoid community in the Gulf of Maine. *Biol. Bull.* **80**:86–110.

1942. The processes determining the concentration of oxygen, phosphate and other organic derivatives within the depths of the Atlantic Ocean. *Pap. Phy. Oceanog. Meteorol.* **9**:1–22.

REDFIELD, A. C., and A. BEALE. 1940. Factors determining the populations of chaetognaths in the Gulf of Maine. *Biol. Bull.* **79**:459–487.

RILEY, G. A. 1941. Plankton studies. III. Long Island Sound. *Bingham Oceanog. Coll. Bull.* **7**(3):1–93.

1941a. Plankton studies. V. Regional summary. *J. Mar. Res.* **4**:162–171.

1944. The carbon metabolism and photosynthetic efficiency of the earth as a whole. *Amer. Sci.* **32**:129–134.

RILEY, G. A., H. STOMMEL, and D. F. BUMPUS. 1949. Quantitative ecology of the plankton of the western North Atlantic. *Bingham Oceanog. Coll. Bull.* **12**:1–169.

RYTHER, J. H. 1954. The ratio of photosynthesis to respiration in marine plankton algae. *Deep-Sea Res.* **2**:134–139.

1955. Ecology of autotrophic marine dinoflagellates with reference to red water conditions. Luminescence of Biological Systems: 387–414.

SETTE, O. E. 1943. Biology of the Atlantic mackerel (*Scomber scombrus*) of North America. Part I: Early life history. *Fish. Bull.* **38**:149–237.

SETTE, O. E. 1950. Biology of the Atlantic mackerel (*Scomber scombrus*) of North America. Part II. Migrations and habits. *Fish. Bull.* **51**:251–358.

STEEMANN NIELSEN, E. 1954. On organic production in the oceans. *J. Con. Internat. Explor. Mer.* **19**:309–328.

STRÖM, K. M. 1936. Land-locked waters. Hydrography and bottom deposits in badly ventilated Norwegian Fjords with remarks upon sedimentation under anaerobic conditions. *Norske Vidensk. Akad. 1. Mat. Naturv. Klasse No. 7*, 85 pp., Oslo.

SVERDRUP, H. U., M. W. JOHNSON, and R. H. FLEMING. 1942. The Oceans, their physics, chemistry and general biology. x+1087 pp., Prentice-Hall, Inc., New York.

THOMSEN, H. 1931. Nitrate and phosphate contents of Mediterranean water. *Danish Oceanog. Exped. 1908–1910.* **3**:14 pp.

WORTHINGTON, L. V. 1954. A preliminary note on the time scales in North Atlantic circulation. *Deep-Sea Res.* **1**:244–251.

Chapter 6

PRECIPITATION OF FISSION PRODUCT ELEMENTS ON THE OCEAN BOTTOM BY PHYSICAL, CHEMICAL, AND BIOLOGICAL PROCESSES

Dayton E. Carritt, *The Johns Hopkins University*

and

John H. Harley, *Health and Safety Laboratory, U. S. Atomic Energy Commission*

Introduction

It has been suggested that naturally occurring processes will remove radioactive waste materials from solution or suspension in the oceans, carrying them to the ocean floor where they will be kept out of the human environment until natural radioactive decay destroys them.

In this section we will attempt to define the processes by which materials may be carried to the bottom, to note the conditions under which these several processes can be expected to operate, and to assess the extent to which these processes have been responsible for the removal of activity to the bottom in cases where bottom accumulation has been measured.

It should be noted that the deposition of fission products on the bottom has not been studied in such a way as to permit an evaluation of the mechanisms responsible for the deposition and retention of the activities. Measurements of bottom-held activities have been made primarily to estimate the total activity. We will discuss later the kind of information that might be obtained in connection with weapons tests and large-scale tracer experiments, and which is needed for a better evaluation of the extent to which deposition processes remove fission product elements from the ocean.

Sources of Fission Products

The oceans may receive fission products from two sources, materials from each of which have unique properties important to deposition. The two sources are:

(1) Radioactivities resulting from bomb bursts, either in weapons testing or military use of bombs in war time. Partial controls can be put on the location and time of weapons tests to take advantage of desirable dispersal or concentrating properties of the oceans.

(2) Radioactivity obtained from nuclear power production plants and released to the oceans for containment or dispersal. The time and location of introduction of wastes of this type can be controlled to obtain optimum oceanic characteristics, and the character of the wastes might be altered by the removal of one or more undesirable active or inactive constituents.

In both cases it can be expected that the fission products will partition into a soluble and an insouble fraction. An estimate of the elements that will appear in each fraction is given in another part of this report.

This division into soluble and insoluble fractions presents essentially two different systems so far as deposition or dispersal processes are concerned.

Deposition and Retention Processes

Deposition and retention of fission product waste on the ocean floor will occur when the waste is sufficiently denser than sea water to permit it to settle to the bottom, and when the stability of a waste-bottom component complex is sufficiently greater than the stability of soluble complexes that might form to prevent its redissolving.

Solid formation

The "denser-than-sea-water" requirement can be met when one of two processes occur: (1) the formation of insoluble substances by interaction of the radioactive components of the wastes with a sea water component, and (2) sorption of the radioactive components of the

wastes by solids naturally occurring in sea water or by solids formed by interaction of non-radioactive components of the wastes with sea water constituents.

Certain generalizations can be made with regard to the formation of a solid phase — a precipitate, by the interaction of radioactive constituents with sea water components. Precipitation may occur when the solubility product of a substance has been exceeded. Fundamentally, in order to be able to predict when this condition has been met, knowledge of the ionic activities of the species involved must be known. Ionic activity is used here in the thermodynamic sense, and is not related to activity in the radioactive sense. Unfortunately practically nothing is known about ionic activities of fission product elements in sea water. The theoretical approach through this route appears, therefore, to be impractical.

The mass of radioactive elements that might be introduced into the ocean from any expected level of power production or foreseeable use of bombs, will be small when compared to the quantities of similar elements already in the ocean. Thus, it is to be expected that chemical precipitation of radioisotopes will occur only in ocean regions where precipitation occurs normally. This process includes precipitation in the usual sense and co-precipitation — the process in which similar elements are simultaneously removed from solution. For example, during the precipitation of calcium carbonate, strontium, a minor element, usually is co-precipitated and carried along with the calcium carbonate.

Sorption processes involving inactive solids provide another set of mechanisms that may produce radioactive solids. The solids that are present in sea water or might be produced from inactive waste components are generally finely divided, have large area to volume ratio, and are charged. The sorption of radioactive and inactive dissolved constituents onto the solids, in the ratio of their relative concentration, is favored by these characteristics. Thus, in cases where an element normally present in sea water is known to be taken up by suspended solids it can be expected that radioisotopes of the same or chemically similar elements will also be taken up.

The oceans contain inorganic and organic, living and dead suspended solids — all have sorption properties and may remove active and/or inactive constituents from solution.

Settling characteristics

The sinking of particles in the sea is usually described in terms of Stokes' Law which assumes, in its simplest form, smooth, rigid, spherical particles of a stated diameter and density, sufficiently widely spaced so as not to impede one another. It provided an adequate description of the behavior of these solids with a restricted particle size range. For particles larger than about 100 microns (0.1 mm) the law must be modified to take into account turbulence around the particle that has a net effect of reducing the settling rate. Also, particles of colloidal and near-colloidal dimensions, less than about a half micron, settle at a rate less than predicted by Stokes' Law, presumably because of charge interaction between particles and dissolved components.

TABLE 1 Settling Velocity of Quartz Spheres (In Distilled Water)

Diameter (mm)	Diameter (microns)	Settling velocity (m/day)	Time to settle 1000 m
1.0	1000	14,000	0.07 days
0.1	100	800	1.25 "
0.01	10	8	125 "
0.001	1	0.08	34 years
1/1024	0.98	0.07	39 "
1/2048	0.49	0.02	137 "
1/4096	0.25	0.004	685 "
1/8192	0.12	0.001	2,740 "

Table 1 gives the settling velocities for particles of a stated size in distilled water, has been calculated from Stokes' Law and is subject to the criticisms noted above.

This table is a highly simplified and idealized picture of the actual settling properties of solids that normally occur in the oceans, and especially of particles in the small size range. Particles in this range probably will be the main concern when considering the deposition of fission products. They are also in the size range that will permit ocean circulation to alter markedly any predicted location of deposition or of time to reach the bottom.

The density and shape factors that effect settling characteristics are important when considering organic solids or living organisms. The density approaches that of sea water which

reduces the settling rate, and the shape may vary considerably from the smooth sphere assumed for Stokes' Law.

The particle-size distribution of solids suspended in the ocean as shown by sediments is broad, varying from over a millimeter in diameter for sands found near shore, to 0.1 micron or less for sediments taken from the open ocean. The median diameter of open-ocean particles is in the range 1 to 8 microns.

The accumulation of solids on the ocean floor is a relatively slow process. Table 2 (Holland and Kulp, 1952) indicates the rate of sedimentation on the several parts of the ocean floor.

TABLE 2 SEDIMENTATION RATES

Type of sediment	Fraction of sea water	Sedimentation rate x 10^{-3} gm/cm^2 per year
Shelf	0.08	40
Hemipelagic	0.18	1.3
Pelagic	0.74	
globigerina pteropod	0.36	0.5
red clay	0.28	0.2
diatom radiolarian	0.10	0.15

A weighted average gives approximately 0.75 mg/cm^2 per year for the oceans. If the area of the ocean floor is 3.6×10^{18} cm^2, the total deposition will be 2.7×10^{15} grams or 2.7×10^9 tons per year.

Retention

Prior to actual deposition on the bottom, radioactive solids that have been formed above the bottom may encounter changes in environment that will tend to return them to solution and prevent or hinder deposition. For example, resolution of precipitates with increasing pressure (calcium carbonate), releases of radioactivity from solids as they fall through uncontaminated water, vertical migration of organisms, and vertical components of circulation are all possible mechanisms that will tend to prevent the deposition of radioactive material on the bottom and, when coupled with horizontal circulation features, will tend to disperse the radioactivity over large areas.

The retention of radioactive material on the ocean floor once it has been deposited there will depend upon the stability of the floor relative to erosion, to further deposition, and to turbidity currents, and upon the chemical features of the bottom relative to those through which the solids have settled.

The deep ocean basins are the regions of greatest stability in all respects. Regions near shores and shelves are subject to the greatest variations in deposition and erosion; in regions where rivers enter the seas, relatively wide changes in chemical properties take place.

Discussion of existing data

Three sources of information give some insight into the probable behavior of fission product elements in sea water. They are: (1) existing information concerning the solution chemistry of the elements in question, (2) the behavior of radioactive debris observed in connection with bomb tests in the Pacific, and (3) information concerning the geochemistry of the elements in question.

In utilizing information from these sources to assess the probable fate of fission product elements in the oceans the chemical properties of the oceans are of major importance. Table 3 lists the elementary composition of sea water together with an estimate of the amounts of natural activities present.

In Table 4 are listed fission product elements, together with their half lives and the equilibrium quantities that would be in existence after 100 days cooling when formed in connection with 10^{11} megawatt hours per year of nuclear power production. Also listed are the specific activities that would result were these activities to be mixed throughout the oceans. It will be obvious from a consideration of oceanic properties, presented in other sections of this report, that under any practical method of introduction of wastes, attainment of uniform specific activity of any given element throughout the oceans will not occur. There will be gradients of radioactivity, decreasing from the region of introduction. The figures for specific activities are, therefore, unrealistic and are included only as a basis for making a better estimate when the effects of circulation and fractionation can be provided.

In a few cases, knowledge of the fraction of an element, that would be normally removed by geochemical processes will permit an estimate to be made of the fraction of a radioisotope that will be removed for a given loading. Con-

TABLE 3 ELEMENTS IN SOLUTION IN SEA WATER (EXCEPT DISSOLVED GASES)[1,2]

Element	mg/kg Cl = 19.00‰	Total in oceans (tons)	Natural activities		
			Nuclide	Total (tons)	Curies
Chlorine	18,980	2.66×10^{16}			
Sodium	10,561	1.48×10^{16}			
Magnesium	1,272	1.78×10^{15}			
Sulfur	884	1.23×10^{15}			
Calcium	400	5.6×10^{14}			
Potassium	380	5.3×10^{14}	K^{40}	6.3×10^{10}	4.6×10^{11}
Bromine	65	9.1×10^{13}			
Carbon	28	3.9×10^{13}	C^{14}	56	2.7×10^{5}
Strontium	13	1.8×10^{13}			
Boron	4.6	6.4×10^{12}			
Silicon	0.02 –4.0	0.028–5.6 $\times 10^{12}$			
Fluorine	1.4	2×10^{12}			
Nitrogen (comp)	0.01 –0.7	0.14 –9.8 $\times 10^{11}$			
Aluminum	0.5	7×10^{11}			
Rubidium	0.2	2.8×10^{11}	Rb^{87}	1.18×10^{11}	8.4×10^{0}
Lithium	0.1	1.4×10^{11}			
Phosphorus	0.001–0.1	0.014–1.4 $\times 10^{11}$			
Barium	0.05	7×10^{10}			
Iodine	0.05	7×10^{10}			
Arsenic	0.01 –0.02	1.4 –2.8 $\times 10^{10}$			
Iron	0.002–0.02	0.28 –2.8 $\times 10^{10}$			
Manganese	0.001–0.01	0.14 –1.4 $\times 10^{10}$			
Copper	0.001–0.01	0.14 –1.4 $\times 10^{10}$			
Zinc	0.005	7×10^{9}			
Lead	0.004	5.6×10^{9}			
Selenium	0.004	5.6×10^{9}			
Cesium	0.002	2.8×10^{9}			
Uranium	0.0015	2.1×10^{9}	U^{238}	2.8×10^{9}	3.8×10^{9}
Molybdenum	0.0005	7×10^{8}	U^{235}	2.1×10^{7}	1.1×10^{8}
Thorium	<0.0005	$<7 \times 10^{8}$	Th^{232}	1.4×10^{7}	8×10^{8}
Cerium	0.0004	5.6×10^{8}			
Silver	0.0003	4.2×10^{8}			
Vanadium	0.0003	4.2×10^{8}			
Lanthanum	0.0003	4.2×10^{8}			
Yttrium	0.0003	4.2×10^{8}			
Nickel	0.0001	1.4×10^{7}			
Scandium	0.00004	5.6×10^{7}			
Mercury	0.00003	4.2×10^{7}			
Gold	0.000006	8.4×10^{6}			
Radium	$0.2–3 \times 10^{-10}$	28 –420	Ra^{226}	4.2×10^{2}	1.1×10^{9}

[1] Sverdrup, H. U., M. W. Johnson, and R. H. Fleming, OCEANS (1942).
[2] Revelle, R., T. R. Folsom, E. D. Goldberg, and J. D. Isaacs (1955).

versely, observations of the behavior of radioactive isotopes would lead to a better understanding of the geochemistry of a given element.

Operational data

Of the fission products listed several are either rare earths or rare-earth-like — such products all have very similar chemical properties. All form relatively insoluble hydroxides of the type $R(OH)_3$. The solubility products of the rare earth elements listed by Latimer (1952) all fall in the range 10^{-20} to 10^{-30}. Although a quantitative comparison of the conditions that actually exist in the sea cannot be made with these constants, it would appear from the scant information available concerning the quantities of rare earth elements in the sea that marine waters are saturated with respect to these elements and that a major portion of the rare earth elements are dispersed in the sea as solids. This is generally confirmed by American and Japanese observations of the distribution of fission product activities in the Pacific following bomb tests. In most cases, however, it is difficult to differentiate between "solid fractions" that have been precipitated as solids by chemical processes, and radioactive solids that have been accumu-

lated by microscopic plankton organisms. Both will be collected by filtration or centrifugation. Goldberg (1956), however, noted that information obtained during Operation WIGWAM suggests a fractionation of a portion of the fission product activities into solids that are collected and concentrated by filter feeding organisms. The activity within the filter feeding

TABLE 4 Fission Product Activity After 100 Days Cooling from 10^{11} Megawatt Hours of Nuclear Power Production [1]

Isotope	Half-life	Tons (metric)	Curies at 100 days	Specific activity curies per ton [2]
Kr^{85}	94 y	7.3	3.3×10^{9}	—
Sr^{89}	55 d	86	2.3×10^{12}	0.128
Sr^{90}	25 y	463	7.5×10^{10}	0.0042
Y^{90}	62 h	—	7.48×10^{10}	178
Y^{91}	57 d	111	2.8×10^{12}	6,660
Zr^{95}	65 d	152	3.2×10^{12}	—
Nb^{95}	35 d	161	6.3×10^{12}	—
Ru^{103} ...	45 d	46	1.3×10^{12}	—
Rh^{103} ...	57 m	—	1.3×10^{12}	—
Ru^{106} ...	290 d	35	1.5×10^{11}	—
Rh^{106} ...	30 sec.	—	5.15×10^{10}	—
I^{131}	8.0 d	—	5.2×10^{9}	0.0743
Cs^{137}	33 y	705	5.63×10^{10}	20.1
Ba^{137} ...	2.6 m	—	5.1×10^{10}	0.728
Ba^{140} ...	12.5 d	2	1.5×10^{11}	2.14
La^{140} ...	1.7 d	—	2.5×10^{11}	595
Ce^{141} ...	28 d	45	1.5×10^{12}	268
Pr^{143} ...	13.8 d	2	1.4×10^{11}	—
Ce^{144} ...	275 d	490	1.6×10^{12}	386
Pr^{144} ...	17 m	—	2.4×10^{12}	—
Pm^{147} ...	94 y	7.3	3.3×10^{9}	—
Sm^{157} ...	73 y	0.7	2.0×10^{7}	—

[1] Adapted from data of Culler (1954b) and Revelle, et al. (1955).
[2] Based on tonnage shown in Table 3.

organisms — ones adapted to the removal of particulate material from suspension — showed a high percentage of rare earth elements that previously were noted as probably being predominantly dispersed as solids in the oceans. These organisms were collected in the mixed layer of the sea.

About a year after the 1954 nuclear tests were completed. Operation TROLL undertook a survey of the region west from the test site, including the region just off the Phillipines and northward off the coast of Japan (U. S. Atomic Energy Commission, 1956). Seventy water and plankton samples taken during this cruise were analyzed radiochemically. When compared on an equal weight basis (1000 gms wet plankton *vs.* 1 liter of water) the plankton contained on the average 470 times the activity of the water. Significantly, 80 to 90 per cent of the activity of the plankton was due to Ce^{144} (and its Pr^{144} daughter). Cerium is a rare earth. No information is yet available concerning the species and the relative quantities of organisms responsible for the concentration of activity. A comparison of the total activity per unit weight of macro- and micro-plankton indicated approximately a one and one half times greater concentration by the micro-plankton.

It is noteworthy that the observations made on Operations TROLL and WIGWAM revealed a system in which the properties, with the exception of radioactive element content, were essentially those of normal sea water. The system can be imagined as being essentially sea water to which had been added the radioactive material — a procedure which because of the extreme dilution of the contaminant, in a chemical sense, would not affect the sea water properties. Furthermore, these observations were made on samples taken in the mixed layer (the upper 100 to 300 m).

These results, though largely qualitative in nature, suggest the following conclusions regarding the behavior of fission product elements in the mixed layer of the open oceans:

1. Radioactive material will be retained in the mixed layer for periods of at least a year during which time horizontal motion may carry them a few thousand miles. (Operation TROLL and SHUNKOTSU-MARU data.)
2. Rare earth elements appear to be dispersed primarily as solids and accumulated by the plankton. (Operations TROLL and WIGWAM.)
3. The initial accumulation of rare earth activities is predominantly by filter feeding organisms, presumably by retention of finely divided solids in their feeding apparatus.
4. The cycle of rare earth activities through the biota is unknown. Nevertheless, biological agencies undoubtedly have an important influence in the deposition mechanisms.

The physical state of fission product elements in sea water is important in all of the processes that have been previously mentioned. Table 5 sets forth several fission product elements, the percent of total activity present one year after removal from a reactor and an estimate of the

physical state of each if dispersed in sea water.

The estimates of physical states have been obtained from oceanographic studies following bomb tests and from considerations of the "solution chemistry" of the elements. It should be emphasized that the terms "solid" and "solution" are relative terms. Measurements made during oceanographic studies invariably base the division upon filterability. Such a division

TABLE 5 AN ESTIMATE OF SOLID AND SOLUBLE FRACTIONS FOR FISSION PRODUCTS IN SEA WATER

Element	Per cent of total activity at end of one year	Physical state in sea water
Sr^{89}	3.8	Solution
$Sr^{90} + Y^{90}$	1.7 + 1.7	Solution + solid
Zr^{95}	7.2	Solid
Nb^{95}	15	Solid
Ru^{106}	2.5 + 2.5	Mostly in solution
$Cs^{137} + Ba^{137}$	1.5 + 1.5	Solution
$Ce^{144} + Pr^{144}$	26 + 26	Solid
Pm^{147}	5.6	Solid

obviously will place soluble elements that are utlized by organisms in the solid or solution+ solid category. The settling characteristics of elements so combined will depend upon properties of the organisms. To what extent anomalies of this kind are in the estimate above cannot be stated. However, the estimates agree qualitatively with those made from knowledge of the behavior of elements in systems where biological activity is not a major variable.

Culler (1954a), has noted that low level activities discharged to White Oak Creek end up primarily with the clay in a retention basin. The character of the waste was not noted. Krumholz (1954), however, found considerable uptake of radioactivity in the biota with subsequent relocation and dispersion in the same region.

Geochemical data

An estimate of the behavior of several sea water constituents can be obtained from the results of geochemical studies. These studies permit an evaluation of the fraction of an element supplied to the oceans that is removed from solution. The removal processes may include one or more of those previously mentioned. The results permit no choice of mechanisms. Table 6 lists several elements found in sea water, the

TABLE 6 GEOCHEMICAL BALANCE OF SOME ELEMENTS IN SEA WATER (FROM GOLDSCHMIDT, QUOTED IN RANKAMA AND SAHAMA, 1950, TABLE 16.19)

Element	Total supplied (ppm)	Amount present in ocean (ppm)	Transfer percentage
Na	16,980	10,560	62
K	15,540	380	2.4
Rb	186	0.2	0.1
Ca	21,780	400	1.8
Sr	180	13	7.2
Ba	150	0.05	0.03
Fe	30,000	0.02	0.00007
Y	16.9	0.0003	0.002
La	11	0.0003	0.003
Ce	27.7	0.0004	0.001

quantities supplied to and present in the oceans and a quantity, the transfer percentage, which is the percentage of "present" to "supplied."

Large values of transfer percentage indicate that relatively large fractions of the elements supplied to the oceans stay in solution — small values of transfer percentage that relatively much is removed.

Using the transfer percentages listed for cesium, strontium, and cerium, and estimates of the specific activities that would occur in the oceans as a result of 10^{11} megawatt hours nuclear power production, the reduction through geochemical processes has been calculated. The figures are given in Table 7.

TABLE 7 ACTIVITY REDUCTION BY GEOCHEMICAL PROCESSES

Element	Specific activity (c/gm) (no removal)	Transfer percentage	Specific activity after removal (c/gm)
Cesium	8.6×10^{-5}	0.005	4.3×10^{-8}
Strontium	6.8×10^{-9}	7.2	4.9×10^{-10}
Cerium	1.8×10^{-5}	0.001	1.8×10^{-10}

Laboratory data

Floccing, possible in the disposal of wastes rich in iron or aluminum, may assist in removal of fission products. Unless settling times of natural or artificial flocs are short, resolution and biological uptake may reduce the settling factor markedly.

Goldberg (1954) has described the coprecipitation processes with iron and manganese. While none of the fission product elements are treated, analyses show that the amounts of trace

elements in the sediments are proportional to the iron or manganese content. In addition, filter feeders show concentrations indicating uptake of undifferentiated particulates.

Several experiments have been reported in which the reactions between fission product activities (mixed and individual isotopes) and suspended solids have been studied. In the following examples both marine and fresh water experiments are noted.

Gloyna in Goodgal, Gloyna, and Carritt (1954) noted that 58 per cent of mixed fission product activity (initially less than 1000 cpm) could be removed from solution during centrifugation of untreated Clinch River water, 70 ppm solids, pH 8.4 and alkalinity 92 ppm ($CaCO_3$). No attempt was made to determine which elements were removed.

Carritt and Goodgal (1954) studied the uptake of phosphate, iodide, iron III, strontium sulphate and copper II on samples of Chesapeake Bay sediments. Measurements were made under controlled but varied pH, temperature, salinity, concentration of solids, and specific activities. Of the elements studied strontium, iodide and sulphate are of interest here — sulphate because of the similar chemical behavior of tellurium. Iodide showed no uptake at concentrations applicable to the present discussion.

Under conditions where strontium carbonate did not precipitate, strontium was absorbed according to the following isotherm:

$x/m = 0.0032\ C^{0.44}$

$x/m = \mu g$ atoms Sr per milligram of solids

$C =$ equilibrium concentration of strontium in μg atoms Sr per liter.

This isotherm was valid over the range 52 to 5200 μg atoms Sr per liter.

The uptake of sulphate showed strong pH dependence. At pH above 4.5 very little uptake was noted. With decreasing pH, uptake increased, suggesting that the bisulphate is more active than sulphate.

At pH 3.3 (an unlikely marine condition) the uptake followed the isotherm:

$$x/m = 0.0013\ C^{0.92}$$

over an initial sulphate concentration range of 10^9.

Several proposals on ocean waste disposal would allow introduction of packaged waste into the bottom by sea burial. Dispersion of activity would be a slow diffusion process as from concreted wastes or would be delayed until rupture of an impermeable container. In either case, the activity released would go into the highly absorptive environment of the sediments.

One form of packaging for the disposal of active waste has been proposed by Hatch (1954). He has described the problems encountered with the absorption of fission products onto montmorillonite clays, followed by firing to 800° C., to produce a high density, high specific activity, insoluble waste. When given appropriate pretreatment, it was estimated that fission products could be removed from reactor wastes to yield clays with an activity of about 10 curies per gram. The practicability of utilizing solids of this kind apparently depends upon the demonstration of long term stability under deep ocean conditions and upon the economics of production and transportation. It should be noted that short term stability tests suggest that the fired montmorillonite clays would be extremely stable.

Deep ocean deposits have appreciable base exchange capacities. Revelle measured this to be in the range 30–60 millequivalents per 100 gram of solids. Soluble waste components can be expected to react with solids on the bottom surface and to be removed from solution by base exchange reactions, and isotopic exchanges. No estimate seems possible of the depth into the sediments that this kind of reaction would take place. Certainly the surface layer of sediments would become saturated and reaction with deep sediments would be controlled by diffusion into the sediments.

Further data required

A survey of available literature reveals many gaps in our knowledge in this field. Basic data on the settling processes of natural sedimentation are few, and the carrying processes by which tracer concentrations of isotopes would be removed from the oceans have been almost entirely neglected. From a practical point of view, the data most needed are measures of the gross sedimentation rate of radioactivity. This would be an integral of the effects of many processes — empirical information that would permit a statement concerning the sedimentation rate of activity without reference to the many mechanisms involved.

Nevertheless, for an understanding of the overall process — so that predictions for conditions other than those existing at the time of observations can be made, and to provide information useful to other studies, many individual processes should be studied. The following studies, grouped according to the primary source of information, and thought to be pertinent to the sedimentation and retention problem, would provide some insight into these processes. Obviously, information obtained from one group of studies may be of value in the solution of problems in others.

Data from weapons tests

1. Measurement of the immediate partition of weapons test debris among large-sized immediate fallout, water-borne activity and the air-borne material which may be quite uniformly distributed over the world.
2. Measurement of partition of individual isotopes in sea water between particulate material and solution. (Dynamic and equilibrium conditions).
3. Mechanism of sorption of radioisotopes on natural suspended solids under the conditions existing in ocean water.
4. Measurement of settling rates of natural inorganic particulates, probably by tracer techniques.
5. Measurement of detrital settling rates, including plankton average life.
6. Measurement of uptake and element differentiation in organisms which may become detrital material.

Data from waste disposal experiments

Certain studies here can be combined with tracer studies, designed primarily to give information on basic oceanographic problems:

1. Life expectancy of burial containers.
2. Diffusion rate from concreted or sintered blocks as a function of size, and the concentration and istopic composition of wastes.
3. Regardless of what disposal system is adopted, there will be liquid wastes produced, and studies must be made of liquid waste dispersal. The pertinent effects will be more related to the weapons test data requirements since this is a surface to bottom transfer.

Tracer experiment data

1. Coprecipitation of individual fission products with their stable isotopes normally occurring in sea water, and the particle size distribution of the solids formed, and their sedimentation rate.
2. Similar data on coprecipitation by isomorphous replacement, for example the carrying of radiostrontium with inactive calcium.
3. Rate of entry of diffused material into the basic biological systems. This includes the bottom to surface movement as modified by sedimentation.
4. Exchange capacities of sediments for the radioisotope ions in sea water medium, and rate of diffusion of these isotopes into the undisturbed bottoms.

In all studies in which dispersion, partition, concentration and localization occur, measurements that would permit a balance sheet to be made (all the activity should be accountable) seem desirable and necessary.

SUMMARY

The only semi-quantitative data relevant to the problem of activity removal from the ocean surface are the geochemical data. These indicate a reduction factor of 14 for strontium, 2,000 for cesium, and 100,000 for cerium (and probably all rare-earth-type elements). No information is available on such elements as ruthenium, rubidium, and iodine. Other mechanisms described may contribute to activity removal, but their effects cannot be evaluated with present knowledge.

The reduction factors are for equilibrium conditions, and the high sea water activity found a year after the Castle tests (Operation TROLL) indicate that equilibrium is reached slowly.

Activity introduced on the bottom through sea burial will be subject to entirely different removal processes. No estimate can be made of their effectiveness.

CARRITT, D. E., and S. GOODGAL. 1954. Sorption reactions and some ecological implications. *Deep-Sea Research* 1:224–243.

CULLER, F. L. 1954a. Unpublished results.

CULLER, F. L. 1954b. Notes on Fission Product Wastes from Proposed Power Reactions. ORNL Central File No. 55–4–25.

GOLDBERG, E. D. 1954. Marine Geochemistry 1. Chemical Scavengers of the Sea. *J. Geol.* **62**:249.

GOLDBERG, E. D. 1956. Unpublished results. Presented at Princeton, N. J., March 3, 4, 5, 1956 meeting of NAS Study Group on Oceanography and Fisheries.

GOODGAL, S., E. GLOYNA, and D. E. CARRITT. 1954. Reduction of radioactivity in water. *Jour. Amer. Water Works Assoc.* **46**, No. 1:66–78.

HATCH, L. P. 1954. Clay adsorption of high level wastes. Ocean dispersal of reactor wastes, meeting at Woods Hole Oceanographic Institution, Woods Hole, Mass., August 5–6.

HOLLAND, H. D., and J. L. KULP. 1952. The distribution of uranium, ionium and radium in the oceans and in ocean bottom sediments. Lamont Geological Observatory Technical Report No. 6.

RANKAMA, K., and T. G. SAHAMA. 1950. Geochemistry. University of Chicago Press.

KRUMHOLZ, L. A. 1954. A summary of findings of the ecological survey of White Oak Creek, Roane County, Tenn., 1950–1953. USAEC-ORO 132.

LATIMER, W. M. 1952. Oxidation Potentials. Prentice Hall, New York.

REVELLE, R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear Science in Oceanography. International Conference on the peaceful uses of atomic energy. A/conference 8/P/277. Scripps Institution of Oceanography contribution No. 794.

SVERDRUP, H. U., M. W. JOHNSON, and R. H. FLEMING. 1942. The Oceans. Prentice Hall, New York.

U. S. ATOMIC ENERGY COMMISSION. 1956. Operation TROLL. U. S. Atomic Energy Commission, New York Operations Office, NYO 4656, ed. by J. H. Harley, 37 pp.

CHAPTER 7

ECOLOGICAL FACTORS INVOLVED IN THE UPTAKE, ACCUMULATION, AND LOSS OF RADIONUCLIDES BY AQUATIC ORGANISMS [1]

LOUIS A. KRUMHOLZ, *Department of Biology, University of Louisville,* Louisville, Kentucky

EDWARD D. GOLDBERG, *Scripps Institution of Oceanography, University of California,* La Jolla, California

HOWARD BOROUGHS, *Hawaii Marine Laboratory, University of Hawaii,* Honolulu, T. H.

Introduction

THIS paper is concerned with the uptake, accumulation, and loss by living organisms, of radioactive materials that may be added to or induced in an aquatic environment. These aquatic organisms may live in either fresh, salt, or brackish water and include vascular plants, algae, protozoans, plankton, all the other invertebrate forms such as aquatic insects, bottom-living crustaceans and molluscs, and representatives of each of the five classes of vertebrate animals.

The accumulation and loss of any radioisotope will depend not only upon its own physical half-life but also upon the biological factors that contribute to its incorporation in, retention by, and disappearance from the organism involved. In general, all isotopes of any one chemical element are similar in chemical behavior, and thus it can be assumed, when tracing the paths of most chemical elements through biological systems, that a radioactive atom will behave in the same way as a non-radioactive atom of the same species. However, relatively little is known about the actual mechanisms of uptake, accumulation, and loss by marine and fresh-water organisms of the elements whose isotopes constitute fission products and other radiomaterials.

For the purposes of this discussion, the following terms will be defined:

Uptake is the amount of material that enters the organism in question and the speed at which the material enters is the rate of uptake.

Loss is the amount of material that leaves the organism, and the speed at which it leaves is the rate of loss.

Accumulation is the amount of material that is present in the organism at a given time, and the rate of accumulation is the amount accumulated per unit time. In practice, the accumulation is the difference between the uptake and the loss.

Metabolic processes include all the chemical changes concerned in the building up and destruction of living protoplasm. During these changes, energy is provided for the vital processes and for the assimilation of new materials.

Specific activity is the ratio between the amount of radioactive isotope present and the total amount of all other isotopes of that same element, both radioactive and stable. Most commonly, it is given as the microcuries of radioisotope per gram of total element.

Although the higher animal forms are dependent upon the primary concentrators, the plants, for their source of energy, these animals may or may not be dependent upon the lower forms for many elements. Some elements may enter the bodies of the higher forms directly from the water, while others must be supplied from the lower trophic levels through the food web. These food webs are not the same for all organisms and may even be different for the same organism at various seasons of the year. In some instances certain elements, although present in the environment, are not in the proper physical and/or chemical state to be utilized by the organisms and thus are not available for metabolism.

Radionuclides may become associated with an organism either through adsorption to surface areas, through engulfment, or through metabolic

[1] Contribution No. 9 (New Series) from the Department of Biology, University of Louisville. Contribution from the Scripps Institution of Oceanography, New Series, No. 901a. Contribution from the Hawaii Marine Laboratory, No. 94.

processes; in some instances assimilation may take place following the engulfment of living or inert particulate matter. A radionuclide may also be incorporated into an organism by simple exchange of the radioactive isotope for the stable isotope of the same species. It is therefore important to know the physical and chemical state necessary for metabolism, the mode of entry, and the ability of all organisms at each of the different trophic levels to concentrate the various radionuclides.

Physical and Chemical Factors Concerned with the Uptake of Radionuclides by Living Organisms

a. *Acute versus chronic exposure*

Chronic exposure of an aquatic organism, even to low concentrations of radiomaterials, usually has a markedly different effect on the organism than an acute exposure; the principal difference lies in the amount of radiomaterial accumulated in the tissues. Because many aquatic organisms have the ability to concentrate radiomaterials from their environments by factors up to several hundred thousand, much radiomaterial may be accumulated during a chronic exposure for a relatively long period of time. A state of equilibrium is ultimately reached at which there is a constant uptake and a constant loss with a resultant constant maximum level of accumulation. Conversely, in an acute exposure, such as a single feeding or a single injection of radiomaterials, only a certain relatively small fraction of the radiomaterial is accumulated in the body and the remainder is lost. In such an instance, the maximum level to which an organism is capable of accumulating the radiomaterial in question is seldom reached and certainly not maintained.

Krumholz and Rust (1954) reported an accumulation of one microcurie of strontium 90 per gram of bone in the entire skeleton of a muskrat (*Ondatra zibethica*) which had been utilizing foods of its own choice in the area contiguous to the Oak Ridge National Laboratory. Certainly this instance can be presumed to represent a chronic exposure inasmuch as the animal was at least two years old and had probably lived in the area during her entire lifetime. Aquatic organisms in the Columbia River below the Hanford Works and those in White Oak Creek, Tennessee, below the Oak Ridge National Laboratory, have all suffered chronic exposures to radiomaterials and have accumulated considerable amounts of those materials in their tissues. Hiatt, Boroughs, Townsley, and Kau (1955) found that the daily feeding of strontium 89 to the fish *Tilapia* for short periods of time (four days) did not increase the level of strontium retention after an apparent steady-state condition had been reached. However, there are no published reports of the results of long-term, controlled experiments of chronic exposures of aquatic organisms to radiomaterials.

The literature contains many reports concerned with acute exposures of aquatic organisms to radiomaterials. Martin and Goldberg (unpublished data), who gave single feedings of strontium 90 to Pacific mackerel (*Pneumatophorus japonicus diego*), found that less than five per cent of the amount fed was retained in the body after 48 hours. Much of the five per cent that was incorporated in the skeleton remained there for the duration of the experiment (235 days). Boroughs et al. (1956) reported that between only one and two per cent of the strontium 89 fed to ten yellowfin tuna (*Neothunnus macropterus*) remained in the body after 24 hours. The small amount retained in the body was largely incorporated into the skeletal structures. However, other fish (*Tilapia*) which had been fed similarly prepared strontium 89 capsules retained about 20 per cent of the ingested material after 24 hours. After four days, the amount retained finally levelled off at values that ranged from 1.5 to 19.5 per cent of the amount ingested; the average amount retained was about 7.5 per cent. Here, again, the retained materials were incorporated mainly in the skeletal structures and integument.

b. *Chemical and physical states of the elements in the environment.*

The chemical composition of the marine environment cannot be rigorously defined. The concentrations of elements depend upon the type and location of the water mass. Although more than 90 per cent of marine waters occur at depths greater than 1000 meters, the majority of chemical analyses have been made for shallower waters. Because of the biological activity of the oceans and the movements and origins of water masses, the abundance of certain elements appears to vary by factors greater than two orders of magnitude. However, as a

first approximation, the chemical constituents may be considered to be much the same in all places. Fairly good approximations of the concentrations of elements in sea water are listed in Table 1 as the numbers of atoms per million atoms of chlorine. The reported values of concentrations of elements on which Table 1 is based frequently fail to distinguish between the solid and dissolved phases.

Whereas the oceans may be considered very roughly as a homogeneous mass, most bodies of fresh water must be examined on an individual basis because of the tremendous range in their physical and chemical characteristics. Many of the elements that occur normally in the oceans are in concentrations too small to be detected by present methods or are present in only trace amounts in fresh water. The pH of fresh waters ranges from perhaps as low as 2.2 to a high of about 10.5 although the pH of most lakes and streams falls somewhere between 6.5 and 8.5. The total dissolved solids in fresh waters ranges

TABLE 1 Chemical Abundances in the Marine Hydrosphere

	mg/l	atoms/10^6 atoms Cl		mg/l	atoms/10^6 atoms Cl
H	108,000	202,000,000	Ag	0.0003	0.005
He	0.000005	.002	Cd	0.000055	0.0009
Li	0.2	50	In	< 0.02	< 0.3
Be			Sn	0.003	0.05
B	4.8	830	Sb	< 0.0005	< 0.008
C	28	4,300	Te		
N	0.5	70	I	0.05	0.7
O	857,000	100,000,000	Xe	0.0001	0.001
F	1.3	130	Cs	0.0005	0.005
Ne	0.0003	0.03	Ba	0.0062	0.008
Na	10,500	850,000	La	0.0003	0.004
Mg	1,300	100,000	Ce	0.0004	0.005
Al	0.01	0.7	Pr		
Si	3	200	Nd		
P	0.07	4	Pm		
S	900	52,000	Sm		
Cl	19,000	1,000,000	Eu		
A	0.6	28.5	Gd		
K	380	18,000	Tb		
Ca	400	19,000	Dy		
Sc	0.00004	0.002	Ho		
Ti	0.001	0.04	Er		
V	0.002	0.08	Tm		
Cr	0.00005	0.002	Yb		
Mn	0.002	0.07	Lu		
Fe	0.01	0.3	Hg		
Co	0.0005	0.02	Ta		
Ni	0.0005	0.02	W	0.0001	0.001
Cu	0.003	0.09	Re		
Zn	0.01	0.3	Os		
Ga	0.0005	0.01	Ir		
Ge	< 0.0001	< 0.003	Pt		
As	0.003	0.07	Au	0.000004	0.00004
Se	0.004	0.1	Hg	0.00003	0.0003
Br	65	1,500	Tl	< 0.00001	< 0.00009
Kr	0.0003	0.007	Pb	0.003	0.03
Rb	0.12	2.2	Bi	0.0002	0.002
Sr	8	160	Po		
Y	0.0003	0.006	At		
Zr			Rn	9.0×10^{-15}	8.0×10^{-14}
Nb			Fr		
Mo	0.01	0.2	Ra	3.0×10^{-11}	2.0×10^{-10}
Tc			Ac		
Ru			Th	0.0007	0.006
Rh			Pa	0.003	0.03
Pd			U		

from very low concentration (less than 5 ppm) in the "battery-water" lakes to very high concentrations (more than 400 ppm) in the "alkali" lakes. The fertility of fresh waters ranges from the almost sterile bog lakes to the highly productive lakes in the midwestern prairies.

The physical states and ionic speciation of elements in sea water cannot be as well defined as their absolute concentrations. However, using the known physicochemical constants, and assuming a pH of 8 and a salinity of 35 parts per thousand for sea water, Krauskopf (1956) postulated that the principal valence states of the ions of a number of metals in sea water are as listed in Table 2. From these data it may

TABLE 2 CALCULATED VALENCE STATES FOR METALLIC IONS IN SEA WATER
(From Krauskopf, 1956)

Element	Ion
Zinc	Zn^{++}, $ZnCl^{+}$
Copper	Cu^{++}, $CuCl^{+}$
Bismuth	BiO^{+}
Cadmium	$CdCl^{+}$, $CdCl_2$
Nickel	Ni^{++}, $NiCl^{+}$
Cobalt	Co^{++}
Mercury	$HgCl_4^{-}$
Silver	$AgCl_2^{-}$
Gold	$AuCl_4^{-}$ (Calculated by Goldberg)
Chromium	CrO_4^{-}
Vanadium	$H_2VO_4^{-}$, $H_3V_2O_7^{-}$
Magnesium	Mg^{++}
Calcium	Ca^{++}
Strontium	Sr^{++}
Barium	Ba^{++}

be concluded that most monovalent or divalent ions, except the noble metals, will occur as cations whereas most metals with valences higher than two, and the noble metals, will occur as anions.

The physical states of a given element under equilibrium conditions depend upon whether or not the solubility product of the least soluble species has been exceeded. Greendale and Ballou (1954) have determined the distribution of elements among the soluble, colloidal, and particulate states by simulating the conditions of an underwater detonation of an atomic bomb. Their data are presented in Table 3.

It is not known whether the elements that occur in colloidal or particulate phases are homogeneous entities or are sorbed in other solid phases. Nevertheless, it appears that elements of Groups, I, II, V, VI, and VII usually occur as ionic forms in sea water, whereas other elements, excluding the rare gases, occur predominantly as solid phases. These generalizations have been confirmed in field tests after underwater detonations where more than 50 per cent of the resultant radioactivity was associated with solid phases retained by a molecular filter of pore size 0.5 micron (Goldberg, unpublished data).

Although the data supplied by Greendale and Ballou (1954) are of value for the physical states of elements following the detonation of an atomic bomb, they are at best only suggestive of the steady-state conditions which might result from the continuous spilling of fission product wastes into the sea on a long-term basis.

TABLE 3 PHYSICAL STATES OF ELEMENTS IN SEA WATER

(From Greendale and Ballou, 1954)

	Percentage in given physical state		
Element	Ionic	Colliodal	Particulate
Cesium	70	7	23
Iodine	90	8	2
Strontium	87	3	10
Antimony	73	15	12
Tellurium	45	43	12
Molybdenum	30	10	60
Ruthenium	0	5	95
Cerium	2	4	94
Zirconium	1	3	96
Yttrium	0	4	96
Niobium	0	0	100

Metabolic processes concerned with the uptake, accumulation, and loss of radionuclides

There are many factors concerned with metabolic processes which are to be considered among the biological aspects of the accumulation of radiomaterials. It has been demonstrated that the metabolism of all forms of life is remarkably similar at the cellular level even though the morphological differences among aquatic organisms range from the bacteria through the vertebrate forms, and from the algae through the vascular plants. Nevertheless, differences do exist. These differences are governed by the complex anatomies, life histories, and physiological processes, and the relationships of the organisms with each other and with their environment. All of these differences must be considered in the light of the physical and chemical states of the elements involved.

In different organisms, ionized or particulate fission-product wastes and other radiomaterials may be either adsorbed, engulfed, or accumulated by metabolic processes. For example, Rothstein and his associates (1951) demonstrated that uranium as the uranyl ion was adsorbed by yeast cells. Hamilton and co-workers (see Hevesy, G., 1948, p. 441) showed that particulate radiomaterials such as various uncomplexed rare earths at physiological pH's were adsorbed by the gut lining of rats. In these experiments practically no accumulation of these particular radiomaterials by the animal was observed. On the other hand, Goldberg (1952) demonstrated with radioactive iron that a marine diatom assimilated particles of hydrated iron oxide, but that these organisms were unable to take up ionic iron in a complexed form.

The first biological experiments in which radioactive atoms were used were performed by Hevesy in 1923. In those classical experiments it was demonstrated that plants could take up lead from solution and translocate it throughout the vascular system.

The accumulation of radioelements is also dependent upon many chemical characteristics of the water in question. Among the parameters affecting accumulation are the salinity, percentage composition of the dissolved solids, pH, the oxygen-carbon dioxide ratio, and the presence of complexing agents.

a. *Chemical composition of marine organisms*

A modern systematic study of the inorganic constituents of marine organisms is yet to be made. The best summary of existing knowledge may be found in Vinogradov (1953).

However, certain generalizations can be drawn from the recent literature on the concentration of metals by marine organisms. Goldberg (in Treatise of Marine Ecology, volume II, edited by J. Hedgpeth, in press) has pointed out that the marine biosphere tends to concentrate such heavy metals as copper, nickel, zinc, etc., over the marine hydrosphere by factors of 100 to 100,000 on a weight-for-weight basis (Table 4). These metals are strongly bound in the organisms and cannot be easily removed by elution. Further, the elements most strongly concentrated in the biosphere are those that form the most stable complexes with organic chelating agents. As an example, copper is concentrated over sea water in the soft parts of most marine organisms by factors of 10^2 to 10^4 whereas calcium shows concentration factors of less than 1 to 50. Copper forms very strong complexes with many organic compounds whereas calcium does not. Although the exact role of most metals in the physiology of organisms is not known, nevertheless, one might *a priori* expect that some heavy metals introduced into the ocean from nuclear reactions would concentrate in the biosphere.

b. *Concentration in the environment*

The concentration of a given radiomaterial by an organism is sometimes proportional to the concentration of that material in the environment. This generalization applies both to aquatic and to terrestrial organisms. The uptake of cesium 137 by the oyster (*Crassostrea virginica*) has been shown to be dependent upon the external concentration of cesium in the sea water (Chipman, et al., 1954). Prosser, et al. (1945), noted that with the addition of strontium to the environment there was an increase in the uptake of that element by goldfish (*Carassius auratus*). Also, it has been demonstrated that as the carrier concentration in the nutrient environment is increased, the concentration factor for a particular fission product in terrestrial plants tends to increase (Rediske, et al., 1955).

c. *Effect of the presence of one element on the uptake of another element*

The uptake of one radioelement by an organism may be altered by the relative abundance of another element in the environment. In instances in which more than one element is involved, one of three phenomena may be observed:

First, elements of similar chemical properties may substitute for one another. For example, it has been shown by Prosser, et al. (1945), that when the amount of calcium in the water was low, there was an increase in the uptake of strontium 89 by goldfish. Conversely, as the amount of calcium was increased, the uptake of strontium decreased. Rice (1956) observed that cells of *Carteria* grown in artificial sea water took up strontium in proportion to the strontium/calcium ratio in the medium. Bevelander and Benzer (1948) have shown that a modification of the constituents of sea water resulted in a change in the constituents of the shells deposited by mollusks.

Second, some elements may have an inhibitory effect on others. A classical example of this

phenomenon is that in which calcium inhibits the stimulatory action of potassium on heart muscle.

Third, there may be a synergistic effect of one element on another. Ketchum (1939) has shown that the uptake of phosphorus by marine diatoms was enhanced with increased concentrations of nitrogen.

d. *Specificity of organisms and tissues for given elements*

The specific activity of a radionuclide in any organism is dependent upon the ability of the organism or any of its parts to concentrate that nuclide. If the stable counterpart of the radionuclide does not normally enter into the physiological processes of an organism, neither will the radioactive material.

It is well known that certain tissues have a predilection for concentrating specific elements. For instance, iodine is concentrated in the thyroid tissue of animals and hence radio-iodine will also be concentrated there. Strontium, like calcium, is a bone seeker and the radioisotopes of both of those elements will be concentrated in the bony skeletons of animals. Similarly, both strontium and calcium are concentrated in certain parts of vascular plants and so are the radioisotopes. Phosphorus is one of the principal constituents of bone and radiophosphorus is also concentrated in that tissue. The compound adenosine triphosphate is commonly present in the flight muscles of some birds and it has been shown that radiophosphorus is incorporated into the flight muscles of migratory waterfowl (Krumholz, 1954).

Although many different kinds of aquatic organisms have the ability to concentrate phosphorus in their tissues, there are few that show such a specificity for that element as the various plankters. The uptake of phosphorus 32 by plankton algae in a lake has been demonstrated by Coffin and his associates (1949) and others, and Whittaker (1953) showed that phytoplankters from the Columbia River concentrated radiophosphorus by factors as great as 300,000. Krumholz (1954, 1956) found that attached fresh-water algae (*Spirogyra*) concentrated radiophosphorus by a factor of 850,000, and that many fresh-water zooplankters concentrated that radionuclide by factors of more than 100,000. Approximate concentration factors for marine organisms are given in Table 4.

TABLE 4 Approximate Concentration Factors of Different Elements in Members of the Marine Biosphere. The Concentration Factors are Based on a Live Weight Basis.

Element	Form in Seawater	Concentration in seawater (micrograms/1.)	Algae (Non-calcareous)	Concentration Factors: Invertebrates Soft	Invertebrates Skeletal	Vertebrates Soft	Vertebrates Skeletal
Na	Ionic	10^7	1	0.5	0	0.07	1
K	Ionic	380,000	25	10	0	5	20
Cs	Ionic	0.5	1	10	—	10	—
Ca	Ionic	400,000	10	10	1,000	1	200
Sr	Ionic	7,000	20	10	1,000	1	200
Zn	Ionic	10	100	5,000	1,000	1,000	30,000
Cu	Ionic	3	100	5,000	5,000	1,000	1,000
Fe	Particulate	10	20,000	10,000	100,000	1,000	5,000
Ni[1]	Ionic	2	500	200	200	100	—
Mo	Ionic-Particulate	10	10	100	—	20	—
V	?	1	1,000	100	—	20	—
Ti	?	1	1,000	1,000	—	40	—
Cr	?	0.05	300	—	—	—	—
P	Ionic	70	10,000	10,000	10,000	40,000	2,000,000
S	Ionic	900,000	10	5	1	2	—
I	Ionic	50	10,000	100	50	10	—

[1] Values from Laevastu and Thompson (1956).

e. *Osmotic and ionic regulation*

Osmotic and ionic regulation are known to occur in a variety of ways. The usual pathways of excretion are through the urine, feces, skin, respiration, and particle ejection, and the method of excretion depends upon the particular organism and element involved. Ionic regulation may also occur by way of the chloride secreting cells in the gills of those fishes that migrate from salt to brackish water (Keys,

1931). Unfortunately, no experiments on such ionic regulation have been performed with radionuclides.

f. *Reproductive processes*

The reproductive processes of plants and animals range from simple fission among the unicellular organisms to the very complex relationships among the gametogenic forms. During reproduction there is a transfer of materials from the parent to the offspring.

In simple fission, the parent cell splits in two and each offspring receives approximately half of the parent material and thus only half of any radiomaterial that may have been present. Under conditions of chronic exposure, the offspring of organisms that reproduce by fission will incorporate usable radiomaterials into their bodies and a state of equilibrium eventually will be reached.

Among the egg-laying forms, most of the material received by the offspring is derived from the contents of the egg. In this form of reproduction, once the egg is laid there will be no further loss of radiomaterials from the mother or gain to the offspring. This applies even when the environment is contaminated and there is chronic exposure of the parents, because the protective coverings of the egg prevent the entrance of radiomaterials.

Among the forms that bear their young alive, however, there is usually some continuous transport of materials between the mother and the embryo. In such an instance it is probable that the embryo will accumulate radiomaterials with a resultant loss to the mother. If chronic exposure of a mother carrying an embryo continues during pregnancy, a state of equilibrium may eventually be reached between the mother and the environment and between the mother and the embryo.

During embryological development of all kinds there is a "biological dilution" of radiomaterials through cell division and growth. This statement applies primarily if there has been an acute exposure to radiomaterials or if the exposure has stopped with the commencement of the embryological development.

g. *Molting*

In instances where the embryos pass through a series of metamorphic stages, there is a loss of radiomaterials from stage to stage as, for example, the loss from instar to instar in insects through molting. Furthermore, it has been demonstrated by Chipman and coworkers (personal communication) that there is an increased accumulation of elemental constituents in crustaceans prior to molting, and a loss of such materials when the carapace is lost.

h. *Age and growth*

It has been established (Olson and Foster, 1952) that younger, more rapidly growing fishes accumulate relatively greater amounts of radiomaterials than do older, more slowly growing individuals. This phenomenon is probably a reflection of the more rapid metabolism that accompanies the growth of the younger fishes. It is not known whether the accumulation of radiomaterials by other aquatic vertebrates and invertebrates is a function of age and growth.

i. *Effect of temperature on cold-blooded and warm-blooded animals*

In general, the body temperatures of warm-blooded animals are more or less constant whereas the body temperatures of cold-blooded animals largely depend upon the temperature of the environment. Similarly, the rate of metabolism in warm-blooded animals is generally independent of temperature changes in the environment while that in the cold-blooded animals is largely dependent upon external temperatures. Changes in temperature affect the rates of chemical reactions and hence chemical processes that involve the accumulation of elements in the body tissues are temperature dependent.

Generally speaking, all cold-blooded aquatic organisms exhibit seasonal changes in the uptake and accumulation of radiomaterials from the environment. Davis, et al. (1953), and Krumholz (1954, 1956) have shown that there is a direct correlation between an increase in temperature and an increase in the accumulation of radiomaterials in fishes of the Columbia River, Washington, and of White Oak Lake, Tennessee, respectively. This increase in accumulation is apparently a reflection of the increase in the speed of the metabolic processes with rising water temperatures. However, Krumholz (1956) suggested that the fishes in White Oak Lake entered a period of dormancy following August 1 and lost about two-thirds of their accumulated radioactivity during the subsequent two months even though the water temperatures were much the same as they were during the earlier part of the summer.

In studies of the uptake of strontium 89 by

oysters and other shellfish at the Radiation Laboratory of the Fish and Wildlife Service (Chipman, unpublished data) it was found that the rate of uptake was slowed down and the retention time was extended when the animals were kept in sea water at low winter temperatures. Conversely, the rate of uptake was speeded up and the retention time was shortened when the animals were kept at summer temperatures. In other experiments at the same laboratory, it was found that larvae of the winter flounder (*Pseudopleuronectes americanus*) took up strontium 89 much more rapidly at higher water temperatures than at lower.

So far as is known, there is no demonstrable seasonal pattern of accumulation of radiomaterials among the warm-blooded aquatic vertebrates. It is generally believed that inasmuch as the body temperatures of those animals remain more or less constant throughout the year there will be no marked seasonal changes in the uptake of radiomaterials based on changes in rates of metabolism.

j. *Effect of light*

Light affects the uptake and accumulation of radioelements by plants. For example, it has been clearly shown by Scott (1954) that the uptake of radiocesium by the algae *Fucus* and *Rhodymenia* was greatly enhanced in the presence of light.

k. *Radiation effects*

Many aquatic organisms have the ability to concentrate radiomaterials in amounts deleterious to their well-being. These deleterious effects range from those in which only the individual is concerned to those in which the population as a whole may be affected. Elsewhere in this series of reports there is a paper on the effects of radiation on aquatic organisms.

Aspects of the accumulation of radionuclides through the ecosystem

For purposes of this paper, the aquatic biosphere can be divided into three trophic levels based on energy sources:

1. Primary producers, such as the photosynthetic plants.
2. Primary consumers, the herbivores, such as water fleas (cladocerans).
3. Secondary consumers, the carnivores, such as the largemouth bass or the tunas.

The community biomass (the total weight of all organisms in the community) is unequally divided between the three trophic levels. Usually there is a progressive decrease in both the biomass and the number of organisms from the first trophic level through the third, and a progressive increase in the size of the organisms. However, most community populations are constantly changing and are affected by seasonal, diurnal, and other cycles of abundance. These changes frequently have a profound effect on the environment and any changes in the environment in turn affect the stability of the community.

Generally speaking, the smaller organisms have a higher reproductive potential, a shorter life span, and a shorter time between generations; the length of the life span and the time between generations usually give a fair indication of the length of the embryological period. Furthermore, the smaller animals usually serve as food for the larger ones.

The discussion will consider the following aspects of the accumulation of radiomaterials in the three trophic levels: (1) the distribution of elements among the three levels, (2) the concentration factors in different organisms within the same level, and (3) the transport of radiomaterials from one trophic level to another.

Problems of the distribution of radionuclides among the trophic levels and the degree of concentration of radionuclides by different organisms can be approached most readily through separate consideration of the effects from an acute exposure and those from a chronic exposure.

A steady-state condition will be approximated when the amounts of radiomaterials introduced into the environment is equal to the amount that disappears through physical decay. Any organisms living in such an environment will suffer chronic exposure to the radioactivity, the level depending, of course, on their ability to concentrate the radiomaterials introduced and on the steady-state concentration of these materials in the surrounding medium. An approximation of the concentration factors for some organisms is given in Table 4.

Davis and co-workers (1952) showed that there was a progressive decrease in the amount of radioactivity found in the aquatic organisms of the Columbia River downstream from the Hanford Works. There, the principal radionu-

clide was phosphorus 32, which has a physical half-life of about 14 days. It is apparent that when following the steady-state transport of radiomaterials through the ecosystem the following parameters must be considered: (1) the physical half-life of the radionuclide, (2) the distance of the organism from the source of radioactive contamination, and (3) the dilution of the radiomaterials between the point of introduction and the area in which the organism lives.

The results from acute exposure cannot be as definitely approximated as for chronic exposure. In such instances, the time element is very important, and the following must be known: (1) the rate of dilution of the radioactive water mass with non-radioactive water; (2) the rate of transfer of radiomaterials from one trophic level to another with the concurrent dilutions and losses or gains in concentration by the organisms; and (3) the life span of the organisms involved.

In general, the radiomaterials taken up by organisms of the first trophic level will be primarily in the ionized state although a certain amount of particulate radiomaterials will be adsorbed to the body surfaces. When uptake occurs, the rate of uptake will probably be more rapid than the rate of uptake in the other trophic levels.

Particulate radiomaterials tend to be concentrated in the second trophic level. Findings from the Wigwam and Castle tests (Goldberg, unpublished data) showed that the principal organisms which concentrated particulate radiomaterials were the mucous, ciliary, and pseudopodial feeders among the zooplankters. These organisms contained much more radioactivity per unit weight than either the algae or the setal or rapacious feeders.

In addition to the differences in concentration of radiomaterials from one trophic level to another, there are marked differences among species in the same level. For instance, it has been shown by Chipman, et al. (1953), that some phytoplankters will concentrate radiostrontium by a factor of about 20 times whereas others will concentrate the radioelement by factors as much as 1500 times. Comparable data have been recorded by Krumholz (1954) for the accumulation of radiophosphorus by the phytoplankters of White Oak Lake.

Differences also exist between individuals of the same species. Very large differences in the amounts of radiomaterials accumulated by individual fishes in White Oak Lake were described by Krumholz (1956). For instance, he reported that the amounts of radiostrontium in the bones of three bluegills (*Lepomis macrochirus*) differed by more than five-fold. These three fish were taken from the same place in the lake on the same day, August 27, 1952. Comparable differences were found in the amounts of accumulated radiomaterials in most other tissues.

The transfer of radiomaterials from one trophic level to another is not only dependent upon the concentration of the radiomaterial in the organism but also is governed by the rate of growth of the organism and the rate of increase in the size of the population. These factors of transfer are of particular importance in the event of an acute exposure because the dilution brought about through cell division and growth may well minimize any radiation effect. In any event, there is always a loss in the total amount of radiomaterials in the transfer from one trophic level to another (though not necessarily a decrease in the concentration in individual organisms). Such a loss may be relatively small or it may be very great depending upon the organism and the particular food web involved.

Not all radiomaterials that enter the first trophic level are passed on to higher levels. At each trophic level there are certain species that, for one reason or another, are not widely used as food by the organisms of higher levels. Also, some of the plants of the first trophic level may die before they are eaten and thus will be returned to the environment as organic matter. In this case the primary producers may be of little or no importance as a source of radiomaterials to the organisms of the second and third trophic levels.

If relatively large quantities of radiomaterials are accumulated in certain hard parts of an organism, such as the shell of an oyster or the bones of a fish, they will, in all probability, remain in those parts during the greater part of the life of the animal concerned, and will not be available to other animals in the biosphere until the animal dies.

Chipman and co-workers (1953) showed that oysters fed on *Chlorella* assimilated only very little of the radiophosphorus from these

algae. On the other hand, oysters fed upon other phytoplankters that contained no more radiophosphorus than the *Chlorella* accumulated relatively large amounts of radiophosphorus and incorporated that element into their tissues as organic phosphorus compounds. It appears that the particular food web used by any organism is of primary importance in the transfer of radiomaterials from one trophic level to another.

Problems for further research

One of the fundamental questions to be answered concerns the mechanism of incorporation of the heavier elements, such as the fission products, in aquatic organisms. To date, no metal heavier than molybdenum has been shown to be necessary for metabolic processes. Specifically, we need to know:

1. How are the radioactive elements passed through membranes and where and why do they concentrate in the organisms?
2. What are their biological half-lives of the different radioactive elements in different organisms?
3. What are the average and extreme concentration levels of these elements in various organisms and in the biosphere?

The revolution in biological thought brought about by the use of labelled atoms is manifest in all branches of biological research today. Radioisotopes have permitted the study of rate processes that could not have been investigated in any other way. Such processes include the pumping rates of water and other biological fluids, and the transfer of molecules or portions of molecules from tissue to tissue, or, on the ecological level, from organism to organism.

REFERENCES

Bevelander, G., and P. Benzer. 1948. Calcification in marine molluscs. *Biol. Bull.* **94**:176–83.

Boroughs, H., S. J. Townsley and R. W. Hiatt. 1956. The metabolism of radionuclides by marine organisms. I. The uptake, accumulation and loss of strontium 89 by fishes. *Biol. Bull.* III:336–351.

Chipman, W. A., T. R. Rice, and T. J. Price. 1953. Accumulation of radioactivity by marine invertebrate animals. U. S. Fish and Wildlife Service Radioisotope Laboratory, Progress Report, April 1953, (Typewritten).

1954. Accumulation of fission products by marine plankton, fish, and shellfish. U. S. Fish and Wildlife Radioisotope Laboratory, Progress Report, July-December 1954, (Typewritten).

Coffin, C. C., F. R. Hayes, L. H. Jodrey, and S. G. Whiteway. 1949. Exchange of materials in a lake as studied by the addition of radioactive phosphorus. *Canad. Jour. Research* **27**:207–222.

Davis, J. J., R. W. Coopey, D. G. Watson, C. C. Palmiter, and C. L. Cooper. 1952. The radioactivity and ecology of aquatic organisms of the Columbia River. *In* Biology Research — Annual Report, 1951, USAEC Document HW-25021, pp. 19–29.

Goldberg, E. D. 1952. Iron assimilation by marine diatoms. *Biol. Bull.* 102, 243–8.

Greendale, A. E., and N. E. Ballou. 1954. Physical state of fission product elements following their vaporization in distilled water and seawater. USNRDL Document 436, pp. 1–28.

Hevesy, G. 1923. Absorption and translocation of lead by plants. *Biochem. Jour.* **17**:439–45.

1948. Radioactive indicators. Interscience Publishers, New York, xvi+555 pages.

Hiatt, R. W., H. Boroughs, S. J. Townsley, and Geraldine Kau. 1955. Radioisotope uptake in marine organisms with special reference to the passage of such isotopes as are liberated from atomic weapons through food chains leading to organisms utilized as food by man. Hawaii Marine Laboratory, Annual Report, AEC project number AT(04–3) 56, pp. 1–29, (Mimeographed).

Ketchum, B. H. 1939. The adsorption of phosphate and nitrate by illuminated cultures of *Nitzchia closterium*, *Am. J. Botany* **26**:399–402.

Keys, A. B. 1931. Chloride and water secretion and absorption by gills of the eel. *Zeitsch. Vergl. Physiol.* **15**:364.

KRAUSKOPF, K. B. 1956. Factors controlling the concentrations of thirteen rare metals in sea water. *Geochim. et Cosmochim. Acta* 9:1–32.

KRUMHOLZ, L. A. 1954. A summary of findings of the ecological survey of White Oak Creek, Roane County, Tennessee, 1950–1953. USAEC Document ORO-132, pp. 1–54, (Mimeographed).

1956. Observations on the fish population of a lake contaminated by radioactive wastes. *Bull. Amer. Mus. Nat. Hist.* **110**(4):277–368.

KRUMHOLZ, L. A., and J. H. RUST. 1954. Osteogenic sarcoma in a muskrat from an area of high environmental radiostrontium. *A.M.A. Arch. Path.* **57**:270–278.

LAEVASTU, T., and T. G. THOMPSON. 1956. The determination and occurrence of nickel in sea water, marine organisms and sediments. *J. du Cons.* **21**:125–143.

OLSON, P. A., JR., and R. F. FOSTER. 1952. Effect of pile effluent water on fish. *In* Biology Research — Annual Report, 1951. USAEC Document HW-25021, p. 41.

PROSSER, C. L., W. PERVINSEK, JANE ARNOLD, G. SVIHLA, and P. C. TOMPKINS. 1945. Accumulation and distribution of radioactive strontium, barium-lanthanum, fission mixture and sodium in goldfish. USAEC Document MDDC-496, October 13, 1954.

REDISKE, J. H., J. F. CLINE, and A. A. SELDERS. 1955. The absorption of fission products by plants. *In* Biology Research — Annual Report, 1954. USAEC Document HW-35917, pp. 40–46.

RICE, T. R. 1956. The accumulation and exchange of strontium by marine and planktonic algae. *Limnology and Oceanography* **1**(2):123–138.

ROTHSTEIN, A., and R. MEIER. 1951. The relationship of cell surface to metabolism VI, the chemical nature of the uranium complexing groups of the cell surface. *J. Cell. Comp. Physiol.* **38**:245–70.

SCOTT, R. 1954. A study of cesium accumulation by marine algae. Proc. Second Radioisotope Conference, pp. 373.

VINOGRADOV, A. P. 1953. The elementary composition of marine organisms. Sears Foundation for Marine Research, Memoir No. 2.

WHITTAKER, R. H. 1953. Removal of radiophosphorus contaminant from the water in an aquarium community. *In* Biology Research-Annual Report, 1952. USAEC Document HW-28636, pp. 14–19.

CHAPTER 8

LABORATORY EXPERIMENTS ON THE UPTAKE, ACCUMULATION, AND LOSS OF RADIONUCLIDES BY MARINE ORGANISMS[1]

HOWARD BOROUGHS, *Hawaii Marine Laboratory, University of Hawaii,* Honolulu, Hawaii

WALTER A. CHIPMAN, *Fishery Radiobiological Laboratory, U. S. Fish and Wildlife Service,* Beaufort, North Carolina

THEODORE R. RICE, *Fishery Radiobiological Laboratory, U. S. Fish and Wildlife Service,* Beaufort, North Carolina

WHAT happens to radioactive materials that are introduced into the oceans may be studied by a marine biologist from at least two points of view. As a physiologist, he will be interested in the uptake, accumulation, and loss of radioelements as a function of the element, and its concentration; in the physical factors of temperature, light, and salinity; and in differences between species of organisms, as well as their age and sex, to mention some of the most important parameters. As an ecologist, he will be interested in these same parameters under a steady-state condition. The physiologist would profit most by exposure of the organism to a single dose of radioactive material, while the ecologist must concern himself with the results of chronic exposure.

Both types of biologists may be interested in tracing the history of an element through the food webs of the various trophic levels. Unfortunately, the experimental data involving the metabolism of radionuclides by marine organisms is extremely meager. In this section some experiments will be described on the uptake, accumulation, and loss of radionuclides by various marine organisms in the three trophic levels. It must be emphasized that the results of these few experiments must be extrapolated with extreme caution in predicting what may happen to radioactive materials introduced into the oceans from nuclear reactor plants, bomb detonations, or from any other sources.

In discussing the uptake of radionuclides by marine organisms, it is sometimes difficult to state exactly what constitutes a single or a chronic exposure. For a unicellular alga, a few hours may represent chronic exposure, while a few weeks may be insufficient for a fish to reach a steady-state condition. No long-term repetitive feeding experiments have been done, so for the purpose of this report, we will discuss the metabolism of the various radionuclides solely on the basis of the trophic level concerned.

The term uptake implies passage through a membrane. Radioactive material may be present in the gut of an organism, but until it enters the organism through a membrane, it can play no role in the metabolism of that organism except by producing radiation effects or by interfering with a chemical reaction occurring within the gut. In some of the experiments to be described, particularly those involving phytoplankton, it was not established whether or not the radioisotope was actually incorporated into the organism, or merely adsorbed to the surface. For simplicity, we will therefore discuss uptake in the sense that the radioisotope is associated with the organ or organism in question.

Isotopes of a given element usually have similar chemical behavior, so that in tracing the path of most elements in biological systems, it can be assumed that a radioactive atom will behave in the same way as a non-radioactive atom of the same species. The only parameters to be considered in the discussion to follow will be the species and age of the organism, the element, the concentration of the element, the temperature, and the duration of exposure or treatment. No work using radioisotopes has been

[1] Work performed at the Fishery Radiobiological Laboratory of the U. S. Fish and Wildlife Service and the Hawaii Marine Laboratory (Drs. H. Boroughs, S. J. Townsley, and R. W. Hiatt).

Contribution No. 95, Hawaii Marine Laboratory.

done on the mineral metabolism of marine organisms relative to sex. The data that will be presented were collected either at the Fishery Radiobiological Laboratory of the United States Fish and Wildlife Service (R.L.F.W.S.) or the Hawaii Marine Laboratory, University of Hawaii (HML).

First trophic level

Experiments performed at the R.L.F.W.S. very clearly show that different species of planktonic algae have remarkably different abilities to concentrate a particular element from the sea water medium. Algae were grown in the presence of radiostrontium obtained from Oak

TABLE 1 The Differential Uptake of Radioactive Strontium and Yttrium By Algae

Species	Percentage activity from strontium	Percentage activity from yttrium
Carteria sp.	100.0	0.0
Thoracomonas sp.	50.4	49.6
Amphora sp.	10.0	90.0
Navicula sp.	8.5	91.5
Chromolina sp.	8.2	91.8
Chlamydomonas sp.	6.5	93.5
Nitzschia closterium	6.0	94.0
Nannochloris atomus	5.7	94.3
Chlorella sp.	5.3	94.7
Porphyridium curentum	4.4	95.6
Gymnodinium splendins	4.1	95.9
Gyrodinium sp.	2.3	97.7

Ridge. The material used contained both Sr^{89} and Sr^{90}; the latter decays to form Y^{90}. By counting the algal samples immediately after they were removed from the culture medium, and again after several weeks, in order to allow the secular equilibrium of the Sr^{90}-Y^{90} pair to be reached, it was possible to determine what percentage of the original radioactivity was due to strontium. Table 1 shows that *Carteria* sp. accumulated strontium 89 and 90 from the isotopic mixture, and that *Gyrodinium* sp. removed almost no strontium 89 or 90, but instead accumulated yttrium 90. It was found that *Nitzschia closterium* under an apparent steady state condition concentrated strontium 17 times over its concentration in sea water (weight of algae/weight of water). The concentration factor for strontium *Carteria* sp. was found to vary with condition of culture but was much greater than for *Nitzschia closterium.*

Experiments using cesium^{137} show that while different species concentrate cesium to different degrees (Table 2) none of the nine species

TABLE 2 Concentration of Cesium By Marine Algae

Algae	Concentration factor [1]
Bacillariaceae	
Nitzschia closterium	1.2
Amphora sp.	1.5
Nitzschia sp.	1.7
Chlorophyceae	
Chlamydomonas sp.	1.3
Carteria sp.	1.3
Chlorella sp.	2.4
Pyramimonas sp.	2.6
Nannochloris atomus	3.1
Rhodophyceae	
Porphyridium curentum	1.3

[1] The concentration factor is reported as the ratio of Cs^{137} in the algae (wet weight) to that in an equivalent weight of sea water at an apparent steady-state condition.

tested from three families showed any marked concentration of this element from sea water.

The effect of the concentration of an element on its uptake by *Nitzschia* cells is shown in Figure 1. *Nitzschia* cells were grown in sea water to which had been added labelled zinc at three different concentrations. From the graph

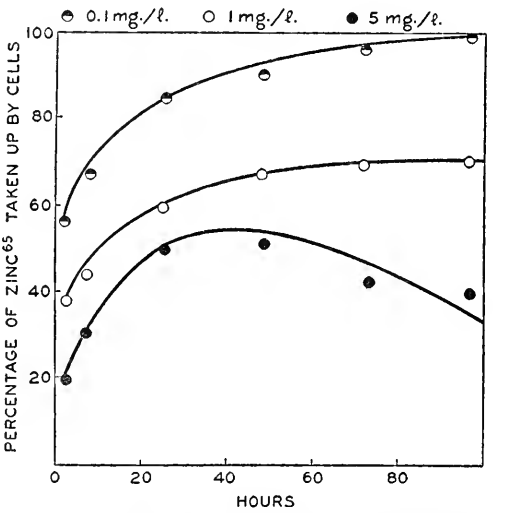


Figure 1. Uptake of $Zinc^{65}$ by *Nitzschia* Cells from Culture Medium Containing Different Concentrations of Zinc.

- 0.1 mg./l
- 1 mg./l
- 5 mg./l

it is evident that at low concentrations all the zinc was removed after about four days. The lowest concentration used was still ten times higher than the average zinc concentration of sea water.

The rate of uptake of zinc^{65} by *Nitzschia* cells is shown in Figure 2. At the normal con-

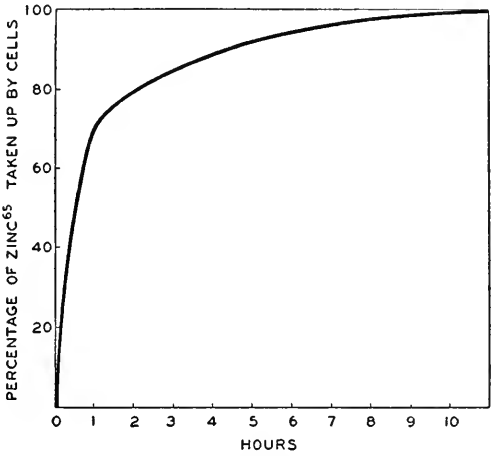


FIGURE 2. Uptake of Zinc^{65} by *Nitzschia* Cells from Culture Medium Containing 10 Micrograms of Zinc/Liter.

centration of zinc in sea water, a dividing culture of *Nitzschia* depleted the zinc^{65} in a closed system in less than one day. Apparently phytoplankton cells concentrate zinc relative to sea water and any radioactive zinc present in the water will be quickly taken up in large amounts.

The radioisotopes so far discussed are very likely always ionic in sea water. Ruthenium solution, however, forms colloids and particles when put into sea water. Ruthenium^{106} obtained as an acid solution from Oak Ridge was added to a sea water culture of *Nitzschia* cells. Figure 3 shows that the cells continued to take up the ruthenium for the 12 days of the experiment. The amount of ruthenium per cell decreased, however, since the cells of the culture were dividing continually. One may conclude from this experiment, that since the ruthenium concentration in sea water is low, dividing planktonic algae would take up large amounts of any radioactive ruthenium present.

Second trophic level

The work reported in this section was also done at the R.L.F.W.S. Larvae of the brine shrimp *Artemia* were put into filtered sea water containing radiostrontium and the daughter

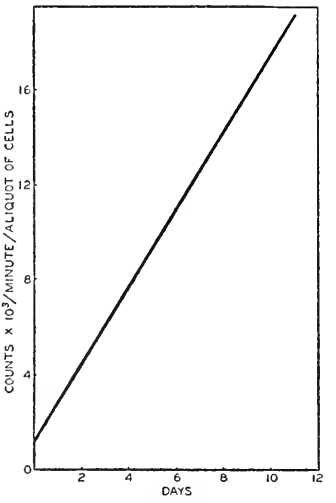


FIGURE 3. Uptake of Ru^{106} by *Nitzschia* Cultures in the Light.

yttrium^{90}. These larvae rapidly took up the SR^{89}-$Sr^{90}Y^{90}$ and reached an apparent steady-state in a few hours. After exposure of the organisms to the isotopes for one day, it was found that the amount of radioactivity/g of *Artemia* was only 70 per cent of that of an equal weight of the sea water. A count of the samples 30 days after their preparation indicated that a considerable amount of Y^{90} was taken up. Other crustaceans used were the shrimp *Penaeus setiferus* and the edible blue crab *Callinectes sapidus*. The molluscan shellfish studied included oysters (*Crassostrea virginica*), clams (*Venus mercenaria*), and scallops (*Pecten irradians*).

All of these organisms accumulated strontium rapidly from sea water. The internal distribution of strontium in oysters is shown in Table 3. This table indicates that the bulk of the radioactivity accumulates in the shell. When

TABLE 3 DISTRIBUTION OF RADIOACTIVITY IN OYSTERS FOLLOWING EXPOSURE TO SEA WATER CONTAINING SR^{89}

Tissues	Per cent of total weight	Per cent of total activity	Per cent of soft tissues	Per cent of activity of soft tissues
Mantle	2.5	4.1	25.0	27.7
Gills	1.7	3.1	17.5	21.2
Adductor muscle	1.9	2.4	19.2	16.2
Other	3.8	5.1	38.3	34.9
Total soft tissues	9.9	14.7	—	—
Shell	90.1	85.3	—	—

the radioactive shellfish were returned to a normal sea water environment, the radioactivity present in the soft tissues declined within one day to 10 per cent or less of the maximum concentration. This residual amount was held by the tissues for several days.

The uptake of radiostrontium by oysters from food was studied by growing *Carteria* cells in sea water to which Sr^{89} was added. Oysters in Sr^{89} sea water served as the controls; the treated oysters were kept in Sr^{89} sea water to which the labelled *Carteria* cells were added. Fresh sea water and plankton suspensions were prepared each day. The curves in Figure 4 show that an

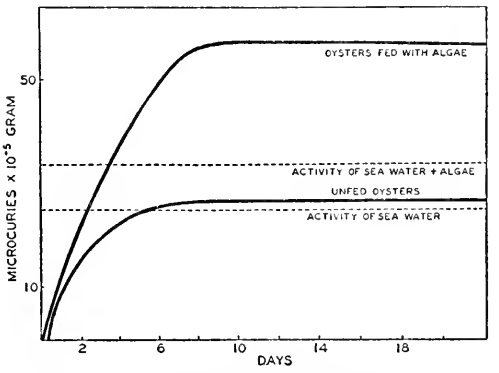


FIGURE 4. The Increased Accumulation of Sr^{89} by Oysters Feeding on Sr^{89}-Fed Algae.

apparent steady-state is reached in eight days. In the unfed oysters the concentration of Sr^{89} in the soft parts is approximately the same as the concentration in the sea water. The oysters which fed on the radioactive algae, however, concentrated the Sr^{89} by a factor slightly greater than two, based on the radioactivity of the suspension per unit of weight. These filter-feeding organisms removed the algal cells from many volumes of water.

The uptake of $cesium^{137}$ by clams, *Venus mercenaria* L., is shown in Figure 5. At the end of 20 days the soft parts of clams had concentrated the cesium by a factor of six over the cesium concentration of sea water. Obviously a steady state had not occurred, so that it is not possible to say what the final concentration factor of clams might be for cesium in solution. Similar experiments using the bay scallop, *Pecten irradians* L., show that the concentration factor of cesium is greater than eight, since the uptake was still increasing at the end of 10 days.

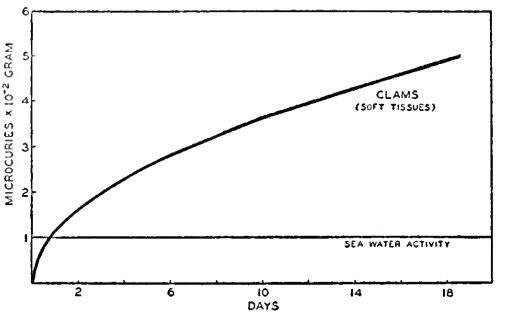


FIGURE 5. The Accumulation of $Cesium^{137}$ by Clams as a Function of Time.

Bay scallops immersed for two hours in sea water containing Zn^{65} very rapidly accumulated this isotope. Table 4 lists the internal distribution of $zinc^{65}$ in the various tissues. The concentration factor for each organ is readily calculated since the activity of the sea water was 10 mμc/g. This means that the figures given in the second column divided by 10 equal the concentration factors. The over-all concentration factor of the soft tissues of the bay scallop was 20 for this short interval. Other observations showed that these scallops contained close to 35,000γ of zinc per gram (wet weight) and thus had a concentration factor for this element of about 3500.

Oysters that were kept in sea water with added Zn^{65} also quickly accumulated the isotope to very high levels. The zinc content of fresh oyster tissue measured almost 170,000γ per gram. This represents a concentration factor of 17,000, since the zinc concentration of the sea water in which the oysters lived was about 10 mcgm/1.

$Ruthenium^{106}$ was one of the separated fission products used to study the uptake of particulate radioisotopes by organisms in the second trophic level. Ruthenium was co-precipitated with calcium carbonate, dried, and ground

TABLE 4 DISTRIBUTION OF Zn^{65} IN THE ORGANS OF THE BAY SCALLOP AFTER A TWO HOUR IMMERSION

Tissue	mμc Zn^{65}/g.	Total mμc
Kidney	1384	824
Liver	243	507
Gills	218	857
Testes and ovaries	138	193
Foot	131	25
Rectum	120	8
Heart	105	13
Adductor muscle	100	375
Mantle	92	321

to a very fine state. Plutei of the sea urchin, *Arbacia punctulata* were put into sea water containing the radioruthenium which was kept in suspension by aerating the culture flask. After 18 hours, the larvae were rinsed and resuspended in fresh sea water. Aliquots of larvae were then removed at intervals and tested for radioactivity (Table 5). A microscopic examination of the larvae at zero time showed that the intestines were filled with the radioactive particulate material, but at 8 hours, very little material was left in the gut. Apparently little ruthenium was actually absorbed through the digestive tract.

TABLE 5 The Decrease of Ru^{106} in Sea Urchin Larvae as a Function of Time in Non-radioactive Water

Hours	Radioactivity in 500 larvae (counts/minute)
1	1413
4	179
8	148

The ingestion of the particulate (co-precipitated) ruthenium by the bay scallop, *Pecten irradians,* also indicated that the radioactivity was mostly associated with the digestive tract. The crystalline style was highly radioactive, although the radioactivity in it decreased during the five days the scallops were kept in running water. The hepatopancreas, on the other hand, showed an increase in radioactivity during this time. No radioactivity was found associated with the internal organs other than those in the digestive tract.

Third trophic level

The uptake, accumulation, and loss of radionuclides has been studied in many fishes by both the R.L.F.W.S. and the H.M.L. These fishes include the skipjack tuna (*Euthynnus yaito*), yellowfin tuna (*Neothunnus macropterus*), dolphin (*Coryphaena hippurus*), papio (*Carangoides ajax*), aholehole (*Kuhlia sandvicensis*), *Tilapia mozambique,* menhaden (*Brevoortia tyrannus*), bluefish (*Pomatomus saltatrix*), little tuna (*Euthynnus allitteratus*), croakers (*Micropogon undulatus*), and king whiting (*Menticirrhus* sp.).

At the H.M.L., strontium^{89} in gelatine capsules was fed to skipjack, dolphin, and yellowfin tuna. These are all fast-swimming pelagic fish. Figure 6 shows that the excretion of strontium is very rapid. In 24 hours, only about two per cent of the dose remains in the fish. Similar experiments with *Tilapia,* a small, sluggish bottom feeder, indicate that the strontium is also mainly excreted, but that the time required to reach a minimum level of about five per cent of the dose requires at least four days. This information is consistent with the idea that the metabolic rates of these fishes are very much different, and the sluggish fish might be expected to retain the strontium for longer periods.

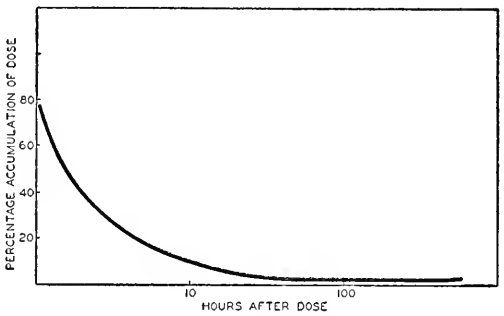


Figure 6. The Percentage Accumulation by Tuna Fish of Sr^{89} Given Orally.

The internal distribution of the total radioactivity recovered is shown in Table 6. By plotting the radioactivity of each organ against time, it is apparent that the soft, visceral tissues rapidly excrete the strontium, but that the bony structures, gills, integument, and muscles retain the strontium for a long period. *Tilapia* show the same behavior. The data are presented in Table 7.

The direct uptake of strontium^{89} in solution by *Tilapia* was also studied at the H.M.L. Figure 7 shows that after about two weeks, the

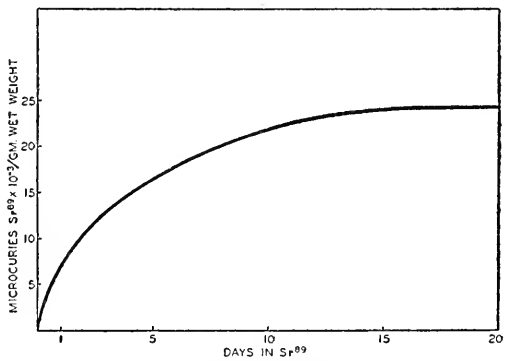


Figure 7. The Uptake of Sr^{89} in Solution by *Tilapia Mozambique.*

TABLE 6 ACCUMULATION OF Sr^{89} IN THE VARIOUS ORGANS AND TISSUES OF TUNA AFTER INGESTION

	Percentage of total activity									
	Dose: 5.55μc	Dose: 480μc	Dose: 240μc	Dose: 51.0μc	Dose: 240μc	Dose: 464μc	Dose: 464μc	Dose: 464μc	Dose: 371μc	Dose: 371μc
Tissue	1 hr	2¼ hr	6 hr	7 hr	11½ hr	24 hr	96 hr	264 hr	480 hr	648 hr
Heart	0.04	0.01	0.03	0.049	0.11	0.05	0.028	0.01	0.014	0.007
Gall bladder	0.05	0.04	0.07	0.10	0.08	0.03	0.0001	0.01	0.004	0.002
Blood	4.21	6.68	15.00	0.85	8.07	2.73	1.14	0.35	0.51	0.12
Gill flesh	12.44	0.18	0.91	8.56	5.06	30.61	25.72	2.42	1.42	2.21
Gill bone		1.34	6.47		26.39			16.80	19.48	22.76
Caecum	37.01	7.67	7.84	2.70	2.64	0.34	0.15	0.05	0.04	0.029
Foregut	0.89	9.32	1.12	0.74	1.03	0.20	0.24	0.04	0.04	0.018
Midgut	2.28	14.16	16.50	1.08	1.48	0.65	0.25	0.05	0.036	0.003
Hindgut	11.78	3.98	21.26	2.26	0.11	0.15	0.024	0.03	0.015	0.016
Gut contents	—	48.32	12.73	0.056	19.65	—	0.10	0.013	—	0.0008
Head, operculum	—	0.41	1.09	24.99	6.28	18.33	24.58	28.18	29.91	24.58
Appendicular skel.	3.60	0.40	1.19	36.21	8.45	23.69	30.32	29.15	30.47	31.43
Liver	3.34	1.48	3.04	0.39	2.46	0.15	0.04	0.03	0.04	0.027
Spleen	0.20	0.32	1.39	0.08	0.60	0.03	0.008	0.03	0.010	0.003
Tail	—	0.42	0.15	—	0.00	—	—	—	—	—
Brain, spinal cord	0.23	0.00	0.01	1.24	0.05	1.66	1.70	1.33	0.030	0.004
Eyes		0.04	0.06		0.60			2.02	1.34	1.34
Integument	5.28	1.69	0.86	10.20	5.89	7.69	11.37	13.73	10.25	10.51
Integument flesh (aliquot)	—	0.01	0.01	—	0.05	—	—	—	—	0.065
Integument scales (aliquot)	—	0.02	0.02	—	0.11	—	—	—	—	0.091
Gonad	2.40	0.09	0.47	0.22	0.08	0.06	0.004	0.03	0.023	0.020
Kidney		0.08	0.16		0.09		0.027	0.07	0.035	0.022
Light muscle	16.23	3.23	8.74	4.19	10.01	12.84	3.94	5.26	5.69	5.79
Dark muscle		0.10	0.86	5.25	0.70	0.72	0.48	0.47	0.63	0.95

TABLE 7 THE INTERNAL DISTRIBUTION AND PERCENTAGE RECOVERY OF A DOSE OF 75 μC OF Sr^{89} BY *Tilapia*

Days after dose	Tissue	Percentage of total recovered
1	Skin	25.27
	Eyes	0.35
	Visceral organs	1.82
	Gills	15.62
	Muscle	5.65
	Skeleton	51.30
	Total	100.01
2	Skin	24.44
	Eyes	0.18
	Visceral organs	1.08
	Gills	8.13
	Muscle	3.10
	Skeleton	63.07
	Total	100.00
4	Skin	24.56
	Eyes	0.25
	Visceral organs	1.01
	Gills	6.42
	Muscle	8.40
	Skeleton	59.35
	Total	99.99

TABLE 7—*Continued*

Days after dose	Tissue	Percentage of total recovered
7	Skin	22.79
	Eyes	0.33
	Visceral organs	1.14
	Gills	10.11
	Muscle	3.01
	Skeleton	62.62
	Total	100.01

uptake apparently levels off at a value which corresponds to a concentration factor of about 0.3. This means that these fish can to some extent exclude the strontium ion in solution. Even the skeleton had not yet come to equilibrium with the radioactivity in the sea water. This may mean that only about 70 per cent of the strontium in the bone is readily exchangeable. The remainder may be firmly bound in a lattice or to an organic matrix which has a slow rate of turnover. It should be emphasized that these fish were mature.

Experiments done at the R.L.F.W.S. with Sr^{89} on post-larval flounders indicate that age

and temperature influence the uptake of elements in solution. One group was kept at 20–22° C in sea water containing Sr^{89}, and another group at 8–12° C. The fish in both groups averaged 0.02 g. each. Figure 8 shows

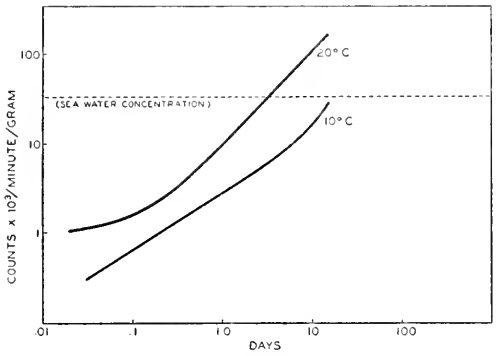


FIGURE 8. Uptake of Sr^{89} by Larval Flatfish.

that strontium was taken up more rapidly at the higher temperature. Thus at one day, the fish at the lower temperature had less than one third of the radioactivity of the fish at the higher temperature. The graph also shows that very young fish continue to take up strontium from solution very rapidly at 14 days, while at 14 days the *Tilapia* had reached an apparent steady state condition.

The uptake of $zinc^{65}$ by croakers was studied at the R.L.F.W.S. These fish were fed the isotope in hardened gelatine. After 12 hours only about 27 per cent of the dose remained in the fish (Table 8). The distribution of zinc is quite different from that of strontium. About 90 per cent of the strontium retained by the various fishes used at the H.M.L. was found in the gills, bones, and integument. Zinc, however, is concentrated mainly in the liver and spleen. The muscle and bone, because of their bulk accounted for a large part of the total $zinc^{65}$ of the body. The turnover times of the zinc-containing compounds of the skin, muscle and bone were slow, whereas those of the internal organs were relatively rapid.

The uptake of radiocesium by fish was studied at the R.L.F.W.S. Table 9 shows the distribution of $cesium^{137}$ which was fed to little tuna. It can be seen that the liver, heart, spleen, and kidney rapidly take up the cesium, but these organs also lose the cesium during the following week. Muscle, gonad, brain, and skin, on the other hand, continue to accumulate the cesium faster than they lose it.

TABLE 8 $ZINC^{65}$ DISTRIBUTION IN CROAKERS (*Micropogon undulatus*) 12 HOURS AFTER ORAL ADMINISTRATION

Tissue or Organ	Per cent of total weight	Weight in grams	$Zinc^{65}$ mμc/gm.	$Zinc^{65}$ mμc per tissue or organ	Per cent of total body $zinc^{65}$
Muscle	80	48.80	1.6	78.1	44.7
Bone	11	6.71	5.5	36.9	21.1
Gills	2	1.22	10.9	13.3	7.6
Liver	0.8	0.49	40.7	19.9	11.4
Gonads	0.4	0.24	17.6	4.2	2.4
Kidney	0.3	0.18	41.5	7.5	4.3
Heart	0.2	0.12	14.0	1.7	1.0
Spleen	0.1	0.06	25.3	1.5	0.9
Remainder ..	5.2	3.17	3.7[1]	11.7	6.7
Skin, scales					
G I tract		60.99		174.8	
Blood					
Brain					
Eyes					
etc.					

[1] Based on skin and scales

Dose per fish—6,100 mμc

Distribution after 12 hours

Tissues	3 percent	
G I tract	24 percent	
Loss	73 percent	(mostly excreted)

The accumulation of cesium in solution was demonstrated by keeping croakers in Cs^{137} enriched sea water. The water was changed daily to maintain a relatively constant concentration. Figure 9 indicates that during the 29 days, the heart, spleen, liver, brain, and muscle continued to accumulate the cesium. The concentration factor for the heart, spleen, and liver, was about 10, but this value is far below an equilibrium value.

TABLE 9 THE DISTRIBUTION OF $CESIUM^{137}$ IN THE TISSUES OF THE LITTLE TUNA AS A FUNCTION OF TIME

	Cs^{137} content in μc/g. wet wt. Days after the dose			
Organ	1	3	6	8
Spleen	3.46	3.29	3.22	1.57
Liver	9.07	4.71	3.45	2.59
Kidney	3.13	2.58	1.78	0.95
Heart	6.17	3.15	2.52	1.57
Bone	0.41	0.40	0.17	0.26
Eye	0.30	0.26	0.31	0.30
Muscle	0.46	0.67	0.87	0.79
Gonad	0.54	0.70	1.33	1.34
Brain	0.30	0.35	0.69	0.66
Skin	0.29	0.46	0.41	1.01

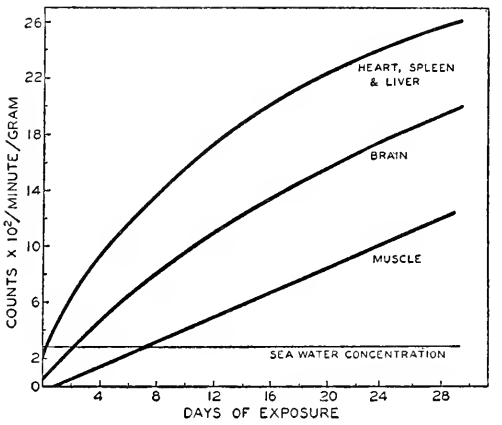


FIGURE 9. Accumulation of Cs^{137} by Croakers Kept in Sea Water Containing 5×10^{-3} Microcuries/Ml.

The relative concentration of cesium by the various organs is roughly the same for croakers, tuna, or bluefish. The same rank order among the organs is maintained both from ingestion, and from direct uptake.

Menhaden, a filter feeder, were put into sea water with ruthenium^{106} that had been co-precipitated with calcium carbonate. Although a considerable amount of particle settling occurred, the menhaden took up the ruthenium in the digestive tract, but the tissues of the fish did not become radioactive to an appreciable extent. Similar experiments using menhaden fed with Ru^{106} labelled *Arbacia* plutei, or Ru^{106} labelled *Nannochloris* cells, gave parallel results. In the latter experiment the fish were allowed to eat the labelled cells for four hours, and then they were put in running sea water. At the time of transfer about 92 per cent of the ingested dose was found in the digestive tract. The gills had 0.64 per cent of the dose, and the remainder of the fish, including the skin, had 0.76 per cent. At 128 hours, only 0.05 per cent of the ingested dose remained in the digestive tract. There was 0.25 per cent in the fish body or on the skin surface, and 0.01 per cent in or on the gills. At no time was there an appreciable increase in the radioactivity of the body of the fish.

CHAPTER 9

ACCUMULATION AND RETENTION OF RADIOACTIVITY FROM FISSION PRODUCTS AND OTHER RADIOMATERIALS BY FRESH-WATER ORGANISMS[1]

LOUIS A. KRUMHOLZ, *Department of Biology, University of Louisville,* Louisville, Kentucky

and

RICHARD F. FOSTER, *Biology Operation, Hanford Laboratories, General Electric Company,* Richland, Washington

Introduction

RELATIVELY little is known about the mechanisms of uptake, concentration, retention, and excretion of fission products and other radiomaterials by fresh-water organisms. These organisms include many biological forms such as the vascular plants, algae and phytoplankton, protozoans, zooplankton and other invertebrate forms, and representatives of each of the five vertebrate classes.

The complex interrelationships of the fresh-water biota, together with their diverse individual anatomies, physiological processes, and life histories indicate the enormous scope of the problem of determining the role of radiomaterials in the metabolic processes of such a community. In addition, there is extreme urgency for obtaining information on many aspects of this problem within a relatively short period of time. Within the next 10 years several power-producing reactors will undoubtedly be in operation; many placed, in all probability, near the large industrial and/or population centers of the United States where the only ready means of disposal of large quantities of liquid effluent will be into fresh waters. Any near-by rivers and lakes may be subject to rather severe contamination by radioactive materials in the event of accidents.

Owing to the complex interactions of the factors involved, any estimates of the levels of radioactive contamination that may occur in a particular situation may be in error by as much as one or two orders of magnitude. For purposes of hazard control, estimates must therefore be based on pessimistic assumptions with the hope that field sampling and experimentation will reveal a more desirable situation.

An estimate of the worst situation can be obtained by comparing the concentration of a particular element in the water with its concentration in an organism or tissue under study. Since the radioisotope of the element will behave in much the same manner as its stable counterpart (for purposes of this paper), there will be no greater concentration of the radioisotope than of the stable form.

Sources of information

At the present time there are three primary sources of information available regarding the uptake, concentration, retention, and excretion of radiomaterials by fresh-water organisms. They are:

1. The long-term program of the Biology Laboratories of the General Electric Company at Richland, Washington. This program has been primarily concerned with the accumulation of radioactive materials in the flora and fauna of the Columbia River. The effluent water released to the Columbia from the plutonium-producing reactors at the Hanford Operation contains radioelements induced when the "impurities" in the cooling water pass through the high neutron flux. The Hanford program was designed as a radiological-ecological study with four main objectives: (1) to determine the geographical distribution of the radioactive materials, (2) to find out how the radioisotopes became distributed in the various kinds of aquatic organisms from the phytoplankton on through the fishes and, to some extent, to the land animals

[1] Contribution No. 10 (New Series) from the Department of Biology, University of Louisville.

which feed on fresh-water organisms, (3) to study the seasonal distribution of the radioactive materials throughout the biota, and (4) to determine whether the aquatic forms were adversely affected.

2. A three-year study at the Oak Ridge National Laboratory, Oak Ridge, Tennessee. That work was performed by the Fish and Game Branch, Division of Forestry Relations, Tennessee Valley Authority, under contract to the Atomic Energy Commission and consisted primarily in an ecological survey of White Oak Creek and its drainage area. In that study, principal emphasis was placed on the effects on the biota and its environment from radiomaterials that consisted of both fission products and wastes with induced radioactivity from the processing of different materials in the preparation of radioisotopes.

The Ecological Survey of White Oak Creek was divided into three main categories: botany, limnology, and vertebrate biology (Krumholz, 1954). Because of a virtual absence of rooted aquatic plants in the area, the fresh-water biology was largely covered in the studies on limnology and vertebrate biology. That program was designed to find out what radiomaterials had accumulated in the biota of the drainage area, in which organisms and tissues they had accumulated, and what, if any, had been the effects of such levels of accumulation on population balances and on the various types of individual organisms.

3. Many studies of lesser magnitude carried on at other installations of the Atomic Energy Commission and at different colleges and universities throughout the United States. Such studies usually are not integrated with one another but are separate studies designed to answer specific questions.

Rather intensive studies of the phosphorus cycle in fresh-water lakes have been carried out by workers at Dalhousie University (Coffin, et al., 1949, and Hayes, et al., 1952), at Yale University (Hutchinson and Bowen, 1950), and at Atomic Energy of Canada, Ltd. (Rigler, 1956). These studies have increased our knowledge of the role of phosphorus in the economy of fresh-water lakes, particularly at the lower trophic levels. Much work has also been done on the economic aspects of such aquatic insects as the mosquitoes (Bugher and Taylor, 1949; Hassett and Jenkins, 1951) and also on such aquatic forms as the frog (Hansborough and Denny, 1951). These animals have been tagged with radioisotopes (usually radiophosphorus) either by direct feeding of substances which contained the radioactive material, or by immersing them in radioactive solutions.

Concentration of radioactive materials in aquatic organisms

The concentration factor $\frac{(\mu c/g \text{ of organism})}{\mu c/ml \text{ of water}}$ for any radioelement cannot exceed the ratio between the normal concentration of that element in the organism and the concentration of the element in the surrounding water. Thus, if the element in question is not normally used by a particular organism, it is unlikely that any of the radioisotopes of that element will be concentrated in the tissues.

Each organism in each environment has specific requirements for the different chemical elements. However, it is necessary to know the chemical composition of the organism and its parts, as well as that of its aquatic environment, in order to understand those requirements and to interpret the role played by each element in the metabolic processes. At present, there is very little information available on the chemical composition of any of the fresh-water organisms or their tissues, and consequently there is virtually nothing known of the concentration factors to be expected for the different elements by the organisms. Some data on the chemical composition of fresh-water lakes and streams are available, but these waters differ so widely from one another that no generalizations can be made. The total dissolved solids in fresh waters range from less than five parts per million to well over 400 parts per million. In addition, the elements which make up these dissolved solids seldom occur in exactly the same percentage composition in any two bodies of fresh water. The concentration of any particular element in the water is directly dependent upon the chemical characteristics in the drainage area. Because of these differences in the requirements of organisms and in the chemical compositions of the different fresh waters, it is necessary to consider each situation as a separate case.

An indication of the differences in the orders

TABLE 1 Concentration (Ppm Wet Weight) of Some Elements in Selected Organisms and in Some Major Rivers of the United States

Element	Organism [1]			Water [2]	
	Algae (*Spirogyra*)	Insect larvae (Caddis fly)	Fish (Minnows)	Low	High
Silicon	1,500	20	10	3	20
Iron	6,500	300	1	<0.01	6.0
Calcium	1,500	300	3,000	2	200
Phosphorus	250	2,000	6,000	<0.001	1.5
Strontium	2	0.2	0.3		
Sodium	1,500	700	1,000	1	200

These values are only estimates of orders of magnitude. They are recorded here to illustrate differences which can exist and are not intended for use in precision work.

[1] Values are from unpublished results obtained by spectrophotometric analysis at the Hanford Laboratories.
[2] Abstracted largely from Moyle (1956) and Clark (1924).

of magnitude of the concentrations of a few of the common elements in some organisms and in water is shown in Table 1. However, the concentrations of particular elements in specific structures or tissues of those organisms may deviate widely from those values. For instance, the concentration of calcium as calcium carbonate in the shells of some molluscs or that of silicon in the siliceous tests of some diatoms may be greater than the listed values by more than one order of magnitude.

Field studies in the Columbia River at the Hanford Operation and in White Oak Lake at the Oak Ridge National Laboratory have provided an opportunity to study the uptake and accumulation of a variety of radioactive materials by organisms in those waters under natural conditions. Omitting those radionuclides which have half-lives shorter than ten hours, there are measurable amounts of Na^{24}, Cr^{51}, Cu^{64}, P^{32}, As^{76}, and rare earths in effluent from the Hanford reactors. The composition of the wastes from the Oak Ridge National Laboratory varies from day to day but there are relatively large amounts of Sr^{89}, Sr^{90}-Y^{90}, Cs^{137}, Ce^{144}-Pr^{144}, Ru^{106}, and other fission products present at all times. In addition, there are relatively large amounts of other radionuclides such as P^{32} and Co^{60} present on occasion. In spite of this large variety of radionuclides available to the organisms of these two aquatic communities, only a few appear to be utilized to any great extent. Observed concentrations of the radionuclides most frequently used by the organisms through their natural food webs in the Columbia River and White Oak Lake are listed in Table 2. From these data it is evident that some elements are utilized in much greater quantities than others. Rather large variations occur from one collecting site to another and between species, however. For example, the concentration factor for P^{32} in filamentous algae of White Oak Lake is listed as 850,000. This figure is for a sample from a large mat of *Spirogyra* that lie on the bottom near the upper end of the lake. In other parts of the lake *Spirogyra* contained less radiomaterial. Furthermore, radioactivity in other filamentous algae, such as *Oedogonium,* was consistently lower than for *Spirogyra.* Comparable differences in the amounts of radioisotopes accumulated by the different phytoplankton and insect larvae were also found.

Very few data have been published which indicate the importance of the physical and chemical states of the various elements in the

TABLE 2 Estimated Concentration Factors for Various Radionuclides in Aquatic Organisms as Observed From Field Studies on the Columbia River and White Oak Lake

Radionuclide	Site	Phytoplankton	Filamentous algae	Insect larvae	Fish
Na^{24}	Columbia River	500	500	100	100
Cu^{64}	Columbia River	2,000	500	500	50
Rare Earths	Columbia River	1,000	500	200	100
Fe^{59}	Columbia River	200,000	100,000	100,000	10,000
P^{32}	Columbia River	200,000	100,000	100,000	100,000
P^{32}	White Oak Lake	150,000	850,000	100,000	30,000–70,000
Sr^{90}–Y^{90}	White Oak Lake	75,000	500,000	100,000	20,000–30,000

physiological processes of fresh-water organisms. Coffin, et al. (1949) and other workers have shown that a large fraction of the P^{32} which was added to fresh-water lakes under natural conditions was quickly fixed in the bottom sediments where it was essentially unavailable to the organisms. Thus it is apparent that elements which are introduced into an environment as insoluble or tightly fixed compounds, or become parts of such compounds shortly after their introduction, may be of little or no use to the organisms even though the particular element involved normally enters into their metabolic processes.

Another factor in the concentration of radiomaterials by fresh-water organisms about which there is only limited information available is the effect of the presence of one chemical on the uptake of another. For example, it was shown by Prosser and co-workers (1945) that the amount of calcium present in the water affected the amount of strontium taken up by goldfish; as the amount of calcium was increased, the uptake of strontium decreased.

The amount of a radionuclide taken up by an aquatic organism is dependent not only upon the concentration of the nuclide in the water (microcuries per milliliter) but also upon its specific activity.[1] As the specific activity is decreased by increasing the concentration of "carrier" over a certain range, the stable form of the element becomes more readily available to satisfy the requirements of the organism, and the amount of radioisotope taken up by the organism will generally decrease. Such isotopic dilution has a non-linear relationship, however, and may be ineffective in instances where low concentrations occur (Whittaker, 1953; Kornberg, 1956).

[1] Specific activity as used here refers to the ratio between the amount of radioisotope present and the total amount of all other isotopes, both radioactive and stable, of that same element.

Methods of accumulation of radiomaterials by organisms

Radiomaterials may become associated with fresh-water organisms in one of three ways: (1) through adsorption to surface areas, (2) through absorption from the surrounding medium, or (3) through ingestion as food. The first of these methods is primarily a physical process whereas the last two are largely biological in nature and make up an integral part of the physiological processes necessary for the metabolism of the population.

In some instances, especially in those organisms which have a large surface-to-volume ratio, adsorption to surfaces is very important. For example, Foster and Davis (1955), working with organisms from the Columbia River, showed that the amounts of radioactivity in sponges and diatoms remained comparatively high at a season when the amounts of radioactivity in other organisms were quite low.

All of the nutrient materials, and thus the biologically important radioisotopes, that are metabolized by plants are absorbed directly from the environment (Rediske, Cline, and Selders, 1955). Direct absorption of a few radionuclides by fresh-water organisms has been observed under laboratory conditions. Gross estimates of concentration factors which appear to have occurred in these studies are listed in Table 3. For the most part, these are short-term tests in which the particular test organism was immersed in the radioactive solution. Entirely different values would result if the organism had also acquired the isotope through the food web.

The principal mode of accumulation of radiomaterials by fishes is through ingestion. Olson (1952) found that young trout which had been immersed in dilute effluent from the Hanford reactors failed to concentrate radiophosphorous, whereas similar fish, which were fed

TABLE 3 Absorption of Various Elements from Solution By Fresh-water Fish

Element	Organism	Probable concentration factor	Investigator
Strontium	Goldfish	150	Prosser, et al., 1945
Barium-Lanthanum	Goldfish	150	Prosser, et al., 1945
Sodium	Goldfish	30	Prosser, et al., 1945
Calcium	Guppy	1000	Estimated from Rosenthal, 1956

organisms that had been grown in the effluent, accumulated substantial amounts of P^{32}. Fish living in the Columbia River downstream from the reactors and which fed on organisms that had assimilated the radioactive materials contained over 100,000 times more radiophosphorus than the surrounding water during the late summer. Krumholz (1954, 1956) attributed the high concentrations of Sr^{90} and Cs^{137} in the fishes of White Oak Lake to the ingestion of contaminated food organisms. In addition, it was shown that the different kinds of animals which served as food for the fishes accumulated different amounts and kinds of radiomaterials. For instance, although a high percentage of the radioactivity in the food organisms, such as larval *Chaoborus,* emanated from radiophosphorus, only a relatively small portion of the radioactivity in the fish was traceable to that radioelement. Similarly, although only a relatively small amount of radioactivity in the plankton organisms was attributable to Sr^{90}, about 80 per cent of the radioactivity in the fish skeleton emanated from that radioisotope. From these findings it is apparent that the ability of the various organisms in the food web to concentrate the different radionuclides is of the utmost importance to the predatory species. If the animals which serve as food were unable to take up the radiomaterials, there would be considerably less chance of the predators becoming contaminated.

The food habits of fishes and other fresh-water organisms determines, to a great extent, which radioelements they may accumulate. In a study of the food habits of the black crappies and the bluegills of White Oak Lake (Krumholz, 1956) it was found that the diets of those two species were considerably different. Marked differences also occurred in the concentration and relative proportions of the radiomaterials in the tissues of the two kinds of fish. Greater amounts of radiomaterials were concentrated in the soft tissues of the bluegills than in the crappies, and greater amounts of radiomaterials were concentrated in the skeleton and other hard parts of the crappies than in the bluegills. Furthermore, there were relatively greater amounts of radiophosphorus in the bones of the bluegills and relatively greater amounts of radiostrontium in the bones of the crappies. These differences may well have resulted from the dissimilar diets or, perhaps, from different physiological demands. Unpublished data of the Hanford Laboratories shows that 50 to 75 per cent of the radiophosphorus ingested by fish is assimilated and retained. Unfortunately, there is virtually no other information available on the efficiency of transfer of radioisotopes from food organisms to aquatic predators.

Concentration of radioactive materials in different organisms

In unpublished results from the studies at White Oak Lake, it was shown that bacteria may have the greatest powers for concentrating radiomaterials of any of the fresh-water organisms, their concentration factors for certain isotopes may exceed 1,000,000. However, it is not definitely known for all radionuclides whether or not they actually enter into the metabolism of the bacteria or are adsorbed to surface areas. Labaw, Mosley, and Wyckoff (1950) showed that the measured radioactivity in *Escherichia coli,* which had been cultured on a medium that contained P^{32} (as $Na_2HP^*O_4$), was not due to adsorption of the P^{32} on the bacterial surfaces nor to residues from the radioactive culture.

The data from the Columbia River and White Oak Lake indicate that the phytoplankton usually concentrate greater amounts of radiomaterials than the zooplankton. Here, again, it is not known for all species whether the radiomaterials actually enter into the metabolism or are adsorbed to surfaces. Some of the filamentous algae are known to concentrate P^{32} at least 850,000 fold (Krumholz, 1954), whereas for other algae the concentration factor may be as little as 300,000. Some zooplankton have concentration factors for radiophosphorus of as much as 250,000 but in others it may be less than 100,000.

Fresh-water invertebrates of all classes studied in the Columbia River and White Oak Lake exhibited maximum concentration factors which ranged from less than 100 to more than 100,000 depending on the radioelement involved. It is believed that most of the radioactive materials accumulated actually enter into the metabolism of these invertebrates. Some of the insect larvae concentrate radioelements by factors upwards of 100,000; some of the microcrustaceans by factors of nearly 200,000; some mollusks may concentrate fission products as ef-

fectively, if not to a greater extent, than some crustaceans. This may be especially true for those long-lived isotopes which are incorporated into the shell.

The length of exposure to water that contains radioisotopes will also greatly affect the concentration in different organisms. The concentration of isotopes in phytoplankton and other micro-organisms will reach equilibrium with the water in a relatively short period of time. For radiophosphorus, this is estimated at about 15 hours (Whittaker, 1953). The larger animals, such as fish, will approach equilibrium much more slowly, however. Coffin, et al. (1949) found that radiophosphorus introduced into an acid bog lake did not appear in the fish until two days later. Several weeks of chronic exposure to an environment containing long-lived, bone-seeking isotopes is undoubtedly necessary before maximum concentrations will result in large fish.

Variations with season, age, and growth

So far as is known, all cold-blooded fresh-water organisms exhibit seasonal changes in the assimilation of radiomaterials through metabolic processes. There is a direct correlation between an increase in temperature and an increase in the accumulation of radiomaterials through metabolic processes in the invertebrates and fishes of the Columbia River (Foster and Davis, 1955) and the fishes of White Oak Lake (Krumholz, 1954, 1956). However, in White Oak Lake it was found that the amounts of radiomaterials in all fish tissues decreased markedly after August 1, even though the temperatures at that time were similar to those during the early summer when there was a rapid increase in the accumulation of radioactive materials. This may well be a suggestion that some warm-water fishes enter a period of estivation or summer dormancy. A decline in radioactivity of Columbia River organisms during the winter months correlates with cessation of feeding.

No seasonal pattern of change in the accumulation of radiomaterials has been demonstrated for any of the warm-blooded aquatic vertebrates, but this may well occur. It is known, for example, that the I^{131} content of rabbit thyroid glands changes markedly with the season (Hanson and Kornberg, 1955).

Among the fishes, it has been established by Olson and Foster (1952) that the younger, more rapidly growing individuals accumulate relatively greater amounts of radioactivity than the older, more slowly growing ones. This phenomenon is probably a reflection of the more rapid anabolism that accompanies the growth of younger fish. It is not known whether any of the other fresh-water vertebrates or invertebrates exhibit this same phenomenon.

Any accumulation of radioactive materials in an organism is subject to biological dilution. Such dilution results from cell division and growth. It is especially manifest in rapidly growing organisms and is particularly noticeable following an acute short-term exposure to the radiomaterials.

Retention and elimination

Radioisotopes will be deposited and retained in the organisms according to the physiological behavior of the particular element involved. Highly mobile isotopes, such as tritium, may be eliminated in a matter of minutes or hours (Foster, 1955), but certain bone-seekers, such as strontium or phosphorus, may be so tightly fixed that little loss occurs, except by radioactive decay, during the life of the organism. The metabolism of the radiophosphorus in trout has been studied by Hayes and Jodrey (1952) and by Watson (Hanford Laboratories, unpublished). Little information is available on the metabolism of other isotopes in other aquatic animals, however.

The recognized methods of elimination of radiomaterials are: (1) through surface exchange, (2) excretion through the natural physiological channels, (3) through moulting where this occurs, and (4) through death. In any of these processes of elimination, the radiomaterials are released into the environment and can be immediately taken up by other organisms.

Discussion

Based on our present knowledge, there can be no broad statement to the effect that "aquatic organisms will concentrate radioactivity in their tissues." Rather, each individual situation must be appraised separately in the light of the following basic considerations which are concerned with the accumulation of radiomaterials by fresh-water organisms: (1) the particular ele-

ment involved and its physiological importance to the organism, (2) the physical and chemical state of the element and its acceptability to the organism, (3) the concentration of the element in the environment and the presence of other elements which may inhibit or enhance its uptake, (4) the morphology of the organism, its life history, and its particular role in the food web, and (5) the physical and chemical characteristics of the environment.

Even though the great majority of research with radionuclides in biological fields has been performed within the past 15 years, enough data have been gathered to serve as a basis for the following general statements.

1. Radioactive materials are taken into the body of an organism either through physiological processes and incorporated directly into the tissues or they are attached to the surfaces of the organisms through adsorption.
2. The concentration of certain radioelements reaches a higher level in many of the lower plant and animal forms, such as bacteria, protozoa, and phytoplankton, than in higher forms, such as the vertebrates. In such instances, there is an inverse correlation between the complexity of body structure and the concentration of the radioelement in question.
3. Certain plants and animals have a predilection for concentrating specific radionuclides in different tissues. For instance, iodine is concentrated in the thyroid tissue, silicon is concentrated in the tests of some diatoms, calcium is concentrated as calcium carbonate in the shells of some mussels and as calcium phosphate in others, calcium and phosphorus are also concentrated in the bony skeletons of vertebrates, phosphorus in concentrated as adenosine triphosphate in the flight muscles of some birds, and potassium and other elements are concentrated in wide variety of tissues.
4. Although certain radioelements may occur in amounts acceptable for drinking water, many fresh-water organisms have the ability to concentrate them to levels which would be harmful. Such deleterious effects could range from those in which only the individual organism is involved to those in which the entire population may be affected.

Little information is available on the tolerances of the various aquatic organisms to different radioactive materials. Recently, however, D. G. Watson at the Hanford Laboratories has determined that a concentration of 65 μc P^{32} per gram of bone was lethal to trout in about six weeks. A concentration of 10 μc P^{32} per gram was not lethal in 12 weeks but caused some radiation damage. This series of experiments is only the first step toward determining the tolerance levels for all radionuclides in each of the animals of the fresh-water fauna.

The use of radiomaterials as a research tool in fresh-water biology has opened new fields which were almost impossible to explore adequately by other means. Determination of the metabolism of many of the elements essential for proper nutrition is now possible. Furthermore, the effects of the radioactivity emanating from isotopes deposited in the tissues can be studied. In the field of fresh-water biology, perhaps the greatest benefits from the use of radioactive materials can be derived from studies of the physiological processes of the organisms.

REFERENCES

BUGHER, J. C., and MARJORIE TAYLOR. 1949. Radiophosphorus and radiostrontium in mosquitoes. Preliminary report. *Science* **110**:146–147.

CLARK, F. W. 1924. The composition of the river and lake waters of the United States. U. S. Geol. Survey, Prof. Paper 135.

COFFIN, C. C., F. R. HAYES, L. H. JODREY, and S. G. WHITEWAY. 1949. Exchange of materials in a lake as studied by the addition of radioactive phosphorus. *Canad. Jour. Research D.* **27**:207–222.

DAVIS, J. J., R. W. COOPEY, D. G. WATSON, C. C. PALMITER, and C. L. COOPER. 1952. The radioactivity and ecology of aquatic organisms of the Columbia River. *In* Biology Research — Annual Report, 1951. USAEC Document HW-25021:19–29.

FOSTER, R. F., and J. J. DAVIS. 1955. The accumulation of radioactive substances in aquatic forms. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, 13 (P/280):364–367.

FOSTER, R. F. 1955. Tritium oxide absorption and retention in the body water of some aquatic organisms. *In* Biology Research — Annual Report, 1954. USAEC Document HW-35917:98–100.

HANSBOROUGH, L. A., and D. DENNY. 1951. Distribution of phosphorus^{32} in the embryo and larva of the frog. *Proc. Soc. Exptl. Biol. Med.* **78**:437–441.

HANSON, W. C., and H. A. KORNBERG. 1955. Radioactivity in terrestrial animals near an atomic energy site. Proceedings of the International Conference on Peaceful Uses of Atomic Energy, 13 (P/281):385–388.

HASSETT, C. C., and D. W. JENKINS. 1951. The uptake and effect of radiophosphorus in mosquitoes. *Physiol. Zool.* **24**:257–266.

HAYES, F. R., J. A. MCCARTER, M. L. CAMERON, and D. A. LIVINGSTONE. 1952. On the kinetics of phosphorus exchange in lakes. *Jour. Ecol.* **40**:202–216.

HAYES, F. R., and L. H. JODREY. 1952. Utilization of phosphorus in trout as studied by injection of radioactive phosphorus. *Physiol. Zoology* **25**:134–144.

HUTCHINSON, G. E., and V. T. BOWEN. 1950. Limnological studies in Connecticut. IX. A quantitative radiochemical study of the phosphorus cycle in Linsley Pond. *Ecology,* **31**:194–203.

KORNBERG, H. A. 1956. Effectiveness of isotopic dilution. *In* Biology Research — Annual Report, 1955. USAEC Document HW-41500:19–28.

KRUMHOLZ, L. A. 1954. A summary of the findings of the ecological survey of White Oak Creek, Roane County, Tennessee, 1950–1953. USAEC Document ORO-132: 1–54.

1956. Observations on the fish population of a lake contaminated by radioactive wastes. *Bull. Am. Mus. Nat. Hist.* **110** (4):277–368.

LABAW, L. W., V. M. MOSLEY, and R. W. G. WYCKOFF. 1950. Radioactive studies of the phosphorus metabolism of *Escherichia coli. Jour. Bacteriol.* **59**:251–262.

MOYLE, J. B. 1956. Relationships between the chemistry of Minnesota surface waters and wildlife management. *J. Wildl. Mgt.* **20**: 303–320.

OLSON, P. A., JR. 1952. Observations on the transfer of pile effluent radioactivity to trout. *In* Biology Research — Annual Report, 1951. USAEC Document HW-25021:30–40 (OFFICIAL USE ONLY).

OLSON, P. A., JR., and R. F. FOSTER. 1952. Effect of pile effluent water on fish. *In* Biology Research—Annual Report, 1951, USAEC Document HW-25021:41–52.

PROSSER, C. L., W. PERVINSEK, JANE ARNOLD, G. SVIHLA, and P. C. THOMPKINS. 1945. Accumulation and distribution of radioactive strontium, barium-lanthanum, fission mixture and sodium in goldfish. USAEC Document MDDC-496:1–39.

REDISKE, J. H., J. F. CLINE, and A. A. SELDERS. 1955. The absorption of fission products by plants. *In* Biology Research — Annual Report, 1954. USAEC Document HW-35917:40–46.

RIGLER, F. H. 1956. A tracer study of the phosphorus cycle in lake water. *Ecology* **37**: 550–562.

ROSENTHAL, H. L. 1956. Uptake and turnover of calcium-45 by the guppy. *Science* **124**: 571–574.

WHITTAKER, R. H. 1953. Removal of radiophosphorus contaminant from the water in an aquarium community. *In* Biology Research — Annual Report, 1952. USAEC Document HW-28636:14–19.

CHAPTER 10

EFFECTS OF RADIATION ON AQUATIC ORGANISMS

LAUREN R. DONALDSON, *Applied Fisheries Laboratory, University of Washington,* Seattle, Washington

and

RICHARD F. FOSTER, *Biology Operation, Hanford Laboratories, General Electric Company,* Richland, Washington

I. *Somatic Effects of Ionizing Radiation*

THE EFFECTS of ionizing radiations on marine and fresh-water organisms have been studied by a few investigators since the early part of the century. The total volume of such work can by no means compare with that which has resulted from the intensive studies with more conventional laboratory animals. The value of much of the early work is impaired by inadequate or imperfect dosimetry. Nevertheless a sufficient block of information has been accumulated to permit several generalizations and at least some well-defined conclusions.

A. Relative sensitivity of different organisms

A broad review of the results obtained with the organisms of different phyla indicates that the lower or more primitive forms are generally more resistant to ionizing radiation than are the more complex vertebrate forms. Welander (unpublished data) has summarized much of the data for which some approximation of dose can be made. Table 1 is a further condensation of these data which were obtained in experiments where whole body doses (usually X-rays) were administered. Owing to the great variety of circumstances under which the experiments were conducted, these data represent only orders of magnitude of effects.

The algae and protozoa are most resistant with LD_{50} values in the order of many thousands of roentgens. The molluscs and crustaceans are somewhat more sensitive, with LD_{50} values of a few thousand roentgens (aquatic insects probably also fall in this category) and the fish are most sensitive with an LD_{50} of about one thousand roentgens — in the same

TABLE 1 RELATIVE SENSITIVITY OF DIFFERENT GROUPS OF ORGANISMS TO RADIATION

Group	(r) Dose which caused 50% mortality	(r) Dose which caused 100% mortality	"Latent" period	Investigators
Algae	8,000–100,000	25,000– 600,000	45 days	Bonham and Palumbo (1951); Crowther (1926) Bonham, et al. (1947).
Protozoa	10,000–300,000	18,000–1,250,000	45 min.–40 days	Ralston (1939); Back (1939); Back and Halberstaedter (1945); Halberstaedter and Back (1943); Powers and Shefner (1950); Feldman-Muhsam and Halberstaedter (1946).
Molluscs	5,000– 20,000	10,000– 50,000	3 weeks–2 years	Bonham and Palumbo (1951).
Crustaceans ..	500– 90,000	5,000– 80,000	5 days–80 days	Bonham and Palumbo (1951).
Fish	600– 3,000	370– 20,000	14–460 days	Corbella (1930); Welander et al. (1948); Foster et al. (1949); Ellinger (1939) (1940); Ssamokhvalova (1938); Solberg (1938).
Insects [1]				

[1] No data except for *Culex* and non-aquatic forms.

order of magnitude as that of other cold-blooded vertebrates.

B. Relative sensitivity of different stages of development

It must be recognized in any consideration of the relative sensitivity to radiation of different groups of organisms that considerable variability exists between similar species. In comparing the sensitivity of two species of snails, Bonham and Palumbo (1951) found that "at 10 kr, approximately one month elapsed before 50 per cent of the *Radix* died, while in the case of the *Thais* it was approximately one-half of a year." Consideration must also be given to the different developmental stages of the same species. Since many investigators (Evans, 1936, Rugh, 1949) have correlated radiosensitivity with metabolic rate of the dividing cell, it is not surprising that dormant eggs of aquatic invertebrates should be especially resistant. Bonham and Palumbo (1951) found that the two-week LD_{50} for dry *Artemia* eggs was about 50,000 roentgens, but after soaking the eggs for a short time in water, so that embryonic development was resumed, the radiosensitivity increased more than twofold.

A review of the results of exposing salmonoid gametes, eggs, fingerlings or adults to X-radiation supports the early concepts (Butler, 1936) that radiosensitivity decreases with age. Table 2 shows that the LD_{50} values range from 50–100 roentgens for gametes within the parent fish to about 1500 roentgens for adult trout. Welander (1954) studied in detail the effects of X-rays on different embryonic stages of rainbow trout. His results with these aquatic forms confirmed the work of Russell and Russell (1954) and others working with mammals that certain critical periods exist during which the embryo is most sensitive to radiation. Table 3 shows some of Welander's data.

Trout eggs were much more sensitive to radiation during the one-cell stage than during later stages of development. Unpublished work by Welander has shown the most radiation-sensitive period of silver salmon (*Oncorhynchus kisutch*) egg development to be a particular stage during the mitosis of the single cell. For the most sensitive period an LD_{50} of only about 16 roentgens was observed.

The change in sensitivity between different stages of development has also been shown with snails. Bonham and Palumbo (1951) showed that eggs of the fresh-water snail *Radix japonica* were more sensitive to radiation than the adults. Further studies of snails (*Helisoma subcre-natum*) eggs by Bonham (1955) showed that, in the one- and two-cell stages, resting eggs withstood from two to four times as much radiation as cells undergoing mitosis, and that later embryonic stages were less sensitive.

TABLE 2 Relative Sensitivity of Different Life Stages of Salmonoids

Stage irradiated	Species	Approximate median effective dose (LD_{50})	Investigator
Gametes [1]	rainbow trout	50– 100 r	Foster, et al. (1949)
Eyed eggs	chinook salmon	1000 r	Welander, et al. (1948)
Fingerlings	chinook salmon	1250–2500 r	Bonham, et al. (1948)
Adult	rainbow trout	1500 r	Welander, et al. (1949)

[1] In parent fish.

C. Pathology of radiation damage

1. Different organs

While effects of exposure to larger amounts of radiation than that sufficient to cause death of the organism have been studied by many investigators, few have studied in detail the physical and pathological syndromes of damaging but non-lethal exposures to radiation.

Retardation in the rate of growth of snails exposed to radiation has been reported by Bon-

TABLE 3 Relative Sensitivity of Different Embryonic Stages of Trout to X-Irradiation [1]

Stage irradiated	Median effective does (LD_{50}) At hatching	At end of yolk stage
One-cell	78.3 r ± 4.4 [2]	57.8 r ± 3.8 [2]
Thirty-two cell	468 r ± 19.4	313 r ± 12.4
Early germ ring	524 r ± 22.1	461 r ± 15.9
Late germ ring	735 r ± 24.7	454 r ± 19.4
Early eye	—	415 r ± 22.0
Late eye	—	904 r ± 38.5

[1] After Welander (1954).
[2] $S_{\bar{x}}$.

ham and Palumbo (1951). Growth in length and weight of fish exposed to radiation is retarded as compared to control populations (Welander et al., 1949).

"The growth increment during the fastest growing period of the experiment was significantly less in a fish irradiated with 750 r or more of X-radiation and proved to be a very sensitive measurement of radiation damage and directly proportional to the amount of radiation given."

The effects of X-radiation upon growth are not confined to the exposed population. Foster et al. (1949), reporting on the growth of rainbow trout fingerlings produced from parent stock exposed to radiation prior to spawning, comment:

"The rate of growth of the young during their first year of life was also found to be directly affected by the amount of irradiation received by the parent fish. While variations in mortality became less with increasing age of the fish, variations in size became greater. Parents treated with 100 r produced progeny in which growth was slightly impeded, while parents treated with 500 or more r units produced progeny which grew appreciably more slowly than normal."

Damage to specific organs and tissues of salmonoid fish as a result of exposure to X-radiation has been studied by the staff of the Applied Fisheries Laboratory, University of Washington.

Adult rainbow trout exposed to X-radiation prior to spawning were examined for gross radiation damage (Welander et al., 1949). The typical syndromes of radiation such as mass hemorrhage, petechiae and ecchymosis have been observed in all trout subjected to 1500 and 2500 roentgens. Gonadal hemorrhage was observed in fish exposed to 500 r of total body radiation. Exposures of 750 r resulted in hemorrhagic areas in the peritoneum, while all exposures of 1000 r or more produced muscular hemorrhage.

The eggs of rainbow trout exposed to radiation during early developmental stages (Welander, 1954) produced fish showing retarded development. The eggs exposed during the 32-cell, late germ ring and early eyed stages tended to have a more juvenile appearance than the controls, viz., a larger eye and head in proportion to the size of the body. Other modifications evidenced in the young produced from radiated eggs were as follows:

"The number of parr marks was significantly reduced in all stages after doses of 300 r or more, with doses as low as 25 r significantly altering the number in embryos irradiated during the 32-cell stage.

"Reduction in number of dorsal and anal fin rays was general after irradiation of 32-cell, late germ ring and early eyed embryos. Doses from 75 to 100 r were significantly effective in reducing the fin ray number in these stages.

"Gross superficial abnormalities observed in X-rayed trout were similar, though usually more numerous, to those found in the controls, with the exceptions of anomalies of dorsal and adipose fin produced by 200 and 400 r X-rays of 32-cell embryos."

The eggs of chinook salmon (*Oncorhynchus tshawytscha*) exposed to X-radiation during the eyed stage with the results reported by Welander et al. (1948), show somatic damage proportional to the amount of exposure.

Histopathological studies on serial sections of the kidneys, with included hemopoietic tissue, the interrenal bodies, the spleen, the gonads and other organs of chinook salmon embryos and larvae revealed first the gonads, then the hemopoietic tissue as most radiosensitive.

Exposure of the eyed eggs to 250 r greatly reduced the number of primordial germ cells in the gonads of the chinook salmon. This sharp reduction (Table 4) in number of cells at 250 r would indicate a measurable reduction at a much lower radiation exposure.

The hemopoietic tissue of the anterior portion of the kidney of the chinook salmon produced from eggs exposed to 250, 500 and 1000 r showed a reduction in number of cells and a temporary retardation in development, roughly proportional to the dose. Temporary cessation of mitosis at 1000 r and permanent cessation at higher radiations was noted.

Counts of the glomeruli in the kidneys of young fish indicated a slight reduction in numbers at 500 r with definite damage at exposures of 1000 r (Table 5).

In general, it is observed that the tissues most sensitive to radiation damage are those in rapid division and growth. Gonadal and hemopoietic tissues that are in rapid division are many times more sensitive than those growing less rapidly. The very early embryonic stages of an organism

TABLE 4 COUNTS OF THE PRIMORDIAL GERM CELLS IN THE GONADS OF CHINOOK SALMON COMPARED BY DAYS AFTER EXPOSURE AND BY DOSAGE [1]

Days after irradiation	Controls	Exposed fish		
	0 r	250 r	500 r	1000 r
9	33	42	25	27
16	46	31	64	27
23	32	35	108	39
30	14	50	46	7
37	21	47	28	42
44	32	124	79	71
51	453	83	65	53
58	2,085	287	55	25
65	1,058	683	286	47
79	6,569	380	131	67
93	7,206	247	94	69
Average	1,595.4	182.6	89.2	43.1

[1] Counts of over 1,000 r were arrived at by first making total counts on all five-micron sections and calculating the actual number of germ cells present using the average size of the cell nucleus (9.2 microns) and actual cell counts of the other fish as a basis. In cases where every fifth section only was used (as in the 65th, 79th and 93rd day series) actual counts were obtained by multiplying original counts by five and then correcting for size of the cell nucleus. Data from Welander et al. (1948).

are more radiation sensitive than the older, mature forms.

In all respects, the damage effects of radiation in fish are similar or identical to the effects seen in other vertebrate animals. In general, the syndromes have a similar pattern throughout the animal kingdom depending on the dose.

2. Relative susceptibility of organs to radioactive material

For most experiments with aquatic organisms conducted to date radiation from external sources has been used. In the work of Chipman (1955) and Hiatt, Boroughs, Townsley, and Kau (1955) radiation from isotopic sources in the body was used, but at such low levels somatic damage was not evident.

The uptake of lethal levels of P^{32} is being studied by Watson (unpublished data). Adult rainbow trout chronically fed P^{32} died in approximately 6 weeks when the concentration of the isotope in bone, the tissue of maximum uptake, reached a level of 18 to 65 μc/g (giving a dose of about 1200 rads per day). Other trout remained alive during the 12 weeks' experiment with concentrations of P^{32} in the bone as high as 10 μc/g. Although these fish showed no external evidence of radiation damage other than a slight reduction in growth rate, subsequent dissection revealed that some damage had occurred. The syndrome was similar to that described for trout damaged by X-irradiation, especially the breakdown of the vascular system as evidenced by hemorrhage of the liver and musculature.

In experiments that have taken place at Eniwetok and Bikini Atolls the resulting radioactive materials that entered the water usually provided three types of exposure to the aquatic organisms: (1) some of the radiation came from contamination of the environment, (2) particulate matter, such as specks of radioactive debris often settled on organisms or adhered to mucus coverings, etc., or (3) the radioactive materials entered the organism through the food chain where it was absorbed and incorporated into the organism or eliminated by the usual biological processes.

Although vast amounts of radiation may be present immediately following a weapons test, amounts that surely would produce measurable changes in the exposed aquatic forms, no specific instances were found in which direct somatic damage could be charged to radiation effects.

It must be realized that in as complex an environment as a coral atoll following the fate of individual populations is very difficult. The most sensitive forms, the fishes, undoubtedly are weakened from somatic and functional damage by radiation. Such weakened forms are usually eaten soon by the large carnivorous fishes that

TABLE 5 COUNTS OF THE GLOMERULI IN THE KIDNEY OF CHINOOK SALMON LARVAE AFTER IRRADIATION IN EYED EGG STAGES [1]

Dose in r	Days									Average
	23	30	37	44	51	58	65	79	93	
0	12	36	42	60	66	98	122	174	282	99.1
250	2	38	16	41	48	86	144	186	288	94.3
500	8	16	38	40	51	86	96	162	260	84.1
1000	0	26	28	25	46	2	70	64	120	42.3

[1] After Welander et al. (1948), counts on 36 fish.

move into an affected area or, if not picked up at once following death, they decay so rapidly in the warm tropical waters as to be undetectable in a few hours, thus escaping notice.

II. *Somatogenic Effects of Ionizing Radiation*

If we consider genetic effects in the strict sense of damage to chromosomes or genes, to the extent the modified characteristics are passed from one generation to the next, there is little to be found in the published literature describing work on marine or fresh-water forms. Some effects on aquatic forms have been described, however, where gametes were exposed to radiation prior to zygote formation. The effects in such cases are due, at least in part, to somatic damage but will be considered here because they represent some changes which may occur in offspring of irradiated parents.

The classical work of Henshaw and his colleagues with the eggs and sperm of the sea urchin *Arbacia* demonstrated that X-radiation of the gametes delayed the first cleavage. Effects of X-rays on gametes of fish have received some attention. In spite of massive doses of X-rays — 100,000 to 200,000 r — (Rugh and Clugston, 1955) to the eggs and sperm of *Fundulus heteroclitus,* fertilization can take place and some embryonic development is possible although this may be parthenogenic from irradiated sperm. Solberg's (1936) work with *Oryzias* indicates that spermatozoa are three to four times as sensitive to radiation as ova, however. Foster (1949) found that

"The mean mortalities of the eggs obtained from parents subjected to 500 or more roentgen units were significantly greater than that of the eggs from the control parents. Most of the eggs which died during the incubation period contained conspicuously abnormal embryos. The abnormalities could be attributed to deficiencies, improper differentiation of cell masses, disproportionate growth, or combinations of these factors. Abnormal types of embryos occurred among the progeny of control parents and of parents which had received low doses of radiation which were almost identical with the types which occurred among the progeny of parents receiving large amounts of radiation. However, as the amount of radiation increased the relative abundance of malformed embryos increased and the degree of development attained decreased. Practically all of the embryos from parents treated with 1500 r and 2500 r were so abnormal that they died before closure of the blastopore. Irradiation of the parent fish thus increased the frequency of occurrence of malformations."

TABLE 6 Effect on Trout Eggs from Irradiating the Parent Fish [1]

(Values are per cent of eggs which died at each stage)

Stage of Development	Number of r units received by parents							
	0	50	100	500	750	1000	1500	2500
No embryo	18.5	32.2	23.0	24.4	42.6	41.5	68.1	83.6
Blastoderm	0.8	0.3	3.2	5.9	0.6	3.2	2.9	4.5
Embryonic axis	4.3	4.6	4.3	8.2	21.1	29.5	22.1	11.5
Blastopore closed	1.0	4.4	3.7	23.6	22.5	15.5	5.5	0.3
Eyed	6.3	9.1	16.2	8.6	3.5	2.1	0.3	0.1
Hatching	8.7	11.4	10.1	14.5	6.3	6.3	0.9	0
Total	39.6	62.0	60.5	85.2	96.6	98.1	99.8	100

[1] Data from means (unpublished) of figure 1 from Foster et al. (1949).

Table 6 illustrates that egg mortality was directly related to the dose received by the parent fish and that the degree of development obtained by the embryo decreased at the higher exposure levels.

Irradiation of gametes prior to "fertilization" is, of course, not the only means of producing abnormal embryos with ionizing radiation. Welander (1954) found that abnormalities increased with dose among trout embryos irradiated at the 32-cell and early eyed stages. The production of phenocopies has been tentatively established. Welander, as stated earlier, found that trout irradiated with 200 and 400 r at the 32-cell stage had abnormal dorsal and adipose fins. Such anomalies arising from irradiation of cleavage stages would appear to result from a disturbance of the precursors.

III. *Other Considerations in Atomic Energy Use*

When potential effects of atomic energy installations upon aquatic life are considered, radiation damage resulting from the release of radioactive isotopes is probably the primary consideration. Conventional types of pollutants must not be overlooked, however. Indeed, the chemical toxicity or high temperature of effluent released into a stream or lagoon could well be of greater concern than the radioactive materials. Olson and Foster (1955) have reported that very high concentrations of effluent from the Hanford reactors are toxic to young salmon and trout, not because of the radioactive isotopes present, but because of the presence of dichromate. Krumholz (1954) states that:

"The waste effluent which enters White Oak Creek consists of a heterogeneous mixture of chemical wastes resulting from laboratory, pilot-plant, and full-scale operations. Some of these wastes are radioactive and some are not."

Since a variety of toxic substances is apt to be present in effluent from atomic energy plants, just as from other types of industry, care should be taken in appraising biological observations. If adverse effects on aquatic populations are observed, one should not immediately conclude that these are a result of radiation damage when, in fact, they may well result from altered chemical or temperature conditions.

Serious radiation damage to aquatic populations is certainly possible, however, under catastrophic or emergency conditions. It could also occur where there is continued release of inordinate amounts of isotopes which are concentrated in the organisms. Such damage appears unlikely, however, in situations where adequate radiation hazard control is extended to the environs of an atomic energy facility. Such control must go well beyond the sole consideration of maximum permissible concentrations for drinking water. Foster (1955) has pointed out that:

"If radiophosphorus were allowed to reach the maximum level permitted for drinking water, organisms living in the water would suffer radiation damage and the fish would be unsafe for human food."

If contamination in the fish and in other edible forms is to remain at a level which is safe for human beings, however, the radiation dose received by the organisms may not be intolerable to the organisms themselves. For example, the International Committee on Radiation Protection recommends maximum permissible concentrations (MPC) for P^{32} in drinking water of 2×10^{-4} μc P^{32} per cc, equivalent to an intake of about 3 μc P^{32} each week. If MPC's were based on a nominal consumption of one pound of fish per person each week, and an additional safety factor of 10 were applied owing to the large populations involved, then the MPC for edible parts (flesh) of the fish would be 7×10^{-4} μc P^{32} per gram. This is only about one per cent of the concentration which Watson (unpublished data) found to be sub-lethal to trout in a 12-week period (although some radiation damage did occur). It seems unlikely, therefore, that significant damage would result to fish if the concentration of P^{32} in the flesh remained below 10^{-3} μc/g.

REFERENCES

BACK, A. 1939. Sur un type de lesions produites chez *Paramaecium caudatum* par les rayons X. *Compt. Rend. Soc. Biol.* **131** (22):1103–1106.

BACK, A., and L. HALBERSTAEDTER. 1945. Influence of biological factors on the form of roentgen-ray survival curves. Experiments on *Paramecium caudatum*. *Am. J. Roentgenol. Radium Therapy* **54**:290–295.

BONHAM, KELSHAW. 1955. Sensitivity to X-rays of the early cleavage stages of the snail *Helisoma subcrenatum*. *Growth* **XIX**:9–18.

BONHAM, KELSHAW, and RALPH F. PALUMBO. 1951. Effects of X-rays on snails, crustacea, and algae. *Growth* **XV**:155–188.

BONHAM, KELSHAW, ALLYN H. SEYMOUR, LAUREN R. DONALDSON, and ARTHUR D. WELANDER. 1947. Lethal effect of X-rays on marine microplanton organisms. *Science* vol. 106, no. 2750.

BUTLER, E. G. 1936. The effects of radium and X-rays on embryonic development. *In*, Biological Effects of Radiation, ed. B. M. Duggar, McGraw-Hill Book Co., Inc., N. Y., pp. 389–410.

CHIPMAN, W. A. 1956. Passage of fission products through the skin of tuna. Fish

and Wildlife Service Special Scientific Report — Fisheries no. 167.

CORBELLA, E. 1930. Influsso delle radiazioni Roentgen sullo svilluppo embrionale dei Teleostei (*Salmo lacustris* L., *Salmo irideus* Gibb., *Perca fluviatilis* L.) *Riv. Biol. Milano* **12**:93–117.

CROWTHER, J. A. 1926. The action of X-rays on *Colpidium colpoda*. *Proc. Roy. Soc.* London, Ser. B, 100, 390–404.

ELLINGER, F. 1939. Note of action of X-rays on goldfish (*Carassius auratus*). *Proc. Soc. Exp. Biol.* **41**:327–9, 2 figs.

EVANS, T. C. 1936. Qualitative and quantitative changes in radiosensitivity of grasshopper eggs during early development. *Physiol. Zool.* **9**:443–454.

FELDMAN-MUHSAM, B., and L. HALBERSTAEDTER. 1946. The effect of X-rays on *Leischmania tropica* in vitro. *Brit. J. Rad.* **19** (217):41–3.

FOSTER, RICHARD F., LAUREN R. DONALDSON, ARTHUR D. WELANDER, KELSHAW BONHAM, and ALLYN H. SEYMOUR. 1949. The effect on embryos and young of rainbow trout from exposing the parent fish to X-rays. *Growth* **XIII**:119–142.

FOSTER, RICHARD F., and J. J. DAVIS. 1955. The accumulation of radioactive substances in aquatic forms. Proceedings of the International Conference on Peaceful Uses of Atomic Energy, 13 (P/280):364–367.

HALBERSTAEDTER, L., and A. BACK. 1943. Influence of colchicine alone and combined with X-rays on Paramecium. *Nature* (London) **152** (3853):275–276.

HENSHAW, P. S., and D. S. FRANCIS. 1936. The effect of X-rays on cleavage in *Arbacia* eggs: evidence of nuclear control of division rate. *Biol. Bull.* **70**:28–35.

HIATT, ROBERT W., HOWARD BOROUGHS, SIDNEY J. TOWNSLEY, and GERALDINE KAU. 1955. Radioisotope uptake in marine organisms with special reference to the passage of such isotopes as are liberated from atomic weapons through food chains leading to organisms utilized as food by man. Ann. Rept., Hawaii Mar. Lab., U. of Hawaii.

KRUMHOLZ, LOUIS A. 1954. A summary of findings of the ecological survey of White Oak Creek, Roane County, Tennessee, 1950–1953. USAEC Doc. ORO-132.

OLSON, P. A., and R. F. FOSTER. 1955. Reactor effluent monitoring with young chinook salmon — 1954. *In* Biology Research, Ann. Rept. (1954), Biol. Sec., Radiol. Sci. Dept., General Electric, Hanford Atomic Products Operation (USAEC Doc. HW-35917):11–18.

POWERS, E. L., and D. SHEFNER. 1950. Effects of high dosages of X-rays in *Paramecium aurelia*. *Genetics* **35**:131.

PROSSER, C. L., C. W. HAGEN, JR., and W. GRUNDHAUSER. 1948. The lethal action of X-radiation, stable isotopes of fission elements Sr^{89} and $(Ba\text{-}La)^{140}$ upon goldfish. (USAEC Doc. ANL-4017).

RUGH, R. 1949. Some prenatal effects of *Amblystoma opacum* larvae exposed to 25,000 r X-radiation. *Anat. Rec.* **103**:500–501.

RUGH and CLUGSTON. 1955. Effects of various levels of X-irradiation on the gametes and early embryos of *Fundulus heteroclitus*. *Biol. Bull.* **108** (3):318–25.

RUSSELL, L. B., and W. L. RUSSELL. 1954. An analysis of the changing radiation response of the developing mouse embryo. *Jour. Cell. Comp. Physiol.* **43**: (Suppl. 1), 103–149.

SOLBERG, A. N. 1938. The susceptibilty of *Fundulus heteroclitus* embryos to X-radiation. *Jour. Exp. Zool.* **78**:441–469.

SSAMOKHVOLOVA, G. W. 1938. Effect of X-rays on fishes (*Lebistes reticulatus, xiphophorus hellerii* and *Carassius vulgaris*) *Biol. Zh.* Moscow **7**:1023–1034.

WATSON, D. G. 1956. Effects of feeding chronic levels of P^{32} to rainbow trout (unpublished data).

WELANDER, ARTHUR D., LAUREN R. DONALDSON, RICHARD F. FOSTER, KELSHAW BONHAM, and ALLYN H. SEYMOUR. 1948. The effects of roentgen rays on the embryos and larvae of the chinook salmon. *Growth* **XII**: (3), 203–242.

WELANDER, ARTHUR D., LAUREN R. DONALDSON, RICHARD F. FOSTER, KELSHAW BONHAM, ALLYN H. SEYMOUR, and FRANK G. LOWMAN. 1949. The effects of roentgen rays on adult rainbow trout. (USAEC Doc. UWFL-17).

WELANDER, ARTHUR D. 1954. Some effects of X-irradiation of different embryonic stages of the trout (*Salmo gairdnerii*). *Growth* **XVIII**:227–255.

CHAPTER 11

ISOTOPIC TRACER TECHNIQUES FOR MEASUREMENT OF PHYSICAL PROCESSES IN THE SEA AND THE ATMOSPHERE [1]

HARMON CRAIG, *Scripps Institution of Oceanography, University of California,* La Jolla, California

I. *Introduction*

THROUGHOUT this report reference has been made to the need for a fundamental understanding, on a long-term basis, of mixing phenomena in the ocean and the atmosphere. In a general sense the ocean and the atmosphere may be regarded as a two-phase system, in which the phases are separated by the fundamental discontinuity of the ocean-atmosphere interface. Each phase is further divided into two parts by a second order discontinuity; the atmosphere, divided by the tropopause at about 12 km, into the troposphere and the stratosphere, etc., and the oceans, divided by the thermocline at some 100 meters, into an upper and lower layer.

The basic problems in determining the effects of both radioactive waste disposal and the dispersal of debris from nuclear explosions may be formulated in terms of a single objective. Given the ocean-atmosphere system under normal steady state conditions, and given some substance introduced at any point in one of the four designated zones, we wish to be able to predict quantitatively the concentration of the substance as a function of latitude, longitude, altitude or depth, and time. These problems thus involve studies of (1) the intra-phase mixing, above, below, and across the second-order discontinuities within each phase, and (2) the inter-phase mixing across the ocean-atmosphere interface, with the aim of predicting the effects of perturbations on the system.

The dominant mixing processes in the various spheres are processes of mass movement or turbulent mixing. In such processes, for elements undergoing no change of phase, there is little or no separation of components, and thus, in general, isotopic tracer techniques may involve a wide range of materials of quite different chemistry. It is this phenomenon of mass movement dominance, and the relative unimportance of diffusive transfer except in special cases, that makes the tracer technique so powerful; a tagged isotope for each element is not required, and one can choose for each particular study the elements most useful for tracing the movement of a mass of heterogeneous material.

As mentioned at several points in this report, both artificial and natural isotopic tracers may be used for the study of transfer phenomena in the sea and the atmosphere. Artificial tracers are of value in such studies because they allow the investigator to introduce perturbations in the system at convenient times and places; they are especially valuable for the study of short term fluctuations in local systems. However, for the general understanding of mass transfer phenomena, artificial tracers are of value mainly as experimental checks on deductions based on other data, with the exceptions of a few special cases to be described below. The reason for this is that mass transfer phenomena are by nature subject to long term periodic fluctuations such as convection, with periods often longer than the time range available for observation. A second reason for this is the high cost of radioisotopes and the large amounts of activity required to tag adequately the large masses of water necessary for ocean studies.

Revelle, Folsom, Goldberg and Isaacs (1955) have discussed in their Geneva report the problems involved in adapting radioisotope tracer techniques to transfer studies in the ocean, and the requirements for usable isotopes. If one introduces some 10 curies of a gamma emitter in solution at some point below the thermocline, it is found that within a reasonable duration of observation time the activity will be concentrated in a layer of the order of 1 meter thick

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 902.

spread over a horizontal area of radius r. They find that with the best of present instruments, the horizontal spread in which the concentration of the introduced radioactivity can be determined corresponds to $r = 1$ km. With the possibility of improved instrumentation, and the use of specially selected nuclides, it may be possible to raise the area of detection and determination of activity concentration to about 100 km^2, an area which is still negligible with respect to oceanic expanses.

The radioisotopes suitable for such measurements must of course have a half-life compatible with the mixing rates to be studied and yet short enough so as not to constitute a permanent hazard, namely of the order of a week to a month. Moreover, they must be available in multi-curie amounts at reasonable cost, should form soluble ionic species in sea-water, have a high specific activity, and, for instrumental reasons, should be gamma emitters with energy between .2 and 1.3 Mev. Revelle, et al., were able to list three such isotopes, which, together with half-life, cost, and other data, are listed in the following table:

Isotope	Half-life	Cost per curie	Specific activity available	Gamma energy Mev
Rb^{86} ...	19.5 day	$1000	9 mc/gram	1.1
I^{131} ...	8.0 day	750	Carrier-free	0.36, 0.72
Ba^{140} ...	12.8 day	500	Carrier-free	0.16, 0.54

Comparison of the cost of these isotopes with the maximum area of detection cited above shows that the study of large-scale transfer phenomena in the oceans, using deliberately introduced artificial radioactivity in the form of specific isotopes, is so costly as to be infeasible with the estimated best instrumentation which will be available in the near future. It is evident that such isotopes are at best adapted only to short-term, small scale studies of local phenomena. The use of mixed fission products on a large scale, discussed elsewhere in this report, is somewhat more feasible but is beset with many difficult problems of transportation and handling.

From these considerations it seems evident that the critical data in studies of atmospheric and oceanic mixing and interaction will come from the use of the naturally occurring isotopic tracers, which reflect in their material balance adjustments the differential rates of transfer from source, through reservoir, to sink, and loss by decay. It is from these transfer rates, adjusted to the steady state geochemical and geophysical cycles of the various elements, that we can hope to gain an understanding of the long period variations in natural transfer phenomena. The importance of gaining a clear understanding of the long period transfer rates, when problems such as storage of potentially hazardous radioactive wastes and cumulative effects of nuclear detonations are considered, cannot be overemphasized.

In the following sections we discuss the present status of our knowledge of the distribution and properties of the various naturally occurring isotopes which are useful for studies of atmospheric and oceanic transfer phenomena. In addition, mention is made of the nuclides produced in nuclear detonations and supplied by reactors which have properties such that they are also useful in such studies and which have been studied to some extent.

II. *Distribution of naturally occurring isotopes of elements adapted for transfer studies*

In this section we discuss the production and occurrence of radioactive and stable isotopes showing measurable isotopic variations, and the distribution factors which determine their relative concentrations in natural materials.

Carbon 14

Carbon 14 is formed in the atmosphere by the reaction of neutrons with nitrogen, i. e.

$$N^{14} + n = C^{14} + p + 620 \text{ kev};$$

the neutrons being the result of the interaction of primary cosmic rays with the atmosphere (Libby, 1955). The carbon 14 is naturally radioactive, decaying by β-emission back to nitrogen 14 with a half-life of 5570 years. Thus the half-life is so short that radiocarbon depends, for its existence, on the continual production in the stratosphere, with which it is presumably in steady state. The assumption of a steady state condition for at least the last 15,000 years is justified by the observation that radiocarbon dates on historic samples agree with the calendar dates. The steady state production rate, which is equal to the steady state disintegration rate, can be calculated from measurements on the neutron flux in the lower stratosphere and compared with the observed specific activity of

carbon. Anderson (1953) who has made the most recent and detailed considerations of the production rate, finds a rate of 2.6 carbon 14 atoms per cm^2 and per sec.

The carbon 14 atoms are oxidized to CO_2 and thus enter the normal geochemical and biological cycles of carbon via the atmosphere. The distribution through the atmosphere and the terrestrial plants is rapid, and the steady state radiocarbon concentration in these reservoirs is taken as the basis for the so-called "modern" specific activity of carbon, namely about 15 disintegrations per minute per gram of carbon.

On the other hand, the transfer of carbon from the atmosphere to the sea is slow enough, compared to the half-life, to produce a significant difference between the predicted and observed activity of carbon in the surface layers of the oceans. Carbon, as one of the lighter elements, is subjected to natural fractionation of its isotopes in the various reactions it undergoes in its biogeochemical cycle (cf. section on stable isotope variations, below). The steady state isotopic separation of the stable isotopes C^{12} and C^{13} produces a C^{13} concentration in surface ocean water bicarbonate and shell carbonate which is about 2.5 per cent higher than the C^{13} concentration in terrestrial plants. It is thus known that the C^{14} concentration in ocean bicarbonate and carbonate should be about 5 per cent higher than the concentration in land plants, namely about 15.75 disintegrations per minute per gram of carbon (Craig, 1954). In actual fact, however, measurements show that the specific activity of bicarbonate and carbonate from the ocean is about the same as the specific activity of land plants. Thus the atmospheric C^{14} activity has been increased 5 per cent by slow exchange of CO_2 between atmosphere and sea, resulting in an "apparent age," relative to wood standards, of 400 years for the bicarbonate and carbonate shells in the surface layers of the ocean (Craig, 1954, 1957 (a)). Some 10 measurements have now been made on marine plants, animals, and sea-water from the Atlantic (Suess, 1954) and from the New Zealand area (Rafter, 1955) which indicate that the radiocarbon "age" of surface marine carbonate is about 400 years; it is thus clear that radiocarbon age determinations made on deep ocean waters must all be referred to this baseline, rather than to the modern specific activity displayed by the terrestrial plants and the atmosphere.

The evaluation of the exchange time of CO_2 between atmosphere and sea from data on the natural distribution of C^{14}, is discussed in Section IV of this paper.

The most recent and accurate measurement of the absolute radiocarbon concentration is that of Suess (1955), based on comparisons with an absolute standard obtained from the National Bureau of Standards. Suess finds a concentration of 1.238×10^{-12} atoms of C^{14} per atom of carbon for average 19th century wood, corrected for decay to the present date and corrected for isotopic fractionation. Based on this value and a half-life of 5568 years, we give below the amounts of C^{14} in metric tons, and the activities in megacuries present in the major reservoirs on the earth (Craig, 1957 (a), calculated from his Table 1). The figures for the atmosphere and terrestrial living matter are normalized for isotopic fractionation, while the organic and inorganic carbon in the ocean was assumed to have an average age of 600 years relative to corrected 19th century wood, or 200 years relative to surface ocean bicarbonate (see Section IV, this paper).

Reservoir	Total C^{14} metric tons	Total activity megacuries
Atmospheric CO_2	0.96	4.4
Terrestrial living matter + humus	2.2	11.0
Ocean: Total organic matter.	3.8	17.6
Ocean: Total inorganic carbon	49.8	228.6
Totals	56.8	261.6

The total activity of radiocarbon present on the earth thus corresponds to some 260 megacuries, practically all of which is in the ocean. Using Anderson's figure for the production rate, cited above, and the decay constant of 3.945×10^{-12} sec^{-1}, the calculated total inventory of radiocarbon on the earth is 78.4 tons, which differs from the figure of 56.8 metric tons, obtained in Table II, by about 28 per cent. However, the production rate, as estimated from cosmic ray data and the counting of atmospheric neutrons, is uncertain to at least 20 per cent because of the uncertainty in the reactor flux from which the neutron counters are calibrated. More recent estimates of the production rate are lower than the figure cited above and all that can be said about the agreement between the calculated

and predicted radiocarbon inventories is that they agree within present limits of error.

Tritium

Tritium (H^3) is made in the upper atmosphere, primarily in the "stars" or nuclear explosions produced by the collisions of primary cosmic ray particles with the atmospheric molecules; it is naturally radioactive, decaying by β^- emission to helium 3 with a half-life of about 12.5 years (Kaufman and Libby, 1954). The T atoms "burn" very quickly to HTO and enter the precipitation — evaporation cycle of water. A very small amount of tritium is produced in rocks by the nuclear reaction of lithium with neutrons produced by spontaneous fission of uranium and from (α,n) reactions (Morrison and Pine, 1955); the production of tritium by this process is insignificant relative to the atmospheric production.

Detailed studies of the distribution of tritium in natural waters have been made by Libby and his co-workers at Chicago. The natural concentration of tritium (before thermonuclear tests) in continental waters averages about 5×10^{-18} atoms of tritium per atom of hydrogen. (Following Libby's usage, such a concentration will hereafter be referred to as 5 tritium units, abbreviated as T.U.) The concentration in oceanic rains is about 1 T.U., while in the surface waters of the ocean itself the concentration appears to be as low as 0.2 T.U. The sea is, of course, the ultimate resting place of the tritium formed in the atmosphere, and the low concentration in the oceanic rains relative to continental rains is principally due to tritium removal by direct molecular exchange with the sea surface (see below).

Kaufman and Libby (1954) calculated the tritium production rate in the atmosphere by equating it with the rate at which tritium disappears from the atmosphere into the ocean, taken as the sum of the tritium entering the ocean by run-off from continental rains and the tritium entering directly via oceanic rains. For this calculation only the average run-off and ocean precipitation figures, and measured average tritium content of such waters, are needed. They obtained a net production rate, averaged over the earth's surface, of .12 T atoms per cm^2 per second. Von Buttlar and Libby (1955) measured many more rain samples, and also analyzed 5 samples of ocean water, from which they could estimate the tritium content of the water vapor which evaporates from the sea surface. Using this latter figure they calculated the production rate over the oceans, assuming that tritium is lost from the atmosphere only by oceanic rain, and gained by production and oceanic evaporation, and obtained a figure of 0.11 to 0.12 T atoms per cm^2 per sec. A similar calculation was made for the production rate over land, assuming tritium is lost from the continental atmosphere only by continental rains running off into the ocean, and gained by production, and by transport of ocean vapor onto the continents. Using the tritium data for average Mississippi Valley rains, they obtained a figure of 0.16. Their estimated world average production rate is 0.14 with a probable uncertainty of less than 20 per cent. This value agrees precisely with the expected world production rate calculated by Currie, Libby, and Wolfgang (1956) from their experimental measurements on tritium production in nitrogen and oxygen by bombarding protons of 450-Mev and 2-Bev energies. Previous experiments and calculations by Fireman and Rowland (1955) gave an expected production rate of 0.2 T atoms/cm^2 sec, also in good agreement with the rate apparently observed.

However, the tritium production rate must be a good deal higher than the figures given above. Von Buttlar and Libby calculated that, with such a production rate, and with the observed surface sea concentration of about 0.24 T.U., then the mixed layer of the sea is about 100 meters deep if one assumes that all the tritium of the sea is in the mixed layer. Though this depth is consistent with observational data on the sea, such a calculation assumes that the mixed layer is sealed off from the deep sea so that no tritium mixes below the thermocline, and the question then arises as to just how much mixing across the thermocline does, in fact, occur.

As discussed by Wooster and Ketchum in a separate paper in this report, various observations on ocean currents and on the heat flux through the ocean floor, indicate that the deep ocean water turns over, or mixes with surface water, in times of the order of a few hundred years. Assuming a generalized two-layer model of the sea, consisting of a shallow mixed layer about 75 meters deep on the average, and a

homogeneous deep sea below the thermocline marking the interface between the layers, Craig (In press (a)) derived equations relating the production rate of a radioactive isotope to the concentrations of the isotope in the two layers of the sea and the mixing time through the thermocline. (These functions are discussed briefly in a separate paper by the writer in this report, in which calculations on the disposal of fission products in the sea and their ultimate steady state concentrations are discussed.) The applications of such calculations to the distribution of radiocarbon in the atmosphere and sea were demonstrated; these results are discussed in Section IV of this paper.

Application of such calculations to the distribution of natural tritium (Craig, 1957 (b) and manuscript in preparation) shows that for reasonable internal mixing rates of the sea, most of the world inventory of tritium must actually be in the deep sea below the thermocline. Thus for a deep water replacement time, or residence time of a water molecule in the deep sea before mixing into the surface layer, of 0 to 1000 years, and with a surface concentration of 0.24 T.U., the tritium flux into the sea must be between 7.6 and 0.3 atoms cm^2/sec. For the most reasonable deep sea residence time of the order of a few hundred years, the flux must be somewhere between 0.4 and 0.8. It is found that about $\frac{3}{4}$ of the total tritium in the sea is below the thermocline, with a deep-sea tritium concentration of about 0.014 tritium units.

The tritium production rate over the North American continent was recalculated (Craig, op. cit.) by taking into account the removal of tritium from the continent by the outgoing water vapor which does not condense over the land. This calculation gives a world average production rate of from 0.6–0.8 after correction for the latitudinal geomagnetic effect on the incoming cosmic rays. A tritium production rate of this order of magnitude indicates an average deep-sea residence time of water of about 250 years, for a simple two-layer ocean. Calculations based on a second-order ocean model in which the deep sea reservoir is exposed to the atmosphere at high latitudes would give a longer residence time relative to the mixed layer of the sea because of direct entry of tritium from the atmosphere to the deep sea. (See the discussion of radiocarbon residence times in Section IV of this paper.)

However, if the bulk of the tritium is not produced by cosmic radiation, but by solar accretion (see below), the world average production may be as high as 1.7 atoms cm^2/sec because the geomagnetic correction applies only to tritium produced by cosmic rays in the troposphere.

The calculated production rate over the oceans of about 0.14 is obtained by considering only the transfer of tritium into the sea by rainfall. Since rainfall appears to account for only about one-tenth of the tritium which actually enters the sea, it appears that the transfer of tritium from atmosphere to sea by direct molecular exchange across the sea surface is about 9 times as effective as the scrubbing action of precipitation.

A production rate of 1.4 atoms of tritium/cm^2sec means that the world inventory of tritium, before thermonuclear tests, was about 20 kg of tritium, or 200 megacuries, essentially all of which is in the ocean. However, from the experimental data obtained by the workers cited above on the production of tritium by the action of protons on nitrogen and oxygen, it appears very doubtful that the cosmic ray production rate can be much higher than about 0.2. In fact it is probably necessary to assume that tritium is produced on the surface of the sun and is directly accreted into the earth's atmosphere, rather than being a secondary result of the action of the cosmic ray protons on the atmosphere, as postulated by Feld and Craig (Craig, 1957 (b)).

From a study of the fall-out rate of strontium 90 pushed into the stratosphere by large atomic detonations, Libby (1956a, b) calculates the stratospheric residence time of strontium to be about 10 years (cf. Section IV of this paper). Since at least half of the tritium production should take place in the stratosphere even if all the production is due to the action of protons on the earth's atmosphere, slow mixing through the tropopause will pile up tritium in the stratosphere in the same way that slow exchange across the sea surface builds up the radiocarbon concentration in the atmosphere. One-sixth of the atmosphere is above the tropopause on the average, but the water vapor concentration is so low that only about 0.3 per cent of the total water vapor in the atmosphere is in the stratosphere; thus the tritium concentration of the stratospheric water vapor will be

much higher than that of the tropospheric vapor, which averages about 1 T.U. From the strontium data we assume that the mixing time of water vapor through the tropopause is at least 10 years.

Assuming a tritium production rate of 1.4, half of which is in the stratosphere, the tritium concentration of stratospheric water vapor is then calculated to be at least 300,000 tritium units. This is an astounding concentration factor relative to tropospheric water vapor. Recently the present writer and F. Begemann analyzed a series of samples of atmospheric molecular hydrogen for deuterium and tritium content respectively. Mass spectrometric measurements showed that all samples contained about 2–10 per cent less D than ocean water, falling just in the range of meteoric waters, and containing far too much deuterium to represent thermodynamic equilibrium with water vapor. These data confirmed a few previous measurements (cf. Harteck, 1954) which showed that the molecular hydrogen in the atmosphere must form by direct photodissociation of water vapor in the region around 70 km altitude, rather than by bacterial decomposition of organic matter which has been shown to produce hydrogen in isotopic equilibrium with water. We may thus assume that the tritium content of stratospheric molecular hydrogen is about the same as that of the stratospheric water vapor.

Assuming that the hydrogen is statistically distributed in the atmosphere, so that $\frac{1}{6}$ is above, and $\frac{5}{6}$ below, the tropopause, and taking again the mixing time through the tropopause as 10 years, we then calculate the tritium content of the molecular hydrogen in the troposphere. This figure is found to be 100,000 tritium units, probably as a minimum figure because of slow vertical mixing from the base of the stratosphere to the 70 km level where the hydrogen is made, and because of the indication that more than half the tritium is found initially in the stratosphere. The tritium contents measured by Begemann on a dozen samples of tropospheric hydrogen range from 50,000 to 100,000 tritium units, averaging about 80,000 T.U., in excellent agreement with the calculated value when the various uncertainities are considered.

It thus appears that the high tritium content of tropospheric hydrogen can be satisfactorily explained by purely geophysical reasoning based on the stratosphere-troposphere exchange time as estimated from Libby's Sr^{90} data, and the known concentration of water vapor in the stratosphere. This explanation seems more likely than the intricate series of photochemical mechanisms proposed by Harteck (1954) which at best may account for a tritium concentration of about 1000 T.U. in the molecular hydrogen.

Beryllium 7

Beryllium 7 is formed in cosmic ray stars, the peak production occurring at about 15 km. It decays by electron capture to lithium 7 with a half-life of about 53 days. The discovery, and the elucidation of the geochemical history, of this cosmic ray produced nuclide is due to Arnold and Al-Salih (1955).

Once formed in the atmosphere, the beryllium burns to the nonvolatile BeO or possibly $Be(OH)_2$, either of which diffuses until encountering a dust particle and adhering thereon. It is thus a tracer for the atmospheric dust, on which it is washed out of the atmosphere by rain, ultimately going into the ocean. Arnold and Al-Salih detected radioberyllium in 22 rain and snow samples from Chicago and Indiana, the average absolute assay being 6×10^6 atoms/liter. The estimated world-wide average production rate is 0.04 atoms per cm^2 per second, based on estimated rates of transfer and mixing in the stratosphere and troposphere. Most of the mixing rates involved are of the order of magnitude of the half-life, which makes calculation of the production rate difficult but greatly enhances the utility of the isotope for studying atmospheric processes, especially when used in conjunction with tritium.

A detailed discussion of the beryllium 7 production rate and atmospheric residence time has recently been given by Benioff (1956). He calculates the production rate to be 5.0 atoms/cm^2min in the stratosphere and 1.3 atoms/cm^2-min in the troposphere, and he finds that a stratospheric residence time of the order of years is required to match these production rates. Thus his stratospheric residence time agrees with that found by Libby for fission products.

Beryllium 10, a β^- emitter with a half-life of 2.5×10^6 years, is also formed in the cosmic ray stars. J. R. Arnold has recently identified this isotope in deep sea sediment samples (manuscript in press); it should be of great

importance, because of the long half-life, for the dating of such sediments.

Deuterium and Oxygen 18

Deuterium and oxygen 18 are stable isotopes of hydrogen and oxygen respectively, and it is now well known that the isotopes of these elements, as well as of other light elements such as carbon, nitrogen, and sulphur, are fractionated, or separated, by chemical and physical processes in natural systems. Since the fractionation factors for stable isotopes are measurable and/or calculable for many separation processes, and since the magnitude of these factors is mainly a function of temperature and process, the stable isotopes are extremely well adapted for the study of natural transfer rates in the geochemical cycles of their elements.

The concentrations of these isotopes show rather wide variations in different natural materials, these variations generally ranging from a few tenths of a per cent to a few per cent. In this report we shall mainly be concerned with the distribution of these isotopes in marine and fresh waters and in the atmosphere. Craig and Boato (1955) have recently reviewed the present status of natural isotopic studies, and reference is made to that paper for a more extended discussion.

VAPOR PRESSURES AND RELATIVE ABUNDANCES OF THE ISOTOPIC WATER MOLECULES

Species	Relative abundance (ocean water)	Mass	p (mm Hg) 30° C	p (mm Hg) 100° C
H_2O	1	18	31.5	760
HDO	1/3230	19	29.4	741
H_2O^{18}	1/500	20	31.3	756

The above table shows the three most prominent members of the family of isotopic water molecules, their masses, relative abundances in average ocean water, and their vapor pressures at two temperatures. Other members of the family are much less abundant and can be neglected. One sees from the table that the vapor pressures are not a direct function of the molecular weight; the vapor pressure difference between HDO and H_2O is 10 times larger than the vapor pressure difference between H_2O^{18} and H_2O, at 30°C. The isotopic separation in an evaporation or condensation process is directly proportional to these vapor pressure differences, so that in water vapor in equilibrium with water at 30°, the percentage depletion in deuterium, relative to the water, is ten times larger than the percentage depletion in oxygen 18.

The natural isotopic variations are customarily given in terms of per mil enrichment or depletion relative to a standard, similar to the way the density parameter is given in an oceanographic temperature-salinity diagram. The data are presented in terms of a function δ, defined as follows:

$$\delta(\%) = [(\text{Rsample}/\text{Rstd}) - 1] \times 1000$$

where R is the isotopic ratio O^{18}/O^{16} or D/H. In the case of deuterium, however, the quantity in the brackets is multiplied by 100 and the δ values are given in per cent, because of the ten times higher isotopic separations encountered. Rstd here refers to the isotopic ratio in average ocean water.

Since H_2O^{16} is the most volatile isotopic species, the water vapor over the oceans is depleted in the heavy isotopes relative to the surface ocean water. As this vapor moves over the continents, the first rain to fall out is enriched in the heavy isotopes relative to the vapor, again because of the higher volatility of the lightest species. Removal of the heavy isotopes, in the form of rain, then causes the vapor to become continually depleted in deuterium and oxygen 18. In general enough rain falls out of an air mass over the oceans so that by the time the mass reaches the continents the rain is already "lighter" in isotopic content than ocean water, and as the air mass moves inland and poleward the rain which falls out becomes more and more depleted in deuterium and oxygen 18.

In a recent study by Craig (ms. in preparation) several hundred fresh water samples from all over the world were analyzed for deuterium and oxygen 18 concentration. The deuterium concentration varies by about 30% relative to mean ocean water, δD ranging from +3 to −27%, while the oxygen 18 concentration varies by only 4%, δO^{18} ranging from +6‰ to −34‰. The delta values for the two isotopes show a linear correlation such that $\delta D = 9\delta O^{18}$, corresponding to the vapor pressure difference ratio at about 25°C. The reason for the high value of the average temperature at which liquid and vapor equilibrate in the atmosphere is as yet unknown; the uncertainty

in the vapor pressure data is such that the value could hardly be less than about 20°. The delta values for fresh waters show a general correlation with latitude or distance from the ocean; there is a general decrease in the heavy isotope concentration as the latitude varies from equatorial to polar, reflecting the continuous loss of vapor from the poleward moving air masses.

Isotopic variations such as mentioned above can be measured quite simply and precisely with the mass spectrometer, and it is evident, from the ranges of variation cited, that such studies on meteoric waters can provide a wealth of information concerning meteorological transfer and mixing phenomena in the atmosphere. The average water vapor of the earth has roughly the composition $\delta D = -10‰$, $\delta O^{18} = -11‰$, but large variations, related to the amount of liquid water which has condensed out of the vapor, occur, and thus such studies are directly adapted to problems of water vapor transport over both the oceans and continents.

The situation in the oceans themselves is somewhat more complicated. The oxygen isotopic composition of ocean waters has been studied by Epstein and Mayeda (1953), and the deuterium variations in the same samples by Friedman (1953); these writers also analyzed nine fresh water samples and first elucidated the D-O^{18} relationship in natural waters. The surface layers of the oceans are in general enriched in the heavy isotopes relative to mean ocean water because of the net storage of H_2O^{16} in the stagnant and circulating fresh water and vapor; the extent of this enrichment reflects the hold up at the boundary of the mixed surface layer, namely the thermocline. On the other hand, the deeper layers of the ocean are depleted in deuterium and oxygen 18, relative to mean ocean water, because of the influx of glacial melt water in polar latitudes, the glacial waters having δ values at the lightest ends of the ranges cited in the preceding paragraphs. Thus the oceans are isotopically upside down with the heavy isotopes concentrated at the surface, and the isotopic composition parameters in general correlate with salinity.

Epstein and Mayeda (op. cit.) showed that the salinity-oxygen 18 variations in marine waters were consistent with a model in which the oceanic precipitation is progressively depleted in the heavy isotopes as a function of the extent of precipitation from the local atmospheric reservoir. Salinity, of course, is uniquely related to the direct amount of fresh water removed by evaporation or added by meltwater dilution, but the relationship in the case of isotopic composition is more complex. This is because the isotopic composition of fresh water precipitating over the oceans, or added by runoff or melting of ice, is variable, depending on the history of the air mass from which it was precipitated. The correlation between isotopic composition and salinity is therefore more or less local, reflecting the particular relations obtaining on the average in the area. As a result, the isotopic composition parameters, rather than being simply transforms of salinity, and thus not inherently very useful for the study of transfer problems, become important parameters for such studies because of the reflection of areal conditions in a manner different from, but related to, the salinity parameter. Examples of this effect are discussed in Part IV, where applications to transfer studies are treated.

The isotopic composition of atmospheric oxygen is an interesting case of adjustment of a reservoir composition to steady state non-equilibrium biogeochemical transfer processes. Oxygen would exist in the atmosphere in the absence of living plants because of photodissociation of water vapor in the atmosphere, with subsequent escape of hydrogen from the earth. However, oxygen is cycled through the biosphere so rapidly that its isotopic composition, rather than reflecting its mode of formation, may be adjusted to a steady state balance between photosynthetic formation and respiratory uptake. The oxygen produced in photosynthesis is in isotopic equilibrium with the water taken up by the plants and is very close in isotopic composition to this water; however the atmospheric oxygen is some 23‰ enriched in oxygen 18 relative to average ocean water. Lane and Dole (1956) have measured the preferential uptake of oxygen 16 by various animals and land plants and concluded that the net fractionation is such as to account quantitatively for the atmospheric oxygen composition. Respiration in the oceans shows a much smaller selective oxygen 16 uptake (Rakestraw et al., 1951; Dole et al., 1954) and the isotopic composition of oxygen dissolved in ocean water is variable and dependent on the amount of oxygen which has been taken up from the local

reservoir. There is some doubt as to whether the data of Lane and Dole can actually yield a material balance without invoking some special mechanisms relating the productivities of the oceans and the land, and a good deal of further study on this question is needed. The intent here is to point out that the isotopic transfer rates involved in this problem of the isotopic composition of atmospheric oxygen, and the variations in the isotopic composition and amounts of oxygen dissolved in ocean waters, may well be important parameters for the study of transfer phenomena in the oceans and the atmosphere and the interaction between them.

Carbon 13

About one per cent of natural carbon consists of the stable isotope C^{13}; the ratio C^{13}/C^{12}, and thus effectively the C^{13} concentration, in natural material shows a range of variation of about 6 per cent. The details of the natural variation have been described (Craig, 1953, 1954), and reference is made to these papers for extended discussion. The delta values for carbon are referred to a standard which has the composition of average limestone; on this scale the characteristic compositions of natural materials are shown below:

Material	δC^{13} (‰)
Limestones and shell	0
Ocean bicarbonate	— 1.5
Atmospheric CO_2	— 7
Marine biosphere	—13
Terrestrial biosphere	—25
Coal	—25
Petroleum	—28
Shales	—28

The difference between the compositions of atmospheric carbon dioxide and ocean bicarbonate probably reflects the isotopic equilibrium constant for the exchange of carbon isotopes between these compounds; the other variations shown in the table are due to kinetic factors which cause a selection of the isotopes in the various processes involved in the biogeochemical cycle of carbon. The carbon 14 variations caused by such processes should be almost exactly twice the C^{13} values shown above, and, as noted previously, the knowledge of the C^{13} variations has been of great value in understanding the transfer rates and mixing phenomena involved in the distribution of radiocarbon.

A particularly fertile field for study is the marine biosphere and the phenomena involved in the isotopic partition of carbon between carbonate and organic matter. One critical parameter in the kinetic processes involved is the rate of uptake of CO_2 by photosynthesis versus the relative rates of CO_2 replenishment by mixing and by reassociation of bicarbonate ions, and such studies may well lead to an improved knowledge of the carbon flux through local ecological systems and the interaction of the local system with the general marine reservoir. Keeling (manuscript in preparation) has studied the isotopic variations in carbon dioxide over the land, and has found that the isotopic parameters are critical indicators of the atmospheric transfer phenomena through local biotopes, as a result of the large difference in isotopic composition between normal atmospheric carbon dioxide and carbon dioxide produced in respiration during the night.

III. *Contribution of radioisotopes to the geosphere by nuclear fission and detonations*

The steady state isotopic distributions discussed in the preceding section have, in the case of radioactive elements, been altered to some extent by contribution to the geosphere of radioisotopes produced in nuclear fission in both reactors and nuclear detonations. Such contributions, rather than being detrimental to the study of natural transfer phenomena, have, on the whole, provided extra parameters of great value for such studies. It is of course obvious that addition of such elements under carefully controlled conditions in selected locations and at planned times would have contributed a great deal more to our knowledge of geophysical phenomena than the actual dispersal of the material has resulted in; nevertheless it is possible, even though working in almost total ignorance of the amounts of material added, to deduce a great deal of valuable information about mixing rates and even to make detailed studies of certain specific problems.

The fission of uranium in reactors and nuclear weapons results in a great variety of elements distributed mass-wise into a spectrum known as the fission yield curve; the proportions of the various masses produced are a unique function of the atomic mass and vary

little with neutron energy or substitution of plutonium for uranium 235. For our purposes, the elements of most interest produced by fission are krypton 85, strontium 90, and cesium 137, ranging in half-life from 10 to 33 years; tritium, which is not a fission product, is also of great importance. Measurable additions to the geosphere of tritium, strontium 90, and krypton 85 have been noted and are discussed in Part IV in connection with general applications to transfer study phenomena. In this section we estimate the total amounts of these nuclides which have been produced; these estimates are also of some interest in connection with the magnitude of the disposal problem, both present and future.

One result of the advent of nuclear fission is that all the krypton in the atmosphere has become contaminated with radiokrypton. De Vries (1956) has measured the specific activity of atmospheric krypton, taken in March of 1955, as 25,000 counts per minute per mole. The activity is due to contamination with krypton 85, which decays by β^- emission with a half-life of ten years. From DeVries' measurement, we readily calculate that 56.4 moles, or 4700 grams, of Kr^{85} are now present in the atmosphere, and in ignorance of the rate of production, we make only a small error by assuming this figure as the total amount of radiokrypton produced and not correcting for decay. From the fission yield of 0.24 per cent for this isotope, it appears that some 23,500 moles, or 5500 kg, of U^{235} and plutonium have undergone fission since the advent of anthropogenetic fission, resulting in an atmospheric krypton activity of 2 megacuries. It is assumed, as seems reasonable, that all fission produced krypton finds its way into the atmosphere.

From the fission yield data, we calculate the total amounts of radiostrontium and cesium which have been produced; the data for the three elements are shown below. Only the strontium and cesium produced by detonation of nuclear weapons will escape into the atmosphere and be deposited over the surface of the earth and sea. Because the krypton figure is uncorrected for decay, and because some krypton must have gone directly into the stratosphere and is not included in the measured activity, the values all represent lower limits and should be slightly larger.

Radioisotope	Half-life	Fission yield (%)	Total activity produced by all fission (megacuries)
Krypton 85	10 years	0.24	2
Strontium 90	28 years	5.0	15
Cesium 137	33 years	6.3	16

Libby (1956a, b) has given detailed discussions of the fall-out patterns of strontium 90 and cesium 137, based on the Project Sunshine measurements on world-wide samples. Geophysically, the most significant finding is that, as mentioned previously, the residence time in the stratosphere of material pushed through the tropopause is about 10 years. The most recent measurements on the distribution of fission products from nuclear explosions (Libby: Address before American Association for the Advancement of Science, Washington, D.C., October 12, 1956) indicate that the amount of strontium 90 scattered over the surface of the earth is now equivalent to an average activity concentration of about 16 millicuries per square mile. In addition, the amount now held in the stratosphere is equivalent to another 12 millicuries per square mile. The total amount so far distributed is thus about 5.6 megacuries of Sr^{90}, of which about 2.4 are still in the stratosphere, and, assuming purely statistical distribution, some 2.3 megacuries have fallen directly into the sea, while about 0.9 megacuries have fallen on the land surface. Because of the similar half-life and fission yield, the figures for cesium 137 will be almost identical to those for strontium 90. Comparing these figures with the ones given in the above table, we see that roughly 5.6/17.4 or $\frac{1}{3}$ of all the solid fission products so far produced, by all fission, have been distributed over the atmosphere, the land, and the sea, by atomic weapons testing.

The most important of these elements for studying mixing rates in the sea should be cesium 137, which being soluble, should be an excellent tracer for the mixing rate of surface ocean water down through the thermocline. Krypton 85 should ultimately prove important for atmospheric mixing studies, especially for comparison of mixing rates of gaseous and solid elements across the tropopause.

Thermonuclear weapons may also be expected to produce some carbon 14 because of the neutrons released into the atmosphere in the explosion. A contemporary sample of grass, collected in the summer of 1955 in S.W. Kansas by the writer, was analyzed for C^{14} content by M. Rubin at the U.S. Geological Survey laboratory. This grass was found to be about 2.5 per cent higher in C^{14} content than the 19th century wood, corrected for age, used as the U.S.G.S. radiocarbon standard (Suess, 1955). The samples and standard were analyzed for C^{13} content by the writer and the results corrected for isotopic fractionation, and the sample was counted twice. Thus the measurement is quite precise, and probably indicates that the atmospheric radiocarbon content has risen about 2 per cent above normal at the present time, due to thermonuclear neutron production. For future geochemical studies with natural radiocarbon it will be important to monitor continuously the activity of contemporaneous plants and atmospheric carbon dioxide, though the effect will be insignificant for radiocarbon dating studies for some time yet.

The situation with tritium is different. The earliest rain ever analyzed for tritium content fell in Chicago in May of 1951; since October 1952 Libby and his co-workers at Chicago have produced an essentially continuous record of the tritium content of Chicago rain, and have analyzed a great many other samples from many parts of the world. Their data show that there was no significant production of tritium in the November 1952 Ivy test (Kaufman and Libby, 1954). However, the March 1954 Castle thermonuclear tests produced an increase in tritium concentration of Chicago rain from an average value of 9 to a maximum value of 450 atoms $T/10^{18}$ atoms H; i.e., a factor of 50 (von Buttlar and Libby, 1955). Even more striking was the discovery that the tritium content of southern hemisphere waters showed no significant increase in tritium concentration, and snow samples collected from the Antarctic as late as February of 1955 showed that during this interval no significant amounts of artifically produced tritium had crossed the equator (Begemann, 1956).

Begemann's recent data show that the tritium rained out of the northern hemispheric atmosphere with a mean-life of about 40 days for the decrease in tritium concentration; as late as the end of 1955 the tritium concentration of Chicago rain was still about three times normal. In Section II above it was concluded that the world inventory of tritium was about 20 kg, with about 5 kg in the mixed layer of the sea, and about 15 kg in the deep sea. The Chicago data show that the tritium content of the surface ocean waters has increased by at least a factor of four, indicating that the order of magnitude of 20 kilograms of man-made tritium has so far rained out into the ocean. Thus the amount of tritium produced by man is now about equal to the natural steady-state inventory.

IV. *Applications of tracer techniques to the study of physical processes in the sea and atmosphere*

In this section we describe a few of the more obvious applications of the tracer techniques and isotopes described in the previous sections to specific problems of transfer phenomena in the oceans and atmosphere. The topics are subdivided in terms of the isotopes discussed, in order to facilitate reference to points in preceding sections and parts of this section.

Carbon 14

Carbon 14 is perhaps the most useful of the isotopic tools available for geophysical and geochemical studies, especially when used in conjunction with oxygen 18 data; the 5700 year half-life and the universal distribution of carbon in organic and inorganic reservoirs make it ideal for such purposes. The most obvious application of immediate interest is the dating of the bicarbonate of the deep-sea waters, in order to determine the mixing rate of the oceans. Unfortunately, only one definitive set of measurements of this type has been made, namely the U. S. Geological Survey laboratory measurements of waters east of the Lesser Antilles at approximately 57° W. and 16° N., made by M. Rubin (personal communication). These data are shown below:

Depth (meters)	Carbon 14 age (years)
Surface	652
640	634
1640	628
1750	841

The absolute values are probably not better than ±150 years, but the relative values are more precise. The age of the surface bicarbonate is somewhat older than the 400-year average age mentioned in Part II as the result of slow transfer of atmospheric carbon into the sea, perhaps as a result of local conditions; however, the important figure is the age difference between surface and deeper waters and it is unfortunate that still deeper waters were not sampled. The importance of a great many vertical profiles of this sort from both oceans, and their fundamental import for knowledge of the mixing rates in the ocean, is obvious.

Because of the requirements of steady state balancing, the amounts of water transferred, per unit time, downward and upward through the thermocline in the sea must be equal, but because the mixed layer contains only about 2 per cent of the sea, this balance requires that a water molecule remain, on the average, some 50 times longer below the thermocline than above. As a consequence of this relationship, an uncertainty of 10 years in the residence time of material in the mixed layer results in an uncertainty of 500 years in the residence time of the material below the thermocline, considering the world average rate of general cross-thermocline mixing of the substance. As we shall see below, the half-life of radiocarbon happens to be so long, that considerations of the extensive data on C^{14} distribution in the atmosphere, biosphere, and mixed layer of the sea, do not yield important information on the internal mixing rate of the ocean itself. In fact, the distribution of tritium above the thermocline of the sea furnishes a much more precise estimate of the general turnover time of water in the deep sea.

Thus the application of radiocarbon analysis to mixing problems within the sea itself can be made only by actually getting below the mixed layer and studying the deep-sea distribution of C^{14} directly; such studies, coupled with chemical analyses and physical data serving as parameters for the identification of continuous water masses, will probably prove to be the most fruitful method for the delineation of large scale mixing phenomena in the sea.

On the other hand, the distribution of radiocarbon in the atmosphere and mixed layer of the sea is strongly dependent on the rate of exchange of carbon dioxide between the atmosphere and sea, and from a study of the relationship between the exchange rates and the radioactive decay rate, it is possible to derive rather precise values for the flux of carbon into the sea and downward through the thermocline. For such calculations it is necessary to assume a model of the atmosphere-sea system based on simplifying assumptions as to the nature of the sea below the thermocline. Calculations of this type, outlining the factors affecting the natural distribution of radiocarbon, have recently been made by Suess (1953), Arnold and Anderson (1957), Craig (1957 (a)), and Revelle and Suess (1957). The conclusions of these papers, though reached by various means, are quite similar, and we shall briefly summarize the general results.

There are two empirically observed effects, of different origin, by which factors affecting the natural distribution of radiocarbon may be evaluated. The first of these is the observation that the carbon in the surface layers of the sea (bicarbonate, shell, and organic matter) has an apparent age of about 400 years relative to the terrestrial wood used as standards for radiocarbon dating. The second is the observation that contemporaneous wood has a radiocarbon activity some 2 per cent lower than the activity of 19th century wood, corrected for age to the present date. This decrease in activity, reflecting the contribution of C^{14} free CO_2 to the atmosphere by the combustion of fossil fuel, was first found by Suess (1953) and we shall refer to it as the Suess effect.

The "apparent age" of carbon in the mixed layer of the sea has been measured on Atlantic ocean samples (and one Pacific sample) by Suess (1954), and on Pacific ocean samples around New Zealand by Rafter (1955). The average age determined by Suess is 430 years, while that of the Pacific samples was reported by Rafter as only 290 years. However, the Suess measurements are relative to the 19th century wood standard, corrected for decay to the present, while the Rafter measurements were made relative to a contemporaneous standard which has suffered a decrease in activity due to the Suess effect. Measurement of the effect in the New Zealand standard shows that 110 years must be added to the ages reported by Rafter (1955) in order to correct for this effect and make the ages comparable to those reported by Suess (Rafter, manuscript in press).

Thus the average ages reported for the two oceans are in almost exact agreement, and we may consider the 400 year apparent age well established as a world-wide phenomenon.

The 400 year apparent age of mixed-layer carbon is simply a less meaningful way of stating that the radiocarbon activity of mixed-layer carbon is 5 per cent lower than the activity in modern wood, uncontaminated by the Suess effect. Actually it is observed that the activities in wood and in surface ocean carbon are measured to be the same, but the measurements must be corrected for natural isotopic fractionation in the physical and chemical processes involved in the carbon cycle (see section on carbon 13 variations). Marine shells concentrate carbon 13 by 2.5 per cent relative to terrestrial wood, and must therefore concentrate carbon 14 by 5 per cent; since this concentration factor is not observed, we see that the activity of carbon in the mixed layer is, in fact, 5 per cent lower than expected. The relationships between carbon 13 and carbon 14 variations expected on theoretical grounds, and on the basis of laboratory measurements, were discussed in detail by Craig (1954) who showed that the 5 per cent discrepancy must be the result of slow transfer of carbon from the atmosphere to the sea, and cannot be explained by any other cause. Rafter (1955) verified the conclusion that the carbon 14 difference between atmospheric CO_2 and wood must be twice the carbon 13 difference, by direct measurement.

The exchange rate of carbon dioxide between atmosphere and sea may be deduced from considerations of the steady state relationships between the exchange rate and the radioactive decay rate; this type of evaluation is independent of considerations based on the magnitude of the Suess effect and the kinetics of the transient state. The general equations governing the transfer of a radioactive isotope between its various exchange reservoirs have been given by Craig (1957(a)) in terms of the relationship between the uniform activity which would be observed if all of the sea and the atmosphere were mixed together at a rate infinitely faster than the radioactive decay rate, and the percentage deviations from this uniform activity which are actually observed in the different reservoirs. Mixing rates are expressed in terms of the residence time of a molecule in a particular reservoir, which corresponds to the operational definition of flushing time or replacement time, used by oceanographers, and, for the first order processes with which we are concerned, to the reciprocal of the exchange rate constant.

The constant radioactive decay rate of carbon 14 furnishes a built-in clock which monitors the transfer rate of carbon between its various reservoirs. For example, if a barrier is interposed between the atmosphere and sea, so that the transfer rate of carbon between these reservoirs is slowed down, the radiocarbon atoms formed in the atmosphere have less probability of getting into the sea and thus of leaving the atmosphere by physical removal. However, the steady state requires that the total number of C^{14} atoms leaving the atmosphere by all mechanisms be equal to the production of radiocarbon atoms by the cosmic rays, and thus the number undergoing radioactive decay in the atmosphere must increase. The number of radioactive atoms decaying per unit time is a constant fraction of the total number present (the exponential decay law), and therefore the piling up of radiocarbon in the atmosphere because of such an exchange barrier results in an increase in the number decaying in just the way required to maintain the steady state secular equilibrium with the production rate. The percentage increase in the C^{14} activity of the atmosphere is a function of the ratio between the exchange rate and the decay rate, or, what is the same thing, between the atmospheric residence time and the radioactive mean life.

Considering then, the percentage change in the radiocarbon activity of atmospheric CO_2 and terrestrial wood, relative to the activity which would characterize these materials in the hypothetical state of infinitely rapid mixing between atmosphere and sea, it is found that for each year of residence time of a CO_2 molecule in the atmosphere as a result of slow exchange, the atmospheric activity will increase by 0.74 per cent. The activity in the sea would, of course, decrease as a result of the slower transfer of radiocarbon into the ocean, but since there is some 60 times as much carbon in the sea as in the atmosphere, the percentage decrease of activity in the sea will be only 1/60 of the atmospheric increase, namely about 0.01 per cent, which is not observable.

We can make a more detailed model of the carbon exchange system by breaking the sea

up into a two-layer ocean, taking the upper layer to be about 75 meters deep corresponding to the average mixed layer as actually observed in the sea. (The 75 meter estimate was made by Dr. Warren Wooster, who kindly studied the question of the average depth of the mixed layer over the year in the various areas of the oceans.) The lower layer, extending to a depth of 4000 meters on the average, is termed for convenience the "deep sea," though it is of course obvious that such a uniform layer has little resemblance to the actual structure of the sea below the thermocline. Nevertheless, it is found that the consequences of such an assumption about the nature of the deep sea are not serious insofar as affecting the validity of the calculations on the atmospheric residence time, and the treatment of the relationships existing between the atmosphere, mixed layer, and main body of the sea, is of course improved immensely by assuming such a model. If we then add a barrier between the mixed layer and the deep sea, representing slow mixing across the thermocline, the radiocarbon is further piled up in both the atmosphere and the mixed layer, in the same manner as previously described. Calculation shows that the activities in the atmosphere and mixed layer are both increased by about 1.2 per cent, relative to the case of a rapidly mixed, uniform sea, for each 100 years of residence time in the deep sea, or, what is almost the same thing, for each 100 years of "age" of the deep water. The activity in the deep sea is reduced by 0.05 per cent for each 100 years of deep-sea residence time.

For deep-sea residence times up to several thousand years, the interpolation of a mixing barrier at the thermocline in the sea causes very close to the same activity increase in both the atmosphere and the mixed layer, and thus the activity difference observed between the atmosphere and mixed layer is sensitive only to the atmosphere-sea exchange rate for internal mixing times of the sea of the order of a few thousand years or less. The physical evidence discussed by Wooster and Ketchum in a separate paper in this report, and the tritium calculations cited previously in this paper, clearly show that the average mixing time of the sea is at least within this range.

Thus the figure cited above of a 0.74 per cent increase in atmospheric activity for each year of atmospheric residence time, indicates that the residence time of a CO_2 molecule in the atmosphere, before entering the sea, is about 7 years, corresponding to the 5 per cent activity difference between carbon in the atmosphere and in the mixed layer of the sea.

An independent calculation of the atmospheric residence time can be made by considering only the steady-state material balance in the atmosphere as a function of the production rate of radiocarbon, taken as $(2 \pm .5)$ C^{14} atoms/cm^2 sec, and the rate at which carbon enters the sea. This calculation gives an atmospheric residence time of about 6 years. Considering the errors to be assigned the numerical values in both these calculations, it appears that the best value of the atmospheric residence time of carbon dioxide may be taken as 7 ± 3 years, corresponding to a rate constant $k_{a-m} = 0.14$, where k is the fraction of the carbon in the atmosphere transferred to the mixed layer per year (Craig, 1957 (a)).

The average annual exchange flux of carbon dioxide, into and out of the sea each year, is thus found to be about 2×10^{-3} moles per square centimeter of sea surface. This rate is lower by a factor of 10,000 than the rate recently obtained by Dingle (1954) from consideration of the various rate constants involved, and the discrepancy thus serves to emphasize the power of natural isotopic studies to yield quantitative data, as compared with more traditional methods.

An entirely independent calculation of the atmospheric residence time, not based on steady-state considerations, may be made from the magnitude of the so-called Suess effect described previously. It is known that since the beginning of the industrial revolution, man has added an amount of carbon dioxide to the atmosphere by fuel combustion equivalent to about 12 per cent of the amount originally present. The degree of dilution of radiocarbon activity in contemporaneous wood by incorporation of C^{14}-free CO_2, measured relative to the activity of 19th century wood, is then a measure of the rate at which the dead CO_2 has been removed from the atmosphere into the sea. The first measurements of this effect, made by Suess (1953), indicated a dilution of about 3 per cent, and from these data Suess deduced an atmospheric CO_2 residence time of 20–50 years.

More recent and extensive measurements by Suess (1955) have shown that the figure of

3 per cent is higher than the average world-wide figure, and represents an increased local contamination in trees growing near sites of industrial activity. The latest measurements indicate a world-wide effect of about 1.7 per cent. Revelle and Suess (1957) have discussed the relationships between the exchange rate, the Suess effect, the effect of an increase in the atmospheric CO_2 content on the atmospheric and oceanic reservoirs, and the buffering effect of the sea water alkalinity on carbon transients. They conclude that, all things considered, the residence time of CO_2 in the atmosphere, relative to exchange with the sea, is of the order of 10 years. Though the uncertainty in their estimate is a good deal larger than in the case of the steady-state considerations discussed above, the close agreement of the figures obtained by these different considerations is gratifying, and indicates that the factors governing the natural distribution of radiocarbon are now fairly well understood.

The size of the terrestrial biosphere and the annual rate of photosynthesis on land have been estimated by Schroeder and Noddack, and from their figures it appears that the terrestrial plants consume about 3 per cent of the atmospheric CO_2 per year, corresponding to an atmospheric residence time before entrance into the biosphere of 33 years. With a residence time of 7 years, prior to exchange into the sea, the total residence time of a CO_2 molecule in the atmosphere is 6 years, after which it goes either into the sea (9 chances out of 11) or into the terrestrial biosphere (2 chances out of 11). Thus the carbon dioxide flux into the sea is about 4.5 times larger than the flux into the biosphere, and about 82 per cent of the CO_2 leaving the atmosphere goes into the sea, while only about 18 per cent goes into the terrestrial plants. This ratio represents a considerable departure from previous estimates, and indicates that the spatial distribution of plants and soils is probably not the dominant factor in determining the steady-state CO_2 concentration in the atmosphere. In fact it appears more likely that the spatial pattern of absorption and release of CO_2 by the sea, and the seasonal variations in this pattern, are the dominant factors.

The various considerations outlined above are all consistent with any deep-sea residence time of carbon up to a few thousand years, and do not yield any closer estimate for this figure. Recent unpublished data by Broecker and co-workers at the Lamont Geological Observatory indicate that the bicarbonate of deep ocean waters probably averages about 8 per cent lower in C^{14} content than the surface mixed layer, corresponding to a radiocarbon "age" of the order of 670 years. However, considerations by Craig (in press), based on a second order oceanic model in which the deep sea reservoir is exposed to the atmosphere in high latitudes, show that about half of the radiocarbon in the deep sea is derived directly from the atmosphere. The other half enters the deep sea from the surface mixed layer of the ocean by the mixing and interchange of water.

Because of this dual source of radiocarbon, the residence time calculated for carbon in the deep sea is only about half of the actual residence time of a water molecule in the deep sea relative to the mixed layer; thus the deep-sea residence time of water relative to the mixed layer is probably of the order of 1000 years as a world-wide average. However the actual interpretation of such residence times in the sea is quite complicated, and reference is made to the paper cited above for a detailed discussion of carbon and water residence times.

Deuterium and Oxygen 18

As discussed previously, the stable isotopes are of great value in the study of ocean water mixing as additional parameters related to salinity. One particular case in which information can be gained from such studies is the problem of meltwater dilution of the oceans in the polar regions. A salinity decrease can be caused by addition of fresh water from river runoff, or from the melting of sea ice, and from salinity data alone these sources cannot be differentiated. However, the isotopic composition of the two sources is quite different; the sea ice should have a composition quite similar to that of the ocean water, while, as shown above, the runoff of rivers in polar areas is greatly depleted in deuterium and oxygen 18 relative to ocean water. Thus from consideration of salinity and isotopic data taken together, a quantitative evaluation of the mixing conditions can be made. Friedman of the U. S. Geological Survey is currently studying such problems with deuterium analyses of Atlantic waters. The isotopic data should also be useful in material balance

studies over various sections of the oceans, because of the latitudinal decrease in deuterium and oxygen 18 concentration of oceanic water vapor, and the known temperature dependence of the isotopic selection in evaporation.

Craig, Boato, and White (1956) have shown how deuterium and oxygen 18 measurements can be usesd to determine the proportions of juvenile or magnetic water to reheated ground water in thermal springs, and volcanic steam. These isotopes, together with tritium, have important applications to practically all hydrologic problems, and the exploitation of such techniques has barely begun.

Tritium and Strontium 90

As described in Part II, the tritium measurements made by Libby and his co-workers furnish an independent value for the mixing rate in the sea; more detailed studies will surely provide important information on the oceanic mixing phenomena. The production of tritium in thermonuclear explosions provides an isotopic tracer for determination of atmospheric mixing times across the face of the earth and storage times in the atmosphere.

The measurement of the world-wide distribution of strontium 90 produced by nuclear detonations has been done by W. F. Libby and E. A. Martell at the University of Chicago. The results of their work have recently been described by Libby (1956 a, b). The radionuclides produced by low-yield kiloton weapons, and part of the activity produced by the higher-yield megaton weapons, are distributed within the troposphere in a belt corresponding to the latitude of the test site. This material has a tropospheric life which is a function of particle size; some of the activity may circle the earth two or three times within the hemisphere in which it was produced before being washed out of the atmosphere. However, the mean life of this tropospheric material is only a few weeks.

More interesting is the fact that Libby and Martell find that half or more of the radiostrontium produced by the megaton weapons is distributed over both hemispheres and falls out much more slowly, the mean storage time in the atmosphere being of the order of ten years. They conclude that this material is carried up into the stratosphere, above the tropopause, where it is mixed horizontally in a time comparable to the storage time at this level.

The contrast between the distribution of megaton weapon produced radiostrontium and tritium is extremely significant. As noted in Part III, Begemann and Libby find that the artificially produced tritium is confined to a single hemisphere and is rapidly washed out of the atmosphere; this material thus follows the pattern of the activities which remain in the troposphere. The tritium and fission product data thus show that over a period of months there is virtually no cross-hemispheric mixing in the troposphere, but that over a period of years the stratosphere is well-mixed horizontally. The failure to detect tritium carried up into the stratosphere with the megaton weapon produced radiostrontium may be due to the instantaneous combustion of tritium to HTO by the catalytic action of the oxides of nitrogen produced in the blast (Harteck, personal communication). As water, the tritium may be frozen out at the lower cold trap, in the tropopause, where the temperature is about $-70°C$, and thus prevented from entering the stratosphere.

On the other hand, Martell points out (personal communication), that the thermal energy of the fireball is still quite large by the time a fireball produced by a megaton weapon has risen to the height of the tropopause. In order for HTO to condense and thus be trapped below the tropopause, it is necessary to assume that the lighter constituents of the fireball have diffused into the cooler outer layers. Martell suggests that if such is the case, then the actual explanation may be that the portion of the cloud containing the HTO may not have sufficient thermal energy to penetrate the tropopause, and as a result, this portion of the cloud merely expands horizontally below the tropopause.

V. *Conclusions*

From the discussion in the preceding parts of this report, it is apparent that the advent of manmade nuclear reactions introduced a series of geophysical and geochemical experiments on a vast scale. It is fortunate that the introduction of such experiments came at a time when geochemists were well underway towards the understanding of natural transfer phenomena by means of studies based on naturally occurring isotopes in their steady state biogeochemical cycles. It should be clear that the need for this knowledge is such that every effort should be

made to prevent irreversible procedures which might eliminate the opportunity to study such mixing at the natural level where evaluation of the long term variables is possible. On the other hand, it is also evident that the introduction of artificially produced radioisotopes into the geosphere has been productive of a great deal of new knowledge that might otherwise not have been obtained.

The importance of continuous monitoring of the levels of such substances as tritium cannot be overemphasized. As an example of this, it may be pointed out that one reason that carbon 14 is such a powerful tool for the evaluation of ocean-atmosphere interaction that we have relatively precise records on just how much dead carbon has been produced by the combustion of fossil fuels; were this information not available the use of radiocarbon in such studies would be exceedingly difficult, if not impossible.

From the carbon 14 inventory discussed in Part II, and assuming an average depth of about 150 meters for the oceanic thermocline, it appears that about 4 per cent of the carbon 14 in the sea lies above the thermocline; this corresponds to an activity of about 10 megacuries. It is thus evident that introduction of artificially produced radiocarbon in 10,000 curie amounts above the thermocline would begin to produce a critical level which would interfere with the natural radiocarbon studies of such fundamental importance. Introduction of 100–1000 curie amounts above the thermocline would produce activity sites which could be traced for years, but such experiments could not be done more than once every decade or so if the natural level is to be preserved. It would thus seem highly desirable that some international body be constituted to record and monitor the material put into the sea and the atmosphere as wastes and for tracer experiments. It is a truism to point out that a contaminated laboratory is rather easily replaced, but that the laboratory of the earth scientists is not easily renovated.

CONCLUSIONS

ANDERSON, E. C. 1953. The production and distribution of natural radiocarbon. *Ann. Rev. Nuclear Science* **2**:63–78.

ARNOLD, J. R., and H. A. AL-SALIH. 1955. Beryllium-7 produced by cosmic rays. *Science* **121**:451–453.

ARNOLD, J. R., and E. C. ANDERSON. 1957. The distribution of carbon-14 in nature. *Tellus* **9**:28–32.

BEGEMANN, F. 1956. Distribution of artificially produced tritium in nature. Nuclear Processes in Geologic Settings: Proceedings of the Second Conference. *National Academy of Sciences — National Research Council Publication* **400**:pp. 166–171.

BENIOFF, P. A. 1956. Cosmic-ray production rate and mean removal time of Beryllium-7 from the atmosphere. *Phys. Rev.* **104**: 1122–1130.

CRAIG, H. 1953. The geochemistry of the stable carbon isotopes. *Geochim. et Cosmochim. Acta* **3**:53–92.

1954. Carbon 13 in plants and the relationships between carbon 13 and carbon 14 variations in nature. *J. of Geology* **62**: 115–149.

1957 (a). The natural distribution of radiocarbon and the exchange time of carbon dioxide between atmosphere and sea. *Tellus* **9**:1–17.

1957 (b). Distribution, production rate, and possible solar origin of natural tritium. *Phys. Rev.* **105**:1125–1127.

In press. The natural distribution of radiocarbon: II. Mixing rates in the sea and residence times of carbon and water. *Tellus.*

CRAIG, H., and G. BOATO. 1955. Isotopes. *Ann. Rev. Phys. Chem.* **6**:403–432.

CRAIG, H., G. BOATO, and D. E. WHITE. 1956. The isotopic geochemistry of thermal waters. Nuclear Processes in Geologic Settings: Proceedings of the Second Conference. *National Academy of Sciences — National Research Council Publication* **400**:pp. 29–38.

CURRIE, L. A., W. F. LIBBY, and R. L. WOLFGANG. 1956. Tritium production by high-energy protons. *Phys. Rev.* **101**:1557–1563.

DE VRIES, H. 1956. Purification of CO_2 for use in a proportional counter for ^{14}C age measurements. *Appl. Sci. Res.* (B) **5**: 387–400.

DINGLE, A. N. 1954. The carbon dioxide exchange between the North Atlantic ocean and the atmosphere. *Tellus* **6**:342–350.

DOLE, M., G. A. LANE, D. P. RUDD, and D. A. ZAUKELIES. 1954. Isotopic composition

of atmospheric oxygen and nitrogen. *Geochim. et Cosmochim. Acta* **6**:65–78.

EPSTEIN, S., and T. K. MAYEDA. 1953. Variation of O^{18} content of waters from natural sources. *Geochim. et Cosmochim. Acta* **4**:213–224.

FIREMAN, E. L., and F. S. ROWLAND. 1955. Tritium and neutron production by 2.2-Bev protons on nitrogen and oxygen. *Phys. Rev.* **97**:780–782.

FRIEDMAN, I. 1953. Deuterium content of natural water and other substances. *Geochim. et Cosmochim. Acta* **4**:89–103.

HARTECK, P. 1954. The relative abundance of HT and HTO in the atmosphere. *J. Chem. Phys.* **22**:1746–1751.

KAUFMAN, S., and W. F. LIBBY. 1954. The natural distribution of tritium. *Phys. Rev.* **93**:1337–1344.

LANE, G. A., and M. DOLE. 1956. Fractionation of oxygen isotopes during respiration. *Science* **123**:574–576.

LIBBY, W. F. 1955. Radiocarbon dating. Univ. of Chicago Press, Chicago: 2nd edition, 175 pp.

1956(a) Radioactive fallout and radioactive strontium. *Science* **123**:656–660.

1956(b) Radioactive strontium fallout. *Proc. Nat. Acad. Sci.* **42**:365–390.

MORRISON, P., and J. PINE. 1955. Radiogenic origin of the helium isotopes in rock. *Ann. New York Acad. Science* **62**:69–92.

RAFTER, T. A. 1955. ^{14}C variations in nature and the effect on radiocarbon dating. *New Zealand J. Sci. Tech.* (B) **37**:20–38.

RAKESTRAW, N., D. RUDD and M. DOLE. 1951. Isotopic composition of oxygen in air dissolved in Pacific ocean water as a function of depth. *J. Amer. Chem. Soc.* **73**: 2976.

REVELLE, R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear science and oceanography. Intern. Conf. on Peaceful Uses of Atomic Energy, Geneva, also Contr. Scripps Inst. Ocean. *N. S.* **794**: 22 pp.

REVELLE, R., and H. E. SUESS. 1957. Carbon dioxide exchange between atmosphere and ocean and the question of an increase in atmospheric CO_2 during the past decades. *Tellus* **9**:18–27.

SUESS, H. E. 1953. Natural radiocarbon and the rate of exchange of carbon dioxide between the atmosphere and the sea. Nuclear Processes in Geologic Settings. *National Academy of Sciences — National Research Council Publication,* pp. 52–56.

SUESS, H. E. 1954. Natural radiocarbon measurements by acetylene counting. *Science* **120**:5–7.

1955. Radiocarbon concentration in modern wood. *Science* **122**:415–417.

VON BUTTLAR, H., and W. F. LIBBY. 1955. Natural distribution of cosmic-ray produced tritium. 2. *J. Inorganic and Nuclear Chem.* **1**:75–91.

CHAPTER 12

ON THE TAGGING OF WATER MASSES FOR THE STUDY OF PHYSICAL PROCESSES IN THE OCEANS [1]

THEODORE R. FOLSOM, *Scripps Institution of Oceanography,* La Jolla, California

and

ALLYN C. VINE, *Woods Hole Oceanographic Institution,* Woods Hole, Massachusetts

FINDING, identifying, and plotting the courses of characteristic masses of water in the oceans are major activities of the physical oceanographer. Assistance from the new techniques that have come with the use of radioactive materials has been welcomed by him; some of these techniques have already been put into service for tracing the water. It is not generally realized how much experience has been gained, beginning with the 1946 tests in Bikini Lagoon, in tracing water masses contaminated with radioactive materials from weapons' tests. And many thoughts are now turning toward the radioactive tagging of ocean water by other means in regions where knowledge of underlying physical processes are meager, especially in the very deep waters. Proposals for the disposal in the sea of atomic energy wastes cannot be properly evaluated until estimates can be improved concerning the motion of this deep water.

Many of the advantages (familiar in the laboratory) of using radioactive identifying tags can be realized at sea, even though rendered difficult by the very large physical dimensions of the oceans. What appeals most to the oceanographer is his new ability, under certain circumstances, to make very rapid identifications of water lying on the surface or deep below his ship; thus allowing large volumes to be surveyed in three dimensions and in more detail than ever before possible.

Three layers in the ocean are distinguished clearly by structure and behavior: the mixed layer near the surface; the intermediate layer lying just below wherein the temperature changes rapidly with depth (this thermal stratification bringing about great stability); and finally, the large, nearly uniform bottom water mass extending to the sea floor with so little variation in density that very stable stratification is not possible. Each water mass reacts differently when disturbed, and therefore, mixing occurs differently in response to currents and mass intrusions. Experiments conducted in any one of these domains must take into consideration the special features that exist, and must call upon equipment most suited to these sections. Equipment specialized for radiological survey work in any of these oceanographic domains is still primitive. However, it can be said that equipment for detecting and measuring radiation is not a bit less highly developed than are the equipment and techniques needed for navigating a ship, and for maneuvering detectors at sea, especially at great depth. So much information now can be reported by radiological means in a short time that a ship now has more reason than heretofore for precise navigation. In some cases, the depth and position of the detector relative to the ship must be known instantly, and almost always must be controlled far better than has been accepted by traditional hydrography. It is difficult to record data in full detail in many cases, and in others it is difficult to evaluate features rapidly enough to alter maneuvers to best advantage; an oceanographer can now expect to be aware of a strongly active layer in less than one second after his electronic probe makes contact, and he even may make use of a fast moving airplane to outline radioactive areas on the surface.

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 905.

Contribution No. 929 from Woods Hole Oceanographic Institution. Part of Table 2 was computed with the collaboration of John Harley of AEC, New York City Operations Office, who gave much other counsel for which the authors are grateful.

Instrument Sensitivity and Natural Backgrounds

Many promising measuring schemes have been proposed. However, it is profitable to compare the equipment and techniques which have been used already at sea. At the top of Table 2 is presented the background radiations coming from cosmic rays and from the natural potassium in the sea, and the activity level now believed tolerable for drinking water also is given for comparison.

At the bottom of Table 2 are listed, in the brief numerical form in which they are commonly stated, the sensitivities of three measuring techniques which actually have been used for radiological exploration at sea. Many aspects of the measurement problem are oversimplified by a comparison of this sort, but the table does indicate that present shipboard beta analysis is capable of measuring beta tracer activity below the background beta activity due to the potassium in the sea water, whereas gamma detectors so far have been limited at levels above the gamma backgrounds of the sea. On surveys covering large distances, such as on Operation TROLL (U. S. Atomic Energy Commission 1956), and on the SHUNKOTU MARU Expedition (Mujoke, Sugiura, and Kameda 1954), there is ample time for water analyses, and advantage can be made of beta techniques. Nevertheless, there are many circumstances where direct measurements by gamma devices are necessary for rapidly locating small contaminated water masses, and it is likely that gamma techniques will be perfected so as to allow use at levels far below their present capability.

There are occasions at sea in which a gamma detector must indicate the presence of tracer activity within a few seconds after making contact. The limitations imposed by this sort of time restriction in the presence of statistical fluctuations in the signals are discussed in Appendix A, and are summarized in Table 3.

Other important details concerning the radioactive background in the sea have not been thoroughly explored. It may be too late to estimate the background level that existed a decade ago for some isotopes, and this should not be forgotten in planning future surveys. Of particular interest are background conditions near the sea floor where radium and thorium activity are known to accumulate in sediments; but little is known in detail about the lateral distribution of bottom activity.

More complete utilization of weapons' tests for the marine sciences

It appears likely that large weapons will continue to be tested in oceanic areas and that radioactive materials will be strewn from time to time over the surface of the sea. Valuable oceanographic data already has come from such sources; for example, direct measurements have been made of the rate of mixing downward from the surface to the thermocline, and also, direct information has been obtained regarding mass motion and lateral mixing. One special feature of benefit in studying weapons tests is the unique initial boundary condition provided by the arrival of fallout activity almost simultaneously over an area having dimensions very large compared with the depth of water involved; downward mixing appears as a relatively simple phenomena following this initial condition, and can be studied under almost ideal circumstances.

Two expeditions mentioned above have proven that further information concerning lateral mixing and flow can be gained for many months after a weapons' test, and obviously this fact should be exploited fully by marine scientists of all nations. Ancillary benefits might come from more or less fixed monitoring stations; if, for example, following the 1954 test, repeated sampling had been done off Guam it would have furnished data of value for interpolating observations made in the two follow-up cruises mentioned.

Bottom exploration following weapons' tests has not been given deserved attention, and insufficient attention has been paid to getting even purely oceanographic information from these sources into the form needed by those people who are charged with making decisions regarding the ominous waste disposal problem.

Hazards involved in the deliberate tagging of ocean waters

Safety of the research staff is always a consideration; at sea because of special circumstances the handling of extremely large amounts of activity is not too difficult or hazardous. Protection can be secured very cheaply by towing the larger sources of radioactivity aft of the

ship, preferably slightly submerged on a suitable barge or special vessel. Bringing large quantities of activity to the waterfront promises to be more expensive, but practical experience in this should be valuable for later planning of large-scale disposals.

The more controversial question of how much radioactivity can safely be introduced into the sea is not without reasonable solutions; but the recommendations depend upon the circumstances, especially, upon the particular part of the oceans to be studied. At the outset, barren areas of ocean rather than those productive of things leading to human food must be selected since the former can yield equally good information regarding purely physical phenomena.

Deliberate tagging of surface waters (Operation PORK CHOP)

Surface waters mix in a turbulent manner due to forces not yet fully understood. Better knowledge of this layer is badly needed justifying the consideration of water tracing experiments involving introduction of fairly large amounts of activity. Greatest care must be exercised here because these waters are those most close to humans, in several senses.

Rate of mixing to the bottom of the mixed layer, and rate and character of lateral motion as functions of the usual parameters of the sea are of most immediate interest, and observations lasting even a few days or few weeks would be of great value at the outset, especially if repeated frequently. A simple surface water experiment now will be proposed in briefest possible outline.

Figure 1 presents schematically some of the procedure which might be used and some of the phenomena to be expected. Guided by suitable navigational aids, here represented by deep-anchored buoys No. 1 and No. 2, the ship A proceeds on a straight course while dropping two quantities of radioactive materials (a and a′) mixed with enough surface water to leave near the surface a small contaminated patch having nearly neutral buoyance. These are essentially point-source initial conditions in this scale of dimensions; although, they are not as convenient as the plane-source initial conditions

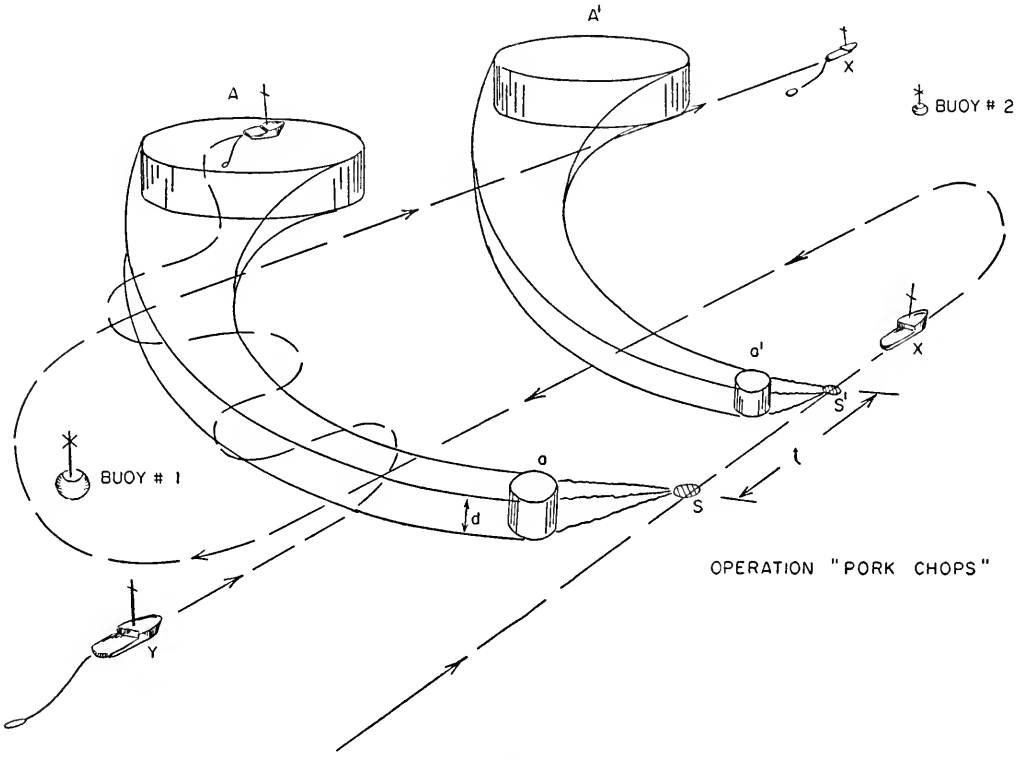


FIGURE 1

provided by fallout, they have some mathematical simplicity. It would appear economical and informative to drop two sources almost simultaneously, some distance apart — say one to ten kilometers; this would permit large-scale advection also to be studied at little extra ship cost.

From the sources s and s′ will grow a larger more dilute patch of water finally ceasing to penetrate rapidly downward at depth d. The rate and lateral spread prior to this time as functions of wind velocity are of special interest. After further downward penetration is retarded, the areas a and a′ move and expand to the larger areas A and A′ conserving most of the original radioactive material, and the product of activity and area should be almost constant after correction is made for the known rates of decay of radioactive constituents.

Dual ship operations

Experience has shown that operations on the scale of this sort can scarcely hope to be successful unless more than one ship is used; even with the best facilities one ship may lose contact with the invisible patch and waste valuable time locating it. One ship, X, must stay in or near the tagged mass while the other one, Y, may survey the area in detail, inspecting sections across the mass, studying the bottom for reference features, and chasing missing buoys if necessary.

Ultimate disposal of hazard in surface waters

Reduction of activity to a level below that of the natural activity of sea water is one criterion which has been used for planning disposals (Glueckoff 1955), and this is fairly reassuring provided the specifically dangerous and the long-lived activities are eliminated, for example, after radiostrontium and radiocesium are removed from raw fission wastes. Present evidence permits the conclusion that in the open ocean, when winds are above the critical white-cap level and under circumstances where mixing ceases at a depth of about 30 to 50 meters, as much as 1,000 curies would mix to a safe dilution in less than 40 days. An example of the dispersal rate in the open sea will now be given.

Brief outcome of an experimental tagging of surface waters in the open sea

Surface water made active by introducing fission products concentrated within a few square kilometers was intercepted by a ship 36 days after inoculation and traversed for 10 days. After corrections were made for the drift of the water during the survey, and for radioactive decay, a synoptic picture could be drawn roughly locating the contours of activity. This estimate of radioactive distribution was referred to the time of 40 days after the start of dispersal.

The contamination had mixed significantly only to about 30 to 60 meters, although the thermocline lay nearer to 100 meters depth. The following tabular description of this synoptic sketch can be made.

TABLE 1 Approximate Distribution of Radioactivity Found in the Surface Waters of the Open Seas 40 Days After Being Introduced Suddenly as a "Point Source." (A Synoptic Picture Computed from Measurements Made on Several Different Days.)

Areas inside contours of equal concentration in square kilometers.	Areas inside the contours as percentages of the area of the maximum contour.	Concentration of radioactivity (as per cent of the maximum concentration measured).
40,000 (km^2)	100%	10%
24,000	65	20
14,000	38	30
8,000	22	40
800	2	60
490	1	80
35	0.1	100

At the end of 40 days, the center of gravity of this distribution was about 120 miles from the point of inoculation and the pattern was about four times longer than broad. The wind was 3 and 4 of Beaufort's scale for the first 20 days, but was much calmer for the last 20 days.

If the average mixing depths are taken as 50 meters, then, 1,000 curies distributed over 40,000 square kilometers would result in an average concentration of $1.5 \times 10^{-10}/\mu c/ml$. This would certainly be safe sea water in most senses; and even in the smaller areas where much less than the average dispersal took place the water should also be safe. In fact, the experiment indicated that it is likely that after 40 days, following the introduction of 1,000

curies of activity into the surface waters of the open sea, only about 0.1 per cent of the total area should retain contamination above the tolerance concentration permitted for potable water, and even in this small region the residual artificial activity would amount to less than the normal natural activity of sea water.

It is evident from Table 2 and Table 3 that shipboard beta measurements would suffice to detect the more radioactive spots if there were initially 1,000 curies of slowly decaying beta activity; it is apparent, however, that direct measurements by gamma detectors might be sufficient for several days or even weeks. Surface experiments are by far the easiest to conduct and implement — they are limited largely by considerations of hazard to humans. Two more difficult experiments will now be described.

Investigations in the thermocline layer by use of radioactivity

The thermocline lying between perhaps 100 meters depth on an average, and 800 meters or more, can be thought of as being a lid which restrains deeper water from reaching the surface. Experiments in this stable region must take into consideration the fact that any liquid introduced here will seek the level of its own density and will then spread out in a very thin layer. An experiment in this layer has been

TABLE 2 APPROXIMATE SENSITIVITIES OF THREE DETECTING AND MEASURING TECHNIQUES PRESENTLY AVAILABLE FOR USE AT SEA COMPARED WITH THE ACTIVITY OF SEA WATER AND WITH THAT OF FRESH WATER.

	d/m/l	curies/l	microcuries/ml	rad/hr [2]	mrad/yr [3]
A *Common background radiation levels:*					
Activity in normal sea water due to potassium: [1]					
Gamma rays 70	3	$\times 10^{-11}$	3×10^{-8}	1×10^{-7}	0.9
Beta rays 660	3.0	$\times 10^{-10}$	3.0×10^{-7}	—	—
Maximum permissible [4] *concentration of unknown mixed beta activities in drinking water:*					
Beta rays 220	11	$\times 10^{-10}$	1×10^{-7}	—	—
Cosmic ray background at sea surface: [5]					
At equator 61	—		—	—	33
At 55°N (mag)	—		—	—	37
B *Sea water activities at which present measurements are significant.*					
Shipboard water analysis [6] *for mixed beta emitters, 60 minutes count after removal of potassium:*					
50 ± 15	2	$\times 10^{-11}$	2×10^{-8}	—	—
Underwater gamma detector, [7] *1956 scintillation rate-meter of AEC-NYOO:*					
220 (approx) (0.6 MEV gammas assumed)	—		—	1.4×10^{-7}	1.2
Underwater gamma detector, [8] *1955 geiger instruments of SIO. (counting pulses):* (See also table 3 for other cases)					
Case A: Used in deep water where net background is 15 CPM, assume photons of 0.6 Mev; assume short measurements required, t = 5 sec.					
6600	3	$\times 10^{-9}$	3×10^{-6}	3.8×10^{-6}	30
Case B: Towed on surface, assume constant background 60 CPM, assume photons of 0.6 Mev; assume long measurements permitted, t = 5 min.					
520	2	$\times 10^{-10}$	2×10^{-7}	0.3×10^{-6}	3

[1] Assuming normal sea water has 3.8×10^{-4} gk/g sea water, that beta activity is 29 d/s/gk and that gamma activity is 3 d/s/gk.

[2] The rad unit is somewhat larger than the more familiar roentgen unit; 1 rad = 1.1 roentgen approximately for gamma rays. Values in this column were computed upon the assumption that the activity was uniformly distributed in the water and that the detector was a meter or more from any boundary.

[3] Referring to beta ray activity in rad units in roentgen units is a dangerous practice—much further specification depending upon the individual experiment is required.

[4] Handbook 52 of the National Bureau of Standards. The values given refer to the case where the nature of the activity is unknown; certain radioisotopes can be tolerated at much higher levels.

[5] See Table 1 in the accompanying paper "Comparisons of Some Natural Radiations Received by Selected Organisms" by T. R. Folsom, and John H. Harley for variation of cosmic rays with depth and altitude.

[6] Cosmic rays are counted by most geiger counters at the average rate of approximately one count/min/sq cm of counter area.

[7] This information was supplied by J. H. Harley from personal communication with H. D. LeVine of the New York City Operations Office of the Atomic Energy Commission who designed this equipment.

[8] This detector was not intended previously for use at low intensities, but rather for measuring a wide range of intensities of gamma rays. Additional geiger tubes might easily be added to increase the sensitivity by at least five fold. Still more sensitive gamma devices are now used in oil well logging.

TABLE 3 COMPARISON OF MINIMUM DETECTABLE CONCENTRATIONS USING SEVERAL MEASURING TIMES AND ASSUMING SEVERAL BACKGROUNDS

(a) Minimum detectable anomolous activity if potassium of the sea produced the only background, i.e., $B = 1.2 \times 10^{-3}$ gammas/sec/ml.

Counting time in secs. t	Minimum detectable concentration γ/sec/ml	$= \gamma$/min/l	Net signal counts/min $C_aVe = 30C_a$	Total net counts $30C_at$	Rads/hour Photons .6 mev	Rads/hour Photons 1.5 mev
	C_a					
3	.19	11,000	5.7	17	6.5×10^{-6}	16×10^{-6}
5	.11	6,600	3.3	17	3.8	9.5
60	.010	600	0.3	18	.3	.8
180	.0039	230	.12	22	.13	.33
300	.0026	160	.078	23	.09	.22
600	.0016	99	.048	30	.06	.14
Very large	$0.025/\sqrt{t}$					

(b) Minimum concentration detectable if backround were 15 CPM, i.e., an actual background signal experienced in deep water.

	C_b					
3	.19	11,000	5.7	17	6.5×10^{-6}	16×10^{-6}
5	.11	6,600	3.3	17	3.8	9.5
60	.010	590	.29	17	.33	.84
180	.0058	350	.17	32	.20	.50
300	.0049	290	.15	45	.17	.42
600	.0032	190	.096	58	.11	.28
Very large	$0.067/\sqrt{t}$					

(c) Minimum detectable concentration if total background were 60 CPM, i.e., an actual background signal experienced at the sea surface.

	C_c					
3	.205	12,000	6.1	18	7.1×10^{-6}	18×10^{-6}
5	.133	8,000	4.0	20	4.6	12
60	.0222	1,330	.67	40	1.9	7.5
180	.0116	700	.35	63	.4	1.0
300	.0087	520	.26	78	.3	.74
600	.0059	354	.17	102	.2	.51
Very large	$0.13/\sqrt{t}$					

described in some detail by Revelle, Folsom, Goldberg, and Isaacs (1955), and discussed in several of the accompanying papers. It will be discussed here only in the matter of difficulty of survey. Although mixing is known to be very slow in the thermocline, it is not certain how direct is the path from this *fringe* biosphere to human food supply, so that the hazard of a long remaining concentration of activity is not easily evaluated. Revelle et al., prefer to suggest the experimental use of the conservative amounts of 10 to 100 curies, and they then show that such small sources of radioactivity might be practical none the less.

Actual field experience has shown that layers as thin as one or two meters thick are extremely difficult to sample for water analyses even after being located by gamma ray detectors. Folsom (1956) has emphasized that future deep surveys with radioactive tags must rely heavily upon discovery of radioactive water by means of gamma detectors, and has urged that specialized forms of these be brought to perfection.

In this particular layer, geometric factors are not adverse for maneuvering a detector into the water mass to be studied; a probe is dropped rapidly and more or less vertically so as to intersect and pierce a rather broad horizontal lamina, sharply confirming the activity. Some difficulty would be encountered in holding the probe in the thin layer long enough to permit accurate measurements after the activity falls to such a low level that statistical fluctuation becomes the predominant source of error; however, the major difficulty even at these depths is holding the ship in the general area of active pools of small size. Any area of less than a square mile below the surface is a tiny detail in the open sea, and oceanographers never before have realized how hard it is to navigate and maneuver to study areas so small. Multiship operation, the use of the best position-

locating gear, and careful crew training and teamwork are necessary for subsurface radiological surveys even at these moderate depths.

Outline of tagging experiment in the thermocline layer

Figure 2 illustrates certain features which must be considered in this region. The ship, A, may lower a gamma sonde through an activated pool and detect its presence by the receiving of a signal like that shown on the right side of the figure; the hydrographer may obtain a water sample by triggering electrically a water sampler at the moment the detector indicates that the sampler is within the active layer. The data in Table 3 make it clear that rapid response is important during this sort of measurement; a statistically significant signal must be accumulated in the short period during which the probe is passing through the active layer.

Attention is called to the need for navigational and maneuvering aids here by including schematically the parachute-drogue C. It is difficult to maneuver a weighted detector horizontally in order to study the lateral distribution in detail. The use is suggested of towed gamma detectors depressed to the desired level by hydrofoils controllable from the surface, more or less as illustrated schematically at the left of Figure 2. By means of a swivel-clamp, SC, a pennant several meters long containing a row of Geiger tubes or other gamma detectors, might be suspended above the depressor so as to present a vertical, linear array, thus giving a high probability of intersecting wide lateral distributions of activity. This sort of gear should not be too awkward nor fragile for deck handling at sea. Signals might be recorded partially, or entirely inside the depressor, or reported to the ship electronically or sonically.

Ship A or a sister ship with similar gear might stay in the pool during the whole experiment, however, if the pool were lost after its depth was established, then Ship B would likely be the first to find it again with its towed detector.

Difficulties in sounding and exploring very deep waters

Bottom exploration so far has been confined largely to sonic plotting and sounding by solid cable; very deep wire casts are very time consuming and difficult; the ship generally is moved laterally by surface currents before the

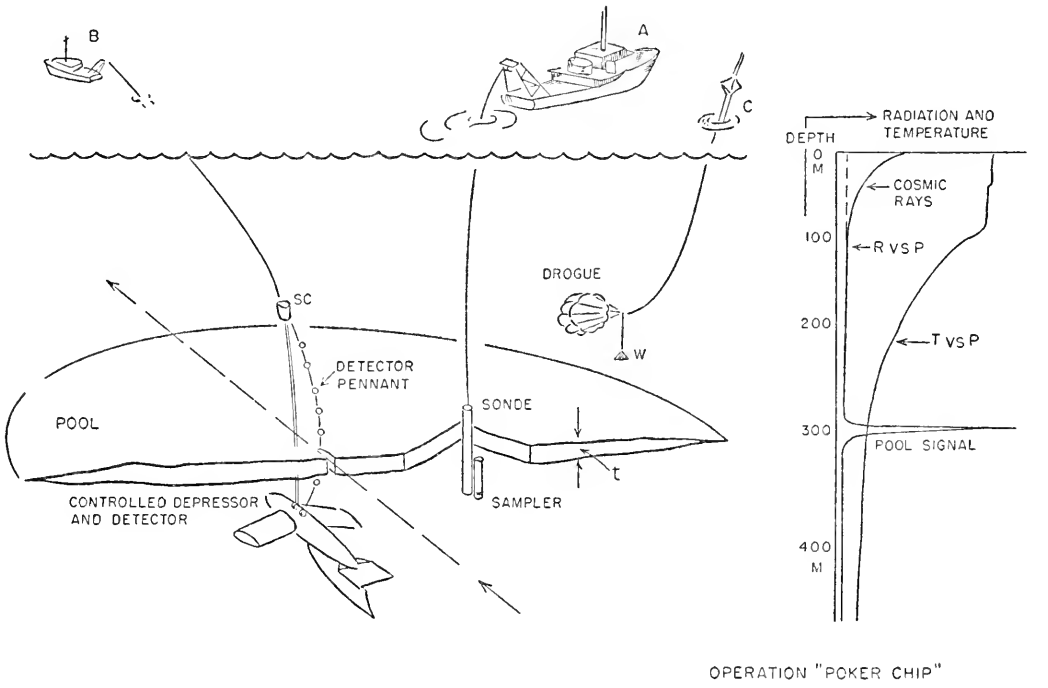


FIGURE 2

wire touches bottom. Oceanographers seldom hope to place their sondes and coring tools upon any pre-selected topographic detail of small area. However, it is quite likely that a technique can be perfected for dragging a detecting instrument along the bottom in many areas of the oceans' floor, and with a dragged detector a large region might be traversed rapidly, and tagged water masses near the sea floor might be located and surveyed. A proposal for tagging bottom waters now will be outlined.

Difficulties in tagging bottom waters

Fortunately, little hazard to human populations would result from putting into the deep bottom waters in certain latitudes almost any amount of activity which might be readily available in the near future, or which would be easy to handle safely ashore and on ordinary surface vessels. After all, these amounts would be only the feeble forerunners of what may have to follow.

The problem is that of displaying even a relatively large radioactive source economically in face of the immensity of the abyssal reaches. One can think of many things which must not be done; heavy, radioactive liquid cannot be merely poured overboard, for example. Matching density at intermediate layers or attempting to insert a strata at a selected depth also would appear experimentally difficult in view of the limited knowledge presently available; an unequilibrated liquid mass might wander about like a sinking dinnerplate — and soon become lost. In the absence of the restraining forces found in more stable waters, the pouring of streams of dense solution downward from a height above the bottom, or alternately the releasing of lighter material upward from the bottom would surely cause mass motion which might not cease until the streams had moved long distances and perhaps had curled into configurations quite unsuited as initial boundary conditions for water tracing experiments. Furthermore, activity spread initially in more or less vertical lines would make very poor targets for detectors trailing on the end of wires three miles long, and would be wasteful in terms of radioactive material and of expedition time.

One might, of course, carefully select a perfect basin, and might gently introduce into it a dense radioactive solution. This certainly should be considered since only a small amount of activity might suffice for tagging the waters in a small basin and valuable information regarding motion and dispersion in basins might result, but results would not lead to a realistic picture of the large scale flow over bottom which may have to disperse the wastes dumped in the future. The results of an experiment set up in this way would be inadequate, and, in fact, might be misleading in a dangerous direction.

Production and use of horizontal line-sources near the bottom

"Operation HARE and HOUND"

It is evident that distribution of activity in a horizontal line near the bottom would be most easy to intercept by a detector dragged along the bottom, and it appears also to be something which would be relatively easy to produce, and economical. It should be possible to hold tagged water near the bottom by mixing it with a very dense solution; and there are two ways immediately evident for effectively spreading streaks of dense solution for long distances over the bottom terrain.

Figure 3 illustrates the two methods proposed for tagging bottom water, and the method proposed for locating the tagged masses later. The Ship B' is shown dragging a "Hare" D, across the bottom leaving behind a streak of contaminated water. Alternately, Ship B is shown just after it has dropped to the sea floor a specialized water blending device which might well be called a "quern" [1], C, which generates for a few minutes or hours, a stream of dense, radioactive solution on the slope of a carefully selected large topographical ridge b — d; this stream flows away very much like one of the submarine currents which are now called "turbidity currents" by geologists. Violence of this sort of free current might theoretically be controlled through wide limits by adjusting the densities of the solution. The essential features of a water-tagging quern are shown in the upper right of Figure 3. Radioactive material, AS, is combined in predetermined proportions with a heavy salt solution by metering pump, P, and the two are then fed to a fan-type mixer, and are there blended with a large volume of

[1] Old English name for a mill for grinding all sorts of things. (Ruggoff, 1949.)

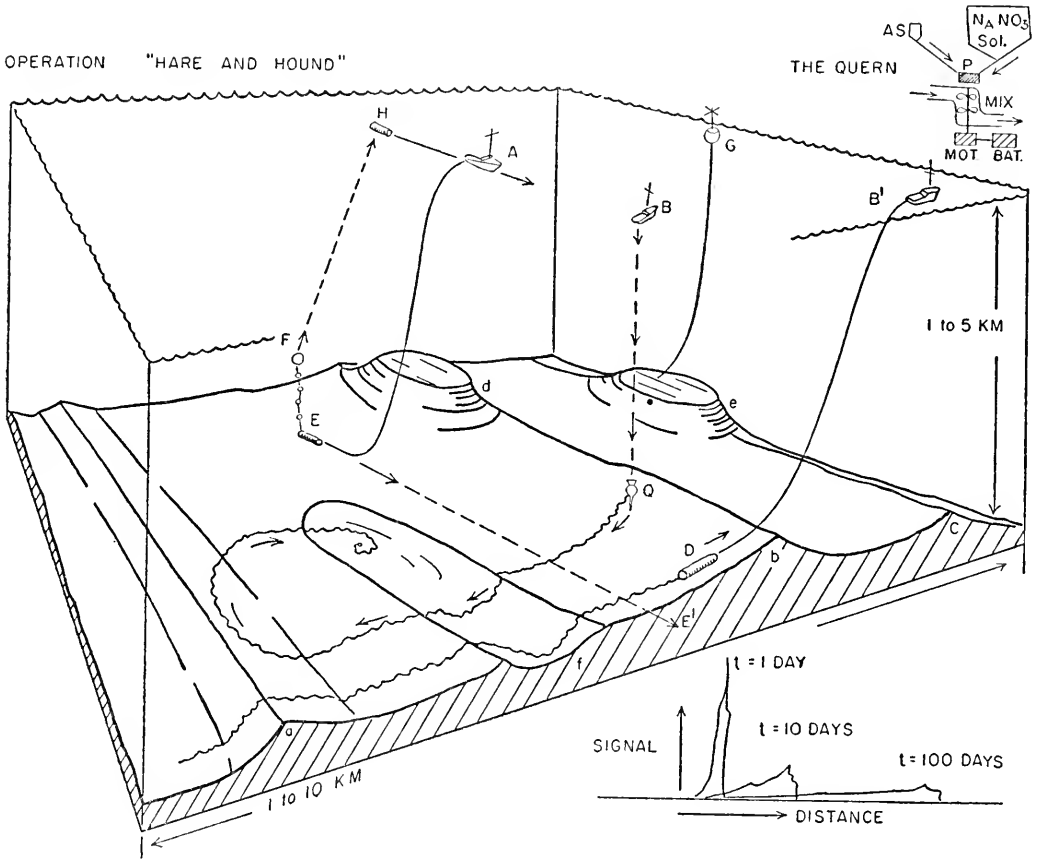


FIGURE 3

local water. There are several reasons for preferring a design leading to inexpensive construction and single use; the cost of decontamination of apparatus of this type would outweigh any benefit from repeated use. Suggestion is made of the use of a salt such as sodium nitrate which has both high solubility, and an endothermic heat of solution which would serve to overcome the adiabatic heat set free during lowering. It would appear that one or more tons of a nitrate salt, mixed into bottom water by use of a few kilowatt hours of energy, stored in oil-sealed accumulators, could produce a compact body of very heavy water which would rush like a freight train across the terrain dropping a streak of traceable radioactive eddies as it traveled.

A fixed, water-mixing quern, of the sort described, might produce a tagged water mass behaving in a manner appearing realistic to both the disposal planner and the submarine geologist; however, its use is not likely to lead directly to the extremely simple results needed for the very first experiments. The employment of a dragged hare might be preferable at the outset — and its metering machinery might be somewhat less elaborate than that of the quern just described.

One might contemplate using 1,000 or more curies for making streaks several kilometers long so that location would not be difficult with a simple gamma device dragged by a ship. In Figure 3, Ship A is shown dragging such a detector which might be called a "hound" for obvious reasons. For very great depths, no electrical wire is presently available with the durability equal to that of an ordinary dredging cable. It would, therefore, be wise to first consider the use of a compact multichannel chart recorder inside the dragged pressure shell E so as to make permanent records of signals picked up by a set of gamma detectors suspended by an

oil-filled float F. Numerous accessories might profitably ornament this sort of gear, but the one which might prove most rewarding would be a sound producer capable of reporting the moment of contact with the tagged water mass; even a crude sonic signal sent from a transducer on the float, F to the ship, A, via the towed hydrophone, H, would suffice. Details of the gamma signals need only be recorded so that they might be inspected later on the recorder chart, however, it would be important for the navigator to recognize instantly when contact was made so that he could maneuver the ship economically.

The operations proposed above are not unlike those used successfully by cable ships when retrieving submarine wires. Careful preliminary surveys of the whole area, the selection of identifying landmarks, and the laying of the marking buoys also appear essential for success in work of this type.

The final results might have the general character of the hypothetical signals shown graphically at lower right in Figure 3. Change in amplitude and displacement, and skewness of the signal records should lead to estimates of both velocity and rate of mixing. If each survey included ten or more intersectings, and if each contact brought separate gamma signals from several detectors distributed along the hound's vertical "tail," then the data of the sort needed would accumulate quickly.

Rough estimate of effectiveness of 1,000 curies for tagging bottom waters

It appears possible to distribute radioactivity uniformly along the course of a device dragged over the sea bottom, and it would appear possible also to deposit the material so gently that it would come to rest within a few meters of the precise course. If, for a rough evaluation, we assume that local diffusion sooner or later produced a uniform distribution within a radius of 10 meters, and that the total activity, M, was 1,000 curies, then the length of the water mass which might be tagged can be stated

$$l = \frac{M}{C\pi r^2}. \qquad (1)$$

where C is the average concentration of activity within the tagged mass.

If now we assume that only 10 seconds can be allotted for traversing 20 meters (that is the ship's speed is about 4 knots), then the equation (9) of Appendix A indicates that a single detector like the 1955 SIO Geiger instrument could detect, in the presence of a realistic deep-water background of 15 cps, a limiting gamma source concentration of 0.061 disintegrations/sec/ml, or $C = 0.061/3.7 \times 10^{-10}$ curies/ml, and the length of traverse which could be tagged with 1,000 curies would be, under these assumptions,

$$l = \frac{1000}{1.65 \times 10^{-12}\pi (1000)^2} = 1900 \text{ Km} \qquad (2)$$

It would appear feasible to locate and allocate by ordinary navigational means a geographical line in the deep sea floor of less than two kilometer's length, so that the hypothetical example just given suggests that 1,000 curies could equally well be used to produce a very concentrated streak of activity having a length of two or three kilometers which might still be detected with ease after it had diffused, mixed, or decayed to less than one percent of its initial concentration. Thus it can be concluded that 1,000 curies, or even less activity, put into bottom water would be quite adequate for tracing movements on a scale large enough to contribute information useful in disposal planning.

SUMMARY AND CONCLUSIONS

1. Consideration has been given some of the problems involved in tagging water masses in the open ocean.
2. The problems are different in the three major strata; the surface layers, the thermocline, and the deep water layer.
3. It appears that under certain circumstances water tagged with even moderate quantities of activity can be followed for at least several weeks; surface waters contaminated by large activities such as result from fallout can certainly be followed for a year or more.
4. Much field experience in radiological oceanography has been gained already. A fairly clear direction for development of instruments has been indicated.
5. The need is seen for attention to the perfection of navigational aids, for use of specialized vessels and gear, and for the use of several vessels simultaneously in oceanic surveys of this sort.

APPENDIX A

In practice, many factors tend to limit the effectiveness of an under sea gamma detector, but the random fluctuation of a feeble radiation may alone prevent its recognition in the presence of a background of similar magnitude. The lowest detectable concentration, limited only by statistical considerations, may be expressed in terms of the strength of the background, the time permitted for measurement, and the measuring efficiency of the instrument.

Let the sea water be contaminated with a concentration of radioactivity N curies/ml, and let this activity cause m counts/sec to be indicated by the instrument, and let the average background be b counts/sec. The relative accuracy, n, of a single measurement made during t seconds will depend upon signal strength and background strength; if the fluctuations are purely random, the error, 95 per cent of the time will be equal to, or less than,

$$n=\frac{2\sigma}{mt}=\frac{2\sqrt{\sigma_M^2+\sigma_B^2}}{mt}=\frac{2\sqrt{mt+bt}}{mt} \qquad \text{A.1}$$

and solving for the net signal gives,

$$mt=\frac{2+2\sqrt{1+n^2bt}}{n^2}. \qquad \text{A.2}$$

Now, the counting efficiency of the instrument logically should be derived from the ratio of counts recorded to the photons striking the instrument. This ratio would be impossible to evaluate, but it is approximated when the instrument is small, and easily penetrated by,

$$e=\frac{mt}{3.7\times10^{10}Nvt} \qquad \text{A.3}$$

that is by the ratio of the net counts recorded to the photons emitted in a volume of liquid, v, equal to that displaced by the detector. Solving this equation for concentration,

$$N=\frac{mt}{3.7\times10^{10}vet} \qquad \text{A.4}$$

curies/ml, and substituting here the value for net count, mt, obtained in equation (2) when the background rate is b, and accuracy is, n, the limiting concentration can be expressed,

$$N=\frac{2+2\sqrt{1+n^2bt}}{3.7\times10^{10}n^2vet} \qquad \text{A.5}$$

curies/ml, wherein b expresses the background rate actually indicated when the instrument is surrounded by clean sea water. If no other background exists except that coming from a surrounding solution having specific activity B, and if the instrument counts this activity with the same efficiency, e, than the limiting detectable concentration becomes, in curies/ml,

$$N=\frac{2+2\sqrt{1+Bn^2vet}}{3.7\times10^{10}n^2vet}. \qquad \text{A.6}$$

Numerical examples applying to an actual undersea instrument

The sensitive portion of the 1955 model of the Scripps Institution of Oceanography's Geiger instrument has a volume of about 1,000 ml. The ratio e, applying to hard gamma rays, was measured directly by submerging the instrument in a tank containing potassium solution of known concentration, and was found to be approximately 0.03.

If by "detection" is meant the measurement of the concentration with an error of not more than 50 per cent, then, $n=0.5$.

Formulas (5) and (6) may now be applied to three characteristic background circumstances:

Case 1: Here no other background is evident except that caused by a solution having specific activity $B=1.2\times10^{-3}$ gammas/sec/ml such as comes from the natural potassium in normal sea water. From (6), the limiting detectable concentration,

$$C_1=\frac{2+2\sqrt{1+0.009t}}{7.5t} \qquad \text{A.7}$$

gammas/sec/ml, and when t becomes very large this approaches,

$$C_1=\frac{0.035}{\sqrt{t}} \qquad \text{A.8}$$

Case 2: In deep water cosmic rays may be neglected, and the S. I. O. probe is likely to indicate a total background of about 15 CPM, or $b=0.25$ counts/sec, therefore, the concentration just delectable is,

$$C_2=\frac{2+2\sqrt{1+0.063t}}{\sqrt{t}} \qquad \text{A.9}$$

gammas/sec/ml, which approaches as t increases to a large value,

$$C_2=\frac{0.067}{\sqrt{t}} \qquad \text{A.10}$$

Case 3: In shallow water where cosmic rays are unattenuated, the background on the S. I. O. probe amounts to about 60 CPM, or $b=1.0$

counts/sec, therefore the minimum detectable concentration becomes,

$$C_3 = \frac{2+2\sqrt{1+0.25t}}{7.5t} \qquad \text{A.11}$$

gammas/sec/ml which approaches for very large values of t,

$$C_3 = \frac{0.13}{\sqrt{t}}. \qquad \text{A.12}$$

Tabulations

Table 3 compares the effect of increasing the period of measurement with the effect of diminishing the background. It is evident that a substantial change in background has relatively small practical effect on any measurement made so rapidly that only a very poor sample is taken out of the fluctuating signal; however, when sufficient time can be alloted for good sampling, the background level becomes the limiting factor. It should not be overlooked that in practical field work, instrument imperfections may contribute to the overall error more or less proportionally with time of measurement, and that measurement time must be spent economically on almost all oceanographic expeditions. It is apparent therefore that efforts should be made towards increasing the counting rate, ve, while reducing the relative value of the background count by all possible means. Technique for cleanliness and for discrimination of background by electronic means have not yet been fully developed for this purpose.

REFERENCES

FOLSOM, THEODORE R. 1956. Problems peculiar to direct radiological measurements at sea. Paper presented at Nat. Acad. of Science Meeting, 29 Feb.-1 Mar. 1956. Washington, D. C. Proceedings (in press).

GLUECKOFF. 1955. Long term aspects of fission product disposal. United Nations Conference on the Peaceful Uses of Atomic Energy, Geneva. Paper No. 398: 11 pp.

MIYAKE, Y., Y. SUGIURA, and K. KAMEDA. 1954. On the distribution of radioactivity in the sea around Bikini Atoll in June 1954. Paper in meteor and geophys., Meteorol. Research Institute, Tokyo, **5**:253–262.

REVELLE, R. R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear science and oceanography. United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva. Paper No. **277**:22 pp.

RUGGOFF, MILTON D. (Editor) Why the sea is salt (an abstract from a translation from the Norse by Sir George Weble) pp 672–676 in Harvest of World Folk Tales. XViii + 734 pages, Viking Press.

U. S. ATOMIC ENERGY COMMISSION and OFFICE OF NAVAL RESEARCH. 1956. Operation TROLL. Health and Safety Laboratory, U.S.A.E.C., New York Operations Office, NYO-4656, Ed. by J. H. Harley: 37 pp.

U. S. DEPARTMENT OF COMMERCE. 1953. Maximum permissible amounts of radioisotopes in the human body, and maximum permissible concentrations in air and water. National Bureau of Standard Handbook 52:445 pp.

CHAPTER 13

LARGE-SCALE BIOLOGICAL EXPERIMENTS USING RADIOACTIVE TRACERS[1]

MILNER B. SCHAEFER, *Inter-American Tropical Tuna Commission, Scripps Institution of Oceanography,* La Jolla, California

ONE OF the major difficulties in evaluating the probable results of the introduction of radioactive materials into the sea is the lack of adequate knowledge respecting the effects of the organisms in the sea on the distribution and transport of such materials. Some information, which has been reviewed in earlier sections of this report, has been obtained on the uptake and excretion of elements by different kinds of marine organisms. This information is, however, not sufficiently extensive. The even more important problems of the quantitative interrelationships and movements of the populations of organisms at the several trophic levels are among the least understood biological phenomena of the oceans. These, together with physical factors, will determine the fluxes of the radioactive materials.

Measurements of the fluxes of materials through physical-biological systems, or ecosystems in the sea are of vast and fundamental importance not only for evaluating the probable distribution of radio-active products introduced into the sea, but also as a basis of evaluating the sea as a source of food and other biological products for the use of mankind. With the approaching full utilization of the land, increasing attention is being directed to the sea as a source of such products, but the basic biological knowledge for realistic evaluation of the potential harvest of the sea is quite inadequate.

The availability of rather large quantities of radioactive materials, as by-products of the development and utilization of nuclear energy, makes possible the study, *in situ,* of the biological and ecological processes in the sea by the use of tracer techniques. A start has been made, in connection with the introduction of radioisotopes into the marine and fresh waters by weapons tests and by the disposal of low-level wastes, but the opportunities for obtaining useful information by these means have not been fully exploited. Also it should be possible by introducing radioisotopes in a planned, controlled, and purposive fashion to obtain even better information than is possible through observation of introductions ancillary to operations having a different primary purpose.

Observation in connection with weapons tests

Observations in connection with weapons tests have the advantages that (1) very large quantities of radioisotopes are introduced into the sea, sometimes over a rather large area, so that radioactivity is sufficiently high to be detected in the sea waters and organisms over a considerable time after the event, and (2) the difficulty of being certain that the organisms have actually remained in the water containing the isotopes is minimized. On the other hand, the determination of exact amounts of isotopes introduced, of their spatial distribution, and of their physical state presents some difficulty.

Biological studies, in connection with the various weapons tests in the Western Pacific ocean, have been primarily directed toward determining the concentration of gross activity in different organisms, the localization of such activity in different parts of the organism, and the rates of decline of activity with time. There has also been limited determination of the isotopes concerned. The most extensive data are from the lagoons of the atolls at and near the test sites. In the open sea, outside the lagoons, usually only limited collections of organisms have been made, incidental to other operations.

Following the test series of 1954, however, two rather extensive surveys were made of the distribution of activity in the sea, and in organ-

[1] Contribution from the Scripps Institution of Oceanography, New Series, No. 903a.

isms at different trophic levels, over a large sea area at intervals of approximately 4 months and 13 months after the test.

These observations have been directed primarily to possible human hazards through contamination of edible marine products. Only minor attention has been given to ecological processes, probably because of lack of facilities for the extensive, systematic collecting required.

Soon after the underwater test in the Eastern Pacific in the spring of 1955, some collections were made that indicate which organisms in the food chain are the primary concentrators of certain radioisotopes, and that give some indication of the time scale in passage to the next step of the food chain. Unfortunately, it was not possible to follow the passage of isotopes farther through the system.

Following a weapons test a series of observations and collections taken in a carefully planned pattern in space and time could provide information on the time scale involved in the passage of material through the system of prey and predators, and on the efficiency of this transfer from one stage to another, two of the little understood basic problems in marine ecology. Data from experiments with radioactive tracers, together with more limited field data, indicate that the transfer efficiencies are different for different elements.

In those situations, following weapons tests, where there is a fairly extensive body of water containing radioisotopes at some particular level, say at the surface, it should be possible by means of collections at various depths over a period of time to obtain worthwhile information on the vertical migrations of organisms, and also to determine how the feeding and excretion patterns of such organisms transport radioisotopes from one level to another.

These and similar studies would require the assignment of a vessel, with necessary equipment and a team of scientists, to the exclusive pursuit of such studies. Since results will depend on systematic, serial observations, the vessel must be available to take them when and where required, which precludes the commitment of the vessel to other activities. Although a sizable cost is involved, it is believed that the results to be obtained are of sufficient value to more than justify it.

It should also be pointed out that effective planning of such studies requires considerable knowledge of the types of organisms to be encountered in the test area, the sizes of their populations, and some knowledge of their migration patterns, as well as data on the currents and other physical parameters to be considered. A pre-survey of the test areas by standard methods of biological investigation is, therefore, an important element in the adequate planning and execution of post-test investigations by means of the radioisotopes produced by the test.

Observations in connection with waste disposal

The disposal of wastes from the fission industry by introduction into the marine environment offers another means of studying the uptake of elements by aquatic organisms, their fluxes in the ecosystem, and their effects on the organisms concerned. Advantages over weapons tests are: (1) the wastes are usually introduced in such a manner that their amount, distribution and physical state can be readily determined, (2) disposal is usually continuous, even though not of constant magnitude, thus permitting systematic study over considerable periods of time.

Disposal in the United States has consisted of relatively low-level wastes introduced into fresh waters by the Hanford works on the Columbia River, the Oak Ridge National Laboratory, and the Plant on the Savannah River. At the first named locality, field observations, supplemented by laboratory experiments, are being made on the uptake of radioisotopes by organisms, their fluxes through the food chain, and their distribution in the river as the result of the combined effects of physical and biological processes. The phosphorous cycle has been investigated in particular detail. At the Oak Ridge Laboratory, observations were made over a period of years on the uptake of fission products by various organisms, the sites of deposition of radioisotopes in the organisms and the effects on some of their populations. Continuous disposal into marine waters is not practiced at present in this country. Reports by H. Seligman, H. J. Dunster, D. R. R. Fair and A. J. McLean at the 1955 Geneva Conference on Peaceful Uses of Atomic Energy describe introduction of low-level wastes into the Irish Sea, and briefly review studies of the uptake of various isotopes by different kinds of organisms.

With the exception of limited work at Hanford and Oak Ridge, it appears in all these cases that primary attention has been concentrated on monitoring aspects, that is measurement of the quantity and distribution of radioisotopes to insure against hazards to human or other animal populations. The work of Richard Foster and others on the radiophosphorus cycle in the Columbia River, and the work of Louis A. Krumholz on seasonal variations in quantities of fission products in different groups of organisms, indicate however, that locations where wastes are being continuously introduced into aquatic environments offer a good opportunity to study the ecological processes of the aquatic populations through the tracers provided by the introduced isotopes. It may be expected with the development of the fission industry in the next few years, that there will be disposal of some low-level wastes into marine waters, which will provide opportunities to investigate the ecology of estuaries and inshore ocean waters by these means.

These introductions also constitute large-scale experiments on both the direct and genetic effects of long-term exposure of marine organisms to atomic radiations. It is important that these effects be carefully investigated, because it is possible that the larger organisms in the sea, which are subjected to much lower rates of natural radiation than terrestrial forms (due to the shielding effects of water on cosmic rays, as well as to the low gamma-ray activity per unit volume of sea water compared with the rock and soil of the land), may show proportionally a greater genetic effect from a given amount of radiation.

Planned experiments

Much useful information may be obtained by well conceived biological observations in connection with weapons tests and routine disposal of industrial atomic wastes. Much more precise information could be obtained, however, by planned experiments introducing measured quantities of known isotopes into the marine environment in a controlled manner. Furthermore, it is evident that the fluxes of different elements through the ecosystem vary according to their abundance in the sea and their physiological roles in the organisms. Some of the most important elements biologically are not fission products, nor are they present in wastes in appreciable quantity. The outstanding example is carbon. The energy which supports most of the life in the sea, as on the land, is fixed as chemical energy of complex carbon compounds synthesized by plants. To study the flux of energy through the different trophic levels of the ecosystem it is necessary, therefore, to measure directly or indirectly the flux of carbon. One of the most promising possibilities, discussed further below, is the use of radiocarbon in tracer experiments on a scale larger than the present laboratory-type experiments.

The need for large scale experiments under natural conditions arises because we require knowledge concerning the quantitative interrelationships of the various populations of organisms, and it is not possible to reproduce natural marine communities, especially the pelagic elements, in the laboratory. It is probably not possible yet to study some aspects of open-sea communities by radioactive tracers, either, but it may be possible to improve on present techniques by larger scale *in situ* experiments than have been attempted.

Large scale experiments, employing either mixed fission products or single isotopes isolated from mixed fission products, appear feasible (at least in selected locations in the open sea) to determine what organisms take up which elements and the quantitative aspects of how these elements are passed through the food chain. It may also be feasible to introduce sufficient quantities of radioisotopes in particular situations to make possible a study of the transport of such elements by migrations of organisms. In general, however, in the open sea, it will be necessary to confine attention to those elements which are naturally present in seawater in very small concentrations, so that the organisms may be expected to take up a relatively large fraction of the isotope in question. In the case of elements such as carbon, only a small fraction of which is taken up by the organisms, experiments in unconfined volumes of open sea would appear to require larger quantities of the radioisotope than are feasible on a cost basis, and experiments therefore will have to be limited, in the near future at least, to small enclosed arms of the sea or artificially bounded volumes of water in the open sea.

In order to conduct experiments in the open sea it is necessary to (1) introduce the radioisotopes into an area sufficiently large so that it can be located and followed, to insure the or-

ganisms under study being in it over a known period of time, and (2) have a sufficiently high radioactivity that it may be followed from shipboard. If we use only fission products which organisms concentrate; then, since longer counting periods are feasible for samples of the organisms than are feasible for the equipment used to locate and follow the water mass, the radioactivity required to determine the position of the contaminated water mass is expected to be the limiting factor in the experiment.

Revelle, Folsom, Goldberg and Isaacs (1955) have indicated that, in the slow-mixing levels of the sea below the thermocline, vertical mixing is almost negligible, so it may be expected that while the area in which the isotopes can be detected spreads over a radius of 4.1 km., vertically it will be limited to about 1 meter. In these circumstances, it has been calculated that 10 curies of gamma emitter may be detected until it has spread laterally to a radius of 4 km., or a mean concentration of about 2×10^{-7} curies per cubic meter. They do not specify the time involved, but it may be presumed to be of the order of one week to one month. For biological experiments, it would be necessary to make observations over a longer period of time, also we cannot commence significant biological observations until the contaminated area is sufficiently large to ensure knowledge of which animals are or have been in the active water. For these reasons the time involved should perhaps be increased by a factor of 10. If the diffusion of the contaminated water, both vertically and horizontally follows the "random walk" law, the volume containing the activity will increase linearly with time, and, in consequence, about 100 curies of gamma activity will be required.

Experiments in the upper mixed layer will require much larger quantities of fission products. Mixing to the top of the thermocline is very rapid; according to the authors above cited the lower boundary of radioactive water moves down at about 10^{-1} cm/second. If we select an area, such as that off Central America where there is a fairly shallow sharp thermocline at a mean depth of about 20 meters, mixing down to the top of the thermocline would be complete in less than ten hours. Thereafter downward mixing should be negligible. Recent experiments suggest that the radius over which the water spreads laterally is increased as about the 0.8 power of time. In Bikini lagoon it has been found that the radius of the radioactive area increased to 4 kilometers in 3 days. If we ran an experiment for 90 days, which is probably the time necessary to follow the flux of radioelements through two or more trophic levels, we would, then, expect the radius to approximate

$$r = 4(30)^{.8} = 60 \text{ kilometers.}$$

The volume would then be (with a 20 meter thermocline)

$$\pi \times 36 \times 10^8 \times 20 \text{ cubic meters}$$
$$\text{or about } 225 \times 10^9 \text{ cubic meters}$$

To be still detectible at this dilution, using the above estimate of 2×10^{-7} curies/cubic meter, an initial quantity of some 4×10^4 curies would be required. The logistics of handling large quantities of fission products will be difficult, but not perhaps impossible.

Because of the smaller volume of water to be dealt with, it may be most desirable, at least initially, to conduct such experiments in a small enclosed arm of the sea. Such an environment is different in many respects from the open ocean, but much useful information about fluxes of radioelements through the several trophic levels could be obtained. It would not be difficult to select a small bay, with a narrow, shallow entrance, which could be cut off temporarily from the sea for this purpose. A body of, say, one square kilometer with an average depth of ten meters might be used, giving a volume of 10^7 cubic meters. Since the problem of locating the water mass is eliminated, and fairly large volumes of water can be filtered for organisms, rather small quantities of fission products, which would not be hazardous, could be employed. One curie would be ample, and the contamination of the water itself would be within safe levels for human hazards.

It was noted earlier that one of the important fundamental ecological problems is to measure the flux of carbon through different trophic levels. Since the fraction of the carbon taken up by plants is a very small part of the total in the sea water, experiments with radio-carbon in the open sea are not feasible. Experiments using samples in bottles have been conducted *in situ* in recent years, but these have two deficiencies: (1) the surface and other effects of the container modify the environment so that the resulting computations for photosynthesis probably are not those that would have occurred

naturally in the sea and (2) only the uptake of carbon at the phytoplankton level is measured. It seems feasible to improve on the experiments in bottles by conducting experiments in small lagoons, or by employing larger partly-enclosed volumes in the open sea.

From experience with such experiments in bottles, it can be shown that there is sufficient uptake of carbon by the phytoplankton, if grown in a concentration of 0.3 micro-curie per liter for one day, to measure it if a one liter sample is filtered and the radioactivity of the filtered plants determined in a counter of 20 per cent efficiency. By increasing either the counting time or the volume of water filtered, the initial concentration of C^{14} can be decreased correspondingly.

For an experiment in a lagoon, we might use a body of water of, say, 500 meters long by 200 meters wide with an average depth of 10 meters, giving a volume of 1×10^6 cubic meters or 1×10^9 liters. By filtering 100 liters of water for phytoplankton, C^{14} at a concentration in the water of 3×10^{-9} curies per liter would suffice, or 3 curies for the experiment. Since there is probably between a 50 per cent and 90 per cent loss at each step up the food chain, correspondingly larger volumes would have to be strained for the higher forms, but this is a simple problem by the use of standard nets, etc.

To get improved measurements of the uptake of carbon by phytoplankton in the open sea, and the passage of carbon to the smaller grazing organisms, it is suggested that a moderately large rubber tank open at the surface be employed to isolate a piece of the top of the sea, yet have a sufficiently small surface-to-volume ratio that the processes will more nearly approach normal conditions than is obtained in bottle experiments. We might employ such an apparatus of 20 meters diameter by 10 meters deep, having a volume of $\pi\, 10^3$ cubic meters, or $\pi \times 10^6$ liters. By filtering 10 liter samples for phytoplankton, with 20 per cent efficient counting equipment, we would need to provide about 3×10^{-8} curies per liter, or a total of about 1/10 curie of C^{14}.

Some cost and logistic considerations

For the two experiments with C^{14}, discussed immediately above, the problems of handling the amounts of activity involved present no particular difficulty. Since C^{14} is a pure beta emitter, the shielding problem for even the experiment requiring 3 curies is a simple matter. The cost of the isotope, however is fairly high; at present about $30,000 per curie. This might be reduced somewhat if the present demand were to increase. The cost, notwithstanding, however, the information to be gained is well worth the outlay.

In the case of an experiment using gamma emitters in the slow-mixing layer below the thermocline, where about 100 curies would be required, it is suggested that mixed fission products from wastes from processing of reactor fuel elements be used. A large quantity of such wastes will be available, probably at no charge. If one used HNO_3 salted waste product from a natural uranium-plutonium reactor, after 100 days "cooling," the reactor waste will contain about 200 curies/gallon. Approximately half a gallon will be needed, requiring about 10″ of lead shielding for transportation and handling. A cubical container will require 10.05 cubic feet of lead, weighing 7,175 pounds. This is feasible to handle by freight and on shipboard.

For the kilocurie quantities required for an experiment in the upper mixed layer of the sea, the handling problem reaches a different order of magnitude. It becomes quite infeasible to handle waste liquids in the volume required. It may be possible, because of the much higher activity per unit volume to employ slugs of U^{235} from a reactor, which, after 30 per cent burning and 100 days "cooling" have about 2×10^5 curies per kilo of fairly long term gamma activity. Even then some 2/10 kilos of "used" U^{235} would be required. The problems of transporting and handling this are somewhat difficult as are methods of dissolving and liberating the material at sea, but probably feasible. Further detailed consideration needs to be given to this problem. It may, of course, be that the use of an explosive reaction — a small nuclear detonation for oceanographic and biological experimental purposes — is the only logistically feasible method.

REFERENCES

REVELLE, R., T. R. FOLSOM, E. D. GOLDBERG, and J. D. ISAACS. 1955. Nuclear Science and Oceanography. United Nations International Conf. on Peaceful Uses of Atomic Energy, Geneva, Paper no. **277**:22 pp.